FINAL REPORT

Savannah River Site Environmental Dose Reconstruction Project

Phase II: Source Term Calculation and Ingestion Pathway Data Retrieval

Evaluation of Materials Released from the Savannah River Site

April 30, 2001

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Submitted to the Centers for Disease Control and Prevention in partial fulfillment of contract No. 200-95-0904

"Setting the standard in environmental health"

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No tables.

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No tables.

Glossary and Acronyms

SRS GLOSSARY AND ACRONYMS

Accuracy – the ability of an analytical method to measure the true concentration of a contaminant (*see bias, precision*).

Actinides – radioactive elements with atomic numbers equal to or greater than that of actinium (i.e., 88). The term refers to the heaviest elements, starting with actinium and continuing to the end of the periodic table. *Transuranic* elements are a subset of the actinide elements and include those with atomic numbers larger than uranium. Actinide elements are all radioactive.

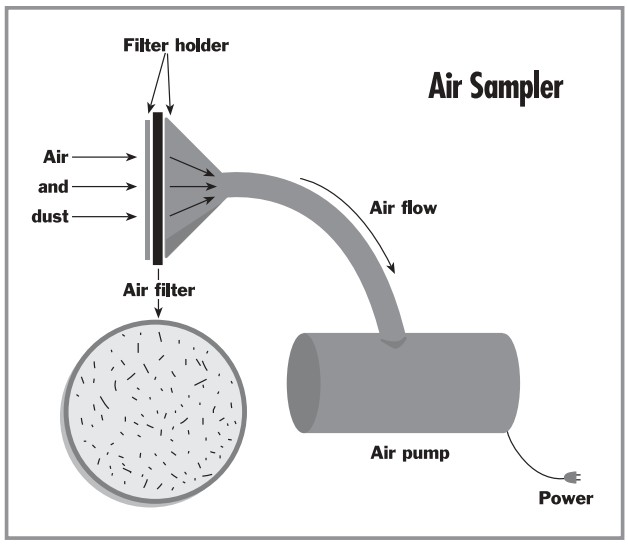
Activation products – radionuclides that result from the absorption of neutrons by uranium, and other materials present in a nuclear reactor. An example is plutonium-239 produced following neutron absorption by uranium-238 and subsequent decays of uranium-239 to neptunium-239 and then to plutonium-239.

Activity – the mean number of decays per unit time of a radioactive nuclide expressed as *disintegrations* per second. Units: becquerel (Bq), formerly curie (Ci).

Aerodynamic diameter, AD – the physical diameter of a particle of unit density (1 gram per cubic centimeter) that has the same gravitational settling velocity as the particle of interest.

Aerosol – a suspension of solid and/or liquid *particles* in a gas (like air).

Air filter – a solid matrix used in an air sampler to collect *particulates* from the air, which is drawn by an air pump through the filter. Air filters are least efficient for particle sizes of about 0.3 *microns* and collect smaller and larger particles more efficiently.



A-line – the facility in the F-Area where uranyl nitrate was converted to uranium oxide**.**

Aliquot – a fraction of a substance taken for sampling purposes.

Alpha particle (*ionizing radiation*) – two neutrons and two protons bound as a single particle (a helium nucleus) that is emitted from the nucleus of certain radioactive isotopes in the process of disintegration. It carries much more energy than *gamma* or *beta* radiation, and deposits that energy very quickly while passing through matter. When alpha particles are being measured in an environmental sample, they can be absorbed by the sample itself unless the sample has been prepared to be very thin. Corrections can be applied for this self-absorption of alpha particles in some samples (e.g. air filters). Plutonium-239 emits alpha particles.

Ambient air monitoring – *monitoring* of the air outside of buildings *(see effluent monitoring)*.

Analytical method – a laboratory test used to detect the amount of a *contaminant*.

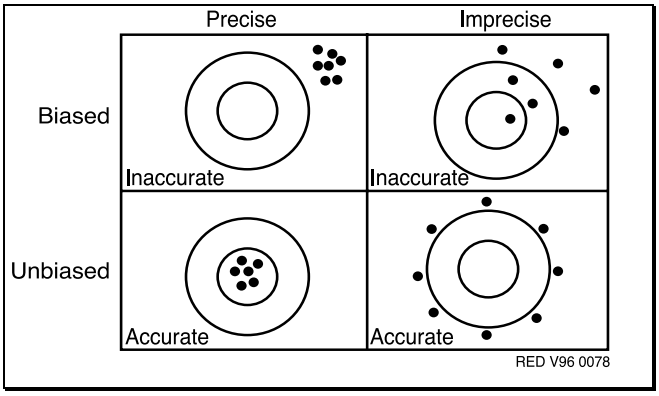
Anisokinetic sampling – a sampling condition that involves a mismatch between the air or \fluid velocity in the sampling probe and that in the stack releasing airborne effluents. It is a source of bias in effluent sampling. In contrast, isokinetic sampling, in which the two velocities are equal, results in an unbiased sample of the stack effluent.

Atomic number – the number of protons in the nucleus of an atom.

Background radioactivity – radioactive elements in the natural environment including those in the crust of the earth (like radioactive potassium, uranium, and thorium isotopes) and those produced by cosmic rays. The term background is also sometimes used in this report to indicate radioactive elements present in the environment that are not a direct result of SRS activities (e.g. atmospheric weapons testing fallout, *see definition for fallout*). The term background can also refer to chemicals, such as heavy metals.

Beta particle (*ionizing radiation*) – a charged particle emitted from the nucleus of certain unstable atomic nuclei (radioactive isotopes), having the charge and mass of an electron. Energetic beta particles penetrate the dead skin layer. The beta particle is not stopped in matter as quickly as an alpha particle.

Bias – a systematic distortion of measurements that makes the results inaccurate. Accuracy is a measure of how close a value is to the true number, or a measure of the correctness of a measurement. Precision refers to the ability of an analytical method to reproduce the same result upon repeated trials.



Biota – living organisms.

B-line – the facility in which plutonium nitrate solution was purified and converted into plutonium metal.

Blanks – samples containing extremely low concentrations of a *contaminant*, which are used to assess *contamination* from laboratory equipment and other steps in an analytical procedure.

Bq – an abbreviation for the SI unit of radioactivity, Becquerel. Equal to one *disintegration* per second. (See *curie.).*

Burial grounds – radioactive waste disposal areas located between the two separations areas, 200-F and 200-H. One disposal area was 76 acres used from 1953 until 1972; the other, 119-acre site, was used from 1972 until the 1990s.

Canyon – the long, narrow, and deep, thick-walled concrete structure in which fissionable materials that had been irradiated in the reactors were chemically separated from fission products and from each other. The hot canyon was the more heavily shielded canyon in which the majority of the fission products were removed and the desired products were separated. The less heavily shielded canyon was referred to as the warm canyon.

cfm – cubic feet per minute, a measure of the rate of flow of a liquid or gas.

Chemical symbols – abbreviations for different elements and compounds. Examples of symbols for elements include U for uranium, Pu for plutonium, O for oxygen, C for carbon and Cl for chlorine. Examples of symbols for compounds include CCl_4 for carbon tetrachloride and PuO_2 for plutonium dioxide.

CIIS (Chemical Information and Inventory System) Database – a database originally developed at SRS to comply with Community/Worker-Right-to-Know legislation and to help organize annual inventory data for EPA reporting requirements. The database inventory contains over 51,000 entries.

CMX – the code letters designating a facility that developed and tested various reactor components**.**

Collection efficiency – the percentage of the total amount of a *contaminant* present in ambient air, which is collected by an air sampler. Collection efficiency is strongly dependent upon the size of the particles that carry the contamination, as well as other factors, such as wind speed.

Compositing – a combining of samples before analysis, usually done to improve sensitivity and/or reduce analytical cost. A quarterly composite of air filters is comprised of all filters collected within a calendar quarter (3 months).

Concentration – the amount of a material of interest in a given volume or mass.

Contamination – unwanted radioactive or other material or the deposition of radioactive material in the environment or other place.

Control rod – a long, slender, cylindrical rod containing a strongly neutron-absorbing material such as lithium or cadmium. The control rods constituted the primary system in SRS reactors for controlling the number of neutrons (flux) and the rate of fissioning power (power level).

Coolant – the fluid that cools the reactor. The primary coolant passes directly through and around the fissioning fuel in a reactor and carries away heat to prevent melting. The secondary coolant carries the heat away from the reactor and releases it to the environment. In the SRS reactors, the secondary coolant was water from the Savannah River or PAR Pond. The heat passes from the primary coolant to the secondary in heat exchangers.

Cooling (radioactive) – the reduction of radioactivity and heat generation of irradiated fuel or target material by radioactive decay.

Counting error – the uncertainty in the measurement of an amount of radioactivity due to the random nature of radioactive decay and electronic noise in the detector (instrument background). Counting error decreases when the sample and background are counted for a long time and when instrument background is minimized.

Critical mass – the minimum mass of fissionable material which can achieve a nuclear chain reaction with a specified geometrical arrangement and material composition.

Curie (Ci) – a traditional unit used to describe an amount of radioactivity. The curie is equal to 3.7×10^{10} *disintegrations* per second (dps). The internationally recognized unit of radioactivity is the Becquerel (Bq), which is one dps. Due to its historical context, this report favors use of traditional units for radioactivity. For environmental samples, the microcurie (10^{-6} Ci), the picocurie (10^{-12} Ci), or the femtocurie (10^{-15} Ci) are often used.

Dana Plant – a facility in Dana, Indiana which produced heavy water by the *GS process*.

Degraded water – heavy water that became diluted with light or natural water.

Degreasers – large vats located in the M Area that contained heated and cooled solvents that were used to remove grease from reactor components.

Detection level – the lowest amount of a *contaminant* which can be detected with a certain degree of confidence by an analytical method. For radioactivity measurements, detection level is the same as minimum detectable activity *(See also minimum detectable concentration, MDC, and lower limit of detection, LLD*).

Deuterium – synonym for ^{2}H, an isotope of hydrogen of double mass (atomic mass = 2).

Disintegration – one decay of a radioactive atom. *(See dpm).*

Deuterium oxide (D_2O) – heavy water.

DOE – U.S. Department of Energy. The DOE is responsible for the sites in the U.S. at which weapons materials have been produced or handled, including the Savannah River Plant. Generally, private *contractor*s, such as Du Pont have operated the weapons facilities for the DOE. *(See ERDA, AEC.)*

Domain - the area considered within the study**.** For this dose reconstruction the domain extends 50 miles in all directions from the SRS boundary; the domain also includes Columbia, SC and an area on either side of the Savannah River to the coast.

Dose – a general term denoting the quantity of *radiation* or energy that is absorbed by the body. There are technical terms with specific definitions, such as absorbed dose, equivalent dose, and effective dose.

Dose reconstruction –a study process in which historical information is used to estimate the amounts of toxic materials released from a facility, how the materials could have moved offsite, and the exposure of the public to those materials. Dose reconstruction involves past releases, not present, or future releases. The study period for this Savannah River Site dose reconstruction is 1951–1992.

dpm – abbreviation for *disintegrations* per minute, a rate of radioactive decay. There are 2.22 dpm per picocurie. (*See curie.*)

DW Process – a separation process which involved the separation of light and heavy water by fractional distillation. The DW (distillation) process uses the difference in volatility between hydrogen oxide (H_20) and deuterium oxide (D_20) in the water to separate them using five stages of distillation in bubble cap tray towers.

Du Pont – the Atomic Energy Commission appointed Du Pont to operate the site in 1950. They ran the site until March 30, 1989 when Westinghouse Savannah River Company took over the responsibility.

Effluent – a gas or liquid that flows from a process, building, or site into the surrounding environment.

Effluent monitoring – the measurement of a *contaminant* or other property (e.g. flow rate) in the effluent (air or liquid discharged) from a building or holding pond.

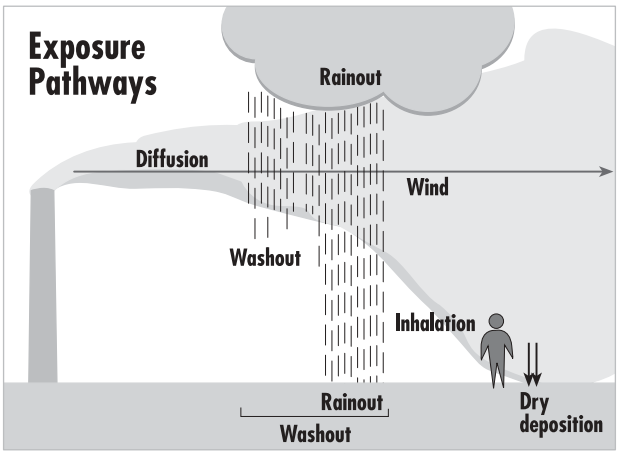
EML – Environmental Measurements Laboratory (*see HASL*).

Environmental monitoring – the measurement of a material in the environment at regular time intervals. Monitoring for *contaminants* often involves the collection of an environmental sample, (like stream water), preparation of the sample in the laboratory, and analysis of the prepared sample using an analytical instrument.

Environmental transport – the mechanisms by which substances can be carried from their source to other points in the environment. Surface water runoff and air dispersion by wind are examples of environmental transport mechanisms.

Exposure (to hazardous substances) – conditions or circumstances causing humans or other living things to come into contact with toxic materials.

Exposure pathways – the means by which humans are exposed to toxic substances. The key exposure pathways are air and water, with most exposures via inhalation, drinking water, crops, other foods, and direct radiation.



Fallout – airborne particles containing radioactive material that fall through the atmosphere and are deposited on the earth's surface following the detonation of nuclear explosives.

femto – a prefix that multiplies a basic unit by 1/1,000,000,000,000,000 or 1 x 10^{-15}. For example, 1 femtocurie equals 1 x 10^{-15} curie.

Fission products – radionuclides that result from the splitting or fissioning of heavy elements like uranium in a nuclear reactor. Examples are cesium-137, strontium-90, technetium, 99, and ruthenium-106.

Fuel assembly – a group of uranium pieces, either short cylinders (slugs) of uranium metal or long tubes of enriched uranium-aluminum alloy. The long tubes were frequently of different diameters, nested together, sometimes with lithium-aluminum alloy, or other, targets.

Fuel elements – aluminum clad uranium rods used in fuel assemblies in SRS reactors.

Fuel element failure – rupture of a fuel element, leading to the release of radionuclides to the cooling or storage water.

Gamma radiation (also gamma rays or *ionizing radiation*) – short wavelength electromagnetic radiation (photon) originating from the nucleus of a radionuclide. Gamma rays are similar to medical x-rays, but are emitted at very specific energies characteristic of their decaying atoms usually of higher energy than about 100 keV. They penetrate tissue more effectively than beta or alpha particles, leaving ions in their path to potentially cause cell damage. Gamma rays travel relatively long distances in air, and leave a low density of ionization damage in their track through tissue. Gamma-emitting radionuclides are hazards from outside the body because their radiation penetrates to living tissue, but they are of less concern than alpha-emitters when ingested or inhaled, because their ionizing energy is deposited less effectively in tissue.

Geometric Mean (GM) – a measure of the central point of a *skewed* distribution. The geometric mean of a set of positive numbers is the exponential of the arithmetic mean of their logarithms. It is typically used to describe *skewed* distributions (e.g. lognormal distributions).

Geometric Standard Deviation (GSD) – a measure of the spread of a *skewed* distribution. A large GSD indicates a wide range of measured or calculated values. The geometric standard deviation of a lognormal distribution is the exponential of the standard deviation of the associated normal distribution.

Grab samples – samples, usually of relatively small volume, which are taken at random or at pre-selected frequencies. These samples define the concentration of a *contaminant* at the specific time when they are collected and differ from continuous or proportional samples that reflect a time-averaged concentration.

Gross alpha – measurements that refer to the total (or gross) amount of *alpha particles* in an environmental sample. Besides materials released from the Savannah River Plant, like *plutonium* and *americium*, naturally occurring substances in the environment, like *uranium* and *thorium*, emit alpha particles and would contribute to the gross alpha measurement.

Gross beta – measurements that refer to the total (or gross) amount of *beta particles* in an environmental sample.

GS process – the method of extracting heavy water containing deuterium from naturally occurring water by the exchange of deuterium between water and hydrogen sulfide (H_2S) at different temperatures. The deuterium will migrate toward a water stream at lower temperatures and to H_2S at higher temperatures.

Half-life, radioactive – the time required for half the atoms of a radioactive substance to disintegrate. During one half-life, the number of radioactive atoms in a material is reduced by one-half. Each radionuclide has a unique half-life. Tritium decays with a half-life of 12.3 years, and plutonium-239 decays with a half-life of about 24,000 years. The term half-life can also be used to describe the time required for the amount of a radionuclide or chemical in a biological compartment (e.g., vegetation, sediment, water, human or other animal tissue) to be reduced by one-half.

Harp – the name of a container used to store a failed fuel element; the container was stored underwater in a reactor basin and vented to the reactor stack.

HASL – the Health and Safety Laboratory, in New York City, operated by the Department of Energy and its predecessors, is known for long-term global monitoring of radionuclides in the environment and for development of analytical techniques for measuring radioactivity in environmental *media*. Later became known as the Environmental Measurements Laboratory (EML).

Health physics – an interdisciplinary science focused on the radiation protection of humans and the environment. Health physics combines the elements of physics, biology, chemistry, statistics, and electronic instrumentation to protect individuals from the effects of radiation.

Heavy water – water in which nearly all of the hydrogen is the heavy isotope, deuterium; deuterium oxide (D_2O).

Heavy Water Plant – a facility in the D Area at SRS that began producing heavy water (deuterium oxide) in 1953 to moderate and cool the site's reactors. The facility stopped production in 1981 because there was a sufficient supply of heavy water.

HEPA filter – a high-efficiency particulate air filter used to remove contaminants from exhaust gases prior to discharge.

HM process – an acronym for "H-Modified," the HM process was a modified *Purex process* , used in the H Canyon to separate uranium from plutonium, neptunium, and other fission products. The Purex process was used in H Canyon prior to the May 1959 startup of the HM process.

Ingestion – radionuclides or chemicals taken into the body by eating or drinking are taken in by ingestion.

Inhalation – radionuclides or chemicals taken into the body by breathing are inhaled.

Inventory – the total amount of a *contaminant* in a defined space, e.g. the amount of *plutonium* in the *sediment* of a reservoir.

Ion exchange – a process for selective removal of a chemical constituent from a particular solution.

Ionizing radiation – radiation sufficiently energetic to dislodge electrons from an atom and thus leave the atom positively charged or "ionized." Ionizing radiation includes x and gamma radiation, electrons (beta radiation), alpha particles (helium nuclei), and heavier charged atomic nuclei. Neutrons ionize indirectly by colliding with atomic nuclei. The creation of ions (ionized atoms, which are chemically active) inside living cells can damage key substances in cells, including the DNA containing the record of the cell's characteristics. Such damage can lead to cancer or other defects.

Isotopes – different forms of elements having the same atomic number (number of protons) but different numbers of neutrons. Different isotopes of a particular element generally have essentially identical chemical properties. Plutonium-239 and plutonium-240 are isotopes of plutonium that can not be distinguished from one another by typical *analytical methods*.

kilo – a prefix that multiplies a basic unit by 1000. For example, 1 kilogram = 1000 grams.

LLD – lower limit of detection. *(See detection level).*

Liter (L) – A metric unit of volume, equivalent to about 1.1 quarts.

Long-lived radionuclides – in this study, radionuclides with *half-lives* greater than 15 days.

MDA –minimum detectable activity. *(See MDC).*

MDC – minimum detectable concentration. *(See definition below).*

MDL – minimum detection limit. *(See detection level).*

Media – a type of environmental sample, such as air, soil, vegetation or water.

Median – the central point of a distribution. Half of the values are larger than the median value and half are smaller. *(See percentiles.)*

Micron (μm) – a micrometer or micron is a unit of length equal to one-millionth (10^{-6}) of a meter. A human hair, for reference, is about 100 microns thick.

Minimum detectable concentration (or activity) – the lowest concentration of a contaminant (or amount of radioactivity) that can be detected with a certain degree of confidence by an analytical method *(see detection level*).

Moderator – a material used in a reactor to slow down neutrons; when neutrons collide with nuclei, they lose speed, making them more likely to be captured by fuel or target materials.

Monitoring – obtaining measurements at regular time intervals.

Monte Carlo procedure – a method that uses computer-generated pseudo-random numbers to make calculations with statistical distributions. In this study, Monte Carlo methods have been used to estimate statistical distributions that represent uncertainties in estimated quantities, such as source term release estimates. This approach contrasts with a deterministic approach in which a calculation is based upon point estimates of the various parameters and yields a single result. The Monte Carlo calculation carries the underlying uncertainty in the parameters forward and displays it in the magnitude of the distribution of results. A statistical risk management computer program, called Crystal Ball™ (Decisioneering 1993) was used in this study for some of the uncertainty analyses.

Naturally occurring radionuclides – radionuclides that are naturally present in the environment and are two general types: primordial and cosmogenic. Most primordial radionuclides are isotopes of the heavy elements of the three radioactive series headed by uranium-238, thorium-232, and uranium-235. Cosmogenic radionuclides are produced by interactions in the atmosphere or in the earth; three of these [tritium (hydrogen-3), carbon-14, and sodium-22] are isotopes of major elements in the body.

Neutron – an uncharged subatomic particle capable of producing ionization in matter by collision with charged particles. Approximately the same mass as a proton it is a constituent of the nuclei of all atoms except hydrogen. The nucleus of deuterium (heavy hydrogen) has one proton and one neutron.

Noble gases – the name given to the following group of elements: He, Ne, Ar, Kr, Xe, and Rn, which all have closed-shell electronic structures that are completely stable.

Nonvolatile beta activity – activity resulting from the presence of radionuclides that do not escape during sample preparation, such as evaporation or wet-ashing (*see beta particle*)

Nuclear materials - materials used to produce a nuclear reaction such as uranium and plutonium.

Nuclide – a species of atom having a specific mass, atomic number, and nuclear energy state.

Outcrop – a place where groundwater is discharged to the surface. At SRS, groundwater outcrops in several places to enter site streams. Also referred to as seepline.

Percentiles – a method for making descriptive statements about a large data set. Percentiles are defined in such a way that a large set of data, arranged from its smallest to its largest value, is divided by its percentiles into 100 classes containing nearly equal numbers of data. The exact rules for defining the percentile numbers are complicated, but the effect is that approximately 5% of the data are less than or equal to the 5th percentile, and approximately 95% of the data are greater than or equal to the 5th percentile (similar statements hold for the other percentiles). The median is defined as the 50th percentile, which divides the data (approximately) into halves (if there are an odd number of data, the middle value is the median; if there are an even number, the

average of the two middle values is the median). In this document, uncertainty distributions are indicated by their 5th, 50th, and 95th percentiles. Observations above the 95th percentile have only a 5% probability of occurrence, as do observations below the 5th percentile. The 50th percentile is presented as the best estimate.

pico – a prefix that multiplies a basic unit by 1/1,000,000,000,000 or 1 x 10^{-12}. For example, 1 picocurie equals 1 x 10^{-12} curie, or one-trillionth of a *curie*.

Plume – the *concentration* profile of an airborne or waterborne release of material as it spreads from its source. A plume from a coal-fired power plant, for example, may be visible for some distance from its stack, with the concentration of its components decreasing with distance from the stack and from the centerline of the plume. After the plume becomes invisible because of dilution, it continues to be diluted with increasing time and distance. Atmospheric dispersion *model*s of this process predict concentrations within a plume far downwind and far beyond the point at which a plume becomes invisible. Similar modeling for releases from nuclear facilities can estimate the impacts of releases long past by reconstructing *exposure* and *dose* estimates.

Plutonium (Pu) – silvery, white radioactive metal (atomic number 94) used in casting, rolling and forming, and machining and final assembly of nuclear weapons components. Its most important *isotope* is plutonium-239, produced by neutron irradiation of uranium-238. Plutonium-239 decays by emitting *alpha particles* and has a 24,065-year half-life.

Precision – the ability of an *analytical method* to reproduce the same result upon repeated trials. (*See bias.*)

Purex process – a process to separate uranium and plutonium from each other and from fission products by means of solvent extraction. The solvent used at the SRS was a solution of tributyl phosphate in "Ultrasene," a high-grade kerosene. The process was used in the F Canyon. The HM process replaced the Purex process in 1959.

Purging – releasing the water from the reactor basins to the seepage basins, allowing the tritium to evaporate.

QA/QC – quality assurance/quality control programs are established to assure accurate and reproducible results from environmental *monitoring*.

RAC – ***Radiological Assessments Corporation***, the contractor selected in October 1992 to conduct the Phase II Source Term Calculation and Ingestion Pathway Data Retrieval. *Radiological Assessments Corporation* changed its name to *Risk Assessment Corporation* in 1998.

Radiation – energy moving in the form of particles or waves. Familiar radiations are heat, light, radio waves, and microwaves. *Ionizing radiation* is a very high frequency form of electromagnetic radiation. It is invisible and cannot be sensed without the use of detecting equipment.

Radioactive contamination – *radioactive material* distributed over an area, equipment or an individual.

Radioactive decay – the disintegration of the nucleus of an unstable nuclide by the spontaneous emission of charged particles or photons of energy.

Radioactive material – material that contains unstable (radioactive) atoms that give off *radiation* as they decay.

Radioactivity – spontaneous transformation of an unstable atom, often resulting in the emission of *radiation*. This process is referred to as decay or *disintegration* of an atom.

Radiological – related to *radioactive materials* or *radiation*. The radiological sciences focus on the measurement and effects of radiation.

Radionuclide – a radioactive *isotope*, for example, plutonium-239 or tritium. Plutonium-239 emits *alpha particle* radiation when it decays; tritium emits low-energy beta particles. The isotope is an element that may make up part of another substance or chemical compound.

Reactor – the nuclear reactors in the 100 areas at the SRS.

Red-oil explosion – an explosion that can result from the presence of organic materials with nitric acid (NO_X) and high temperature; the rapid exothermic nitration of the organic material can lead to a "red-oil explosion."

Sand filters – at SRS, these underground filters were rectangular concrete structures with beds made of layers of coarse stone and succeeding layers of finer and finer gravel and sand for a total filter depth of about 8.5 feet. They were designed to filter the air exhausted from the canyon buildings.

SCRAM – an acronym for Safety Control Rod Ax Man. A SCRAM entails dropping the safety rods in a reactor to shut down the reaction.

Seepage basins – unlined excavated bowl-shaped areas for receiving liquid wastes from numerous facilities onsite. They were designed to allow infiltration of the liquid into the ground, thus decreasing the total volume of liquid released to onsite streams. The first seepage basins were put into operation in 1954.

Seepline – see outcrop.

Sensitivity – ability of an analytical method to detect small *concentration*s of a *contaminant*.

Separation areas – the F-Area and H-Area where fissionable materials that had been irradiated in the reactors were chemically separated from fission products and from each other.

Septafoil – clusters of seven rods in the reactors.

Slugs – irradiated uranium in the form of solid or hollow cylinders, encased in aluminum cladding.

Source term – the quantity, chemical and physical form, and the time history of *contaminant*s released to the environment from a facility.

Spatial trend (or spatial distribution) – a description of how a contaminant is distributed in the environment, for example with distance away from the facility. Two-dimensional trends in measurements are sometimes illustrated with *isopleths*.

Spiked samples – samples to which a known amount of the *contaminant* has been purposefully added to assess the accuracy of an *analytical method*.

Strike – a type of precipitation (head end) process in the separations area in which manganese nitrate and potassium permanganate were added to a treatment tank to form a manganese oxide precipitate.

Tank farm – series of interconnected underground tanks used at SRS for storage of high-level radioactive liquid wastes.

Time trend (or temporal trend) – a description of how the *concentration* of a *contaminant* changes over time at the same place.

Toxicity assessment – an evaluation of the types of health effects usually caused by specific substances, and the quantity (or *dose*) required to cause the effects.

Toxicologic review – an evaluation of the presence, use, and possible releases of toxic substances and the resulting potential for *exposure* or hazard to occur.

Transuranics – nuclides having an atomic number greater than uranium (i.e., greater than 92); all known transuranium elements are radioactive.

Tritium (T) – synonym for ^{3}H, a radioactive isotope of hydrogen of triple mass (atomic mass = 3).

Tritium reservoirs – small pressure vessels of various shapes that were filled with tritium gas under high pressure for use as components of a thermonuclear weapons. These are the only weapons components that were produced at the SRS.

Uncertainty – a general term used to describe the level of confidence in a given measurement or estimated quantity. Uncertainty depends on the amount and quality of the evidence (data) available. Uncertainties in the results of this study arise primarily from *bias* and imprecision in available measurements, absence of measurements at some times and places, lack of knowledge about some physical processes and operational procedures, and the approximate nature of mathematical models used to predict the transport of released materials.

Uranium (U) – a naturally occurring radioactive metal with atomic number 92, the heaviest natural element. Small amounts are present in soil, coal and rock materials, water, plants, and animals.

Validation – the process of comparing predicted *concentration*s of a material in the environment, based on *source term* reconstruction and environmental transport *models*, with historical measured concentrations to demonstrate that the models, within their domain of applicability, adequately represent the system they are intended to describe.

Water table – ground water that is held by a natural basin of clay overlain with sand or sandy clay and isolated from principal aquifers.

ACRONYMS

ACGIH – American Conference of Governmental Industrial Hygienists

AEC – Atomic Energy Commission

AIRS – Air Information Reporting System (database)

ATSDR– Agency for Toxic Substances and Disease Registry

BTU – British Thermal Unit

CAS – Chemical Abstract Service (number)

CCI – Company Chemical Inventory (Du Pont's)

CMP – Chemical, Metal and Pesticide (Pits)

CIIS – Chemical Information and Inventory System (Database)

CERCLA – Comprehensive Environmental Response, Compensation, and Liability Act

CMX – code letters designating a facility at SRS for the development and testing of various reactor components and auxiliaries.

cpm – counts per minute

DOE – Department of Energy

DP – Savannah River Laboratory research and development reports, originated by the Atomic Energy Division, Engineering Department, and or contractors

DPS – Savannah River Laboratory internal documents issued jointly by Technical Information Service (SRL) and Plant Records Division (SRP)

DPSOL – Du Pont Savannah River Plant Operating Log

DPSOP – Du Pont Savannah River Plant Operating Procedure

DPST – Savannah River Laboratory internal correspondence, memos, reports

DPSTL –Savannah River Laboratory Operating Log

DWPF – Defense Waste Processing Facility

EIS – Environmental Impact Statement

EPA – U.S. Environmental Protection Agency

EML – Environmental Measurements Laboratory (*see HASL*).

ERDA – Energy Research and Development Administration

FDA – Food and Drug Administration

FMF – (Naval) Fuel Manufacturing Facility

GIS – Geographic Information System

GM – Geometric Mean

GSD – Geometric Standard Deviation

GWQCB – Georgia Water Quality Control Board

HAW – high activity waste

HLW – high-level radioactive waste

HEPA – high-efficiency particulate air (filters)

HEAST – Health Effects Assessment Summary Tables

IARC – International Agency for Research on Cancer

ISCST – Industrial Source Complex Short Term (model)

K_{ow} – octanol-water partition coefficient

LAW – Low Activity Waste

LETF – Liquid Effluent Treatment Facility

LLD – Lower Limit of Detection

MCL – Maximum Contaminant Levels (Drinking Water Standards)

MSDS – Material (or Manufacturer) Safety Data Sheets

NAAQS – National Ambient Air Quality Standards

NCRP –National Council on Radiation Protection and Measurements

NESHAPs – National Emission Standards for Hazardous Air Pollutants

NPDES – National Pollution Discharge Elimination System

NIOSH – National Institute of Occupational Safety and Health

NTP – National Toxicology Program

ORNL – Oak Ridge National Laboratory

OSHA – Occupational Safety and Health Administration

PCBs – Polychlorinated biphenyls

PDWS – Primary Drinking Water Standards

PEL – permissible exposure limit

pCi – picocurie (*see curie, pico*)

Pu – plutonium

PVC – Polyvinyl chloride (pipe)

RBOF – Receiving Basin for Offsite Fuels

RCRA – Resource Conservation and Recovery Act

RfC – reference concentration

RfD– reference dose

RM – river mile

SARA – Superfund Amendments and Reauthorization Act of 1986

SCDHEC – South Carolina Department of Health and Environmental Control

SREL – Savannah River Ecology Laboratory

SRL – Savannah River Laboratory (named Savannah River Technology Center, SRTC in 1992)

SRP – Savannah River Plant (former designation for SRS and the production facilities)

SRS – Savannah River Site

SRTC – Savannah River Technology Center formerly called *Savannah River Laboratory*

SWDF – Solid Waste Disposal Facility

TBP – Tributyl phosphate or tri-n-butyl phosphate

TCDD – Trichlorodibenzodioxin

TLLa – total long-lived *alpha* activity

TLV – Threshold Limit Value

TNX – code name for the first pilot or semi-scale works facility at the SRS for the development and testing of equipment for the chemical separations processes

TRI – toxic release inventory

TRU – transuranic (refers to nuclides with atomic number greater than that of uranium

TSCA – Toxic Substances Control Act

TWA – time weighted average

U – uranium

UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation

USGS – U.S. Geological Survey

WSRC – Westinghouse Savannah River Company; took over the site operations from Du Pont in March 1989.

MEASUREMENT ACRONYMS

g – gm

kg–kilogram

mg– milligram (10^{-3} g)

ppb – parts per billion

ppm – parts per million

ppt – parts per thousand

µg–micrograms (10^{-6} g)

y–year

d–day

m–meter

L–liter

Gal–gallon

CHAPTER 1

EXECUTIVE SUMMARY

The Centers for Disease Control and Prevention (CDC) is studying the potential human health risks to people exposed to chemicals and radioactive releases to the offsite environment from the Savannah River Site (SRS). The SRS is a U. S. Department of Energy (DOE) complex occupying about 300 square miles along the Savannah River in South Carolina, on the Georgia border. The SRS has produced plutonium, tritium, and other materials for the national defense and some civilian purposes. The study is evaluating past releases leading to human exposure and dose, and is called a "Dose Reconstruction." Figure 1-1 shows important steps in the dose reconstruction process. The term dose is used regularly throughout this report and refers to the committed effective dose, which refers to the organ and tissue weighted dose to a person received from an intake of radioactive material during the 50-year period following the intake. Beyond this, risks associated with these dose values are calculated to put the exposures to contaminants in perspective to other risks, such as the risk from smoking or the risk associated with natural background radiation.

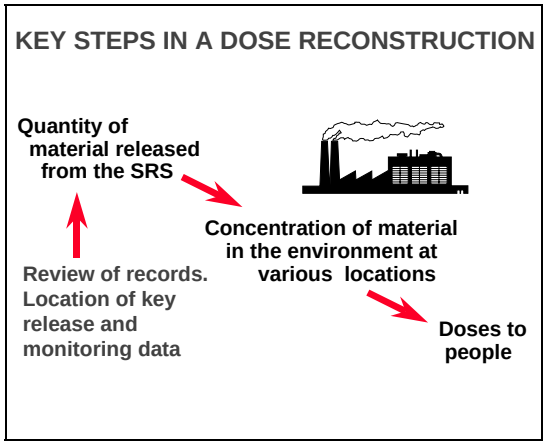


Figure 1-1. The key steps in a dose reconstruction, which evaluates the effects of past releases to the environment on offsite residents.

The CDC contracted with *Radiological Assessments Corporation* (*RAC*)[1] to conduct Phase II of this health risk study. Phase I was a search of SRS and other areas to find and copy documents and other records of potential value to the dose reconstruction project. Phase I was finished in June 1995, and an electronic document database was the primary product. Phase II began in October 1995 to estimate the releases of radionuclides and chemicals from the SRS to the environment during past operations from 1951 through 1992. Future phases of the dose reconstruction will include assessing the movement of the contaminants into the environment, estimating radiation doses and chemical exposures, and evaluating health risks to those living offsite.

The project Study Area is shown in Figure 1-2. It includes all communities within a distance of 50-miles from the SRS boundary, Columbia, South Carolina, and communities along the Savannah River south of the SRS. The CDC, the SRS citizens' Health Effects Subcommittee, and others helped *RAC* set up the Study Area, and find population, food crop, and similar information for this Study Area. The initial scope of this part of the project was cut back early in Phase II because of Federally mandated funding limitations, but with the help of South Carolina State University, *RAC* and CDC have collected a great deal of useful information, stored as a set of

[1] *Radiological Assessments Corporation* changed its name to *Risk Assessment Corporation* (*RAC*) in 1998.

computerized maps that will be available to those people who are interested. These types of data will be useful during later phases of the project.

This report and its computer files are part of the results of the Phase II study, along with a collection of about 5500 documents, declassified for this project and available to the public. The documents are the result of *RAC*'s systematic review of *all* records at the SRS. *RAC* opened and examined documents from about 50,000 boxes, and recorded their findings in a computer database. CDC received the document database, which will also be available to the public.

The Phase II work has been open to the public, and has offered opportunities for people to become involved. *RAC* has conducted hundreds of interviews and dozens of public meetings and visits to the site to learn about the SRS and potential sources of data.

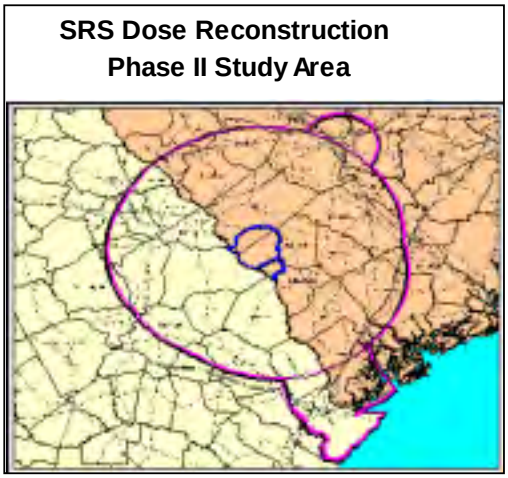


Figure 1-2. The project Study Area surrounding the SRS.

The study results are available as a report and as computer files with hypertext links to references, and to Excel® files containing the data sets summarized via graphics within the chapters. The document files are in Word2000® format, and the associated Excel files are in Excel2000® format. Eleven appendices to the report present procedures and supporting data. In addition, several of these appendices contain information specifically to support future stages of the dose reconstruction. Appendix D summarizes the meetings, fact sheets, newsletters, and other means used to communicate interim results to members of the public. Appendix F describes the large set of map-based data (Geographic Information System data) and demographic data collected by *RAC* and South Carolina State University during the course of the study. The GIS data were provided to CDC as part of the report, on CD ROM disks.

This report presents our results in this order: 1) radionuclide releases, 2) environmental monitoring data, 3) chemical releases and environmental monitoring, and 4) appendices and computer files. This current report summarizes the site history that was described in detail in the Phase I, Task 3 report, and includes diagrams of process areas onsite. *RAC* compiled and reviewed declassified production information for the five production reactors and the two reprocessing canyons, and estimated tritium production from the records (Figure 1-3). The release estimates presented here may be used to estimate doses to people living near the facility during its production history (1954-1992). The CDC will decide later whether such a dose reconstruction is needed. The scientific community and the

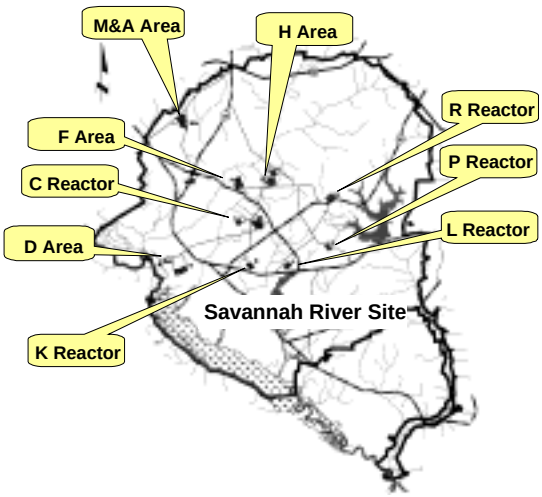


Figure 1-3. Diagram of the Savannah River Site showing the main facilities.

public have had the opportunity to review the report and have provided comments, which have been addressed and incorporated into this Final Phase II Report.

SELECTING MATERIALS OF CONCERN

Because there are numerous materials potentially released from a facility like the SRS to the environment, screening calculations helped to make objective decisions about the focus of the source term work. Through the screening methods, *RAC* identified certain chemicals (see Chapter 16) and radionuclides (see Chapter 3) through a phased approach. This approach did not eliminate any materials from consideration but rather focused first on those that are most important for public health. For screening radionuclides, we used the National Council on Radiation Protection and Measurements (NCRP) screening methods that have been revised and updated over the years and used extensively in other projects. This method involves a phased approach to screening and does not eliminate any radionuclides from consideration; rather, it focuses first on those radionuclides that are most important for public health (Table 1-1). For radionuclides, the screening process also evaluated how important each radionuclide was with respect to the exposure pathway (air or water), and produced a list of key radionuclides for which detailed source term, or release, estimates were developed

For chemicals, *RAC* reviewed their use in site processes and their potential for release from the SRS in the screening process. This evaluation of their hazards and magnitude of the quantities of the chemicals onsite led to the selection of key chemicals for further evaluation (Table 1-2). Chemicals that were detected in the environment around the SRS were added to the list of chemicals subjected to the ranking, shown here.

Table 1-1. Radionuclides identified in our screening process as the important contributors to dose

Key Radionuclides	
■ Releases to air	■ Releases to water
– Iodine-131	– Cesium-137
– Tritium	– Tritium
– Argon-41	– Strontium-90
– Iodine-129	– Cobalt-60
– Plutonium-239,240	– Phosphorus-32
	– Iodine-131

Table 1-2. Chemicals identified as potentially important from an initial screening of SRS chemicals

arsenic	benzene
cadmium	chromium
coal	coal ash
hydrogen sulfide	hydrazine
gasoline	diesel fuels
lead	manganese
mercury	nickel
nitric acid	nitrogen dioxide
sulfur dioxide	tetrachloroethylene
trichloroethylene	trichloroethane
uranium	zinc

RELEASES OF RADIONUCLIDES FROM THE SRS

Sources of Radionuclide Contamination

When evaluating the effects of radionuclides released from a DOE weapons facility, it is important to be aware of the radionuclides present in the environment that were released from other sources. Figure 1-4 shows that one of the main sources of radioactivity in the environment was fallout from the nuclear weapons testing program in the 1950s and 1960s. *RAC* evaluated these sources, including fallout from nuclear weapons tests in the 1950's and 1960's, to help place SRS releases in the appropriate context of these sources and of background levels.

Particular emphasis was given to examining a March 1955 incident during which atmospheric and ground surface contamination appeared at the SRS, apparently caused by the rainout of radionuclides from a Nevada nuclear weapon test. This case provides a detailed example of the methods used to identify SRS-released vs. offsite contamination, and also provides a reevaluation of an incident that still concerns some individuals who lived near the site at that time.

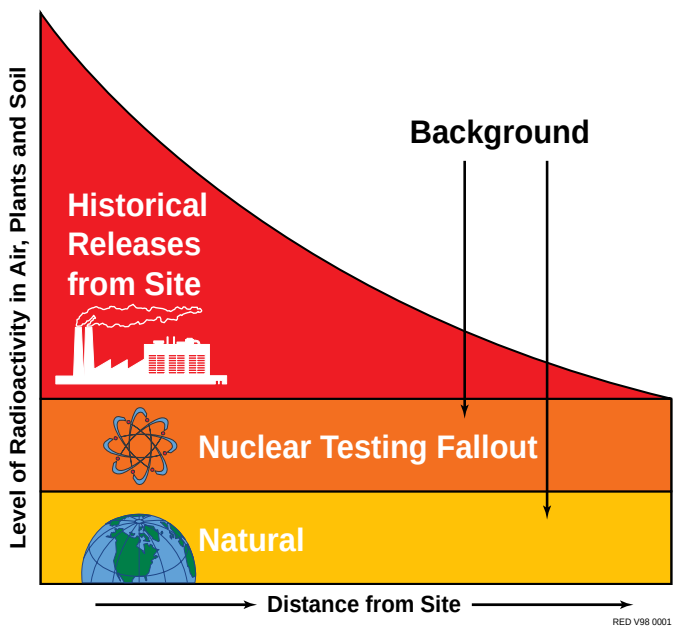


Figure 1-4. Other sources of radionuclide contamination.

Some radionuclides would be expected to come almost exclusively from SRS releases. These include: ^{85}Kr, ^{41}Ar, ^{32}P, ^{35}S, ^{60}Co, ^{129}I, ^{99}Tc, and ^{51}Cr. In addition, ^{238}Pu is mainly released to the environment from facilities that produce or process nuclear materials (such as the SRS), although weapons or satellite launch accident fallout have contributed lesser amounts. Radionuclides in weapons fallout, including ^{241}Am, ^{137}Cs, ^{3}H, ^{131}I, ^{90}Sr, ^{239,240}Pu, ^{65}Zn, and ^{95}Zr/Nb were also released in significant quantities from the SRS. A careful examination of quantities and trends is necessary before an informed judgment can be made about radionuclides arising from multiple sources.

Releases of Radionuclides to Air

Releases of Tritium To Air

Tritium (^{3}H or T) was one of the principle nuclear materials produced at the SRS to multiply the firepower of plutonium in nuclear weapons. It is the heaviest and only radioactive isotope of hydrogen, with a physical half-life of 12.5 years. Tritium processing operations began at the SRS

facility in October 1955 in the F-Area (Building 232-F). In July 1957, the process was moved to the H-Area (Building 232-H) and operations doubled by 1958. The key processes that lead to tritium releases at the SRS were:

- Reactor operations (C, K, L, P, and R Reactors)
- Recovery of transuranic elements in the separations facilities (H-Area and F-Area)
- Recovery of tritium in the Tritium Facilities (H-Area)
- Laboratory research area
- Heavy water rework facility.

The releases of tritium from the SRS are generally very well documented in published reports. Records of releases have been kept since the beginning of operations. In general, there is quite good agreement between the different release values reported in the different sources. Because in most cases the summary values were within 10% of the published values, it is likely that most "unchecked" monthly values are also valid. The key processes that have lead to atmospheric tritium releases are reactor operations, recovery of transuranic elements in the separations facilities (F-Area and H-Area), recovery of tritium in the tritium processing facilities, laboratory research area, and heavy water rework facility. The majority of the tritium releases came from the reactors and the tritium facilities through routine operations. In addition, there were about 3000 release incidents involving tritium releases. Figures 1-5 and 1-6 show the annual tritium releases to air from the reactors and from the tritium facilities in the F-Area and H-Area. Table 1-3 lists our annual median release estimates for tritium to air from SRS sources. Our tritium release estimates to air are described fully in Chapter 4.1.

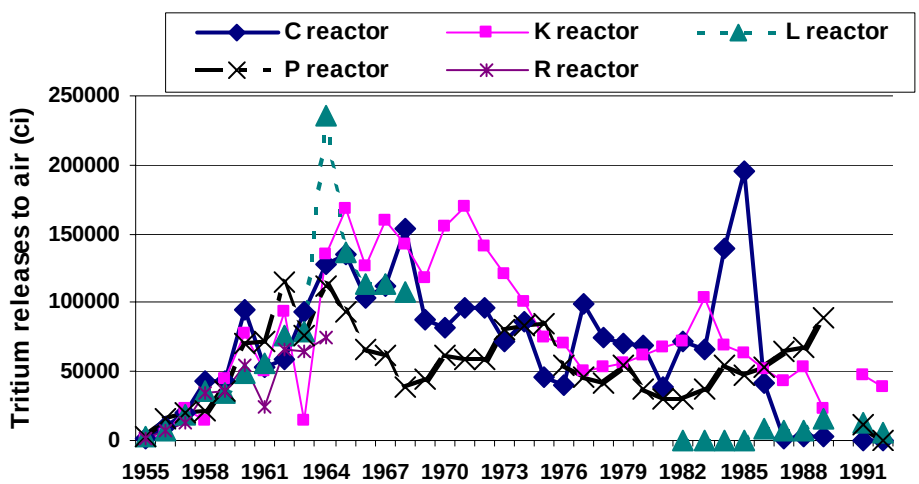


Figure 1-5. Annual releases of tritium to air from the 5 production reactors for all years of operation. The L Reactor released over 235,000 curies in 1964, the highest annual total of any of the reactors onsite. The L reactor was shut down after 1968 and restarted in 1982. The R reactor operated only through 1964. The C Reactor, which operated from 1954 through 1987, had high releases in 1984 (150,000 Ci) and 1985 (200,000 Ci) due to leaks in the reactor vessel. (See Figure 1-6 for comparison with releases from the tritium facilities in the H-Area and F-Area).

Table 1-3. *RAC* Median Release Estimates of Tritium to Air (Ci)

Year	C reactor	K reactor	L reactor	P reactor	R reactor	232F	232H	234H	238H	244H
1955	1044	1694	2745	3075	783	17860	10997			
1956	10037	4967	6936	15250	6652	436662	19990			
1957	19441	22820	19279	20595	13235	665665	665231	498102		
1958	43007	14856	35629	22182	34442	868285	1349157	249945		
1959	43007	44515	34820	35916	35557		596399	280047		
1960	94200	77443	48303	70632	54417		426420	250241		
1961	53209	49609	55558	72497	23855		451435	217586		
1962	59214	93494	75957	114893	66786		376473	390601		
1963	93788	14686	78591	76067	64331		430499	338132		
1964	127339	134994	235424	111557	75240		345264	652547		
1965	135144	168067	136136	93526			182393	144144		
1966	103764	125843	112807	66754			147635	165242		
1967	112399	158956	113617	61267			193225	130930		
1968	153151	141998	107220	38165			226760	202871		
1969	87297	118276		45246			147072	118236	2045	
1970	81939	155790		61386			136798	122507	1902	521
1971	96018	169705		58719			194523	204444	6226	82
1972	95785	141291		58825			299624	252260	3429	
1973	71512	120422		80632			198546	124097	4192	
1974	86137	101168		83914			131235	508598	3512	642
1975	45432	74662		84904			65262	256696	3320	367
1976	39682	69934		55206			56640	70283	3255	71
1977	98776	50295		46661			86916	106696	6770	1097
1978	74871	52709		41156			120247	63154	8339	373
1979	70380	56609		54794			76262	84139	7898	
1980	69334	61904		37548			71414	68933	5284	1450
1981	39430	67809		30042			92438	144738	3762	
1982	71607	71443	149	30611			122771	143252	3839	507
1983	66259	104078	491	36692			172842	253529	4646	1286
1984	139132	68511	127	54677			140483	388456	4739	1271
1985	195877	63303	683	47664			136764	231312	3889	
1986	41565	51817	7943	53252			127007	132209	3191	
1987	2040	43456	7009	64289			283003	183954	2332	
1988	2656	52496	6711	67757			95080	167944	3902	
1989	2329	22687	15713	89685			60436	122182	2984	
1990							48573	131522	4145	
1991	553	46805	12261	10872			49837	86557	3722	
1992	368	38427	6017	307			68742	62223		

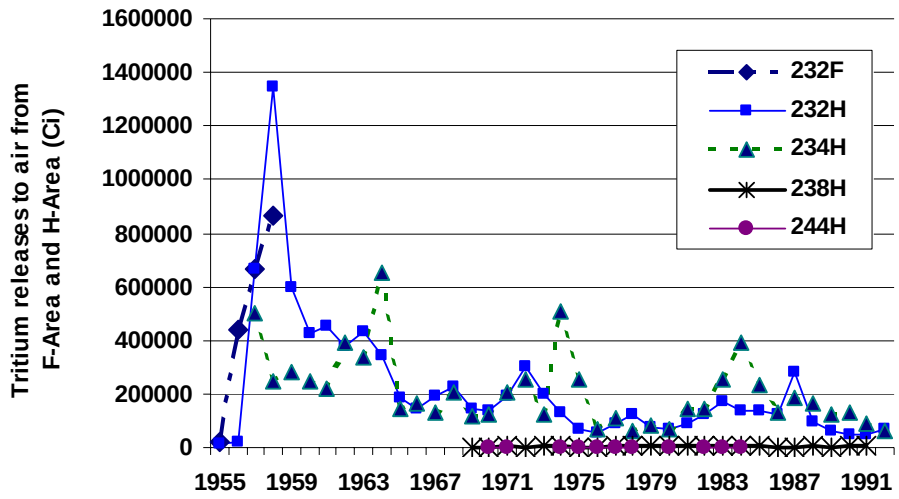


Figure 1-6. Annual releases of tritium to air from the tritium facilities in the F-Area and H-Area for all years of operation. The original tritium extraction facility in 232-F operated from 1954 through 1958. It was replaced by the H-Area tritium facilities (232-H and 234-H) that have operated continuously since 1957. The largest releases of tritium occurred from the H-Area facilities from 1957 through 1959, with over 1.3 million curies released from 232H in 1957. (See Figure 1-5 for comparison with tritium releases to air from the 5 reactors).

Releases Of Radioiodines and Beta-Gamma-Emitting Particles to Air

Radioactive iodine and other beta-gamma-emitting materials are produced during fission of uranium and plutonium in reactor fuels. Because most of the releases of radioiodines and beta-gamma-emitting particles occurred when the fuel was processed (usually many days after removal from the reactor), the radionuclides of greatest interest are those produced in large quantities and that have half-lives that exceed 1 day. Reprocessing facilities were the largest sources of airborne radioiodine releases. From the point of view of human health, the most important discharges are those of elemental iodine (I_2), which are most important for human thyroid. Releases of organic iodides (like CH_3I), which are relatively unimportant for food chain pathways, contribute to exposure by inhalation. Releases of ^{131}I were highest in the 1950s, and our estimates for that time for elemental ^{131}I are about four times those that were reported by the SRS. This is due mainly to a correction for deposition in the sampling line. Only elemental iodine was measured at that time; however, organic iodides accounted for 70-90% of the ^{131}I release from the separations areas. Thus, the organic iodide releases are roughly 4 times higher than the elemental iodine releases. This leads to a big addition in the early years because no contribution to the total from the release of organic iodides was previously considered. After 1961 when total iodine was being measured there are smaller differences between previous reports and the revised estimates. Figures 1-7 and 1-8 show the annual releases from the SRS of elemental and organic forms of ^{131}I, respectively. See Chapter 4.2 for full details on iodine release estimates.

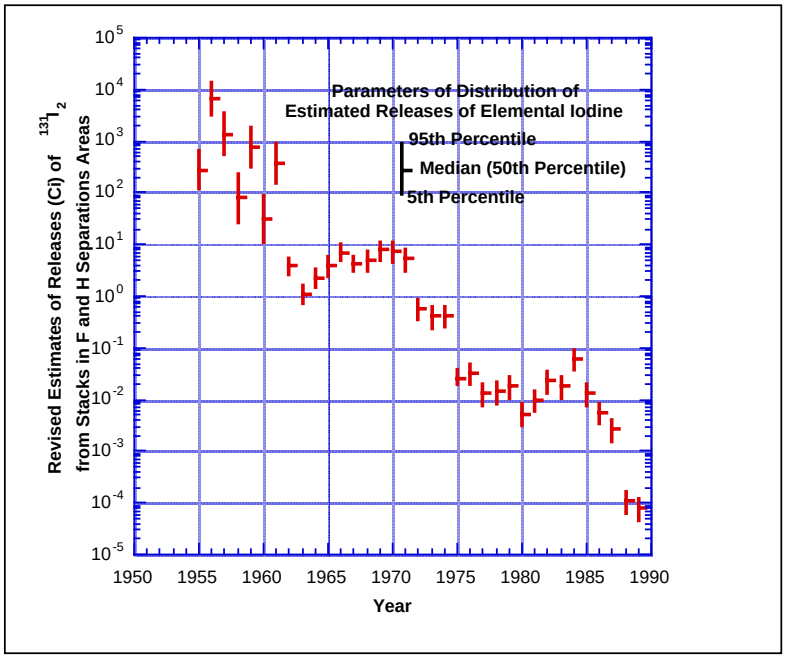


Figure 1-7. *RAC* releases estimates of ^{131}I in elemental form (I_2) from stacks in the F and H separations areas. For each year, the length of the vertical line shows the range (5th to 95th percentiles) of the distribution of release estimates and the median estimate is indicated by the horizontal bar.

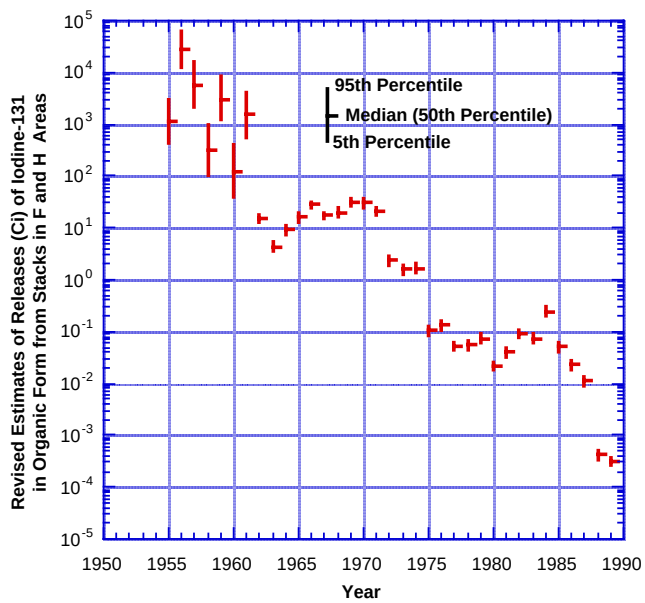


Figure 1-8. *RAC* estimates of releases of ^{131}I as organic iodides from stacks in the F and H separations areas. For each year, the length of the vertical line shows the range (5th to 95th percentiles) of the distribution of release estimates and the median estimate is indicated by the horizontal bar.

Releases of Activation Products to Air

While most of the radioactivity produced by a reactor involves the products of fission, some isotopes are produced as activation products, when neutrons interact with materials in the reactor. The quantities of activation products are smaller but are created outside the fuel area of the reactor so that they are closer to the outside environment. Several SRS locations had the potential to release activation products. The largest releases occurred during the 1960s, and consisted primarily of ^{41}Ar from the production reactors. Figure 1-9 presents annual data for ^{41}Ar releases. For additional details on the production and release of activation products, see Chapter 4.3.

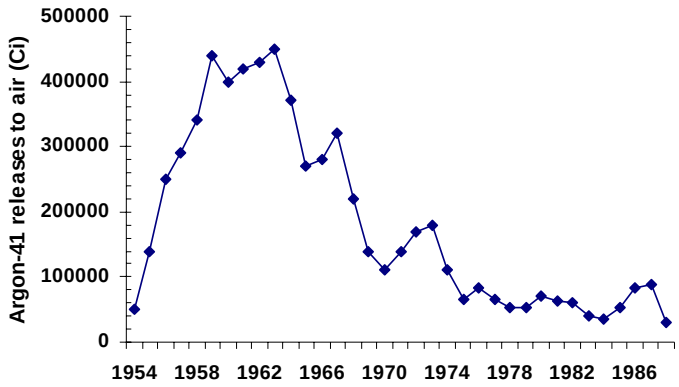


Figure 1-9. Annual releases of ^{41}Ar to air from reactors at the SRS.

Releases Of Alpha-Emitting Radionuclides to Air

The alpha-emitters, primarily plutonium and uranium at the SRS, were released from the fuel fabrication facilities in the M-Area, the reactors, and the reprocessing facilities in the H-Area and F-Area. Releases from the separations facilities were monitored since December 14, 1954, shortly after startup. Continuous sampling was accomplished by passing a portion of stack effluent through a filter designed to trap particulates. The filter was changed weekly and analyzed for specific isotopes. Measured releases from the M-Area and reactor facilities (R reactor, P reactor, K reactor, L reactor, and C reactor) were quite small. The data indicate that the majority of plutonium emissions from both F-Area and H-Area stacks occurred during 1955 and 1969. The data also indicate that the majority of uranium emissions from H-Area stacks occurred during 1955, 1968, and 1969, and that the majority of uranium emissions from F-Area stacks occurred during 1955, 1956, and throughout the 1960s. Evaluation of potential impacts resulting from alpha-emitting radionuclide releases should be focused on these years because the relative magnitude of total emissions during other years (including all years since 1970) appears to be quite small. Figure 1-10 shows our annual release estimates for plutonium from the main F-Area and H-Area stacks. See Chapter 4.4 for details on release of alpha-emitting radionuclides to air from the SRS.

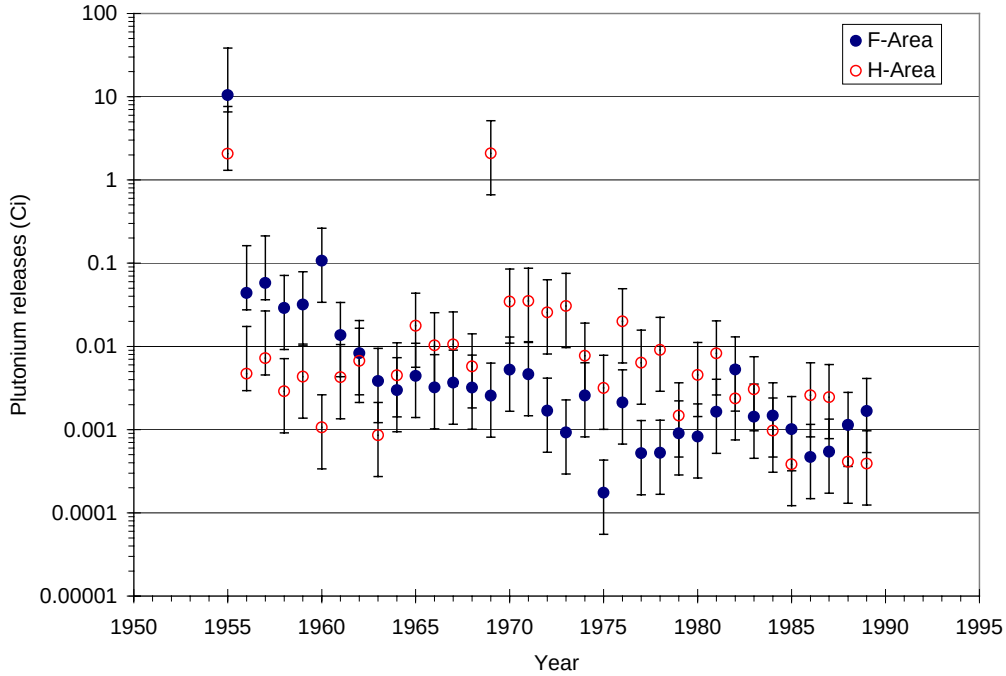


Figure 1-10. Annual median plutonium (including ^{238}Pu and ^{239,240}Pu) release estimates for F-Area and H-Area. The upper and lower error bars represent the 95th and 5th percentile values, respectively.

Releases Of Radionuclides to Surface Water

The controlled fission process within the reactors produced enormous amounts of heat. Heavy water (water made from the hydrogen isotope deuterium) was circulated in a closed system through heat exchangers to cool and moderate the reactors. Incoming Savannah River water was stored in holding basins at each reactor, passed through the heat exchangers, then was discharged to Site streams. In 1958, Par Pond, a 2,700-acre lake, began providing cooling water for the P and R Reactors. The water was pumped back to the pond, allowing more river water to be pumped to the L, K, and C Reactors. In 1985, L Lake was formed by damming Steel Creek, to provide cooling water for the L Reactor. Before Par Pond and L Lake were created, water was discharged directly to Lower Three Runs Creek (from P and R Reactors), to Steel Creek (from L Reactor), to Pen Branch (from K Reactor), or to Four Mile Creek (from C Reactor).

The Disassembly Basins were the primary sources of radionuclide releases to surface water from the reactors. Spent fuel and irradiated target elements were removed from the reactor and stored in these large water-filled basins adjacent to the reactor building. The water in the basins cooled the components and provided shielding. Contamination from these elements gradually moved into the basin water. From startup to the mid-1960s, visual clarity was maintained in the basins by continuously purging them with fresh, filtered river water. These purges released radioactivity dissolved or suspended in the basin water to the SRS streams.

Complex chemical and physical processes in the F- and H-Canyon Buildings separated uranium, plutonium, and fission products. Fission products were placed in underground high-level waste tanks nearby. Cooling water for portions of the F-Area separations process line was

pumped from deep wells and discharged to Four Mile Creek. At times, this water contained some radioactivity because of cooling coil leaks. The F-Area liquid effluent included process cooling water, sanitary wastewater treatment effluents, and spill runoff; H-Area effluent was similar.

The SRS had a fairly broad effluent monitoring program for releases of key radionuclides from the main facilities onsite to the streams and seepage basins. Tritium and ^{137}Cs were the main radionuclides of concern for releases to surface streams and the Savannah River. We developed detailed source terms for those radionuclides (see Chapter 5 for details). Other radionuclides of importance include ^{90}Sr, ^{131}I, ^{60}Co, ^{32}P, and uranium releases from the M-Area to Tim's Branch Creek. Surface water releases of radionuclides were highest in the early to mid-1960s and decreased into the 1980s. Figure 1-11 shows annual release estimates of tritium to surface water.

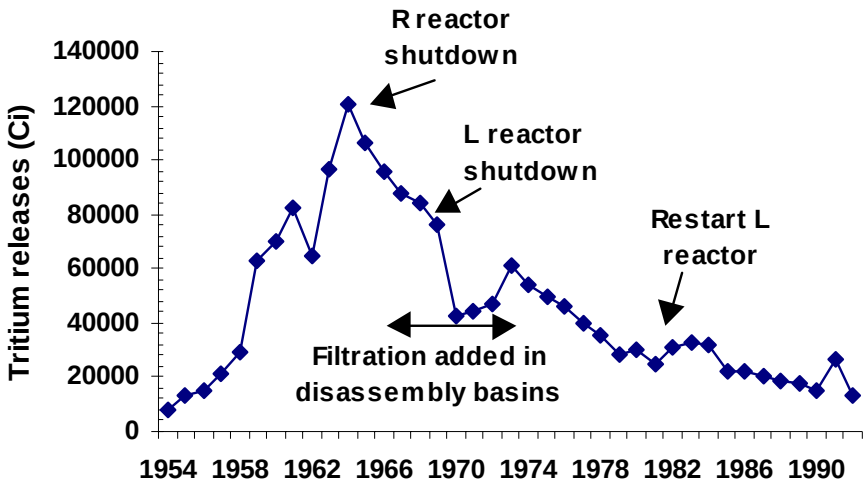


Figure 1-11. *RAC*'s annnual tritium release estimates to surface water. Some important events that affected releases of radioactivity to surface water at the SRS are shown. The highest releases of tritium to surface water occurred in the mid-1960s.

Figure 1-12 shows our annual release estimates of ^{137}Cs to surface water with uncertainty estimates. There is overall general agreement between our reconstructed release estimates to the Site and the annual total reported by SRS.

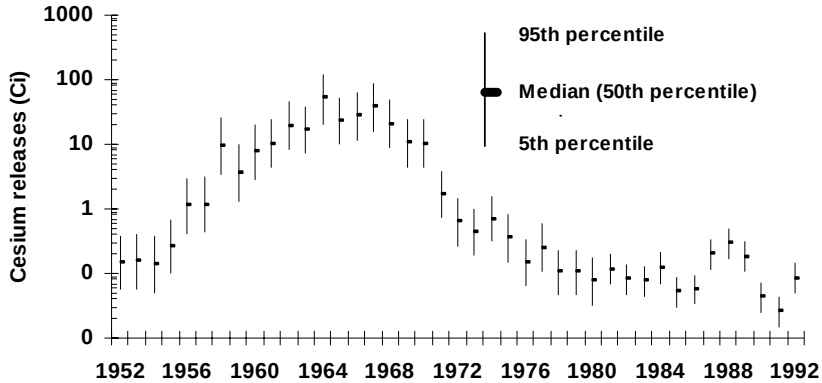


Figure 1-12. *RAC* estimates of ^{137}Cs releases to surface water from the SRS with uncertainty estimates, shown on a logrithmic scale. Each year is represented by a vertical line that represents the 95th (top) and 5th (bottom) percentiles of the distribution of releases with the median or 50th percentile shown as the filled shape in the center.

Environmental Monitoring For Radionuclides

Environmental monitoring data are valuable resources for dose reconstruction because they provide direct information about the concentrations of radionuclides in air, water, vegetation, and foods at particular places and times. These data may be used to independently check the source term estimates or environmental transport model calculations, for example. In some cases, the data may also be useful for direct exposure assessment. The current report describes the history, scope, and results of the environmental monitoring programs for:

- Air and rainwater
- Vegetation and agricultural products
- Milk
- Wild game
- Sediments and soil
- Savannah River water, community water supplies, and water treatment plants
- Fish.

Air and rainwater have been sampled at various locations on or in the vicinity of the SRS since background measurements began in 1951. Air represents a major pathway for transport of SRS releases and exposure of offsite individuals. The key contaminants detected in air and rainwater during the period of interest (1951-1992) are radioiodine and tritium. Their concentration decreases with distance from the SRS. Air monitoring and other environmental data may be used to validate airborne source terms and dispersion models.

The report describes the extent and analytical results of the radiological monitoring program for water supplies of 14 communities surrounding the SRS, for one upstream and two downstream drinking water treatment plants supplied by the Savannah River, and for offsite locations from the Savannah River. For dose reconstruction, the potential impacts of SRS

operations on both the surrounding community water supplies and the downstream water treatment plants are of greatest concern because the safety of drinking water is of great interest to the public. Figure 1-13 shows tritium concentrations in water from the treatment plants near the SRS. The Savannah River monitoring data can be used evaluate the impact of SRS releases of radionuclides on the downstream drinking water supplies, and will be useful to evaluate other exposure pathways, such as the ingestion of fish and the recreational use of the river.

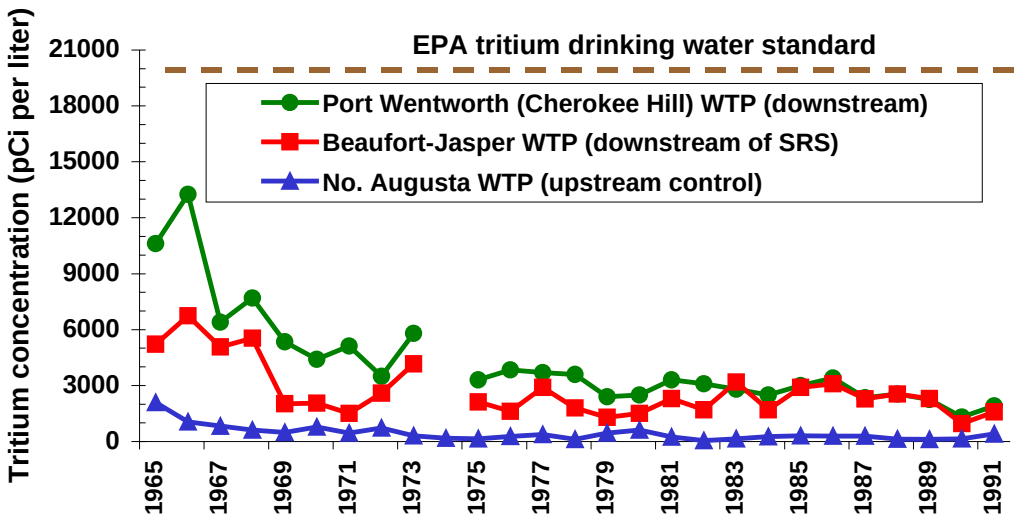


Figure 1-13. Tritium concentrations in water from the water treatment plants.

RELEASES OF CHEMICALS FROM THE SRS

While radionuclides released from the SRS are important in evaluating potential risks to individuals living offsite, it is also necessary to understand that the SRS used, produced, and released large quantities of chemicals. The extent of releases of these chemicals must also be considered when evaluating the potential risk to offsite residents. Source term, or release, estimates were determined using inventory or usage estimates, knowledge of processes, information currently required by regulatory agencies, and monitoring data. There is sparse or no monitoring data for chemicals used at the SRS, especially before 1980. A limited amount is available from ambient air monitoring, water quality monitoring, and special studies. For some chemicals, extrapolating back in time from the 1980s monitoring data was the best way to estimate a source term, assuming that the amounts and types of materials used and the processes did not change greatly over the years.

RAC used emissions estimates from the air emissions inventory to estimate chemical releases to air. Large amounts of sulfur dioxide, nitrogen dioxide, and particulates (ash) were released from seven coal-fired power plants. We estimated releases of mercury, lead, manganese, nickel, nitric acid, chromium, cadmium, and hydrogen sulfide to air, as well. We found enough information to calculate the uncertainty associated with the release estimates for the chemicals listed in Table 1-4.

Table 1-4. Release Estimates and Uncertainty Ranges for Chemicals Released to the Air

Chemical	Median average annual release (ton y^{-1})	5th–95th percentile values on the median (ton y^{-1})
Coal ash	4200	2300–7100
Mercury	0.3	0.18–0.51
Nitrogen dioxide	6050	4320–8480
Sulfur dioxide	11000	8470–14400

Uncertainty calculations were not made for the chemicals listed in Table 1-5 because there was a lack of information; however, a range of releases was estimated.

Table 1-5. Release Estimate Ranges for Some Chemicals Released to the Air

Chemical	Range of release estimates (ton y^{-1})
Benzene	1.8–18
Lead	0.05–0.12
Manganese	0.07–1.9
Nickel	0.11–0.42
Nitric acid	30–150

Table 1-6 shows that thousands of tons of chlorinated solvents were released from the M Area. The table displays our median release estimates of solvents to air and to Tim's Branch Creek from the M-Area. Almost all of the chlorinated solvents evaporated during use or after being discharged to surface water. Until 1979, waste solvent was released to the M-Area settling basin or to a stream called the Tim's Branch, where most evaporated. Table 1-6 summarizes the release estimates for trichloroethylene, tetrachloroethylene, and trichloroethane. Chapter 17 has details on releases of chemicals to air.

Table 1-6. Trichloroethylene, Tetrachloroethylene, and Trichloroethane Releases to Tim's Branch from the M-Area

Solvent	Source	Time period	Median
Trichloroethylene	M-Area use	1952–1970	1700 ton (average of 90 ton/y over 19 y)
	M-Area air strippers	1985–1990	70 ton (average of 12 ton/y over 6 y)
Tetrachloroethylene	M-Area use	1962–1975	4055 ton (average of 289 ton/y over 14 y)
	M-Area air strippers	1985–1992	30 ton (average of 3.8 ton/y over 8 y)
Trichloroethane	M-Area use	1979–1988	2200 ton (average of 220 ton/y over 10 y)

For releases of chemicals to surface water, we described the sources and potential releases of arsenic, cadmium, chromium, coal and coal ash, gasoline, hydrogen sulfide, lead, manganese, mercury, nickel, nitrates, uranium, and zinc to water. Mercury was used for several purposes at

the SRS, and in the early years was often discarded after use. In summary, historical releases of chemicals to the Savannah River are difficult to determine and quantify. Release estimates are summarized in Table 1-7. In addition, an undetermined amount of chromium used to treat high level waste tank cooling coil water was released.

Table 1-7. Summary of the Estimated Releases of Chemicals to Surface Water

Release estimate (maximum or estimated range)	Released to
1 kg y^{-1} of cadmium	To Tim's branch
900 kg y^{-1} of hydrogen sulfide	To Beaver Dam Creek
8–50 kg y^{-1} of lead	To Tim's Branch
15–623 kg y^{-1} of lead	To the Separations Area Seepage Basins
16–70 kg y^{-1} of mercury	To the Separations Area Seepage Basins
0.1–8 kg y^{-1} of mercury	Entering Four Mile Creek in Groundwater
116–2000 kg y^{-1} of nickel	To Tim's Branch
0–1383 tons y^{-1} of nitrate	To the Separations Area Seepage Basins
Up to 108 tons y^{-1} of nitrate	To Four Mile Creek in Groundwater
27–200 tons y^{-1} of nitrate	To Tim's Branch

Although large amounts of some chemicals were released to seepage basins and SRS streams, the impacts to surface water extending beyond the Site boundary do not appear to be measurable. The available environmental monitoring information does not support an appreciable source term from the SRS for nonradiological contaminants for all identified chemicals and heavy metals of concern measured in surface water, groundwater, and ambient air.

CONCLUSIONS

The review of some 50,000 boxes of records associated with the SRS has produced a large set of data pertinent to the reconstruction and validation of radionuclide and chemical source terms for the Site's operational history. We conclude that, for the key radionuclides, the available data are adequate to develop estimates of dose to individuals living offsite during past SRS operations. For the key chemicals, information prior to the 1980s is very sparse. However, we have reconstructed approximate ranges of key chemical releases for SRS operations, and it may be feasible to develop conservative, bounding risk estimates for these chemicals to members of the public living offsite.

CHAPTER 2

BRIEF HISTORY OF FACILITIES AND OPERATIONS RELEVANT TO THE RELEASE OF RADIONUCLIDES

ABSTRACT

The Savannah River Site (SRS) Dose Reconstruction Project, a research effort sponsored by the Centers for Disease Control and Prevention (CDC) is a study of historical releases, doses, and risks to members of the public living offsite during past operations. *Radiological Assessments Corporation* (*RAC*) has performed Phases I and II of the dose reconstruction. During Phase I, *RAC* gathered the records necessary for a dose reconstruction. During Phase II of the project, *RAC* staff have used that information to estimate historical radionuclide and chemical release rates (the source term) from the SRS. Also during Phase II, environmental monitoring records, map-based information and other records have been collected and evaluated. Information from these additional records is presented in this report and its associated document and data sets. This chapter provides background information in support of the subsequent source term development and environmental monitoring information chapters. It contains a brief history and description of the SRS, then presents SRS reactor and reprocessing data collected by *RAC* during the study and used in later chapters during source term development efforts.

The first section of this chapter discusses key areas and processes at the SRS to help the reader understand the factors contributing to offsite releases and potential public health impacts. The most likely scenarios for offsite impact involve releases of particulate and gaseous materials to the air, with subsequent atmospheric transport offsite, and releases of liquids to surface streams that flow offsite. The SRS areas and processes, organized by location, are described in detail in "History and Descriptions of Key Processes at SRS," in the SRS dose reconstruction Phase I, Task 3 report (Meyer 1995).

The second section of this chapter discusses the methods used by *RAC* to identify, locate, extract and declassify monthly reactor and reprocessing canyon records for use in the study. Some of these data have not previously been released to the public domain.

HISTORY AND DESCRIPTION OF THE SAVANNAH RIVER FACILITY

Creation and Development of the Savannah River Site

With the signing of the Atomic Energy Act of 1946, the Congress provided for the Atomic Energy Commission (AEC) to take over the nation's nuclear programs that had been administered by the Manhattan District of the United States Corps of Engineers. Early in 1950, a study group of Du Pont personnel who had been involved in the Hanford work in the late 1940s, began considering the design, construction, and operation of new production facilities. Of nearly one hundred possible sites, four sites were identified as favorable, including a site on the Red River in Texas, a site on the Wabash River in Indiana/Illinois, a site on the shore of Lake Superior in Indiana, and the current Site in South Carolina below Augusta, Georgia (Figure 2-1) (Joseph and Bannick 2000).

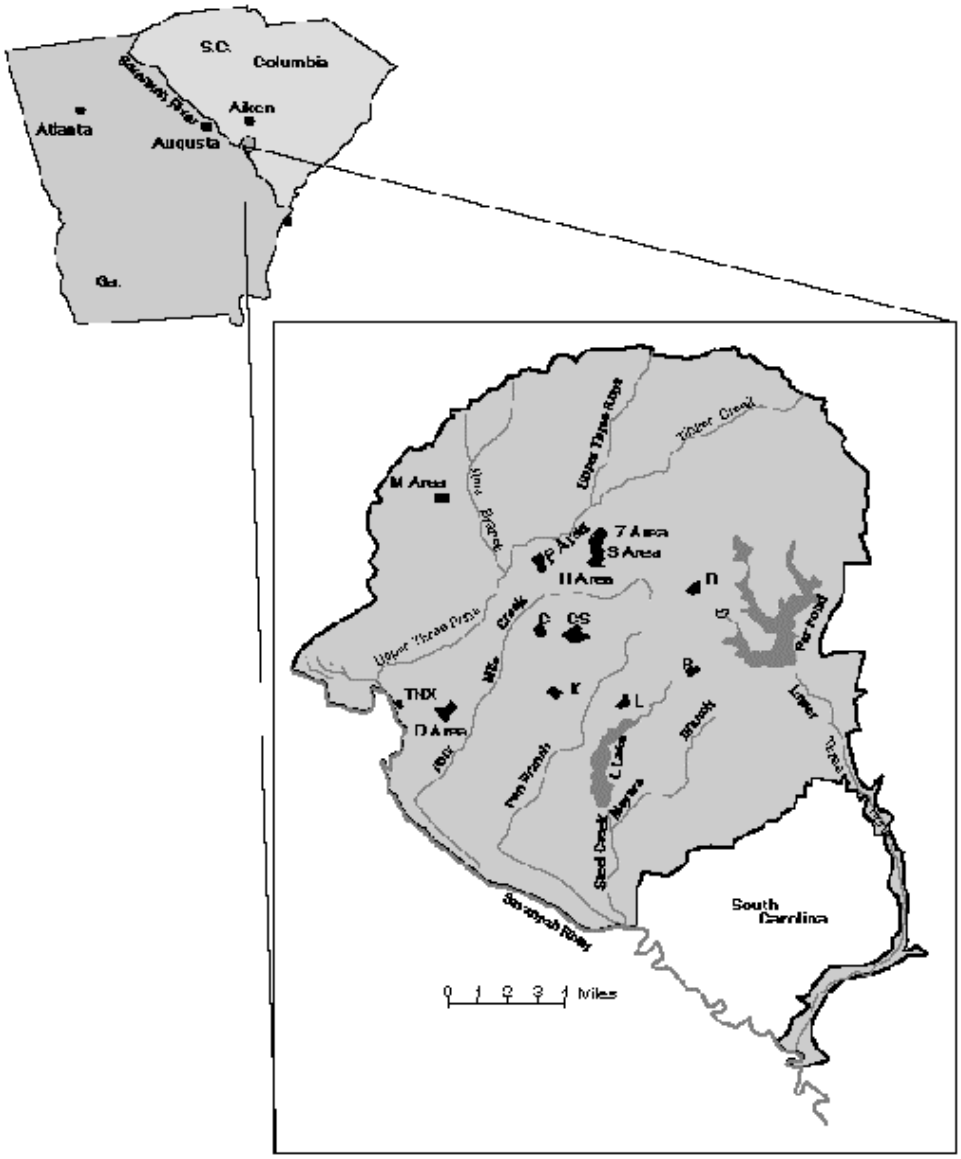


Figure 2-1. The Savannah River Site, showing its location in South Carolina on the Georgia border. Five Site streams flow into the Savannah River to the southwest. The Site occupies approximately 300 square miles. Figure 2-3 shows the Site in more detail.

The SRS, known as the Savannah River Plant (SRP) until 1989 when Westinghouse took over operations, was chosen because of its proximity to the Savannah River, a large source of water needed to remove the heat generated in the reactors, and a source for heavy water extraction. The large 300-mi^2 tract allowed builders to space production facilities at distances that would ensure security and safety (Bebbington 1990). Five major streams on the SRS feed into the Savannah River: Upper Three Runs Creek, Four Mile Creek, Pen Branch, Steel Creek, and Lower Three Runs Creek. Chapter 5 of this report presents detailed information on these streams and their involvement in releases from the SRS.

Figure 2-2 is a sketch of SRS processes and waste flows. Although SRS is divided into six major operational areas, each having a specific function, the key areas for our study were the set of five nuclear production reactors (100-R, -P, -L, -K and -C); the two chemical separations plants (200-F and -H) and their associated tritium production and waste management facilities; M-Area, where fuel and target elements were fabricated and cleaned; and D-Area, where heavy water was produced and processed. These process areas, spaced 2 to 3 mi apart along a rough circle centered within the Savannah River Plant (SRP) Site, were constructed in the early 1950s. They supported the primary industrial operations at SRS. The main activities were

- Fuel and target fabrication
- Operation of large-scale gas and liquid processes to extract and purify heavy water
- Reactor operation to create plutonium and tritium
- Production and purification of plutonium and tritium.

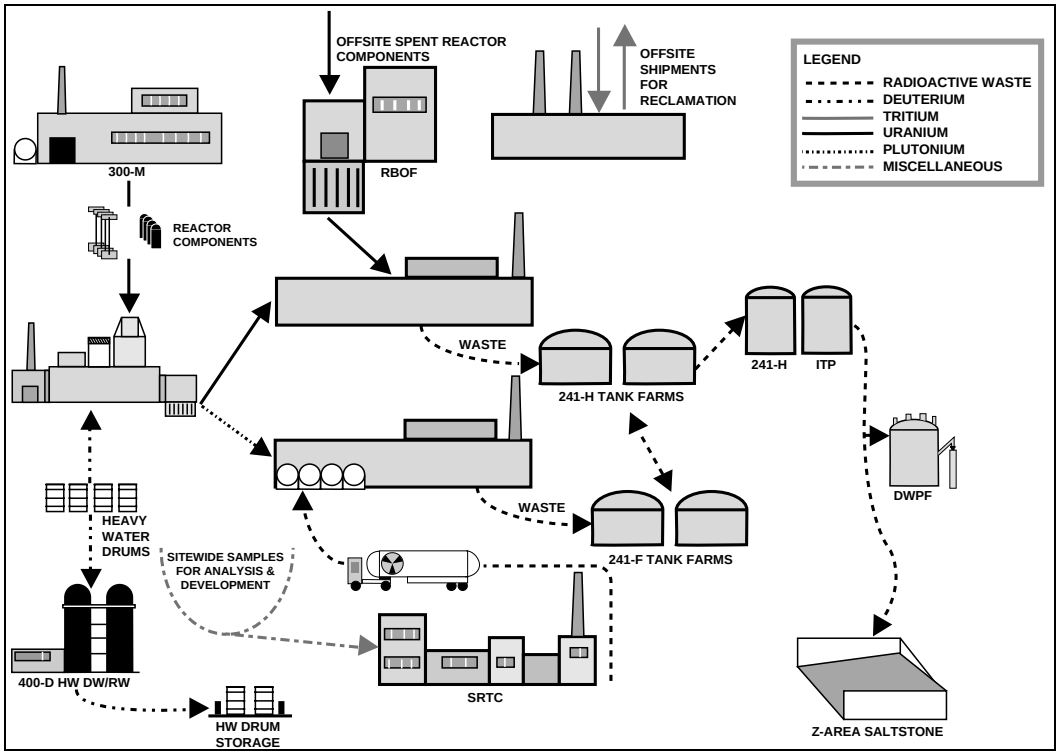


Figure 2-2. Process flow summary diagram of the SRS. The heart of the Savannah River Plant was the group of five nuclear production reactors and the F-Area and H-Area chemical separations plants (221-H Canyon and 221-F Canyon). The 400-D or Heavy Water (HW) area provided deuterium oxide or heavy water as a moderator to the reactors; its also was used to reconcentrate diluted heavy water. Fuel and target components, fabricated in the 300-M Area, were sent to the reactors. Cleaning solvents and wastes from the M-Area work remain as subsurface contamination problems. Fuel and target elements irradiated in the reactors were sent to H-Area and F-Area to be dissolved and the products extracted. Liquid waste was transferred to various tank farms, seepage basins, the Defense Waste Processing Facility (DWPF), or the Z-Area saltstone depending upon the level of radioactivity in the waste (Meyer 1995).

Table 2-1 provides a general overview of the key operational areas at the SRS and important release points, and more detailed descriptions of key SRS areas follow.

Table 2-1. Overview of SRS Facilities[a]

Facility	Dates of completion or operation	Notes
Heavy Water Plant-400-D Area	First of 24 GS units operating by Oct 1952; all by May 1953	Concentration of heavy water, and reconcentration after contamination in reactors with light water. Large quantities of H_2S were used at high pressure in the GS process; consequently, H_2S monitoring procedures were in place early because of its corrosive and toxic nature. Some potential for tritium release.
Fuel and Target Fabrication - 300-M Area	Jan 1953	Produced reactor fuel and target assemblies. Reactor assemblies became more complex as different products were desired and power levels were increased. M-Area used about 13 million pounds of chlorinated solvents to degrease items since mid-1950s. Uranium releases.
CMX and TNX technical development facilities. Savannah River Technical Center (SRTC)	1953	Technical support and pilot plant data and training.
Test “pile”; graphite-moderated reactor	Dec 1952	The reactor was used to calibrate monitors and to assay fuel and target slugs; dismantled in 1980
R-Reactor (100 Area)	Dec 1953–1964	First SRS production reactor; first fuel discharged June 1954; plutonium and tritium production. Liquid effluent to Lower Three Runs Creek, Par Pond, one seepage basin. Airborne releases, primarily gaseous, from all five reactors.
P-Reactor (100 Area)	1954–1988	Plutonium and tritium production. Liquid effluent to Steel Creek, Par Pond and three seepage basins
C-Reactor (100 Area)	1955–1985	Plutonium and tritium production. Liquid effluent to Four Mile Creek and three seepage basins
K-Reactor (100 Area)	1954–1988	Plutonium and tritium production. Liquid effluent to Pen Branch and two seepage basins
L-Reactor (100 Area)	1954–1968; 1985–1988	Plutonium and tritium production. Liquid effluent to Steel Creek, L Lake and a seepage basin; L Lake built in early 1980s by damming Steel Creek
H-Area and F-Area Canyon Buildings (200 Area)	November 1954, July 1955	Reactor products were separated chemically in the reprocessing canyons. Very large quantities of solid, liquid, gaseous wastes produced.
Waste Management Areas	1953	Includes various seepage basins, disposal pits; tanks and burial grounds.

[a] Source: Stetson et al. (1963), Bebbington (1990), Du Pont (1957), Meyer (1995), Wahl (1967).

Heavy Water Production and Reprocessing: D-Area

A heavy water production plant, in D-Area, began operation early in SRS history to concentrate heavy water from Savannah River water to moderate and cool the Site's reactors. The first of 24 girdler sulfide (GS) units was installed in October 1952, with all in place by May 1953. Tremendous quantities of hydrogen sulfide (H_2S) were used at high pressure in the GS process, which concentrated heavy water (D_2O) from its natural 0.015% level in river water (Bebbington 1990). Detailed procedures to monitor for H_2S gas in and around the operations areas were developed because of its corrosive, toxic nature and the possibility of accidental releases (French 1975). The facility stopped production in 1981 because there was a sufficient supply of heavy water. A heavy water reprocessing facility (built to reconcentrate heavy water that had become contaminated with light water during use in the reactors), a coal-fired power plant, and a laboratory to analyze process effluent samples were also located in D-Area. The concentration of tritium in heavy water was a function of the neutron flux in the reactor and the length of the irradiation, with tritium concentration is the moderator building up slowly over the years. Some of the tritium was lost to air and to liquid effluents by evaporation of moderator leaks and carry over of tritium oxide on fuel and target elements during reactor discharge. Chapter 4.1 discusses significant release points for tritium to the atmosphere at the SRS. Any tritium releases to the atmosphere resulting from heavy water processing were measured as stack effluent and are reported in Chapter 4.1. Chapter 5 discusses tritium releases to surface water.

Reactor Materials: 300-M Area

The facilities called the 300-M Area, the 300 Area or M-Area, produced fuel and target elements for the reactors. Control rods and other reactor components were manufactured here as well.

Over time, changes in the fuel rods and target elements were made to support higher levels of production, increased emphasis on tritium production, and the creation of other products for military, research, and satellite purposes (Pelfrey 1987). The reactor assemblies became increasingly complex as these different products were emphasized and higher reactor power was required. Since 1952, it is estimated that M-Area has used about 13 million pounds of chlorinated solvents to degrease the reactor components produced in the facility (Christensen and Brendell 1981). Much of this solvent material was disposed onsite, and it remains as underground contaminants. Chapter 15 discusses these materials in detail.

Reactor Areas

There are five nuclear production reactors located at the SRS; they are in the locations designated K-Area, L-Area, P-Area, C-Area, and R-Area. Plutonium and tritium—the primary products of the SRS reactors—were created in these reactors by uranium and lithium absorption of neutrons. The controlled fission process within the reactors produced those neutrons and enormous amounts of energy. The reactors were operated at relatively low temperatures (less than 100°C) and pressure (near 5 psi) for safety and to optimize product formation. These were heavy-water moderated reactors, which means that heavy water was circulated in a closed system

through heat exchangers to moderate and cool the reactors. The heavy water slowed fission neutrons, greatly increasing the number of additional fissions occurring. The energy produced was discarded as heat via the heavy water coolant/moderator flowing through the reactor cores. The moderator flowed through tubular heat exchangers, cooled by ordinary water. This heat exchange step greatly reduced the quantity of reactor radioactive materials directly entering the outside environment. The Savannah River and two onsite lakes provided cooling water for the heat exchangers. Reactor power levels were increased greatly during the early years.

During the construction of the SRS, the reactor tanks were fabricated by New York Shipbuilding Company at Camden, New Jersey, and shipped by barge via the Savannah River to SRS (Du Pont 1954; Bebbington 1990; Du Pont 1976–1984). All five reactors were operating by early 1955. By 1963, high-efficiency filters had been installed in the reactor ventilation systems to remove particulates, and charcoal beds had been installed to remove radioiodine (Wahl 1967). From 1956 through 1971, the reactors were also used to convert thorium to ^{233}U for the thorium breeder reactor program, as well as to produce numerous other special products (Stetson et al. 1963). Reactor shutdown began in 1964 with R-Reactor; C-Reactor was shut down in 1987. L-Reactor was shut down in 1968, restarted in 1985, then shut down again by 1988. K-Reactor and P-Reactor operated with few interruptions until 1988. A restart of the K-Reactor began in 1991, but it was not completed. All SRS reactors are now shut down.

F and H Separations Area, or 200 Areas

Products produced in the reactors were separated chemically in the F and H Separations Canyons, located in an 8 km^2 area near the center of the SRS (Figure 2-3). Operations in the separations area also include the unloading and storage of offsite fuel at the Receiving Basin for Offsite Fuel (RBOF).

The two chemical separations facilities, 200 F and 200 H, are very similar in construction. Each has two parallel lines of process cells, known as the hot and warm canyons, with a central system of corridors. Complex chemical and physical processes in the F- and H-Canyon buildings separated uranium, plutonium, and fission products. Processing capacities during the 1950s and 1960s were increased significantly.

The separated plutonium and uranium were transferred to other facilities in the F-Area and H-Area and processed into solid forms. Fission products were stored in high level waste (HLW) tanks in the separations areas. Detailed descriptions of the separations facilities, processes, and safety analyses can be found in Du Pont (1957). Bebbington (1990) contains a photographic history of Site construction.

Originally, the tritium received in the separations areas was a by-product of plutonium production. By 1955, greater production of tritium was required so reactor fuel rods and targets were modified, and a second tritium production line became operational in 1957. The key processes leading to tritium releases (detailed in Chapter 4.1) at the SRS included

- Production reactor operations
- Recovery of products in the separations facilities
- Recovery of tritium in the tritium facilities
- Laboratory research processes
- Heavy water rework facility.

Administration Area, or A-Area, and the TNX/CMX Areas

Organizations that supply direct support for SRS operations, including the U.S. Department of Energy (DOE) office for the Site, the Savannah River Ecology Laboratory (SREL), the Savannah River Laboratory (SRL), and other administrative offices are located in the Administration Area.

Although not located in the A-Area, the TNX and CMX Semiworks were considered a part of the SRL, and were some of the first facilities to operate at SRS. CMX and TNX were code designations and had no logical derivation according to Bebbington (1990). CMX investigated problems associated with using Savannah River water for cooling, and it housed the river water pumps and a pressure facility for the testing of reactor elements. CMX was shut down in 1984. The TNX facility, one of the first to operate at the Site, provided technical support, pilot data, and personnel training. In later years, the facility was involved in waste processing research and development.

Waste Management Areas

SRS operations generated hazardous, radioactive, and mixed (radioactive and hazardous) wastes that were handled in a number of ways. Methods included the use of seepage basins for liquids, disposal pits and waste piles for solids, and burial grounds for solid radioactive wastes. Site records report that the historical disposal of waste materials occurred at over 150 individual waste sites, at over 100 areas around the plant. Over 100 of the waste sites contain nonradioactive waste materials, and 20 have been used as disposal sites for radioactive wastes. Fifteen sites have been used as disposal locations for mixed wastes (Christensen and Gordon 1983; Looney et al. 1986).

Waste management facility development was part of the initial construction phase at the F-Area and H- Area. Twelve underground carbon steel tanks were initially built to hold fission products separated from the irradiated uranium (Thomas and Robnett 1981a, 1981b). The tanks were located in reinforced concrete vaults, which had 5-ft-high steel liners. Three other series of tanks were installed in the mid-1950s (Goslen and Mcguire 1981, 1983a, 1983b). The first HLW tank was full in June 1955 (Bebbington 1990). Additional HLW tanks were constructed into the 1980s.

Two burial grounds in the separations area have received radioactive waste since 1953. Although most of the waste in the burial ground originated from onsite operations, a number of other facilities sent waste to the SRS, including

- Knolls Atomic Laboratory—the Naval Reactors Program
- Los Alamos National Laboratory
- Mound Laboratory
- Shippingport Atomic Laboratory
- Westinghouse—Bettis Atomic Laboratory
- Plutonium-contaminated debris from two U.S. military airplane accidents that occurred in other countries.

By the end of the first 5 years of SRS construction and operation, all of the basic production facilities were in operation and the products (plutonium metal and tritium gas) were being delivered. Following the division of the AEC into the Nuclear Regulatory Commission and the

Energy Research and Development Administration (ERDA), the SRS was operated for ERDA, then for the DOE.

Production Data Needs

While reconstructing facility releases during Phase II of the SRS dose reconstruction project, the CDC and *RAC* decided it was necessary to develop a detailed history of reactor and reprocessing facility production during the years of SRS operations. These data have been used to develop estimates of certain types of radionuclide and chemical releases linked to reactor and reprocessing canyon production rates, where more accurate rate data are not available. While partial information related to production was available from various sources, *RAC*'s discussions with SRS staff did not identify a complete source of data for all key facilities for all years of operation. Therefore, in 1997, we undertook an additional records review to create the following production history. While this research has been summarized in a project newsletter and during public meetings and presentations to the CDC Health Effects Subcommittee, it has not been published in full detail elsewhere.

SRS REACTOR POWER, CANYON, AND TRITIUM PRODUCTION DETAILS

Introduction

A key element of the Phase II research involves estimating releases from the SRS. Most of these releases occurred from a few major facilities onsite: the five reactors and the reprocessing and tritium production areas. Figure 2-3 maps the location of these facilities. To assist in developing an understanding of the historical operation of these key areas and to provide data necessary to estimate routine releases of radionuclides and chemicals, we developed a history of reactor and fuel processing at SRS. Detailed data in spreadsheet format are supplied with this report.

Details: Reactor and Reprocessing Area Power/Production History and Data Compilation

On December 28, 1953, the R-Reactor went critical and began producing plutonium. Scrams (unplanned reactor shutdowns) because of various problems averaged one per day for the first 2 months, one per 3 days in March 1954, and one every 15 days by June. In June, the first irradiated fuel was removed from the reactor. After a cooling period, plutonium was extracted from this fuel in the 221-F reprocessing canyon beginning in November 1954. The 221-H reprocessing canyon began operating in July 1955. Tritium was extracted from LiAl targets in a small facility in the 200-F Area beginning in October 1955 (Stetson et al. 1963).

Initial Production Levels

From 12/28/53 to 2/3/54, R-Reactor power was raised in steps to its initial rating of 375 megawatts (MW). Continuing modifications (described later in this section) increased reactor power ratings; near the end of its first fuel cycle, R-Reactor was operating at about 640 MW. By March 28, 1955, all five reactors were in operation. After initial startup and adjustments to processes and equipment, the 221-F Canyon design throughput of 3 metric tons of uranium per day was reached. The 221-H Canyon operated at 7 tons per day during this period. Figure 2-3, created by *RAC*, displays the rough circle within which the primary production facilities at SRS were located (Stetson et al. 1963).

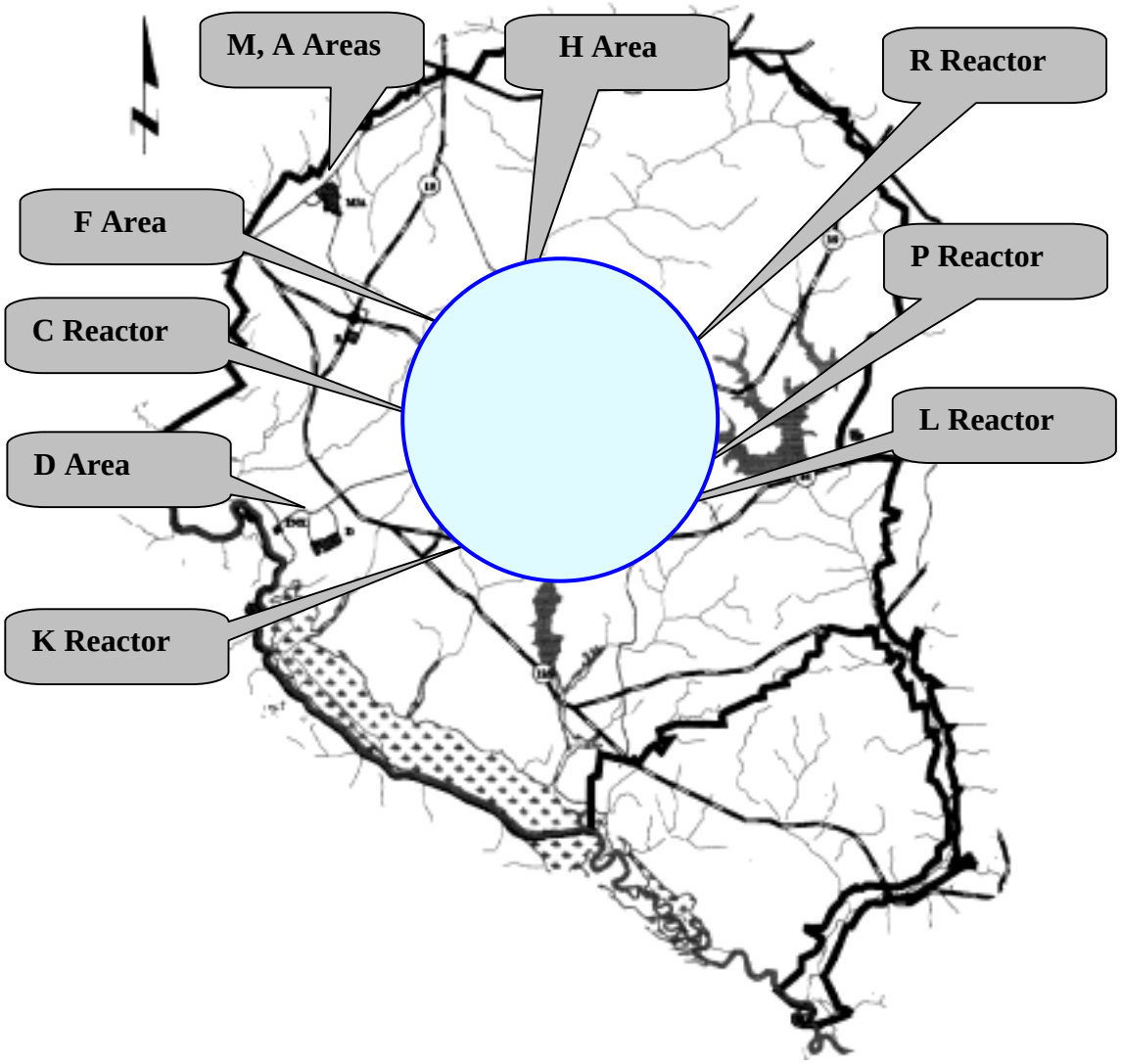


Figure 2-3. The SRS showing the reactor (C-, K-, L-, P-. and R-Reactors) and processing areas (F-Area and H-Area) in a rough circle toward the center of the Site, and D-Area and M-Area near the Site perimeter (Meyer et al. 1995).

Reactor Operations, Modifications, and Incidents

The heat output of a reactor is proportional to the rate of fission and, thus, to the creation of fission products in the fuel. Heat output is also proportional to the rate of production of ^{239}Pu and tritium for a given reactor/fuel configuration. Reactor thermal output at SRS was gradually increased beginning in 1954. In 1955, the first enriched (5% ^{235}U) uranium fuel was used to allow increased loadings of LiAl for tritium production, with LiAl rods inserted into fuel spaces (earlier only in control rod spaces). Solid enriched uranium slugs alternated with LiAl slugs in the four-element tubes (Bebbington 1990).

C-Reactor power was increased from 378 to 877 MW by December 1955. Various factors contributed to reactor increased power capacities. The allowable upper limit for fuel slug surface temperature was raised from 80 to 160ºC without harm to the cladding. Additional heat

exchangers were added as the supplier's production capacity permitted; in 1956, the number of heat exchangers for the R-, P-, L- and K-Reactors was doubled (C-Reactor was provided with 12 exchangers before startup). Reactor water flow distributions were adjusted and river water pump impellers were increased in size. A peak of 1400 MW was reached in R-Reactor by the end of 1956. Higher-capacity heavy water circulating pumps were installed in L-, K-, and C-Reactors in 1957. By the end of the year, peak power in C-Reactor had reached 2250 MW (Stetson et al. 1963; Meyer et al. 1995; Bebbington 1990).

Construction of Par Pond was completed in 1958; it received all R-Reactor cooling water. Water from the pond was pumped to R- and P-Reactors, making more river water available to L-, K-, and C- Reactors. The overall effect increased cooling water flow from 650,000 to 775,000 gpm, increasing total five reactor power by 850 MW. In 1960, larger impellers in the river pump house and the addition of three pumps at Par Pond raised cooling water flow to 900,000 gpm. This raised peak power in C-Reactor to 2575 MW (Du Pont XX)[a].

Fuel slugs evolved as well. Beginning in January 1960, hollow slugs to improve cooling capacity were being produced at SRS. Hollow slugs were used as early as 1955, produced elsewhere by Sylvania. Longer and more complex fuel elements were developed to increase power levels and neutron availability for tritium production. In 1963–1964, reactor powers were increased 100–200 MW by increasing pressure within the reactor tanks by 5 psi (Stetson et al. 1963, Du Pont XX).

In 1964, after 10 years of operation, R-Reactor was shut down; L-Reactor shutdown followed in February 1968. Both were shut down because of reduced production requirements; R-Reactor also had longstanding leakage problems. L-Reactor was later refurbished for operation. The C-Reactor peaked at 2915 MW in 1967, the maximum power level reached by an SRS reactor. This is comparable to the thermal output of a commercial electric power reactor. Figure 2-4 shows peak power levels from 1954–1989 (Bebbington 1990).

The SRS reactors operated near atmospheric pressure and the normal boiling point of water, precluding the explosive release of latent energy during an accident. Ventilation of reactor buildings was continuous, with air passing around the reactors and out to the atmosphere through tall stacks. Tritium from deuterium neutron capture and ^{41}Ar from neutron activation of natural ^{40}Ar were detected at low concentrations in the reactor building air as early as 1956 (Hall and Coombs 1956; Hoy 1962).

[a] The SRS vaults onsite contain several series of records with monthly reactor and canyon production data. These records are identified as SRS CXXIX-XX-X from 1955, as DPSP-XX-717 in 1962, and as DPSP-XX-307 beginning in 1963. Beginning in June of 1979, reactor monthly reports were deleted from the 307 series and reported separately as DPSP-XX-14-XX. This series remained classified through May 1982, then was published monthly, internally, as unclassified reports. These records have been extracted and deleted/declassified as necessary, and are in *RAC* and CDC possession offsite. They are referenced throughout this document as Du Pont XX.

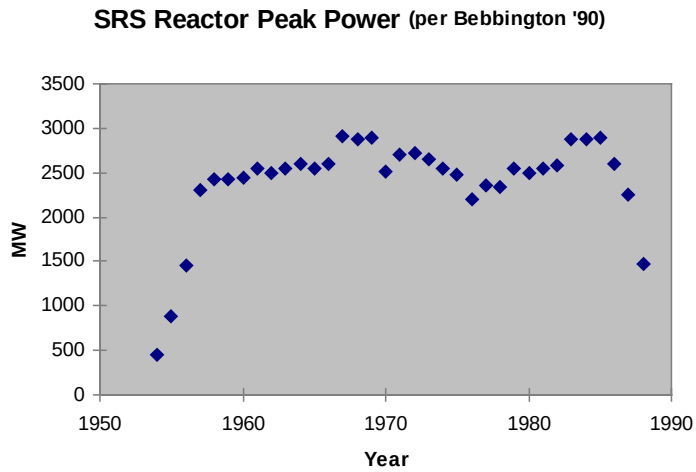


Figure 2-4. SRS reactor peak power (in MW) (Bebbington 1990).

The potential for the dropping and melting of a fuel element during fuel handling, with consequent release of volatiles such as ^{131}I, lead to installation of deluge sprays in the reactor rooms and filters and activated carbon adsorbers installed ahead of the stacks in the R-Area, P-Area, and K-Area in 1962. In 1963, similar facilities were installed in L-Area and C-Area. In the following 3 years, retention basins were constructed to contain water from the deluge systems. Later, 500,000-gal tanks were added in these basins to catch deluge runoff. These systems were never used according to Bebbington (1990).

In 1964, the first process-control computer was given a trial run in K-Area, monitoring some 3600 sensors of process variables. By the end of 1966, similar first-generation computers were installed in all four operating reactors. They scanned the sensors, compared observations with allowable limits, and presented analyses to the operators. Scram capability linked to over-temperature was added to the computer system in late 1966. Additional new controls and capabilities were added over the years (Bebbington 1990).

From 1954 to 1974, Advisory Committee on Reactor Safety reactor physicists and engineers performed 30 reviews of SRS reactor operations. In January 1960, the most serious SRS reactor incident occurred, when a rapid restart of L-Reactor (following a scram) resulted in over-temperature and the potential for fuel melting within 40 seconds if shutdown had not been performed. Rapid restarts were prohibited in all SRS reactors following that incident (Stetson et al. 1963). A 1970 incident involving a neutron source held too long without cooling after removal from K-Reactor, resulted in significant contamination inside the reactor building. Personnel dose during cleanup totaled 600 person-rem, with a maximum dose of 3.3 rem and an average dose of 0.7 rem. The reactor was down for 3 months for cleanup. In 1982, a new Reactor Safety Advisory Committee was formed.

Stress cracking of stainless steel welds at complex shape locations on the reactor vessels eventually led to leaks. The first was discovered in R-Reactor in 1960 after about 7 years of operation. This leakage problem was chronic but controlled. Only R- and C-Reactors were found upon detailed inspections to have developed serious cracks. R-Reactor cracks were related to early welding fabrication technology (largely inadequate thermal stress relief). C-Reactor cracks were associated with a curved transition section (the knuckle) with high residual stresses. Smaller

cracks discovered in C-Reactor in 1985 were caused by helium embrittlement in stainless steel; repair attempts were not successful (Bebbington 1990).

Reactor heat exchanger tube cracks and leakage appeared as early as 1956; modification of water chemistry controlled the problem by 1961. Disassembly basin operations, involving fuel elements with failed cladding, regularly resulted in fission product escape to basin water then to seepage basins. Redesigns later reduced releases, particularly of ^{137}Cs.

A 15-year program was initiated in 1956 to produce ^{233}U for the thorium breeder reactor program. Some 630 kg of ^{233}U was eventually produced at the SRS (Bebbington 1990). The ^{232}U daughter high-energy gamma radiation eventually made the program unattractive.

About 66 million curies of ^{60}Co was deliberately produced, beginning in 1955, for potential use in food sterilization programs. Some 275 kg of ^{238}Pu was produced from 1959–1978 for various heat source and related programs. Other production programs, including creation and extraction of ^{252}Cf during the period 1965–1970, were also conducted at the SRS (Stetson et al. 1963; Du Pont XX). During this period, SRS produced a variety of radionuclides ranging from tritium to ^{252}Cf in a series of reactor campaigns. The program to produce ^{252}Cf, which was used mainly for research purposes, required the modification of the reactor core because the heat generated per foot of fuel element was about three times normal. During these high-flux operations, over 250 target capsules, comprised of 150 nuclides of 66 chemical elements from nine research laboratories for Oak Ridge National Laboratory, Brookhaven National Laboratory, the University of Illinois, and Denmark, were irradiated (Meyer et al. 1995).

Determining Reactor Power and Canyon Production Levels

During Phase II of the Dose Reconstruction Project, researchers determined that detailed reactor and canyon operational data would be useful to estimate radionuclide and chemical release data for periods when measurements were unavailable. Monthly F- and H-Canyon input and production data were not available from SRS staff in summary form, and monthly reactor power data summaries were not found during an extended search by *RAC* and Site staff. *RAC* then examined the detailed monthly production summaries to collect these data. Tritium production data remains classified, but *RAC* and CDC were able to reach a compromise with SRS and DOE staff allowing the declassification of an adequately detailed history of tritium production, discussed below. *RAC* and CDC research to access the reactor power and canyon production data included the following steps:

- A Search of the *RAC/CDC* Phase I document database for records of potential value
- Discussions with SRS staff to identify potential data sources
- An SRS visit in April 1997 to inspect records, conduct interviews, and search for data
- Conferences with SRS declassifiers to format data to allow release to the public domain
- Extraction of key data from classified and unclassified records
- Declassification review by SRS staff
- Conversion of declassified data to spreadsheet format
- Quality assurance review; graphical analysis of results
- Additional Site visits to locate more records
- *RAC* report preparation, including internal and external reviews.

Searching the Phase I Document Database

During Phase I of the SRS Dose Reconstruction, *RAC* and CDC devoted a great deal of effort to a hands-on review of records stored onsite and offsite. To document this record location and review, *RAC* developed an electronic database to track boxes and individual documents as they were reviewed and to categorize their likely value to the study. Documents determined to be potentially useful to the study were copied and sent through the SRS classification review process. The declassified or unclassified records and the *RAC* database describing their contents are available at the University of South Carolina public reading room in Aiken.

The Phase I database reflects the results of our review of more than 40,000 boxes of records, including material found in all formal repositories located onsite. The search for these records was comprehensive, involving hundreds of interviews and many dozens of Site visits.

To locate Phase I database records related to Site production levels, we searched the Phase I database for the words “reactor,” “power,” “production,” “canyon,” “tritium,” and “plutonium.” Additional keywords were applied based on the results of the initial search. The resulting sets of document descriptions were printed for use during the initial Site visit. Approximately 200 documents of potential interest were identified via this search.

Discussions with SRS Staff

Dr. Robert Meyer of *RAC* contacted Mr. Greg Peterson, the SRS onsite coordinator for *RAC*’s research, and asked for assistance in identifying historical reports on reactor and canyon production levels. Other SRS staff members were contacted to determine whether existing summaries of reactor and canyon production levels were available in sufficient detail. No detailed summary documents were found during these discussions, and *RAC* proceeded to review detailed classified and unclassified records in Buildings 773A and 773-52A to create such a summary, published here.

Site Visit - April 13–18, 1997

RAC spent the week onsite, initially searching for records and interviewing SRS staff, then focusing on extracting data from the pertinent records. A key *RAC* Category 1 record was located during research in the 773A classified vault. This document led to a series providing monthly reactor and canyon production data of adequate detail. These records are identified as SRS CXXIX-XX-X in the earlier years, as DPSP-XX-717 in 1962, and as DPSP-XX-307 beginning in 1963. Each classified monthly summary consists of 20 or more pages detailing the canyon, tritium area, and reactor production record for the month. A great deal of production information is available in these reports; only a subset was necessary for the purposes of this specific research project. Data sheets were used to collect that subset, and about 250 pages of data were extracted from the reports during the week. Copies of all document pages used during creation of this SRS production history were declassified as necessary by SRS staff, are now held offsite by *RAC*/CDC, and are available for public review.

In June 1979, the classified report series previously containing both reactor and canyon data was converted by SRS to a classified, canyon-only production summary. The reactor power summaries were continued as separate, unclassified monthly reports. Reactor power data for the

period April 1982 through the end of 1987 were located during the summer of 1997 and are presented in this chapter. *RAC* continued to search for monthly reactor power data for the period June 1979 to March 1982, eventually discovering the final data sets under a new report title. All necessary data, with minor exceptions, were eventually located during this research.

Tritium Data Declassification

After several discussions, SRS staff determined that all needed data could be declassified, with the exception of tritium exact production data. To circumvent this remaining problem, tritium production data were sorted by *RAC* into 15 "bins," consisting of ranges of tritium monthly production rates. This approach obscures the data, allowing Site declassifiers to treat the results as unclassified, while maintaining sufficient accuracy for the purposes of dose reconstruction. The results of this conversion are reported here as "estimated" tritium production data. As is the case with all records of value to the dose reconstruction, the original monthly report sections were submitted by *RAC* to the SRS declassifiers and were eventually released to the public domain as "deleted versions" (with classified information removed).

Extraction of Key Data

The monthly SRS DPSP reports contain estimates of future production, actual reactor power and production data, and information related to specific campaigns at SRS to produce other products, including ^{233}U, ^{238}Pu, ^{252}Cf, and ^{60}Co. Most of this information was not needed for our purposes. For the records period January 1955 through October 1967, *RAC* extracted peak power, average power, megawatt-days per month, estimated ^{239}Pu production, estimated tritium production, and heavy water accumulated losses for each reactor. For the same period, *RAC* extracted F-Canyon and H-Canyon uranium-to-dissolver, tritium, and plutonium production. For H-Canyon after mid-1959, "U235 tubes to dissolver" data (enriched uranium fuel element data), were extracted. These fuel elements were used to enhance reactor tritium production beginning in June 1959. On the data sheets, *RAC* also captured notes concerning certain campaigns, reactor heavy water inventory, error corrections, and SRS production of additional radionuclide products. For the period following October 1967, we reduced the amount of data extracted, capturing reactor megawatt-days per month, heavy water accumulated losses, uranium to dissolver, ^{239}Pu production, tritium production, and U235 tubes to dissolver. The *RAC* data sheets were declassified by SRS staff, and they are available for public review (Du Pont XX). The tritium production estimates and actual tritium production data were deleted from the *RAC* data sheets by the SRS classifiers (as agreed), but the "binned" tritium production data were cleared to the public domain and are reported within this chapter. These data have not been previously released.

Transcription of Declassified Data to Spreadsheet Format: QA Review

RAC received the declassified data sheets after our Site visits, and we transcribed power and production data into an Excel® spreadsheet. The data entries were reviewed and corrected during a second pass through the data sheets. (Time onsite did not permit similar review of the data extracted during the Site visit.) These data are available offsite now, in the declassified records held by *RAC*/CDC. The plotted data were inspected for outliers and trends. A change in trends

observed for the heavy water accumulated releases after July 1974 indicated the need to correct for a units change from pounds to kilograms at that point. Later data retrieved from SRS validated this correction.

Report Preparation and Review

A *RAC* Technical Memo detailing the power and production study was transmitted for review by additional *RAC* researchers and to evaluate the adequacy of the data collected. Several report versions were prepared over time to incorporate reviewer comments and to add late-discovered reactor power data sets. *RAC* asked SRS declassifiers to additionally review the potentially sensitive information. Results of the power/production study were presented to the CDC's SRS citizens' Health Effects Subcommittee in August 1997; they were updated at a later meeting.

Reactor Power Output

The following graphs reflect monthly reactor power output data for the five SRS reactors. Reactor power for each month is presented below in thousands of megawatt-days. This is a measure of the reactor's thermal power output daily average during the month, in millions of watts, times the number of days in the month. A reactor averaging 2000 MW thermal power output for 30 days would produce 60,000 megawatt-days (MWd) of power in that month. Thermal power levels reached by SRS production reactors are similar to those reached by commercial electric power reactors. On the following plots, zero power for a month means that the reactor was not operating for that month, except for March and April 1985. We could not locate data reports for that 2-month period. P-, K-, and C-Reactors were operational at that time. Detailed data supporting the following graphs are available in an associated spreadsheet.

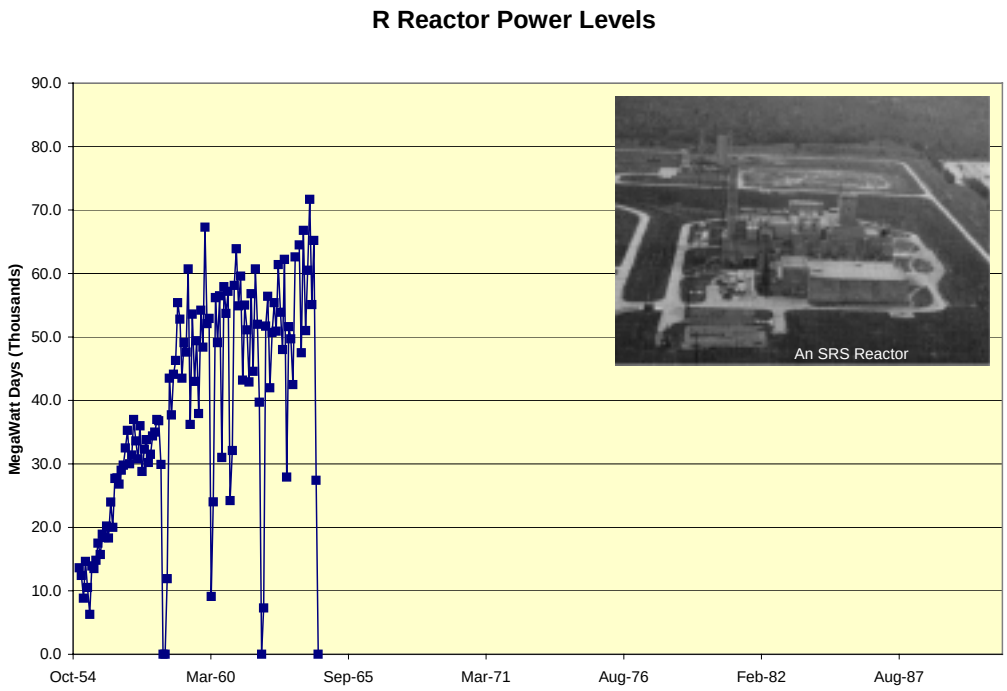


Figure 2-5. R-Reactor operated from December 1953 to June 1964, initially reaching 375 MW thermal power. Modifications increased power levels over time. R-Reactor reached its maximum of 71,700 MWd in March 1964. The reactor developed significant vessel cracks, discovered in mid-1960 (Du Pont XX).

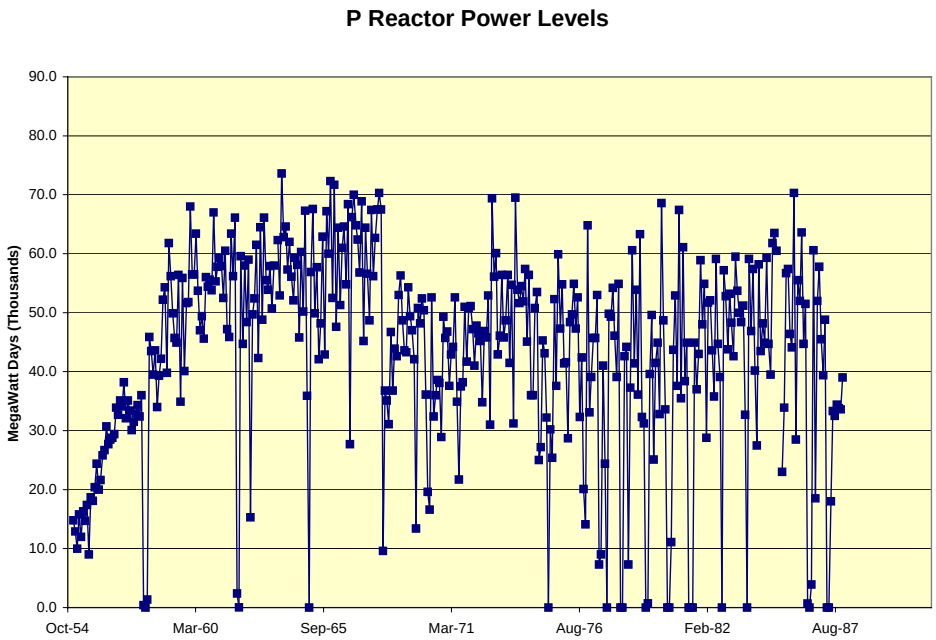


Figure 2-6. P-Reactor operated for nearly 35 years, reaching its maximum monthly power output of 73,600 MWd in December 1963 (Du Pont XX).

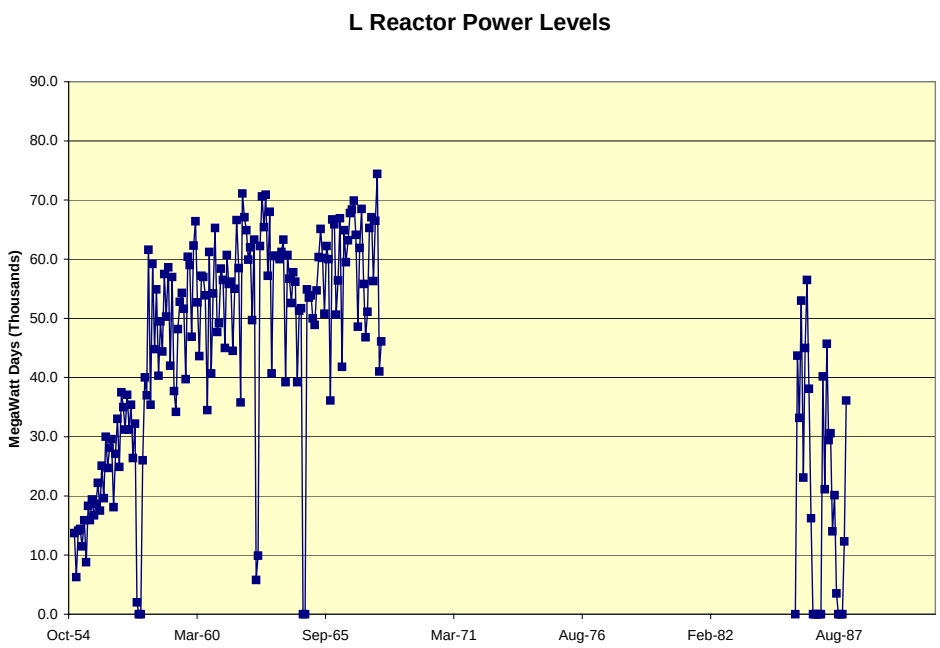


Figure 2-7. L-Reactor operated from 1954 through February 1968, reaching its maximum monthly output of 74,400 MWd in December 1967. It operated again from November 1985 through December 1987. A January 1960 rapid restart lead to a serious incident in which fuel temperatures could have reached the melting point. SRS procedures were modified to prohibit rapid restarts as a result of that incident (Du Pont XX).

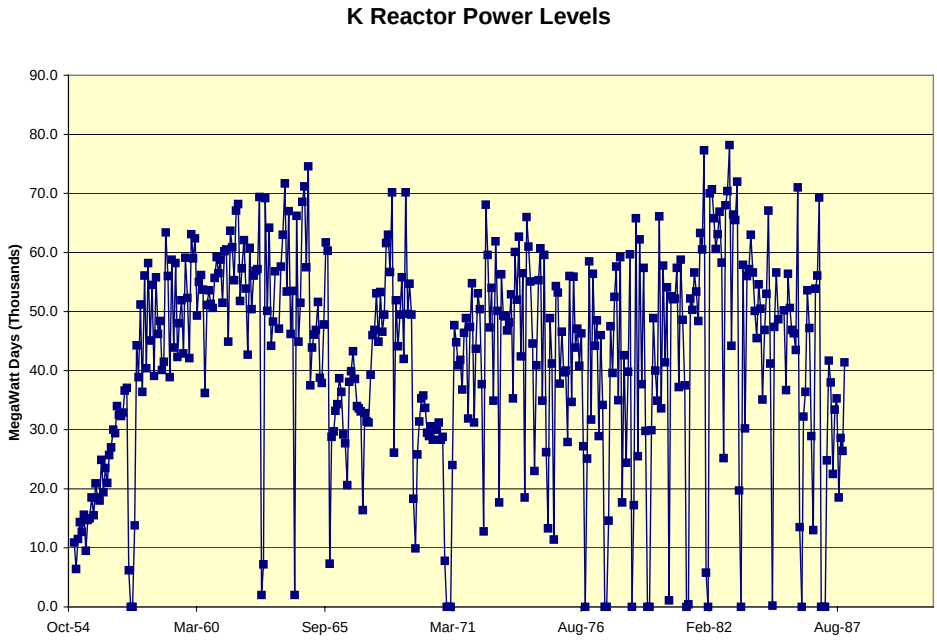


Figure 2-8. K Reactor operated for more than 30 years, reaching its peak monthly power output of 78,200 MWd in January 1983 (Du Pont XX).

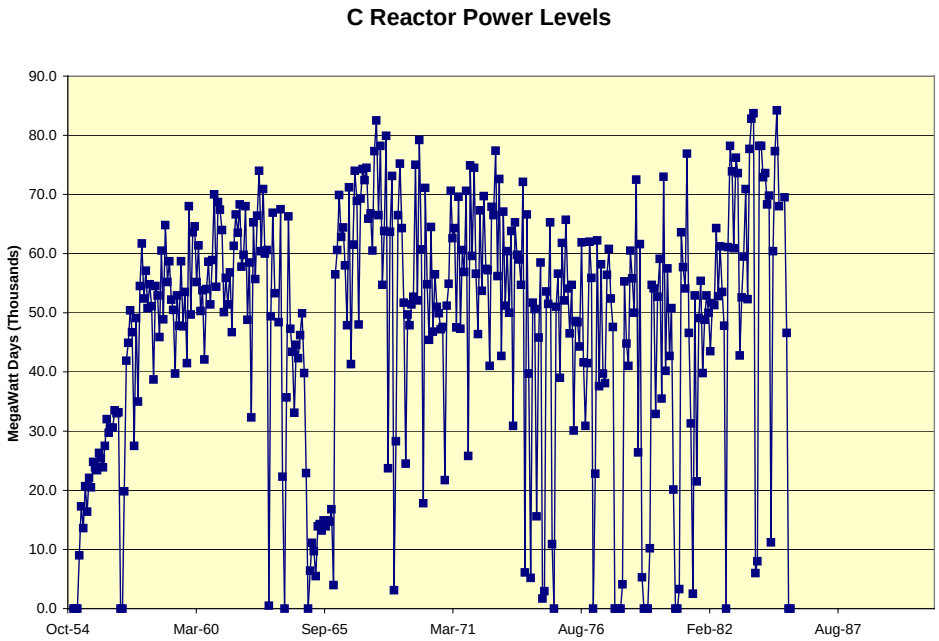


Figure 2-9. C-Reactor also operated for more than 30 years, reaching a peak monthly power output of 84,200 MWd in January 1985. Inspections in the early 1960s indicated cracking in a highly stressed section of the vessel. In 1985, additional cracks caused by helium embrittlement were found (Du Pont XX).

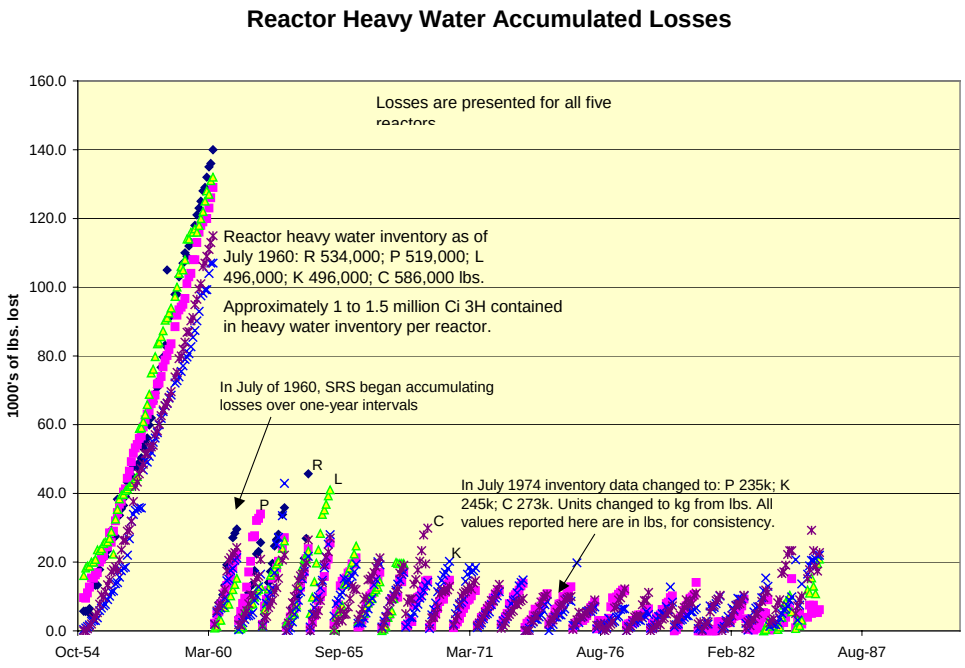


Figure 2-10. Because reactor heavy water moderator was an expensive SRS product, its loss rate was monitored routinely. Tritium (as tritiated water) was present in the moderator and was released offsite via moderator leakage. Routine tritium loss from the reactors may be estimated using records of moderator loss (Du Pont XX).

Canyon Processes, Modifications, and Incidents

Bebbington (1990) notes that early in 1955 efforts were concentrated on increasing F-Canyon throughput to work off the accumulation of irradiated fuel in the reactor cooling basins. Throughput increased from 1.6 tons uranium per day at the beginning of 1955 to 4 tons per day by May. The cooling time of stored fuel had by then been reduced to 118 days. The throughput increase was managed by increasing the amount of uranium in solutions entering solvent extraction and increasing the tributyl phosphate concentration in the solvent. Dissolver batch size was increased from 5.9 to 9.4 tons uranium. Less than 0.2% plutonium and 0.1% uranium were being lost to HLW. The first carbon steel, underground HLW tank was full in June 1955. Following this event, reagent concentrations were more carefully controlled, reducing the amounts sent to HLW by 22%. By the end of 1956, H-Canyon was up to 6.6 tons per day input and F-Canyon to 4.5 tons per day. Rates were then limited by the available quantity of adequately cooled fuel. By the end of 1956, a second tritium production line in 232-H was operating.

At the end of February 1957, F-Canyon was shut down for enhancements. New mixer-settlers as large as could fit into the canyon cells replaced the existing units. After beginning tritium recovery in 232-F in October 1955, in July 1957, a larger tritium facility began operation in 232-H. Uranium processing was moved to the H-Area where 8 tons per day throughput was soon attained. The 232-H capacity was doubled in 1958. In 1958, plutonium demand slumped and H-Canyon operation dropped to three shifts, 5 days per week. Hollow (Mark VII) fuel slugs dissolved much more rapidly but with some increase in waste (Meyer 1995; Wahl 1967).

Also in 1957, SRS began compressing tritium into steel reservoirs, components of enhanced nuclear weapons. The 234-H facility began operation in August 1957 to produce these filled (high-pressure) tritium reservoirs. These were regularly returned to SRS over the years to extract ingrown ^{3}He, the tritium decay product. Equipment to separate ^{3}He was installed in 234-H in August 1958. Reservoir recycling was added subsequently in a new building, 238-H.

Two of the three large heavy water plants in D-Area were shut down and scrapped in 1957–1958. Adequate supplies of heavy water were in place by then, and D-Area reconcentration equipment was used to recover from dilution of reactor moderator (Bebbington 1990; Wahl 1967).

In August 1958, tritium processing began in 232-H-2 (plus existing processing in 232-F and 232-H). In October 1958, the F-Area tritium facility was shut down permanently.

A new plutonium-finishing facility, the JB line, was built on the roof of 221-F in the late 1950s. In March 1959, Purex operations resumed with hollow slugs as feed. Process capacity in 221-F was then 14 tons of uranium per day. Plutonium processing in H-Area then ceased; Building 221-H was shut down for 3 months in 1959 and converted to process the enriched uranium tubes (U-235 tubes; the HM process) used to enhance tritium production. That operation began in May with a throughput of 25–53 fuel tubes per day. During the preceding 3 years, these tubes had been processed at the Idaho Chemical Processing Plant for uranium recovery. The 221-H dissolver was extended to handle long fuel tubes, and neutron-sensing instruments were added to monitor ^{235}U concentrations to prevent criticality. Processing and upgrading were done remotely using the canyon cranes. Operations in H-Canyon were suspended from May to October 1960, limited by irradiated fuel availability. From 1961–1965, F- and H-Canyons were operated alternately (Du Pont XX).

In 1959, the high-neutron-flux potential of SRS reactors led to the production of high atomic weight research and industrial materials, including ^{252}Cf and isotopes of curium and berkelium. Stepwise, recirculating ion exchange processes were developed to first produce precursors of these high atomic weight particles and then final products. Solvent extraction was eventually added to this process. About 2 g of ^{252}Cf was eventually produced in the SRS reactors. Both ^{244}Cm (a heat source) and ^{241}Am were produced/extracted at SRS, with the major production activity in the 1970s.

The Multipurpose Processing Facility (MPPF) was completed in 1972, but it was not operated until 1978 when it converted a stock of ^{241}Am to oxide for shipment to Oak Ridge National Laboratory. SRS produced americium/beryllium neutron sources, and some ^{241}Am was used in smoke detectors produced elsewhere. Some ^{252}Cf was used in well logging, industrial radiography, and activation analyses. About 2 g ^{252}Cf still in its target elements was eventually shipped to Oak Ridge National Laboratory for storage (Bebbington 1990).

Beginning in 1964, SRP increasingly reprocessed uranium fuels from experimental power reactors and research reactors at universities and AEC laboratories. Early Candu (Canadian power reactor) fuels were included in this program. The RBOF was built to inspect and prepare these variously encapsulated fuels. An electrolytic dissolver (enhancing corrosion/dissolution rates) was built to deal with stainless-steel-clad offsite fuels. This dissolver operated through 1979.

Neptunium-237 was purified and converted to ^{238}Pu in SRS reactors. Recovery of neptunium began in 1960, first by ion exchange but later, in 221-H, by solvent extraction. Neptunium was purified and converted to oxide in the HB line, which was no longer being used for plutonium finishing. To enhance ^{238}Pu production, uranyl nitrate solution returned to Oak Ridge for reenrichment in ^{235}U was kept segregated for return to SRP so that the concentration of ^{236}U built up (^{236}U is part of the ^{237}Np creation chain). Building 235-F, never used for production of weapons shapes, was converted in 1961 to producing neptunium oxide-aluminum target slugs for irradiation. Building 235 was modified further to produce neptunium oxide-aluminum billets for extrusion in Building 321-M. The 321-M Building extruded powders into target tubes later. The first separation of ^{238}Pu was done in the SRP High Level Caves. The process was then scaled up and transferred to the 221-H-Canyon. The Plutonium Fuel Facility (PUFF) was constructed in Building 235-F in 1978 to produce ^{238}Pu oxide spheres, about 1.5-in. diameter, sealed in iridium shells. These each were capable of producing 7 W electrical. Several hundred spheres were produced between the late 1970s and the mid-1980s (Bebbington 1990; Meyer 1995).

The first of several Thorex campaigns was run in 221-H beginning in late 1964. This work continued intermittently through 1969. Some of the ^{233}U produced was used in the Navy's Shippingport, Connecticut, experimental light-water breeder reactor.

Solvent deterioration lead to the need to separate solvent purification stages early in the canyons' processing years. First stage solvent eventually deteriorated to the point that it was burned in an open pan in the low-activity waste burial ground. A great deal of smoke was produced, but the process "did not release significant radioactivity to the air" (Bebbington 1990, p. 120). Changing from ultrasene (highly refined kerosene) to the more stable hydrocarbon adakane in 1962 ended most of the solvent degradation problems. Contact time was reduced by developing a centrifugal contactor in F-Canyon—this reduced solvent damage and startup and shutdown time.

One of the "more serious" F-Canyon incidents occurred in 1960. About 5000 Ci of "radioactivity" (Bebbington 1990) escaped from the HLW evaporator and flooded down a

stairwell and through normally nonradioactive sections of the building. Contamination in the corridor was reduced to operable levels in about 3 months; some lower-level spaces were out of service for several years.

In 1969, after 15 years of operation, the H-Canyon sand filters failed because of concrete corrosion by acidic vapors. New sand filters were installed in H-Area and F-Area, this time with steel support structures under the filter beds to reduce further collapse. In 1975, a chemical explosion in the A-line of F-Area (uranyl nitrate solution processing) caused no injuries, but the facility was shut down for about 6 months. Refer to the dose reconstruction Phase I report for additional details concerning the reprocessing canyons (Meyer 1995). Detailed data supporting the following graphs are available in an associated spreadsheet.

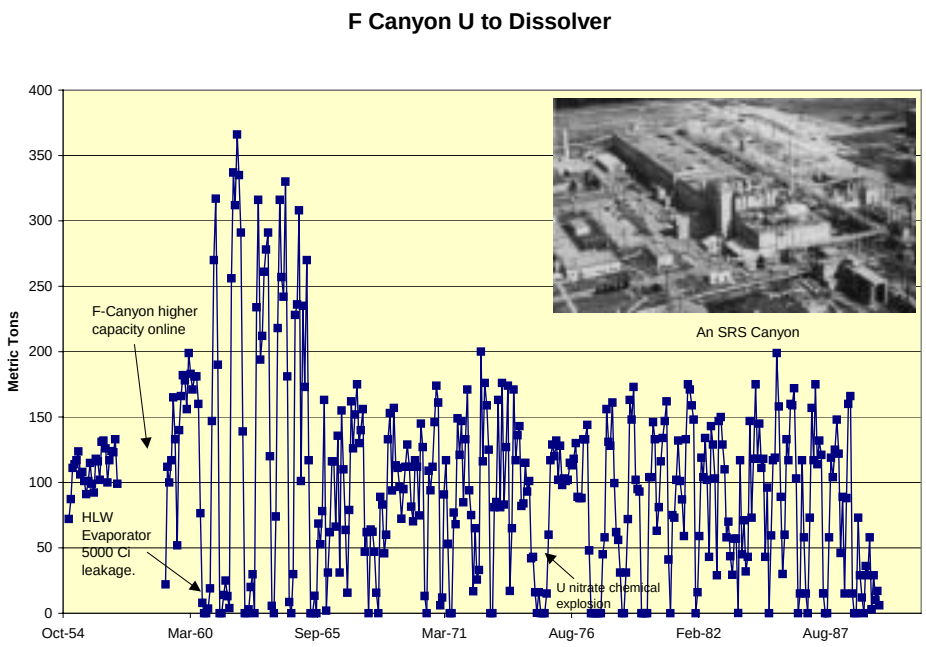


Figure 2-11. F-Canyon uranium to dissolver. The first plutonium extraction at SRS took place in November 1954 in the F-Canyon. The canyon reached its peak capacity in April 1962. A leak from a HLW waste evaporator in mid-1960 caused internal flooding and contamination (Du Pont XX).

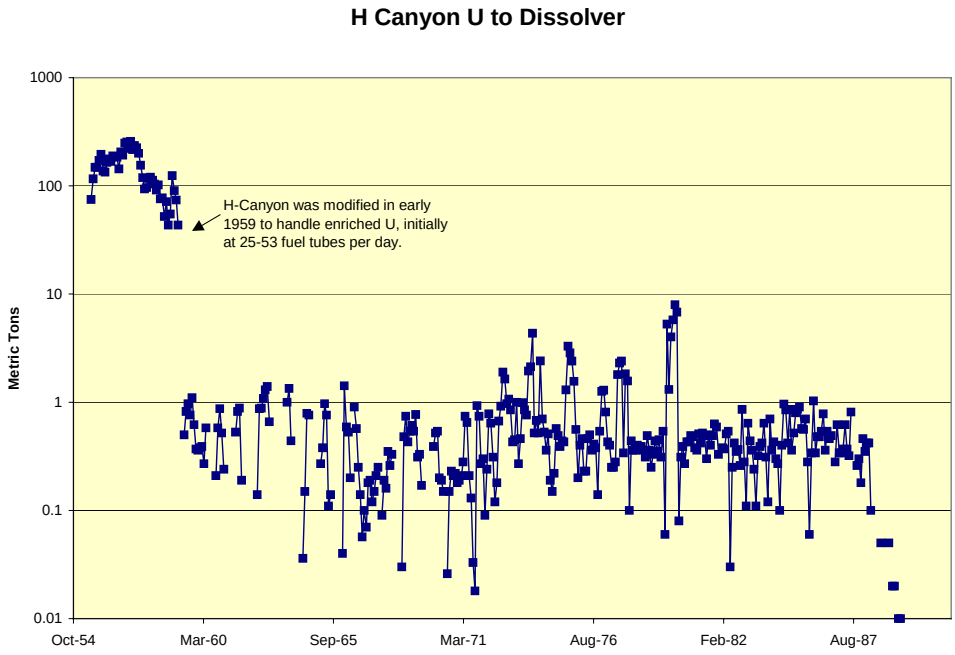


Figure 2-12. H-Canyon uranium to dissolver. In later years, H-Canyon throughput was reported both in metric tons of heavy metal (uranium) and as the number of (enriched) ^{235}U tubes charged to the dissolvers (Du Pont XX).

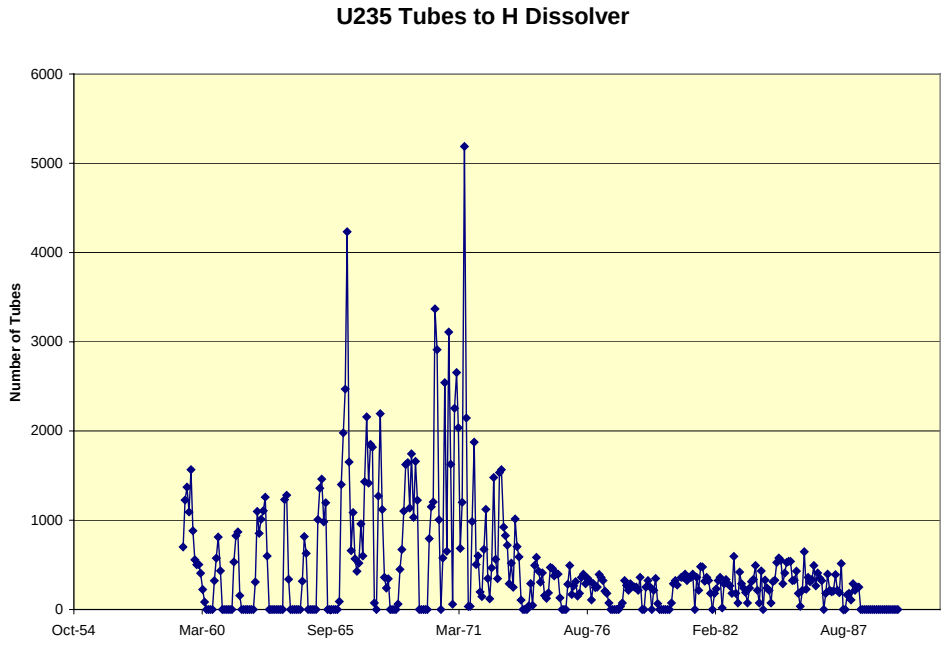


Figure 2-13. Uranium-235 tubes to the H-Canyon dissolver. Uranium-235 tube input peaked in June 1971 (Du Pont XX).

Estimated Tritium Production

Exact or "accurate" levels of tritium production at SRS remain classified. For purposes of dose reconstruction, ranges of production levels by month are adequate to estimate routine releases. The tritium production data were sorted into ranges as shown in Table 2-2, allowing presentation of the data in an unclassified document. The tritium production graph (Figure 2-14) plots these sorted data. These binned data are called "estimated" values here. Detailed data supporting the following graphs are available in an associated spreadsheet.

Table 2-2. Tritium Production Rate Data Sorted into Bins (Du Pont XX).

Production rate range (g mo^{-1})	Bin number	Average of range (g mo^{-1})
0—50	1	25
50—100	2	75
100—200	3	150
200—300	4	250
300—400	5	350
400—500	6	450
500—700	7	600
700—900	8	800
900—1100	9	1000
1100–1400	10	1250
1400–1700	11	1550
1700–2000	12	1850
2000–2300	13	2150
2300–2600	14	2450
2600–2900	15	2750

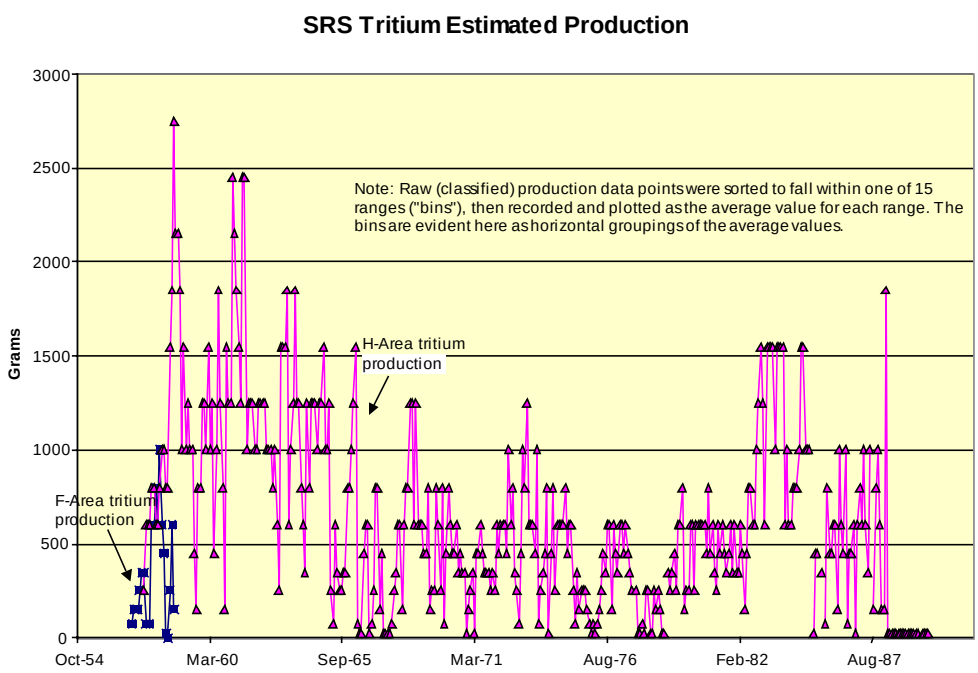


Figure 2-14. SRS estimated tritium production (Du Pont XX).

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CHAPTER 3

SELECTION OF KEY RADIONUCLIDES FOR SOURCE TERM STUDIES

ABSTRACT

This chapter describes the screening process we used to determine the key radionuclides released from the Savannah River Site (SRS) that are most important to public health. The screening process is an objective and efficient way to identify the radionuclides that are potentially important contributors to offsite radiation dose to local residents and for which detailed source term or release estimates will be done. We used the National Council on Radiation Protection and Measurements (NCRP) screening methods that have been used extensively in similar projects. The NCRP screening techniques are based on documented and well-known radiological assessment principles that combine environmental transport mechanisms, exposure pathways, and dosimetry components into a few calculational steps that require a minimum of site-specific data for the initial screening approach. This method does not eliminate any radionuclide from further consideration; rather, it helps focus resources first on those radionuclides that are most important for public health. The result of the screening process is reported as a screening value for each radionuclide. We summed the screening values and calculated the relative contribution of each radionuclide to the total screening value for all pathways of exposure. We established our screening criterion at a level of 0.1% of the total screening value. The screening method involved two steps. In the first step of screening, the results showed that the important contributors to the total screening value are somewhat different for the air and water pathways. For the air pathways, the radionuclides from the SRS contributing greater than 0.1% to the screening value for all potential pathways of exposure are ^{241}Am, ^{41}Ar, ^{14}C, ^{137}Cs, ^{3}H, ^{129}I, ^{131}I, ^{238}Pu, ^{239,240}Pu, ^{103,106}Ru, ^{89,90}Sr and uranium. For releases to surface water, the radionuclides that contributed 0.1% or more to the total screening value are ^{137}Cs, ^{60}Co, ^{3}H, ^{129}I, ^{131}I, ^{32}P, ^{238}Pu, ^{239,240}Pu, ^{89,90}Sr, ^{35}S, ^{99}Tc, uranium, ^{91}Y, ^{65}Zn, and ^{95}Zr,Nb. In the second step of screening, we further ranked the radionuclides identified in the first step by determining the relative importance of these radionuclides by exposure pathway. The percent contribution of each radionuclide to the total screening value exposure pathway for each exposure pathway was the basis for ranking the radionuclides. The radionuclides that ranked among the top three for at least two of the seven exposure pathways were selected for detailed source term development. Based on this second screening step, detailed source term estimates were developed for ^{131}I, ^{3}H, ^{41}Ar, ^{129}I, and ^{239,240}Pu released to air. For surface water pathways, detailed source terms for ^{137}Cs, ^{3}H, ^{90}Sr, ^{60}Co, ^{32}P, and ^{131}I were developed. Release estimates for uranium were also determined.

INTRODUCTION

Dose reconstruction is a staged process involving important steps where very detailed work must be done. Some initial, critical steps must be taken early in a dose reconstruction study if scientific and public credibility is to be achieved. One of these critical steps is the use of screening methods to focus later work on the important contaminants released from the facility to

the environment. Screening is a method for ranking the radionuclides and chemicals released to the environment in terms of their importance to human health. Because there are numerous materials potentially released from a facility like the SRS to the environment, screening calculations provide an objective basis for making decisions about priorities. It is important to carry out a screening process before beginning in-depth dose reconstruction because it is essential to allocate resources for and focus efforts on those that were most important to public health. For radionuclides, for example, there are differences in amounts released to air and water, half-life, behavior in the environment and biological uptake. It is impossible, under common operating constraints, to give equal attention to each radionuclide in the early stages of dose reconstruction.

The screening process helps to focus our research efforts and provides a relative ranking of the radionuclides released to air and water in terms of their importance to human health. We used the National Council on Radiation Protection and Measurements (NCRP) screening methods that have been revised and updated over the years and used extensively in similar projects. The NCRP screening techniques are based on documented and well-known radiological assessment principles that combine environmental transport mechanisms, exposure pathways, and dosimetry components into a few calculation steps that require a minimum of site-specific data for the initial screening approach. The first step in applying the screening methods to atmospheric or surface water releases from the SRS was assessing the releases of particular radionuclides during their operational history.

The output from this screening is a list of the radionuclides and their relative contribution to the screening values[1] from all pathways (or for individual pathways) of exposure to a nearby individual. We compiled the release estimates and results of the NCRP screening methodology in Excel spreadsheets. The result of the calculational steps is reported as a screening value, and provides a basis for comparing and ranking the radionuclides by their contribution to the total screening value. While the screening value is reported in units of dose (millirem or sievert), this value does not represent a "true dose" because the many conservative assumptions used in the screening analysis tend to maximize the screening value. It is reasonable to assume that the true dose under more realistic situations would be lower than the screening value.

Only Site personnel monitored effluents at the points of release onsite at the SRS. Because the majority of historic monitoring and record keeping came from the Site, we must rely on available Site records for the input release estimates for our screening analysis because onsite effluent monitoring at the points of release has been performed only by Site personnel. In addition to Site monitoring and process records, however, we can also rely on the basic chemistry and nuclear physics of the reactor and reprocessing operations the SRS. The process engineering for the separations areas and the nuclear reactors are quite well understood. Therefore, it is possible to understand the types and relative quantities of materials that might be expected from a particular process or reactor

The purpose of screening is to identify and focus resources on radionuclides released to air and water that are the most important contributors to radiation dose to those living offsite.

[1] The screening value is reported in units of dose, but does not represent a "true dose" because of many conservative assumptions used in the screening analysis for both routine and episodic releases.

operation run. For this reason, we tried to locate, compile, and evaluate the historic production and reactor power operations because these data can provide the foundation for other types of analysis during this project. This chapter describes the methods we used to focus on the key radionuclides of concern. The selection of key chemicals of concern is presented in Chapter 16.

RADIONUCLIDE RELEASE SUMMARY

During the early stages of this project, we thoroughly evaluated the process history, unplanned releases, and effluent monitoring programs, reviewed numerous documents, and conducted interviews and workshops (Meyer et al. 1995). We compiled information on radionuclides using a variety of early monthly reports and memoranda (Du Pont 1954, Du Pont 1958, Du Pont 1965, Du Pont 1966, Du Pont 1973), as well as series of reports such as *Radioactive Releases at the Savannah River Site 1954–1989 (U)*, (Ashley and Zeigler 1982, Cummins et al. 1991), annual *Audit of SRP Radioactive Waste* reports (e.g., Ashley 1962), and environmental impact statements for the reactors, which list inventory of materials generated during reactor operation (DOE 1984, DOE 1990). Recently, SRS published a series of reports describing the use and releases of key radionuclides at SRS, including tritium (Murphy et al. 1991, Murphy and Carlton 1991), radioiodine (Kantelo et al. 1993), cesium (Carlton et al. 1992a), plutonium (Carlton et al. 1992b), technetium (Carlton et al. 1993), and uranium (Evans et al. 1992). The initial focus of the document review was on radionuclides documented in SRS environmental reports (e.g. Ashley 1966; Cummins et al. 1990), periodic radionuclide release reports (e.g., Ashley and Zeigler 1974; Cummins et al. 1991), and early Site survey and monitoring reports (Albenesius 1954, Horton and Mealing 1956) described special monitoring techniques or problems that existed. While radionuclides potentially released at SRS were generally created onsite in the production reactors and processing areas, most chemicals were initially shipped to the Site from various sources.

These reports helped (1) identify those radionuclides that were generated during processing, and potentially released from the Site, and (2) provide an historical perspective on process history and radionuclide effluent monitoring at SRS. Based upon this broad range of documents, we compiled a "master list" of ~ 100 radionuclides (Table 3A-1, Addendum 3A) which had been:

- Present in SRS reactor fuel assemblies,
- Estimated to have been released, either to air or water, from the SRS,
- Measured in high-level sludge, supernate or reconstituted waste, or
- Present in low-level radioactive waste.

Several report series provided summaries of effluent data collected over the history of operations at the Site for the radionuclides in our list (e.g., Ashley 1959–1967). The Site compiled data for radioactive releases to water and to air from both routine and special monitoring at effluent sampling locations (Stephens and Ross 1984). Each year a compilation of the release data gathered from 1954 through the current year was published for the Site (e.g., Ashley and Zeigler 1982; Cummins et al. 1991). Periodically, the radiochemical separation of selected samples was used to determine quantities of a specific radionuclide of interest. For the annual radioactive release reports, releases of the long-lived radionuclides ^{60}Co, ^{90}Sr, ^{106}Ru, ^{137}Cs, and ^{147}Pm were quantified in the early years using isotopic distributions (Cummins et al. 1991). Special studies reports or historic memoranda provided information on other specific

radionuclides (Johnson 1957; Marter 1965; Smith 1965; Johnson 1968; Fowler and Simmons 1985). Understanding the way in which radionuclides were produced and released from specific processes provided information when direct effluent measurements were not made or were not available (Du Pont 1966, 1973, 1980; Fisk and Durant 1987). For example, during the 1960s and 1970s, the Site developed and used more advanced counting instrumentation that allowed for the routine detection of radionuclides at lower levels. In July 1974, the Health Physics Environmental Monitoring Group assumed responsibility for making "official release calculations for radioiodine and radioactive particulates" so that the Site would have independent release estimates made by a single group using the same techniques and equipment (McClearen 1975).

We compiled estimates for releases of radionuclides to air and water in Excel spreadsheets associated with the electronic version of this report. The spreadsheet identifies the references used in determining release estimates for the radionuclides. For release estimates for the surface water screening, we combined reported releases to streams and to seepage basins to ensure that no radionuclide of potential importance would be missed. In some cases, where releases were not reported, we assumed releases based on those reported for other years.

RADIONUCLIDE SCREENING

The relative importance of releases of radionuclides to the environment depends upon the quantities released, differences in the potential for nuclide concentration in the environment, and the relative toxicity of the radionuclides, as measured by their dose conversion factors. The method used to screen radioactive contaminants potentially released from the Site to the environment is one developed by the NCRP. It was initially developed for evaluating releases to the atmosphere (NCRP 1989) in terms of their relative importance as potential contributors to radiation dose. The methods were reviewed and revised over the years and, in 1996, the NCRP revised and expanded the methods to cover releases to surface water and to the ground (NCRP 1996). The NCRP screening methodology is a valuable tool because it provides a compilation of effective dose factors and screening factors for exposure pathways of more than 800 radionuclides and generic environmental transport parameters, including uptake, bioaccumulation, and environmental transfer factors. The information for each radionuclide is encapsulated in the total screening factor, which is the sum of committed effective doses received from inhalation; plume immersion; external irradiation from ground contamination; and ingestion of soil, vegetables, milk, or meat assumed to be locally produced during 1 year for a unit concentration of radioactivity in air. Screening factors for a radionuclide are also provided by pathway so the dominant exposure pathway for that radionuclide can be evaluated. The screening factors assume an average annual air concentration and a 30-year buildup time to account for accumulation in the environment. The resulting screening value applies to a period of 50 years following the release.

The results of the calculations are reported as screening values. Cautious or conservative calculations that overestimate the importance of radionuclides produce a ranking of radionuclides in terms of the screening value, a measure of the potential impact on local public health. While the radionuclides ranked low on the basis of the first screening calculation are not likely to be important in terms of public health, no radionuclides are completely eliminated. In a subsequent step, more realistic calculations focus on better estimates of releases and give a better measure of the priorities for the most highly ranked contaminants, based on the various exposure pathways.

The NCRP screening methodology is a valuable tool because it provides a compilation of effective dose factors and screening factors for exposure pathways for more than 800 radionuclides, along with generic environmental transport parameters, uptake and bioaccumulation factors, and environmental transfer factors. Human consumption rates and usage factors that are used in the screening methods are quite cautious or conservative and tend to overestimate the parameters used in the screening calculations. Table 3-1 provides examples of some individual usage factors in the screening and illustrates the use of conservative values. For example, the NCRP methods assume an individual consumes 20 kg y^{-1} of fish, a value that represents the 99th percentile of adult fish consumption in the U.S. Although this consumption rate is the 95th percentile for adults for freshwater fish, fishing represents an important potential exposure pathway for a portion of the SRS population. Another example of conservatism in the screening approach is milk consumption, assumed as 0.87 quarts per day, while the EPA reports that average milk consumption in the U.S. is about 0.26 quarts per day.

Table 3-1. Annual Individual Values Used in the NCRP Screening Models

Exposure pathway	Selected parameters	NCRP value
Inhalation pathway	Breathing rate	8000 m^3 y^{-1}
	Resuspension factor	2 x 10^{-8} m^{-1}
External exposure	To contaminated ground surface (Assume exposed most of the year)	8000 h y^{-1}
Ingestion pathway	Vegetable, fruits, grains (Assume root uptake and soil adhesion)	100 kg y^{-1}
	Water (Assume drinking water from area)	800 L y^{-1}, or 2.5 qt d^{-1}
	Fish (Consumption of fish flesh only)	20 kg y^{-1}
	Milk (Assume no milk is imported from other areas)	300 L y^{-1}, or 0.82 qt d^{-1}
	Soil	0.25 g d^{-1}

Screening SRS Releases: Step 1

The NCRP approach considers environmental transport mechanisms, exposure pathways, and radiation dosimetry in a few simple steps. In the first step, the concentration of the radionuclide in the environment is calculated by using environmental transport screening models with the release quantity from the facility. The environmental concentration is then multiplied by a *screening factor*(*SF*) for that particular radionuclide to obtain a *screening value (SV)* that can be compared with screening values for other radionuclides. The screening factors are based on effective dose factors. In our first screening step, we used the most cautious screening approach for both air and water pathways, which considered the combined effects of all significant potential pathways of exposure. The results of the preliminary screening done in the first step showed that the important contributors to the screening value are somewhat different for the air and water pathways. For the initial, most conservative approach to screening air releases, the atmospheric concentration, C_a, is calculated as follows:

$$C_a = fQ/V \tag{3-1}$$

where

C_a is the atmospheric concentration of the radionuclide (Bq m^{-3})
Q is the release rate from the facility (Bq s^{-1})
V is the volumetric flow rate of the exhaust vent (m^3 s^{-1})
f is the fraction of time the wind blows toward the receptor (dimensionless).

The release rate, Q, for each radionuclide is based on estimates of the amount released in a 1-year period from the facility. For the first step in the screening process, we calculated an average annual release for each radionuclide based on compilations of the release data that the Site published (e.g., Ashley and Zeigler 1982; Cummins et al. 1991). For, V , the volumetric flow rate of the exhaust vent, the default value (0.3 m^3 s^{-1}) was assumed as a conservative estimate in place of detailed knowledge of vent flow rates for all main stacks. For f, the most conservative screening approach assumes that the wind blows only 25% of the time toward the potentially exposed individual, a conservative estimate because as the frequency of wind increases, the air concentration decreases.

For the initial, most conservative approach to screening surface water releases, the radionuclide concentration, C_w, is:

$$C_w = W_o / Q_o \tag{3-2}$$

where

C_w is the radionuclide concentration in the receiving surface water (Bq m^{-3})
Q_o is the flow rate of an effluent discharge at the point of release (m^3 s^{-1})
W_o is the radionuclide release rate at the point of release (Bq s^{-1}).

After the concentration of radionuclide, i, in air or water ($C_{a\ or\ w}$) is determined for each radionuclide, , I, then the screening value (SV_i) for each radionuclide is:

$$SV_i = C_{i(a\ or\ w)} \times SF_{i(a\ or\ w)} \tag{3-3}$$

where

SV_i is the screening value for the radionuclide, I (Sv or mrem)
$C_{i(a\ or\ w)}$ is the radionuclide concentration of radionuclide, i, in the air or water (Bq m^{-3})
$SF_{i(a\ or\ w)}$ is the screening factor for the radionuclide, i, in air or water (Sv per Bq m^{-3}).

The output from the first screening step was a list of the radionuclides and their relative contribution to the screening value considering all pathways of exposure for those living nearby. We summed the screening values and calculated the relative contribution of each radionuclide to the total screening value for that year for all pathways of exposure. We established our screening criterion at a level of 0.1% of the total screening value. This criterion means that radionuclides that contributed 0.1% or more to the total screening value were considered further in the second screening step. The input release estimates and results of the NCRP screening methodology have

been compiled in the Rad-Screening workbook. In the electronic version, double-clicking on the following hyperlink provides access to this workbook: Rad-Screening.xls.

For the air pathways, the radionuclides that contribute 0.1% or higher to the total screening value are ^{241}Am, ^{41}Ar, ^{14}C, ^{137}Cs, ^{3}H, ^{129}I, ^{131}I, ^{238}Pu, ^{239,240}Pu, ^{103,106}Ru, ^{89,90}Sr and uranium. For surface water pathways, the radionuclides that contribute 0.1% of the total screening value are ^{137}Cs, ^{60}Co, ^{3}H, ^{129}I, ^{131}I, ^{32}P, ^{238}Pu, ^{239,240}Pu, ^{89}Sr, ^{90}Sr, ^{35}S, ^{99}Tc, uranium, ^{91}Y, ^{65}Zn, and ^{95}Zr,Nb. Figures 3-1 and 3-2 display the relative contribution to the screening value by each radionuclide for the air and water pathways. This first step in the NCRP screening for radionuclides released to air showed that ^{131}I and ^{3}H were the major contributors to the screening value (~50% and 35%, respectively) with ^{41}Ar contributing about 7%. Two other radionuclides released to air (^{129}I and ^{239,240}Pu) each contributed less than 5% each to the total screening value. The other radionuclides released to air that met our screening criteria of contributing at least 0.1% to the screening value (^{241}Am, ^{14}C, ^{137}Cs, ^{238}Pu, ^{103,106}Ru, ^{89,90}Sr and uranium) jointly contributed about 5% (Figure 3-1).

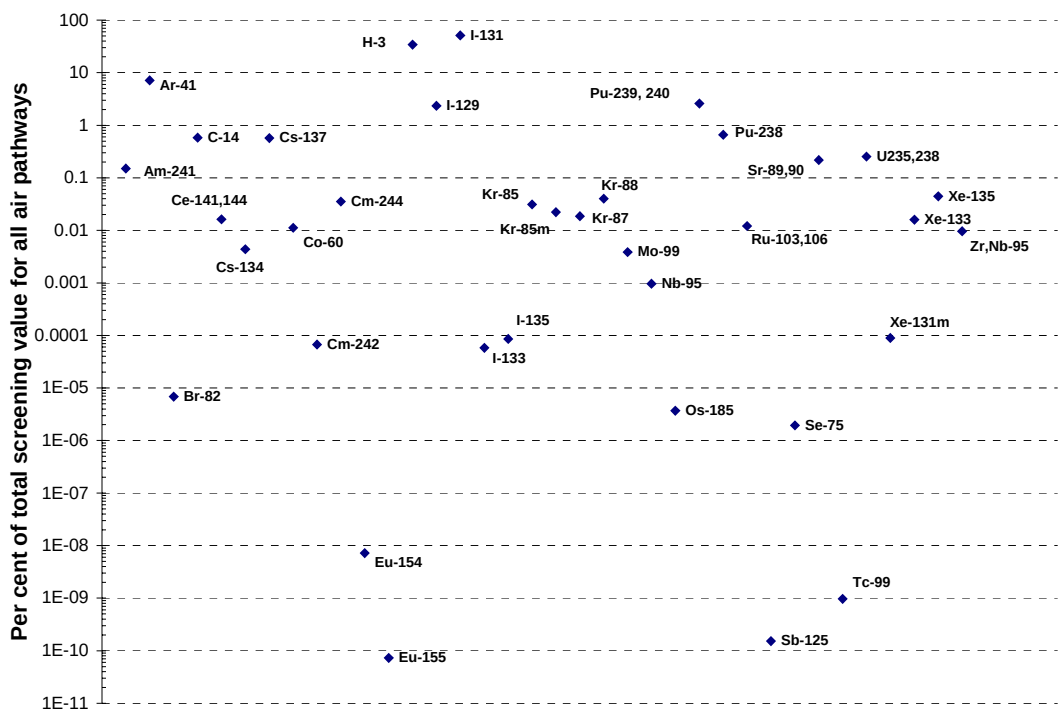

Figure 3-1. Relative importance of airborne radionuclides released from the SRS as potential contributors to the total screening value for all air pathways. For example, ^{131}I and tritium contribute about 50% and 30% of the total screening value, respectively, and ^{239,240}Pu contributes about 2% to the total screening value. The screening analysis used NCRP methods and helped to identify the radionuclides for detailed source term development.

The first step in the NCRP screening for radionuclides in surface water showed that ^{137}Cs would be a major contributor to offsite dose via the surface water pathways (~75% of the

screening value), with ^{60}Co, tritium, ^{131}I, ^{32}P, and ^{90}Sr also important contributors (jointly contributing about 20% to the screening value) (Figure 3-2). Sulfur-35 and ^{65}Zn together contributed less than 4% to the screening value via the surface water pathway, and all other radionuclides meeting our screening criteria (^{129}I, ^{239,240}Pu, ^{99}Tc, uranium, and ^{95}Zr,Nb) jointly contributed less than 2% to the screening value.

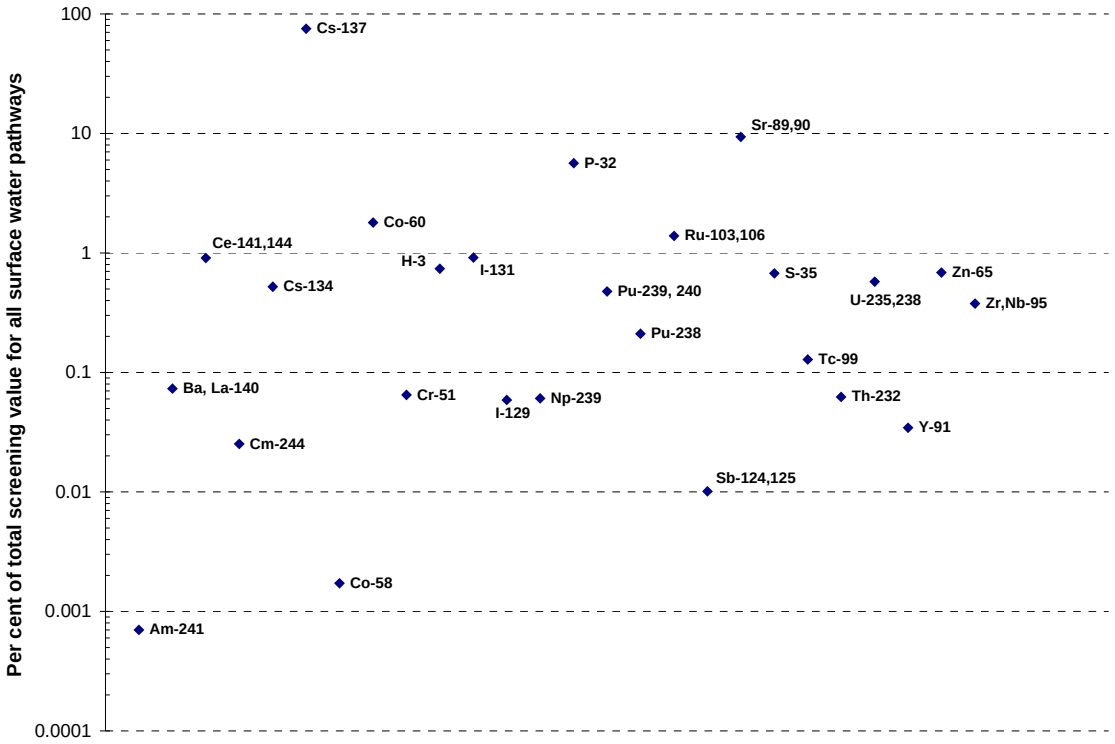

Figure 3-2. Relative importance of waterborne radionuclides released from the SRS as potential contributors to the total screening value, a measure of the potential public health impact. The results show the relative contribution to the screening value for all water pathways. For example, ^{137}Cs and ^{90}Sr contribute about 75% and 5% of the total screening value, respectively. This screening, using the NCRP methods, helped to identify the radionuclides for detailed source term development.

Screening SRS Releases: Step 2

Next, additional screening was carried out to provide a more realistic ranking of the radionuclides that were identified in the first step. This second step of screening evaluated the importance of the exposure pathway for the radionuclides meeting our screening criteria. The NCRP screening models consider the following pathways for radionuclides released to air: inhalation of contaminated air, direct radiation exposure from immersion in the airborne plume, direct radiation exposure from ground contamination and deposition of radionuclides on vegetation and ground. Radionuclides in air deposit on soil, pasture land, and vegetation and can

lead to further potential exposure pathways: ingestion of contaminated vegetables, and pasture and soil consumption by cows lead to ingestion of contaminated milk and meat. From releases of radionuclides in liquid effluent, two modes of radiation exposures can occur: external and internal. External exposure can occur from ground contamination, shoreline activities or swimming. Internal exposures can occur when the radionuclides are ingested into the body, either directly in water, or indirectly through ingestion of fish, agricultural or garden produce, animal products or through inadvertent ingestion of soil. River or stream water used for irrigation or for watering animals may introduce radionuclides into food crops and animal products such as milk or eggs that people ultimately consume. For surface water pathways, the NCRP screening includes ingestion of water, vegetables, fish, milk, meat, soil, and the contribution from both swimming and boating, and ground irradiation from garden soil and shoreline deposits.

Exposure pathways are the ways by which humans are exposed to radionuclides or other toxic substances. The key exposure pathways are air and water, with most exposures via inhalation, drinking water, ingestion of crops and other foods, and by direct radiation. The relative importance of a pathway depends on the particular radionuclide and how it was released from the facility.

In the second step of screening, we further ranked the radionuclides identified in the first step (those contributing greater than 0.1% to the total screening value) (Figures 3-1 and 3-2) by determining the relative importance of these radionuclides by exposure pathway. The percent contribution of each radionuclide to the total screening value for each exposure pathway was the basis for ranking the radionuclides. The radionuclides that ranked among the top three for at least two of the seven exposure pathways were selected for detailed source term development. For the air pathway, those radionuclides are ^{241}Am, ^{41}Ar, ^{14}C, ^{137}Cs, ^{3}H, ^{129}I, ^{131}I, ^{238}Pu, ^{239,240}Pu, ^{103,106}Ru, ^{89,90}Sr and uranium. For the water pathway, those radionuclides are ^{137}Cs, ^{60}Co, ^{3}H, ^{129}I, ^{131}I, ^{32}P, ^{238}Pu, ^{239,240}Pu, ^{89}Sr, ^{90}Sr, ^{35}S, ^{99}Tc, uranium, ^{91}Y, ^{65}Zn, and ^{95}Zr,Nb. For the air pathways, exposure could occur through inhalation, immersion in the plume of radioactivity, and eating produce, drinking milk from local dairy cows, or eating meat after deposition of radionuclides on vegetation and ground.

The results of the second step screening calculations are summarized in Table 3A-2 and Table 3A-3 in Addendum 3A for the air and water pathways, respectively. Tables 3-2 and 3-3 summarize the results of that assessment for air and water, respectively, and identify the radionuclides that ranked among the top three for at least two of the seven exposure pathways. Detailed source term development was undertaken for those radionuclides, shown in bold in the first column of each table. For radionuclides released to air from the SRS, the screening shows that ^{3}H and ^{131}I are ranked high in at least 5 of the 7 exposure pathways. Argon-41 contributes almost 100% of the screening value from the plume immersion pathway. Based on this second screening step, detailed source term estimates were developed for ^{131}I, ^{3}H, ^{41}Ar, ^{129}I, ^{239,240}Pu and uranium released to air. More detailed assessments of release estimates for ^{3}H, ^{131}I, ^{129}I ^{41}Ar, plutonium, and uranium can be found in Chapters 4.1, 4.2, 4.3, and 4.4.

Screening shows that tritium and iodine are the major contributors to dose from all airborne pathways.

Table 3-2. Results of Screening Radionuclides by Air Pathways[a]

Radionuclide[b]	All Pathways	Inhalation	Ingestion			Ground contam.	Plume immersion	No. of pathways[c]
			Milk	Produce	Meat			
^{241}Am								0
^{41}Ar	✓						✓	2
^{14}C								0
^{137}Cs						✓		1
^{3}H	✓	✓	✓	✓	✓			5
^{129}I	✓		✓		✓			3
^{131}I	✓	✓	✓	✓	✓	✓		6
^{239,240}Pu	✓	✓		✓				3
^{238}Pu		✓						1
^{103,106}Ru						✓		1
^{89,90}Sr								0
uranium						✓		1[d]

[a] Results of the second step in the screening process; the radionuclides that ranked among the top for at least two of the seven exposure pathways were selected for detailed source term development. See Table 3A-2 for details.

[b] The radionuclides identified in the first screening step that contribute greater than 0.1% to the screening value for all potential pathways of exposure.

[c] The number of times a particular radionuclide ranked high in at least two air exposure pathways. These radionuclides are shown in bold and source terms were developed for them (see Chapter 4).

[d] Uranium was included in the final source term development because of its concentrated use in the M-Area and its potential chemical toxicity (see Chapter 16). Understanding the kidney toxicity of uranium may be an important step in future SRS studies if health risks are calculated

For surface water pathways, ^{137}Cs dominates all pathways except the ingestion of drinking water, where tritium is equally important. Strontium-90 also emerges as an important radionuclide for exposure through surface water pathways. Phosphorus-32 is important when the ingestion of fish is a key exposure pathway, while ^{131}I should be considered when the ingestion of drinking water and meat are major pathways of exposure. When ground contamination is a major exposure pathway, then ^{60}Co and uranium should be considered. More detailed assessments of release estimates for ^{3}H, ^{137}Cs, ^{90}Sr, ^{60}Co, ^{32}P, ^{131}I, and uranium can be found in Chapter 5, *Releases Of Radionuclides To Surface Water*. Uranium was included in the final source term development because of its potential chemical toxicity (see Chapter 16). Understanding the kidney toxicity of uranium may be an important step in future SRS studies if health risks are calculated.

Table 3-3. Results of Screening Radionuclides by Surface Water Pathways[a]

Radionuclide[b]	All Pathways	Ingestion					Ground contamination	No. of pathways[c]
		Water	Fish	Milk	Produce	Meat		
^{137}Cs	✓	✓	✓	✓	✓	✓	✓	7
^{60}Co	✓						✓	2
^{3}H	✓	✓						2
^{129}I								0
^{131}I	✓			✓				2
^{32}P	✓	✓	✓					3
^{239,240}Pu								0
^{238}Pu								0
^{89,90}Sr	✓	✓	✓	✓	✓	✓		6
^{35}S						✓		1
^{99}Tc					✓			1
uranium							✓	1[d]
^{91}Y								0
^{65}Zn	✓							1
^{95}Zr,Nb								0

[a] Results of the second step in the screening process; the radionuclides that ranked among the top for at least two of the seven exposure pathways were selected for detailed source term development. See Table 3A-2 for details.

[b] The radionuclides identified in the first screening step that contribute greater than 0.1% to the screening value for all potential pathways of exposure.

[c] The number of times a particular radionuclide ranked high in at least two surface water exposure pathways. These radionuclides are shown in bold and source terms were developed for them (see Chapter 5).

[d] Uranium was included in the final source term development because of its concentrated use in the M-Area and its potential chemical toxicity (see Chapter 16). Understanding the kidney toxicity of uranium may be an important step in future SRS studies if health risks are calculated

SUMMARY

This chapter describes the screening process we used to focus our efforts and resources most effectively on the radionuclides important to public health. The first step in the NCRP screening process showed that the radionuclides released to air and contributing greater than 0.1% to the total screening value are ^{241}Am, ^{41}Ar, ^{14}C, ^{137}Cs, ^{3}H, ^{129}I, ^{131}I, ^{238}Pu, ^{239,240}Pu, ^{103,106}Ru, ^{89,90}Sr and uranium. For releases to surface water, the important contributors emerging from the first screening step are ^{137}Cs, ^{60}Co, ^{3}H, ^{129}I, ^{131}I, ^{32}P, ^{238}Pu, ^{239,240}Pu, ^{89,90}Sr, ^{35}S, ^{99}Tc, uranium, ^{91}Y, ^{65}Zn, and ^{95}Zr/Nb. The second step of screening evaluated the importance of exposure pathway for each of these radionuclides. For radionuclides released to air from the SRS, the screening shows that tritium and iodine are the major potential contributors to radiation dose from all airborne pathways except plume immersion. Detailed assessments of release estimates to air for

^{3}H, ^{131}I, ^{41}Ar, ^{129}I, plutonium, and uranium can be found in Chapters 4.1, 4.2, 4.3, and 4.4. For surface water pathways, ^{137}Cs dominates all pathways except the ingestion of drinking water, where tritium is most important. Strontium-90 is relatively important when considering the ingestion of vegetables and meat as major exposure pathways, and ^{32}P is important when considering the ingestion of fish from local streams. Detailed assessments of release estimates to water for ^{3}H, ^{137}Cs, ^{90}Sr, ^{60}Co, ^{131}I, ^{32}P, ^{35}S, and uranium can be found in Chapter 5.

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ADDENDUM 3A

ADDITIONAL RADIONUCLIDE DATA FOR SCREENING CALCULATIONS

Addendum 3A—Tables

Table 3A-1. Comprehensive List of Radionuclides Associated with Fuel, Releases or Waste at SRS

Symbol	Half-life	Present in SRS fuel assembly [a]	SRS releases estimates [b]	High level sludge [c]	High level supernate [c]	Reconstit. Waste [c]	Low-level waste [d]
Am-241,243			X				
Am-241	432.y	X		X	X	X	
Ar-41	1.83 h		X				
Ba-140	12.8d	X					X
Ba,La-140			X				
Br-82	35.3 h		X				
C-14	5730 y		X				
Ce-141	32.6 d	X	X	X	X		X
Ce-141,144			X				
Ce-143	33.0h	X					
Ce-144	284.d	X	X				X
Ce,Pr-144				X	X	X	
Cm-242	163.d	X	X				
Cm-244	18.1y	X	X	X	X	X	
Co-58	70.8d	X	X				
Co-60	5.27y	X	X				X
Cr-51	27.7 d		X				X
Cs-134	2.06y	X	X	X	X		X
Cs-135	2.3E7 y			X	X		
Cs-136	13.0d	X	X				
Cs-137	30.1y	X	X	X	X	X	X
Eu-152	13.3 y			X	X		
Eu-154	8.8 y			X	X		
Fe-59	44.5 d						X
H-3	12.35 y		X				X
I-129	1.57E7 y		X	X	X		
I-131	8.04d	X	X				X
I-132	2.28h	X					
I-133	20.9h	X	X				
I-134	52.5m	X					
I-135	6.61h	X	X				
Kr-85	10.7y	X	X				
Kr-85m	4.48h	X	X				
Kr-87	1.27h	X	X				
Kr-88	2.86h	X	X				
La-140	40.3h	X					X
Mn-54	312.5 d		X				X
Mo-99	2.75d	X	X				

Table 3A-1. Comprehensive List of Radionuclides Associated with Fuel, Releases or Waste at SRS (cont'd)

Symbol	Half-life	Present in SRS fuel assembly [a]	SRS releases estimates [b]	High level sludge [c]	High level supernate [c]	Reconstit. waste [c]	Low level waste [d]
Na-24	15 h						X
Nb-95	35.0d	X	X	X			X
Nb-98	51.5 m				X		
Nd-147	11 d	X					
P-32	14.3 d		X				X
Pd-107	6.5E6 y			X	X		
Pr-143	13.6 d	X					
Pr-144	17.28 m						X
Pm-147	13.6 m		X	X	X	X	X
Pu-238	87.7y	X	X	X	X	X	X
Pu-239, 240			X			X	
Pu-239	2.4 E4y	X		X	X		X
Pu-240	6.6 E3y	X		X	X		
Pu-241	14.4y	X		X	X		
Pu-242	3.76E5 y			X	X		
Rb-86	18.8d	X					
Ru-103	39.4d	X	X	X	X		X
Ru-105	4.44h	X					
Ru-106	1.00y	X	X	X	X	X	X
Rh-106	29.9 s						X
Rh-105	1.48d	X					
S-35	87.4 d		X				X
Sb-124	60.2 d		X				
Sb-124,125			X				
Sb-125	2.77 y		X				
Sn,Sb-126	3.91 h	X		X	X		
Sb-129	4.41 h	X					
Se-75	119.8 d		X				
Se-79	6.5E4 y			X	X		
Sm-151	90 y			X	X	X	
Sr-89	50.6d	X	X	X	X		X
Sr-90	28.8 y	X	X	X	X	X	X
Sr-91	9.48 h	X					
Tb-158	150 y			X	X		
Te-127	9.35 h	X		X	X		
Te-127m	109. d	X					
Te-129	1.16 h	X		X	X		
Te-129m	33.5 d	X					

Table 3A-1. Comprehensive List of Radionuclides Associated with Fuel, Releases or Waste at SRS (cont'd)

Symbol	Half-life	Present in SRS fuel assembly [a]	SRS Releases estimates [b]	High level sludge [c]	Hi-Level supernate [c]	Reconstit. waste [c]	Low level waste [d]
Te-131m	1.25 d	X					
Te-132	3.26 d	X					
Tc-99	2.13E5 y			X	X		
Tc-99m	6.01 h	X					
Th-232	405E10 y		X				
U			X				
U-233	1.59E5 y			X	X		
U-235	703E6 y			X	X		
U-238	4.47E9 y		X	X	X		
Un-ID-alpha			X				
Un-ID-B+G			X				
Xe-131m	11.9 d		X				
Xe-133	5.25d	X	X				
Xe-135	9.10h	X	X				
Y-90	2.67d	X					X
Y-91	58.5d	X	X	X	X		
Zn-65	243.9 d		X				X
Zr,Nb-95			X				
Zr-93	1.53E6 y			X	X		
Zr-95	64.0d	X	X	X	X		X
Zr-97	16.9h	X					

[a] From Bauer et al. 1986, DOE 1984, DOE 1990
[b] From Cummins et al, 1991, Ashley and Zeigler 1982
[c] From DOE 1979, Looney et al. 1989
[d] From Stone and Christensen 1983

Table 3A-2a. Ranking Radionuclides by Air Exposure Pathway Based on their Contribution to the Total Screening Value: All Pathways

Radionuclide[e]	Atmospheric release rate[a] (Bq y^{-1})	Atmospheric concentration[b] (Bq m^{-3})	All paths Screening Factor[c] (Sv per Bq m^{-3})	Screening value[d] (Sv y^{-1})	Percentage of total screening value
I-131	2.6E+12	6.8E+04	2.80E-02	1.9E+03	50
H-3	2.5E+16	6.7E+08	1.90E-06	1.3E+03	35
Ar-41	6.6E+15	1.7E+08	1.50E-06	2.6E+02	7
I-129	5.9E+09	1.5E+02	5.60E-01	8.7E+01	2
Pu-239, 240	3.1E+09	8.2E+01	1.00E+00	8.2E+01	2
Ru-103,106	1.6E+11	4.3E+03	9.60E-03	4.1E+01	1
Pu-238	1.0E+09	2.7E+01	8.90E-01	2.4E+01	1
C-14	3.1E+12	8.1E+04	2.60E-04	2.1E+01	1
Cs-137	3.6E+09	9.5E+01	2.20E-01	2.1E+01	1
U235,238	9.3E+08	2.4E+01	3.40E-01	8.3E+00	0.2
Sr-89,90	2.1E+09	4.1E+01	1.90E-01	7.8E+00	0.2
Am-241	2.1E+09	5.5E+00	1.00E+00	5.5E+00	0.1
Total screening value from radionuclides released to air				3.7E+03	100

a The atmospheric release rate and atmospheric concentraton in the next column are the input values for the assessment of each exposure pathway (inhalation, ingestion of milk, vegetables, meat, and ground contamination) in subsequent portions of this table.
b Concentration at the point of release from the facility
c From Table B.1, NCRP 1996
d Product of the screening factor times the atmospheric concentration.
e The radionuclides that are the main contriubotrs to the screening value are shaded.

Table 3A-2b. Ranking Radionuclides by Air Exposure Pathway Based on their Contribution to the Total Screening Value: Inhalation Pathway

Radionuclide	Inhalation Screening Factor (Sv per Bq m^{-3})	Screening value[d] (Sv y^{-1})	Percent of inhalation scrn value
H-3	1.40E-07	9.3E+01	53
Pu-239, 240	5.50E-01	4.5E+01	26
Pu-238	5.00E-01	1.4E+01	8
I-131	1.20E-04	8.1E+00	5
U235,238	2.60E-01	6.3E+00	4
Ru-103,106	1.00E-03	4.3E+00	2
Am-241	5.70E-01	3.1E+00	2
C-14	4.50E-06	3.7E-01	0.2
Sr-89,90	2.80E-03	1.1E-01	0.07
I-129	6.20E-04	9.6E-02	0.06
Cs-137	6.80E-05	6.5E-03	0.004
Ar-41	a		
Total screening value for inhalation		1.7E+02	100

[a] No screening factor for this radionuclide for this pathway.

Table 3A-2c. Ranking Radionuclides by Air Exposure Pathway Based on their Contribution to the Total Screening Value: Milk Ingestion Pathway

Radionuclide	Milk Screening Factor (Sv per Bq m^{-3})	Screening value (Sv y^{-1})	Percent of milk ingestion screening value
I-131	1.50E-02	1.0E+03	62
H-3	8.60E-07	5.7E+02	35
I-129	2.20E-01	3.4E+01	2
C-14	6.30E-05	5.1E+00	0.3
Cs-137	4.10E-02	3.9E+00	0.2
Sr-89,90	4.60E-02	1.9E+00	0.1
U235,238	1.80E-03	4.4E-02	0.003
Ru-103,106	2.50E-06	1.1E-02	0.0007
Pu-239, 240	9.90E-05	8.1E-03	0.0005
Pu-238	8.90E-05	2.4E-03	0.0001
Am-241	2.00E-04	1.1E-03	0.0001
Ar-41	a		
Total screening value for milk		1.6E+03	100

[a] No screening factor for this radionuclide for this pathway.

Table 3A-2d. Ranking Radionuclides by Air Exposure Pathway Based on their Contribution to the Total Screening Value: Produce Ingestion Pathway

Radionuclide	Vegetable Screening Factor (Sv per Bq m^{-3})	Screening value (Sv y^{-1})	Percent of produce ingestion screening value
H-3	5.80E-07	3.9E+02	54
I-131	4.00E-03	2.7E+02	38
Pu-239, 240	2.70E-01	2.2E+01	3
I-129	8.00E-02	1.2E+01	2
C-14	1.30E-04	1.1E+01	1
Pu-238	2.50E-01	6.8E+00	1
Sr-89,90	7.30E-02	3.0E+00	0.4
Cs-137	2.30E-02	2.2E+00	0.3
Ru-103,106	3.60E-04	1.5E+00	0.2
Am-241	2.80E-01	1.5E+00	0.2
U235,238	1.40E-02	3.4E-01	0.05
Ar-41	a		
Total screening value for veg		7.2E+02	100

[a] No screening factor for this radionuclide for this pathway.

Table 3A-2e. Ranking Radionuclides by Air Exposure Pathway Based on their Contribution to the Total Screening Value: Meat Ingestion Pathway

Radionuclide	Meat Screening Factor (Sv per Bq m^{-3})	Screening value (Sv y^{-1})	Percent of meat ingestion screening value
I-131	9.50E-03	6.45E+02	73
H-3	2.90E-07	1.93E+02	22
I-129	2.20E-01	3.41E+01	4
C-14	7.20E-05	5.87E+00	0.7
Cs-137	5.10E-02	4.85E+00	0.5
Sr-89,90	5.80E-02	2.38E+00	0.3
Ru-103,106	5.70E-05	2.45E-01	0.03
Pu-239, 240	2.50E-03	2.05E-01	0.02
Pu-238	2.20E-03	5.98E-02	0.01
U235,238	9.20E-04	2.24E-02	0.003
Am-241	1.30E-03	7.12E-03	0.0008
Ar-41	a		
Total screening value for meat		8.86E+02	100

[a] No screening factor for this radionuclide for this pathway.

Table 3A-2f. Ranking Radionuclides by Air Exposure Pathway Based on their Contribution to the Total Screening Value: Plume Immersion Pathway

Radionuclide	Plume immersion Screening Factor (Sv per Bq m^{-3})	Screening value (Sv y^{-1})	Percent of plume immersion screening value
Ar-41	1.50E-06	2.61E+02	99.9
I-131	4.70E-07	3.19E-02	0.01
Am-241	2.30E-08	1.26E-07	0
U235,238	1.30E-10	3.17E-09	0
I-129	1.00E-08	1.55E-06	0
Pu-239, 240	1.10E-10	9.02E-09	0
Pu-238	1.10E-10	2.99E-09	0
Ru-103,106	6.00E-07	2.57E-03	0
C-14	a		
Cs-137	a		
H-3	a		
Sr-89,90	a		
Total screening value for plume imm		2.62E+02	100

[a] No screening factor for this radionuclide for this pathway.

Table 3A-2g. Ranking Radionuclides by Air Exposure Pathway Based on their Contribution to the Total Screening Value: Ground Contamination Pathway

Radionuclide	Ground contamination Screening Factor (Sv per Bq m^{-3})	Screening value (Sv y^{-1})	Percent of ground contamination screening value
Cs-137	9.80E-02	9.31E+00	43
I-131	1.20E-04	8.15E+00	38
Ru-103,106	7.00E-04	3.00E+00	8
U-235,238	4.60E-02	1.12E+00	5
I-129	5.10E-03	7.90E-01	4
Am-241	6.80E-03	3.73E-02	2
Pu-239, 240	1.90E-04	1.56E-02	0.07
Pu-238	1.80E-04	4.90E-03	0.05
Ar-41	a		
C-14	a		
H-3	a		
Sr-89,90	a		
Total screening value for ground cont.		2.62E+02	100

[a] No screening factor for this radionuclide for this pathway.

Table 3A-3a. Ranking Radionuclides by Water Exposure Pathway Based on their Contribution to the Total Screening Value: All Pathways

Radionuclide[e]	Half-life	Releases in liquid effluents[a] (Bq y^{-1})	Liquid effluent concentration[b] (Bq m^{-3})	All pathways Screening factor[c] (Sv per Bq m^{-3})	Screening value[d] (Sv/y)	Percent of total screening value
Cs-137	30 y	6.3E+11	8.1E+01	1.10E-06	8.96E-05	60
Sr-89,90		6.4E+11	8.3E+01	4.30E-07	3.56E-05	24
Co-60	5.271 y	6.8E+10	8.8E+00	6.10E-07	5.37E-06	4
P-32	14.29 d	3.7E+10	4.8E+00	8.20E-07	3.94E-06	3
I-131	8.04 d	3.1E+11	4.0E+01	8.60E-08	3.48E-06	2
H-3	12.35 y	1.5E+15	2.0E+05	1.40E-11	2.80E-06	2
Zn-65	243.9 d	1.4E+11	1.9E+01	1.30E-07	2.43E-06	2
S-35	87.44 d	1.5E+12	2.0E+02	1.10E-08	2.20E-06	1
Zr,Nb-95	63.98 d	1.0E+11	1.3E+01	7.40E-08	9.88E-07	1
Tc-99	2.13e5 y	1.0E+11	1.3E+01	6.90E-08	9.28E-07	1
U-235,238	4.468e9 y	3.1E+10	4.0E+00	1.80E-07	7.21E-07	0.5
Pu-239, 240	24065 y	1.2E+09	1.6E-01	1.70E-06	2.72E-07	0.2
I-129	1.57e7 y	1.2E+09	1.6E-01	1.40E-06	2.24E-07	0.2
Y-91	58.5 d	1.2E+11	1.5E+01	8.20E-09	1.27E-07	0.09
Pu-238	87.74 y	4.1E+08	5.3E-02	1.50E-06	8.01E-08	0.05
Total screening value from radionuclides released in liquid effluents					1.49E-04	100

a The release rate and liquid effluent concentraton in the next column are the input values for the assessment of each exposure pathway (drinking water, ingestion of fish, milk, vegetables, meat, and ground contamination) in subsequent portions of this table.
b Concentration at the point of release from the facility
c From Table C.1, NCRP 1996
d Product of the screening factor times the concentration in liquid effluents.
e The radionuclides that are the main contriubotrs to the screening value are shaded.

Table 3A-3b. Ranking Radionuclides by Water Exposure Pathway Based on their Contribution to the Total Screening Value: Drinking Water Pathway

Radionuclide	Drinking water Screening factor (Sv per Bq m^{-3})	Drinking water screening value (Sv/y)	Percent of drinking water screening value
Cs-137	1.10E-08	2.9E-06	30
H-3	1.40E-11	2.8E-06	29
Sr-89,90	2.50E-08	2.1E-06	21
I-131	1.90E-08	7.7E-07	8
Pu-239, 240	4.50E-07	4.8E-07	5.0
Pu-238	4.10E-07	2.2E-07	2.27
U-235,238	2.20E-08	1.2E-07	1.3
Co-60	5.70E-09	6.4E-08	1
Zn-65	3.00E-09	6.0E-08	1
S-35	2.30E-10	5.4E-08	1
P-32	2.00E-09	5.2E-08	1
Y-91	3.00E-09	4.6E-08	0.48
Zr,Nb-95	9.70E-10	1.9E-08	0
I-129	9.90E-08	1.6E-08	0.2
Tc-99	5.30E-10	7.1E-09	0
Total screening value for drinking water		9.6E-06	

Table 3A-3c. Ranking Radionuclides by Water Exposure Pathway Based on their Contribution to the Total Screening Value: Fish Ingestion Pathway

Radionuclide	Fish Screening factor (Sv per Bq m^{-3})	Fish screening value (Sv/y)	Percent of fish ingestion screening value
Cs-137	5.40E-07	1.4E-04	83
P-32	8.10E-07	2.1E-05	13
Sr-89,90	3.70E-08	3.1E-06	2
Zn-65	6.60E-08	1.3E-06	1
S-35	4.20E-09	9.8E-07	1
Co-60	4.20E-08	4.7E-07	0.3
Pu-239, 240	3.40E-07	3.6E-07	0.2
Pu-238	3.10E-07	1.7E-07	0.1
I-131	4.00E-09	1.6E-07	0.1
Zr,Nb-95	4.90E-09	9.5E-08	0.06
H-3	3.40E-13	6.8E-08	0.04
U-235,238	5.50E-09	3.1E-08	0.02
Y-91	1.50E-09	2.3E-08	0.01
I-129	9.90E-08	1.6E-08	0.01
Tc-99	2.70E-10	3.6E-09	0.002
Total screening value for fish ingestion		1.7E-04	100

Table 3A-3d. Ranking Radionuclides by Water Exposure Pathway Based on their Contribution to the Total Screening Value: Produce Ingestion Pathway

Radionuclide	Vegetables Screening factor (Sv per Bq m^{-3}	Vegetables screening value (Sv/y)	Percent of vegetable screening value
Sr-89,90	1.50E-07	1.2E-05	46
Cs-137	4.60E-08	1.2E-05	45
Tc-99	5.70E-08	7.7E-07	3
Pu-239, 240	5.50E-07	5.9E-07	2.2
I-131	8.10E-09	3.3E-07	1
Pu-238	5.00E-07	2.7E-07	1.0
U-235,238	2.70E-08	1.5E-07	0.6
Co-60	9.30E-09	1.0E-07	0.4
Zn-65	4.30E-09	8.6E-08	0.3
S-35	2.70E-10	6.3E-08	0.2
Y-91	2.90E-09	4.5E-08	0.2
P-32	1.30E-09	3.4E-08	0.1
I-129	1.60E-07	2.6E-08	0.1
Zr,Nb-95	1.10E-09	2.1E-08	0.08
H-3	a		
Total screening value for produce		2.7E-05	

[a] No screening factor for this radionuclide for this pathway.

Table 3A-3e. Ranking Radionuclides by Water Exposure Pathway Based on their Contribution to the Total Screening Value: Milk Ingestion Pathway

Radionuclide	Milk Screening factor (Sv per Bq m^{-3}	Milk screening value (Sv/y)	Percent milk ingestion screening value
Cs-137	8.50E-08	2.2E-05	68
Sr-89,90	9.30E-08	7.7E-06	24
I-131	3.30E-08	1.3E-06	4
S-35	1.80E-09	4.2E-07	1
P-32	9.70E-09	2.5E-07	1
Zn-65	1.20E-08	2.4E-07	1
Tc-99	1.10E-08	1.5E-07	0.5
Co-60	7.70E-09	8.6E-08	0.3
I-129	4.60E-07	7.4E-08	0.2
U-235,238	4.00E-09	2.2E-08	0.1
Y-91	6.40E-11	9.9E-10	0.003
Pu-239, 240	2.10E-10	2.2E-10	0.001
Pu-238	1.90E-10	1.0E-10	0.0003
Zr,Nb-95	2.90E-13	5.6E-12	0.00002
H-3	a		
Total screening value for milk		3.2E-05	100

[a] No screening factor for this radionuclide for this pathway.

Table 3A-3f. Ranking Radionuclides by Water Exposure Pathway Based on their Contribution to the Total Screening Value: Meat Ingestion Pathway

Radionuclide	Meat Screening factor (Sv per Bq m^{-3}	Meat screening value (Sv/y)	Percent meat ingestion screening value
Cs-137	1.10E-07	2.9E-05	69
Sr-89,90	1.20E-07	9.9E-06	24
S-35	4.20E-09	9.8E-07	2
I-131	2.20E-08	8.9E-07	2
Zn-65	3.10E-08	6.2E-07	1
Co-60	2.90E-08	3.3E-07	1
P-32	4.80E-09	1.3E-07	0.3
I-129	4.60E-07	7.4E-08	0.2
U-235,238	2.10E-09	1.2E-08	0.03
Y-91	5.00E-10	7.7E-09	0.02
Pu-239, 240	5.20E-09	5.6E-09	0.01
Tc-99	2.70E-10	3.6E-09	0.009
Pu-238	4.70E-09	2.5E-09	0.01
Zr,Nb-95	8.60E-14	1.7E-12	0.000004
H-3	a		
Total screening value for meat ingestio		4.2E-05	100

[a] No screening factor for this radionuclide for this pathway.

Table 3A-3g. Ranking Radionuclides by Water Exposure Pathway Based on their Contribution to the Total Screening Value: Ground Contamination Pathway

Radionuclide	Ground contamination Screening factor (Sv per Bq m^{-3})	Ground contamination screening value (Sv/y)	Percent ground contamination screening value
Cs-137	3.50E-07	9.1E-05	91
Co-60	5.20E-07	5.8E-06	6
U-235,238	3.10E-07	1.7E-06	2
Zr,Nb-95	6.70E-08	1.3E-06	1
Zn-65	1.70E-08	3.4E-07	0.3
Pu-239, 240	6.90E-09	7.4E-09	0.007
Y-91	2.40E-10	3.7E-09	0.004
Pu-238	6.40E-09	3.4E-09	0.003
I-131	4.20E-11	1.7E-09	0.002
I-129	1.80E-09	2.9E-10	0.0003
Sr-89,90	5.90E-13	4.9E-11	0.00005
Tc-99	5.20E-14	7.0E-13	0.000001
H-3	a		
P-32	a		
S-35	a		
Total screening value for meat ingestion		1.0E-04	100

[a] No screening factor for this radionuclide for this pathway.

CHAPTER 4.1

RELEASES OF TRITIUM TO THE ATMOSPHERE

ABSTRACT

Tritium (^3H or T) is one of the principle nuclear materials produced at the Savannah River Site (SRS). The key processes that have lead to atmospheric tritium releases are reactor operations (100 Areas), recovery of transuranic elements in the separations facilities (200 Areas), recovery of tritium in the tritium processing facilities (200 Areas), laboratory research area, and heavy water rework facility. The majority of the tritium releases came from the reactor facilities and the tritium processing facilities. This chapter presents brief histories and descriptions of operations at these facilities and the SRS source term.

The releases of tritium from the SRS are generally very well documented in published reports. Records of releases have been kept since almost the beginning of operations. The main sources of information were SRS weekly and monthly reports. In the cases where daily or shift data were found in logbooks, we used them to check the monthly published data. In general, there is quite good agreement between the different release values reported in the different sources. Because in most cases the summary values were within 10% of the published values, it is likely that most "unchecked" monthly values are also valid.

The research on the quality of the atmospheric tritium release data reported by SRS has given us no indication that there were serious or consistent errors made in carrying out the tritium release monitoring. However, it is important that the best estimates presented in this report are used in conjunction with the uncertainty associated with each of the values.

INTRODUCTION

Tritium is one of the principle nuclear materials produced at the SRS. It is the heaviest and only radioactive isotope of hydrogen with a physical half-life of 12.5 years. Tritium is used to multiply the firepower of plutonium in nuclear weapons. Tritium processing operations have been performed at the SRS facility since October 1955, beginning in Building 232-F. In July 1957, the process was moved to Building 232-H and operations doubled by 1958. This section discusses the atmospheric releases of tritium to the environment, called the atmospheric source term. Chapter 5 presents the liquid releases.

Tritium is produced naturally by cosmic ray interactions and as a fission product in nuclear reactors. The nuclear reactions that produce tritium are

- Fission of lithium by neutron irradiation of targets
- Reaction of neutrons with the heavy water moderator used in reactors
- Ternary fission of transuranic elements in the reactor fuels and targets.

At SRS, tritium was originally produced as a by-product of producing plutonium in the reactors. Lithium-aluminum control rods were used in R-Reactor and other reactors from the first day of operation to produce tritium and control reactor power. When additional tritium was needed beginning in 1955, enriched uranium was used in the reactors, which allowed for increased loading of lithium-aluminum targets for tritium production.

Gaseous T_2 at room temperature tends to form elemental tritium (HT) by reaction with gaseous hydrogen. Tritium in this form does not constitute a significant radiation health hazard because the beta particle given off during its decay is of low energy and has low penetrating power. Tritium closely follows the reactions of ordinary hydrogen when it exists as tritiated water or vapor (T_2O; tritium oxide [HTO]) and is taken into the body quite easily, resulting in a higher factor of risk. Tritium is released from SRS in both elemental and oxide forms.

The key processes that have lead to tritium releases at the SRS are

- Reactor operations (100 Areas)
- Recovery of transuranic elements in the separations facilities (200 Areas)
- Recovery of tritium in the tritium processing facilities (200 Areas)
- Laboratory research area
- Heavy water rework facility.

The majority of the tritium releases came from the reactor facilities and the tritium processing facilities. This chapter presents brief histories and descriptions of operations at these facilities and the reconstructed SRS atmospheric tritium source term. Appendix E presents a detailed description of the monitoring equipment used to monitor atmospheric tritium releases and an uncertainty analysis to be applied to reported releases from the different monitors.

REACTOR FACILITIES

History

Heavy water moderated and cooled reactors using natural uranium fuel were used at the Savannah River Plant (SRP). The modified reactors were called pi-Pile and were from Argonne National Laboratory (Stetson et al. 1963). The first reactor came online in December 1953, and by March 28, 1955, all five reactors were in operation. Reactor locations were designated as the 100 Areas (e.g., 100-R was the name of the R-Reactor area).

The R-Reactor went into operation on December 28, 1953. Full power was added stepwise until the spring of 1955 when all reactors were at their design limit of about 500 MW. During the second period (from July 1955 to November 1956), there was an extension of power ascension to about twice the original design limit. During the third period (from December 1956 to October 1958), there were major increases in the capacity of the primary and secondary cooling systems. During this period, all reactors were shut down for 2–3 months each to replace D_2O pumps and piping. The pumps were replaced from December 1957 to July 1958. Cooling water supply from the Savannah River was augmented by the construction of Par Pond (Stetson et al. 1963).

From October 1958 to January 2, 1962, there were no dramatic increases in reactor power levels (only about 2 to 10%). From January 2, 1962, until the end of 1963, there were extended shut downs to install containment facilities, which were built in phases during that period (Stetson et al. 1963).

The reactors were devoted primarily to the production of plutonium and tritium. On September 5, 1956, L-Reactor changed to 100% tritium production and operated with enriched uranium fuel most of the time. C-Reactor converted to enriched uranium fuel March 3, 1957, and K-Reactor converted on August 5, 1957. After that time, the operating reactors alternated between plutonium and tritium production. Tritium production peaked in 1958, when three

reactors operated on enriched fuel. By January 1959, K-Reactor and C- Reactor began to alternate between enriched and natural uranium fuel (Stetson et al. 1963).

During Fiscal Year (FY) 1959 and FY 1960 (during this period of time, the government FY ran from July 1 to June 30), about 40% of reactor operation was devoted to tritium production and about 60% to plutonium production. From July 1960 to late 1963, about 25% of reactor operations was devoted to tritium production (Stetson et al. 1963). The R-Reactor was placed on standby status in 1964 and was never restarted. The L-Reactor was placed on standby in 1968 and later restarted. The other three reactors operated in the 1970s, producing both tritium and plutonium.

In 1982, it was planned that at startup L-Reactor would devote 3% of its capacity to ^{240}Pu production, which would involve five sets of targets to be discharged during each fuel cycle. These conditions were expected to increase the tritium evaporation from the disassembly basin and seepage basin (Dukes and Benjamin 1982). Table 4.1-1 summaries reactor operations.

Production reactors processes at SRP included reactor operation, an analytical laboratory, and facilities for reworking the degraded heavy water moderator (D_2O). The major reworking facilities consisted of vacuum fractional distillation equipment, minor rework operations, and a facility for cleaning the drums in which the heavy water was stored and transported (Longtin et al. 1973; Jacober et al. 1973).

Table 4.1-1. Summary of Reactor Operations

Reactor	Dates of operation	Comments
C	1955–1987	Placed in cold standby[a] in 1987; now shutdown
K	1954–1988	1991 put in cold standby; now shutdown
L	1954–1968	Now shutdown
	1985–1988	
P	1954–1988	Now shutdown
R	1953–1964	Now shutdown

[a] Cold standby: available to restart after refueling and upgrading.

The major part of the work to restore isotopic purity to the D_2O was done by vacuum fractional distillation in the 400 Area. When not in use, the heavy water was stored in drums. The drum wash facility was used to remove the small amounts of tritiated D_2O that remained when the drums were emptied (Longtin et al. 1973; Jacober et al. 1973).

Releases of tritium from the production reactors were due almost entirely to releases of moderator, either as liquid or vapor from the reactor systems and from the heavy water rework facility. Small amounts could have been released from the reactor blanket gas system and from irradiated lithium-bearing control rods and targets stored underwater in the disassembly basins (Longtin et al. 1973; Jacober et al. 1973).

Reactor Stacks Tritium Monitoring

Early years (pre 1970s)

During the early years, before 1970, the two main types of monitoring systems employed to measure airborne tritium releases from SRS reactors were the silica gel and dehumidifier monitors. These methods were used to monitor the total releases of tritiated water from the reactor stacks. Initially, direct measure of the loss of moderator from the reactor was used to determine tritium losses, but this measured both atmospheric and direct liquid releases, so tritiated water releases in the stack were monitored.

Silica gel was used to measure tritium vapor release from the reactors and to determine the amount of moderator that was lost from the reactor stacks (Miller 1956a). This method was difficult and inaccurate, so there was a shift to dehumidifiers (Miller 1956a; Kiger 1955).

A bit later, Kanne ionization chambers were also employed for online monitoring (Caldwell 1958). Kanne chambers cannot differentiate between tritium and other radioactive gases, so they were used as real-time monitors to alarm at preset activity levels and not to determine the quantitative atmospheric tritium releases (Longtin et al. 1973).

In the late 1950s, the two methods of estimating total tritium releases in ventilation air discharged from the reactor buildings (e.g., 105-R) were cited in Caldwell (1958) as follows:

1. Inventory losses derived from observed reductions in moderator inventory. This was considered accurate within a factor of 2 because of the difficulty in differentiating stack release from other moderator losses through sump collection, losses to the disassembly basin water during reactor discharge, or other mechanisms.
2. Computing stack releases based on the percent of moderator (D_2O) contained in the water samples condensed from the stack stream. A dehumidifier was operated three times a week for a total of 24 hours during normal operations, but it was not used during reactor outages. This method provided an estimate of the amount of moderator that was lost and, therefore, the total tritium released during the measurement period, but it did not provide rate or time of releases. For this reason, it was considered a good method of monitoring only during continuous routine operations.

Kanne ionization chambers were considered the most practical systems available. They were used in reactors for online alarm systems and at the Tritium Facilities for personnel monitoring and for stacks and central air ducts monitoring (Conway 1956). A central system connecting sampling lines to numerous points, controlled by valving, was considered ideal. By 1965, Tyson (1965) noted the need for the new and improved method, the stack tritium monitor (STM).

1970s

For each reactor process, releases were monitored in the stack at +148 ft and in three exhaust ducts. A 1-in. diameter sample probe in the stack at +148 ft carried an aliquot of stack effluent to the Kanne chamber, which was set to alarm at a predetermined point. Because any noble gas activity present in the stack effluent would be added to the tritium activity measured, the Kanne chamber was not used to obtain quantitative measures of total tritium released.

The daily stack tritium discharge was calculated based on a sample of water collected by a continuously running dehumidifier (apparently no longer run only 3 times a week). The water

sample was counted in a liquid scintillation counter with a sensitivity of 3×10^{-7} µCi mL^{-1} (±10%). The daily tritium stack loss was calculated based upon these sample results, humidity, and effluent airflow rate (Longtin et al. 1973; Jacober et al. 1973; Reinig et al. 1973).

A new effluent air monitoring system, designed in the 1960s (Tyson 1965), was evaluated in the early 1970s—the STM. This system consisted of two ionization chambers in parallel with a moisture trap ahead of one chamber to remove HTO. The difference in the signals from the two chambers was proportional to the HTO concentration. This system eliminated background and noble gas interference. It measured only tritium in the oxide form, but nearly all the tritium released from the reactors was in that form (Longtin et al. 1973; Jacober et al. 1973). This system became the primary monitoring method used in the reactors.

In the disassembly bay (in 105-P), no tritium stack monitoring appeared to be implemented. Atmospheric releases were estimated based on area, not the exhaust vent, monitoring (Reinig et al. 1973; Longtin et al. 1973). For the purification makeup room exhaust (105-C and 105-K) no monitoring for tritium was done. In 1973, it was recommended that the air be rerouted to the purification exhaust, which was monitored (Reinig et al. 1973; Longtin et al. 1973). For the purification resin preparation and cold pipe space exhaust (105-P), no tritium monitoring was carried out; it was also recommended that the exhaust be rerouted to the purification exhaust. In the heat exchanger decontamination facility (105-C), no tritium monitoring and grab sampling was recommended (Reinig et al. 1973).

1980s

Documentation from the 1980s showed that losses from the process water system were directly detected by process water loss (volume or pressure drops) and measurement of tritium in the heavy water vapor that evaporated from the leaks in the cooling system to the stacks. The tritium in the D_2O ranged from 3000 to 13,000 µCi mL^{-1}. Tritium concentration in the moderator was measured weekly by laboratory analysis, and daily airflow in the stack was measured by taking the average pressure drop across the carbon filters (Smith 1989).

STM and a new Berthold tritium monitor (BTM) were both being used by the end of the 1980s. Since September 1985, two BTMs had been operating in L-Reactor and K-Reactor for a combined total of 2.5 years of in-service operation (Merz 1988). The BTM was recommended as the primary method to measure tritium releases.

Smith (1989) reported on a leak before break program (LBB), which was established to provide a technical and operational basis for the new design basis loss-of-coolant accident for the emergency cooling system in the reactors. It dealt with the tritium monitors, airflow measurements, and moderator tritium analysis. The leak detection instrumentation consisted of the BTMs, STMs, and their associated equipment. These instruments were used to indicate a leak in the primary coolant system with a 35% error in measurement. The error in the measurements was the result of a 15% error in the detection of the tritium by the tritium monitors, a 15% error in the stack airflow measurement, and a 5% error in the moderator tritium analysis. The BTMs were checked for accuracy every 6 months. Daily comparisons between the BTM and STM results were made to ensure that the instrument readings were comparable (Smith 1989).

In a 1986 review of radiological effluent monitoring, analytical techniques, and reporting, Zeigler (1986) reported on the current status of tritium monitoring at the SRS and the factors contributing to the uncertainty of the measurements, stating that:

> There are a few minor radioactive effluents that are monitored by Area Survey HP personnel, but the releases are not tabulated and reported because they are small . . . Overall the most important improvements needed in SRP effluent monitoring relate to measurement of volumes of air and liquid that are released. In many cases, volumes are estimated based on fan or pump capacities or on measurements that were made at some time in the past. Frequently, there is no documentation of the methods used to obtain the volumes that are used.

In the 1980s, the following measurements were made in the reactor areas:

105-P, 105-L, and 105-K Stacks and Disassembly Area. Department personnel collected a daily dehumidifier sample from the 195-ft stack and a silica gel sample weekly from the disassembly area for tritium analysis (no direct effluent monitoring was done for the disassembly stack). The tritium stack losses were calculated using the STM results to confirm losses determined by the dehumidifier sample. The 195-ft stack flow rate was determined from daily readings of flow measuring instruments. The disassembly building exhaust flow was determined from the manufacturer-cited capacity of the fans, checked by periodic measurements of actual flow. The releases from the disassembly area were based on the area air tritium concentration.

The uncertainty resulting from this method arises because the disassembly area does not have a stack and, therefore, no well-defined exhaust. The facility was maintained under positive air pressure, so the air was exhausted through doors and other openings in the facility. This factor would have introduced some error in the estimated releases. The HT/HTO ratio from these areas was not documented.

105-C Stacks and Disassembly Area, Stack Areas Decontamination Facility, and Sand Blast Facility in 717-C. Silica gel samples were taken 1 day a week in the disassembly area and were assumed to represent the whole week (value multiplied by 7). A daily dehumidifier sample from the stack was analyzed. Stack airflow was determined from weekly readings of flow measuring instruments. Disassembly airflow was determined based on the capacity of the stack fans with periodic measurements. By only periodically measuring the exhaust air, some additional uncertainty was introduced. In addition, there was no characterization and documentation of the HT/HTO ratio.

A schematic of 105-C Reactor shows the airflow to the 200-ft stack and the location of the sampling system (Figure 4.1-1).

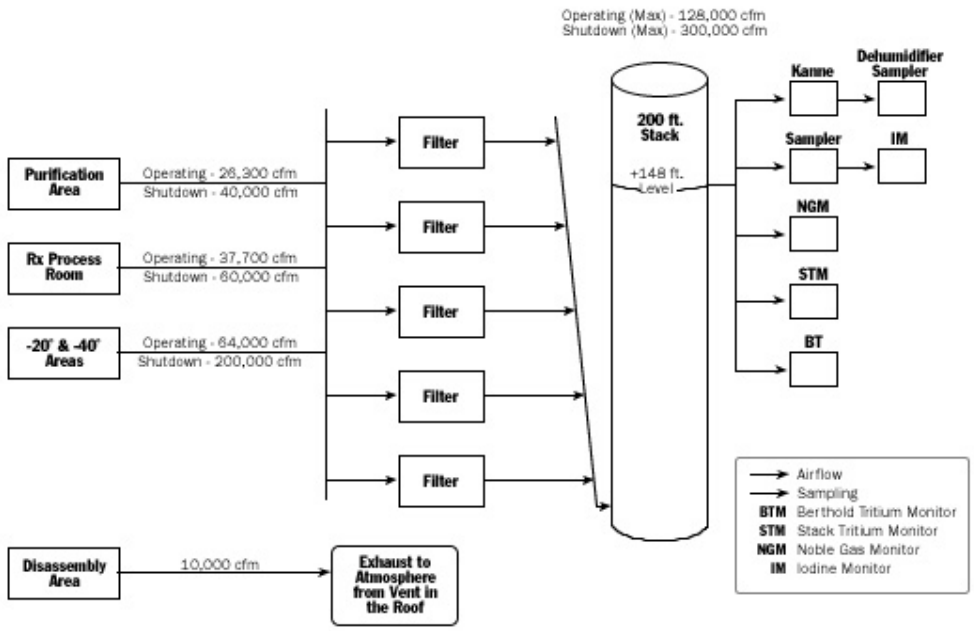


Figure 4.1-1. Airflow schematic of 105-L-Reactor (Westinghouse 1991a).

Reactor Stack Releases

In 1956, it was believed that tritium releases were largely a result of tritium in the moderator and that there were practically no losses from lithium-aluminum target slugs during reactor operations. Nonmoderator losses were believed to occur only on the rare occasions when the slug was cut into accidentally when cutting through control rods. In this event, the release is a small fraction of what is contained in the slug and dissipates rapidly; it could not be measured (Miller 1956a). At that time, the 100-Area tritium releases to the atmosphere were considered low enough to be neglected except in the case of a major reactor failure. Releases of T_2O or DTO averaged 590 curies per month (Ci mo^{-1}) per area during May–August 1956. An increase was expected due to power ascension, but the routine releases were not expected to exceed 5000 Ci mo^{-1}. Compared to the expected Separations Area (200 Area) releases, this increase was considered negligible (Denham 1956).

Original estimates of tritium production in the moderator (from the 100-Area Technical Manual) were 0.5 mCi/mL/y production in the moderator at 700 MW because of neutron capture by the deuterium. Actual results showed a 20% fluctuation about the expected value, so the expected rate was 0.002 µCi/mL/MWd. This estimate could not be checked daily, but monthly it was reported to match reasonably well with tritium concentration estimates (Miller 1956a).

The atmospheric release points from the reactors are the 200-ft stack and ground-level evaporation from the disassembly basin water shown in Figure 4.1-1. An additional source of atmospheric tritium release is the ground-level evaporation of water purged from the disassembly basin to a seepage basin. Tritium in the form of DTO (from the moderator) is released to the ventilation system and, therefore, to the stack by evaporation of process water exposed to air

flowing through the process area. During reactor operations, D_2O containing DTO is evaporated from small leaks in pipe flanges, valves, and from the D_2O process water exposed in the control rod guide tubes. During reactor shutdown, DTO is evaporated when pipes or valves are opened for inspection and maintenance work and when wet fuel, target, and control rods are discharged from the reactor (Dukes and Benjamin 1982).

Although the discharged fuel and target assemblies are rinsed with water before being placed in the disassembly basin, some tritium is transferred to the disassembly basin with process water that adheres to the assemblies and from DTO contained as water of hydration in the aluminum oxide on the assemblies. Disassembly basin water was discharged to the seepage basin after being recirculated through filters and deionizers to clarify the water and remove radionuclides. The tritium concentration in the disassembly basin water increased following each reactor discharge. When the set level reached 0.2 to 0.4 μCi mL^{-1}, the water was purged to the seepage basin through filters and deionizers. Tritium was not removed during this process. It was estimated that about 30% of the tritium purged evaporated from the seepage basin each year (based on average atmospheric conditions) (Dukes and Benjamin 1982).

By 1971, atmospheric tritium releases were 210,000 Ci from the reactors, 400,000 Ci from the processing operations, and 9,000 Ci from D_2O recovery (Jacobsen 1972); 94% of the tritium was released to the atmosphere and 6% to aqueous effluent streams. In 1973, the situation was about the same; 63% of SRP tritium releases resulted from processing the irradiated lithium and uranium and 37% from various sources associated with the reactor moderator.

The possibility of decreasing the loss of D_2O (coolant) to offset the increase of tritium with time was discussed in 1972. The heavy water loss reduction was shown to have partly offset the increasing tritium concentration in the moderator. Some of the activities taken to reduce heavy water losses were described. The average tritium concentration in the reactor stack air was noted as 5×10^{-4} μCi cc^{-1} (Jacobsen 1972).

In the 1980s, further efforts were made to reduce the reactor releases with an intensive leak reduction program and provisions for recovery of D_2O carried on discharged fuel. The D_2O loss rate was reduced, but the increase of tritium in the moderator caused a lesser reduction in the tritium releases. It was too expensive to introduce fresh D_2O or remove the tritium from the moderator, so improvements were planned to the leak detection and recovery systems for the processing of irradiated lithium. About a 25% reduction in tritium releases was expected to be accomplished at a reasonable cost (Dukes and Benjamin 1982). Further reference of this program was not found in the document database.

In 1981, the reactor areas released 1.3×10^5 Ci to the atmosphere. Of this number, 1.2×10^5 was from stack releases, 5.4×10^3 Ci (92%) was from disassembly basis, and 5.6×10^3 Ci (4%) was attributed to seepage basin evaporation (Crawford and Roggenkamp 1983).

Westinghouse (1991b) indicates that although the reactors were not working, stack monitoring of tritium, ^{41}Ar, and particulates continued at all times. Releases from the reactors were due to the evaporation of tritiated water; therefore, tritium released from these facilities was in the form of tritiated water (HTO). Virtually all the tritium was released to the atmosphere through the stack. Some very small fraction (approximately 1%) was released through evaporation from fuel and target storage basins (Murphy et al. 1991). Tritium losses from the reactor area were estimated to be 100% oxide (Miller and Patterson 1956).

SEPARATIONS AREAS

History

Tritium target processing operations began in October 1955 in a small facility (232-F) and moved to larger quarters in July 1957 (232-H and 234-H). In 1957, tritium was first loaded into reservoirs that are components of nuclear weapons. Tritium was also recovered, purified and reloaded into weapons components. By 1958, the capacity of the recovery operation was doubled. Today, there is no tritium production at the SRS. Tritium that was produced in the past is recycled, mixed, and reloaded in the new Replacement Tritium Facility, which began operations in 1994. This new facility has replaced most of the SRS facilities that processed tritium during the last 35 years.

Tritium Processing Facilities Stack Monitoring

Early years

In the mid 1950s, the Health Physics Group expected that stack monitoring in Building 232-F would be an easy task, but they soon found that more attention to detail was needed to produce good results (Landon 1960). The original system consisted of 1-in. stainless steel sampling line running from the stack plenum into a Kanne system in the air monitor room. The Kanne system monitor measurement output (in curies per meter) was recorded on a paper strip chart that moved at a constant rate. The amount of gas leaving the stack was determined by manually integrating the area under the curve (Landon 1960). Miller and Patterson (1956) reports that based on experimental results (which are not presented in this memo), the figure of 60% oxide and 40% tritium gas is used for the releases from the 232-F facility slow leak in Building 232-F, which went undetected for about 12 days in July 1956. As a result, the reliability of the monitors was carefully investigated and the representativeness of the sampling locations for air sampling points was checked (Miller 1956b). By mid-1956, the adequacy and performance of the system was in question and there was an interest in a more quantitative measurement of stack losses. A test of the system with a series of known releases showed that the system varied as much as ±80% because of two sources of error: the manual integration and inadequate sampling of unmixed air streams. Adding more sampling points showed that activity in different quadrants varied by as much as a factor of 2, so the sampling point was relocated at the 50-ft level in the stack. Comparison between the two sampling points showed that the values were 7% lower at the 50-ft point (Landon 1960).

To improve the accuracy of the quantitative integration of the curve, the paper strip chart speed was increased by a factor of 5 to enlarge the area under consideration and more accurately integrate brief releases. An electronic integrator was installed on July 24, 1957, to digitally integrate the cumulative curies of tritium discharged from the stack. A comparison of the old and new methods showed that electronic integration results were 9% higher than manual integration results. The relatively large releases from furnace changes and discharges were in closer agreement at 2% (Landon 1960).

The electronic integrator, with seven-decade dynamic range, was built to track the total amount of radioactive material released from the 232-F stack. The output current from the Kanne

chamber was recorded by a Beckman logarithmic electrometer and a Brown strip chart recorder. The control shaft followed the position of the Brown recorder indicator by a servomechanism. An oscillator attached to the control shaft oscillated with a frequency that was exponentially proportional to the vertical position of the control shaft. This output frequency was linearly proportional to the Kanne chamber current. A 13-decade counter totaled the cycles from the oscillator to integrate the amount of radioactivity released. Because the oscillator had a dynamic range of only three decades (20 to 20,000 counts per second [cps]), an automatic switching circuit was provided to inject the signal pulses at different decade positions in the counter chain to perform the function of frequency multiplication (Du Pont 1957a).

This integrator was used in 232-F until a few months before shutdown, when it was moved to 232-H. Originally, its maximum range was 6300 Ci min^{-1}. This was expanded after two stack releases exceeded that value (Landon 1960). After the electronic stack integrator was installed in 1958, a 2-week calibration period followed. Manual and electronic integration were compared, after which a controlled tritium release was used to check the monitor (Du Pont 1958a: August 30, 1957–May 9, 1958). The stack integrator was placed in routine operation, and it agreed with the manually integrated values to within 20% (Du Pont 1958a: September 13–19, 1958). Accuracy of these measurements through time was unknown, and it was concluded that the electronic integrator was not yet to be totally trusted (Miller 1959). Unfortunately, no further details were provided. It was not until the mid-1970s that the stack monitor integrator (SMI) was introduced as a primary method of monitoring stack releases (Kilpatrick 1975). Calibration of the stack monitor with known quantities of tritium was discussed in the January 1958 Monthly Progress Report (Du Pont 1958b). It was indicated that the reported losses were 15 to 20% below the actual values. In the July 1958 Weekly Progress Report (Du Pont 1958a: July 1–11, 1958), the accuracy of stack monitor results was questioned during one 2-day period when the Beckman amplifier was known to be drifting as much as 25%. After the amplifier was repaired, the integrated and analyzed results from a controlled tritium release agreed within 2%. By the third week in August, the total discharge to the stack was measured and agreed within 1% of design flow (Du Pont 1958a: August 23–29, 1958).

The stack monitor with controlled releases was not calibrated during the third week in September 1958 because of problems with leaks in sampling equipment (Du Pont 1958a). A controlled tritium release to the stack, carried out during the second week in October 1958, showed that calculated and integrated results agreed within 10%. Stack monitor response to the probe source was within limits the entire week (Du Pont 1958a; Du Pont 1958c).

Miller (1959) is a response to a report by the Atomic Energy Commission (AEC), the U.S. Government agency overseeing the national laboratories, that reviewed the accountability of nuclear materials at SRS. This memo indicates that in February 1959, the amount of tritium that was captured in the zeolite (stripper) beds in 232-H and 234-H was not known. AEC had recommended that better methods were needed to determine the quantities contained in the beds. (Zeolite beds were installed to capture tritium that was released during routine operation. In addition to providing protection to the environment, it enabled the capture and recharged of the tritium so it was not lost.) The method would attempt to complete a mass balance of all tritium produced; the losses determined by pressure, volume, and temperature measurement; and mass spectrometric analysis of the remaining tritium contents of process vessels before discharge to the stack.

The Health Physics Department reported tritium releases hand-integrated from the Kanne monitor paper strip charts (considered to have a ±10% uncertainty) (Miller 1959). When the results of the stack monitor and the spectrometric analysis of the material believed to be released were compared, there appeared to be a discrepancy. The difference resulted from the lack of measurement of all process vessels by the mass spectrometer (resulting in lower values than the Kanne monitor). It was concluded that the releases were recorded by the stack gas monitor (Miller 1959).

In a report on the accidental release of tritium gas from the tritium processing facility on December 31, 1975, the FORMS sampler, designed to differentiate between the forms of tritium released, was used. In addition to the Kanne monitors for total tritium releases, it was indicated that they have been collecting separate samples of elemental tritium gas and tritium oxide since August 1974 (Jacobsen 1976). In 1978, Johnson recommended that the HT/HTO atmospheric release data should be monitored from 232-H and 234-H continuously (Johnson 1978).

No other specific information on changes in the tritium monitoring equipment at the Tritium Facilities was found for this era, other than the full implementation of the SMI in the 1970s.

1980s

In June 1982, it is reported that a 6500 ft^3 hold volume tank was installed in 300-H to contain accidental releases of tritium from 234-H (Du Pont 1982). Sensors in the process hoods were designated to automatically trigger diversions to the hold volume tank. Before this change, the monitoring capabilities of tritium diverted to the hold volume were limited to approximately 0.05 g (about 470 Ci). A monitor-integrator was installed to provide sufficient information on the activity in the hold tank to decide whether to recover the tritium.

Zeigler (1986) presented a review of radiological effluent monitoring, analytical techniques, and reporting. This included the current status of tritium monitoring at the SRS and the factors contributing to the uncertainty in the measurements. He stated that,

> There are a few minor radioactive effluents that are monitored by Area Survey HP personnel, but the releases are not tabulated and reported because they are small. Overall the most important improvements needed in SRP effluent monitoring relate to measurement of volumes of air and liquid that are released. In many cases, volumes are estimated based on fan or pump capacities or on measurements that were made at some time in the past. Frequently, there is no documentation of the methods used to obtain the volumes that are used.

Zeigler (1986) reported the following measurements being made at the Tritium Facilities in 1986:

> To determine the total tritium releases from the Tritium Facilities stacks (232-H [2 stacks], 234-H, and 238-H stacks), the peak areas on Kanne recorder charts were integrated using empirically determined factors. Automatic integration of these releases was also available. The form of tritium is determined by an on-line FORMS monitor in 234-H and by molecular sieve samplers in 232-H. Back-up molecular sieve samples were also collected in 234-H. Molecular sieve samplers were also maintained

in the 232-H stacks by the Environmental Transport Division of SRL. All tritium released from 238-H was assumed to be in oxide form. On-line tritium FORMS monitors are also being installed in 232-H stacks and were expected to be operational by the end of 1986. Stack flows were determined manually when requested specifically, although there is no frequency set for these measurements.

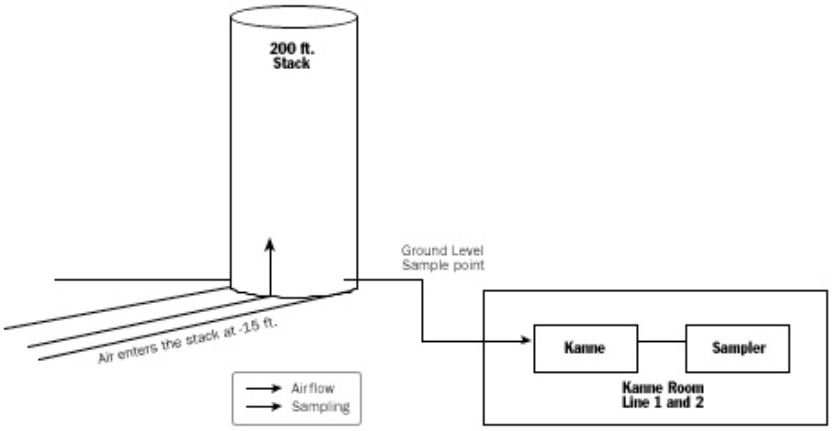
The 772-F stack is not monitored for tritium at this time, but a new system is being installed. The tritium releases from the 244-H, RBOF stacks, are determined from the stack Kanne chamber by integrating areas on the recorder chart. The releases are reported in monthly radioactivity release reports. The F and H areas tank farm waste tanks, diversion boxes and pump pit exhausts are not routinely measured for tritium releases. A HP survey done in 1983 indicated that the annual tritium releases from 11 of the most likely tanks to contain tritium were less than 100 Ci y^{-1}.

An Air Effluent Monitoring Procedure for the FORMS monitor indicated that in the 1990s, FORMS monitors were operating on Lines 1, 2, and 3 in 232-H and in 234-H (Westinghouse 1991c). Further explanation of how the monitoring results from the molecular sieves and FORMS monitors were used was not found. Figures 4.1-2, 4.1-3, and 4.1-4 show the airflow schematics indicating the air sampling points in facilities 232-H, 234-H, and 244-H, respectively.

Releases

In 1956, 232-F tritium loss was determined based on material balance from product input after extraction (Miller 1956b). It seems that there was considerable interest in limiting the releases because of economic considerations. Stack losses from 232-F averaged 30,000–40,000 Ci mo^{-1} under routine operating conditions during 1955 and 1956. These releases were larger than had been predicted. The primary sources identified were miscellaneous process leaks and the extraction furnaces. It was reported that the experience in 232-F showed that ≥60% of the tritium released was quickly converted to the oxide through exchange with water vapor, but no data were presented to support this report (Denham 1956).

Start up of tritium processing in 232-H was scheduled for July 1, 1957 (Du Pont 1957b). In August 1957, a zeolite bed for absorption was installed in 232-H Line 1. It was estimated that a 33% reduction in tritium losses occurred after installation (Harris 1957). Harris (1957) speculated that based on this result, 80% recovery of product could be attained. Estimates were made for the minimum amount of product that would be lost from the 232 Buildings: 232-F 1.5 l d^{-1} or 12 g mo^{-1} (or about 117 kCi mo^{-1}); 232-H 4.5 l d^{-1} or 36 g mo^{-1} (or about 350 kCi mo^{-1}); and 234-H 7.5 l d^{-1} and 60 g mo^{-1} (or about 585 kCi mo^{-1}).



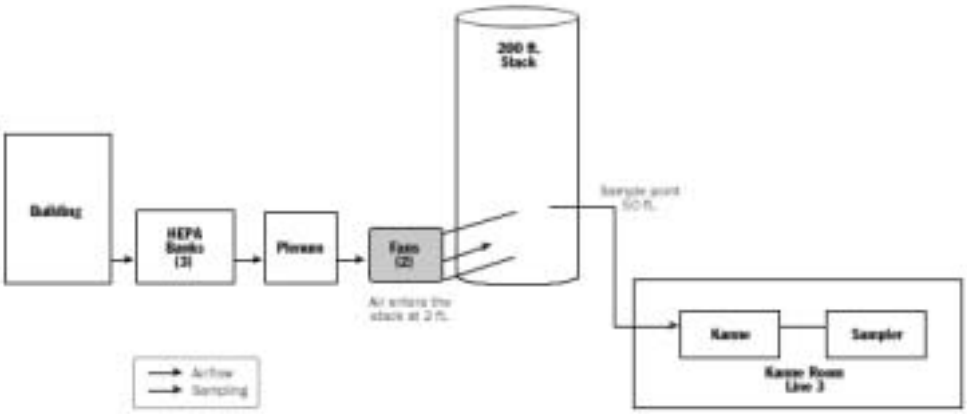


Figure 4.1-2 a and b. Airflow schematic for the lines in Building 232-H (Westinghouse 1991a).

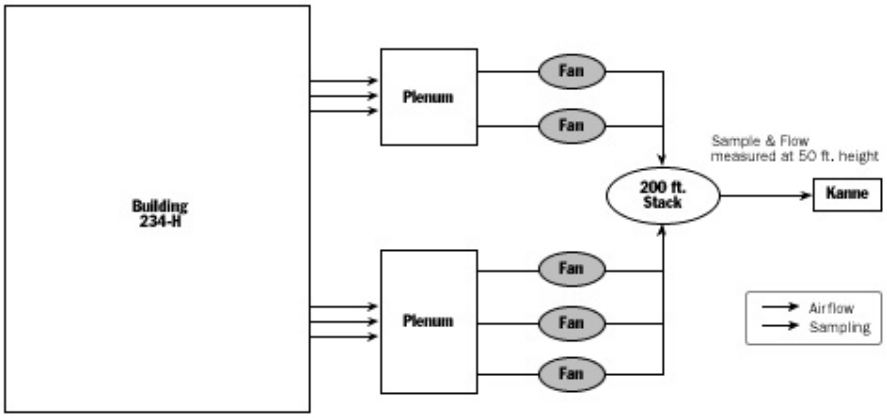


Figure 4.1-3. Schematic of air flow in Building 234-H.

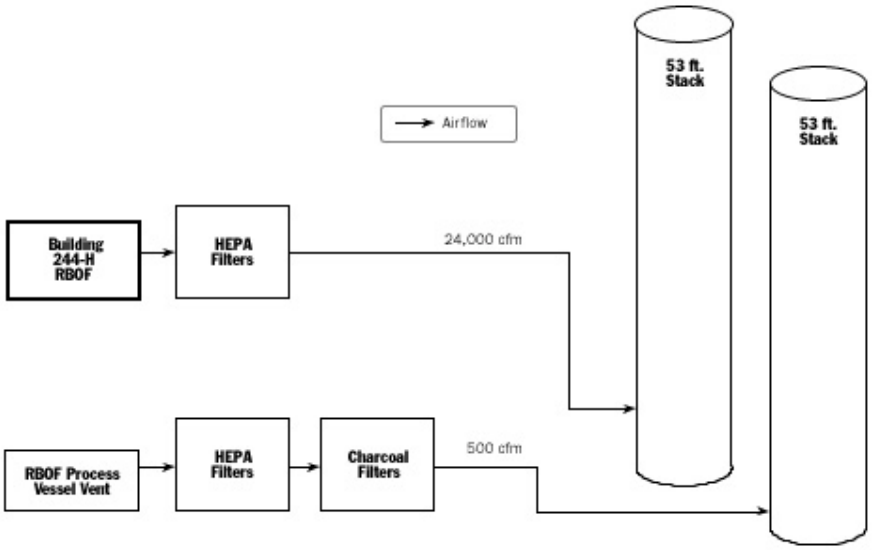


Figure 4.1-4. Schematic of airflow in Building 244-H.

In September, stack losses from 232-H were reported to be high during the first 20 runs of the month, but they were reduced (McKeller 1957). Most of the releases were due to by-product and first evacuation gas. McKeller (1957) indicated that the use of a stripper brought about a 70% reduction in the first evacuation and a lesser reduction in the by-product loss. This appears to be referencing the zeolite beds installed, but the memo also indicates that "the process design for a permanent stack stripping system is being developed."

During the early operation of 232-H, the product losses shown by the stack monitor were higher than expected, so the accuracy of the stack monitor's response was questioned (Butler et al. 1957). A series of tests was carried out to look for possible reasons for the discrepancy. These tests included

- Surveys for particulates
- Effects of argon introduction
- Release of known quantities
- Measurement of the presence of oxides during different operations.

Known releases of 25 Ci and 130 Ci were introduced into the main air exhaust and showed that the monitor results were within 10% of the amount released, confirming the calibration of the monitor. Additional work showed that about one-half of the product released was in oxide form. It is not clear if this determination of the chemical form of the tritium released was a result of the same work mentioned in other documents. Tests showed that the addition of argon gas acted only as a dilutant in the Kanne chamber (Butler et al. 1957).

Moisture in the furnace appeared to be a main cause of high stack losses in 232-H. An additional zeolite bed was added to the test stripper assembly. It had one copper oxide (CuO) and two zeolite beds in series. Data from one run indicated that 50% of the losses came from the last part of the run when the effluent had not normally been stripped. The memo also indicated that the following determinations were planned as part of the permanent stripper evaluation; however, documentation of the results was not found:

- Oxide conversion efficiency
- Oxide absorption efficiency
- Measurement of total water from the system
- Recovery form zeolite
- Oxide decomposition.

The largest losses occurred during the charge-discharge operation in 232-F. The drop in releases is attributed to cleanliness of the system. Increased losses during zeolite bed trials could have been associated with gradual buildup of condensed metal distillate on the furnace cover. The losses in 232-H gradually decreased after the zeolite beds began operation (Bird 1957a). In December 1957, it was reported that an efficient permanent stack stripper was installed on Line 2 (Bird 1957b).

The variation of the form of the release of tritium (elemental, HT, or tritiated water vapor, HTO) depended on the nature of the work being done in the facilities. Murphy et al. (1991) cites studies that show the fraction of HTO, the form of greater concern to this study, was 56% over a 3-year period in the 1970s. In a 3-year period in the 1980s, the fraction of HTO was cited to be 48% (Murphy et al. 1991). The fraction HTO released was likely dependent on the type of operation carried out at the facility. Because this information is not available, it would not necessarily be valid to assume the fraction HTO released on a given day, week, or month; however, at least some data are available to assume about 50%. Unless other data are found, it

can be assumed that about one-half of the tritium releases from these facilities was HTO, which is representative of operations after 1970. The section on inadvertent releases in this report presents cases of large inadvertent or accidental releases where the ratio of tritium to tritiated water (HT/HTO) was known.

TRITIUM SOURCE TERM RECONSTRUCTION

The releases of tritium from the SRS are generally very well documented on a monthly basis in published internal reports. Records of releases have been kept since almost the beginning of the operations. Yearly data are very well summarized for all main release points in two summary documents published in 1991 (Cummins et al. 1991; Murphy et al. 1991). In an effort to corroborate the summarized data, we conducted a record search to find original data to reconstruct the monthly releases wherever possible. We reviewed hundreds of documents, including numerous facility report series. The main sources of information were SRS weekly and monthly reports. The best estimate of the source term has been compiled on monthly where possible (in some of the earlier years, it was compiled biannually). Atmospheric tritium releases were separated into reactor releases and tritium facility releases. Detailed data are available electronically via hyperlink by selecting either release type above.

The documents used to develop the source term compiled in the above data files have not been cited in connection with the individual release values, but they are listed in the reference list at the end of the chapter by year, in the ”Reference - Source Term” section. Lee (1998b) contains even more detailed tritium release data.

Whenever possible, we checked the release values with shift or daily data found on datasheets or handwritten laboratory logbooks for both the reactors and tritium facilities.

In general, there is quite good agreement between the different release values reported in the different sources. In fact, in most cases, the published aggregated values are larger than the sum of the logbook or datasheet values reported for shorter time scales. Because in most cases the summary values were within 10% of the published values, it is likely that most “unchecked” monthly values are also valid. It is possible to conclude that other small sources were not published independently in regular reports, but they were included in the Site-wide totals. It is also possible to speculate that the reported values were “rounded-up” when published. It is, however, impossible to accurately reconstruct a daily or weekly history of releases.

No consistent monthly release trends across the year were found for the reactor data, so it was not considered prudent to artificially break down the 6-month and annual totals by month (when monthly data were unavailable). When data for releases from the reactor disassembly area were available, they were included (at the end of each reactor section). The total annual releases from each reactor are plotted over time in Figures 4.1-5, 4.1-6, 4.1-7, 4.1-8, and 4.1-9. Figures 4.1-10, 4.1-11, 4.1-12, 4.1-13, and 4.1-14 provide plots of the annual releases from the Tritium Facilities.

Lee (1998a) concluded that the SRS reactor data found were insufficient to reproduce the releases reported by SRS using original raw data. Based on the comparisons that could be made, evaluation of equipment and methods, and the uncertainty analysis, we have concluded that it was possible to reconstruct a credible set of best estimate tritium monthly release data from the reactors and Tritium Facilities. These best estimates should be used as a range suggested by the

uncertainty analysis and should be considered when performing out future steps in the dose reconstruction process.

Based on the review of the available data sources, we concluded that the SRS reported the monthly tritium release data that were measured at the facilities. The review of the quality of the atmospheric tritium release data reported by SRS has given us no indication that serious or consistent errors were made in monitoring Site tritium releases. Details of this review are provided in Appendix E.

The releases are reported by month because it was not possible to reconstruct daily releases for the whole operational period of SRS for all facilities. During the next phase of the SRS dose reconstruction, it will not be possible to model daily environmental dispersion and human exposures. The next section summarizes the uncertainty calculations that we performed for the atmospheric tritium source term. See Appendix E for details of these calculations.

Table 4.1-2 provides an overview of the data that were used to develop the tritium source term. All documents used to develop the source term are included in the reference list at the end of this chapter (organized by year).

Table 4.1-2. Tritium Release Documentation Used To Reconstruct the Source Term

Year	232-F	232-H	234-H	238-H	244-H	Reactors	Others
1954	monthly; beginning in Nov.					monthly, start in July	
1955	monthly; Jan–Nov					monthly Jan–Sept, biannual	
1956	biannual	biannual (July–Dec only)	biannual (July–Dec only)			biannual	
1957	biannual; monthly (except Nov.–Dec.)	monthly (except Aug.)	monthly			biannual	
1958	CLOSED	monthly	monthly			biannual	
1959	⇓	monthly	monthly			biannual	biannual
1960		monthly	monthly			biannual	biannual
1961		monthly	monthly			biannual	biannual
1962		monthly	monthly			biannual	biannual
1963		monthly	monthly			monthly	
1964		monthly	monthly			monthly (R until July)	biannual
1965		monthly	monthly			monthly	biannual (July–Dec only)
1966		monthly	monthly			biannual	biannual
1967		monthly	monthly			monthly	
1968		monthly	monthly	START OF OPERATION		biannual (L from Jan-July only)	annual
1969		monthly	monthly	monthly (beginning in May)	START OF OPERATION	biannual	annual
1970		monthly	monthly	monthly	monthly (for some months)	annual	annual
1971		monthly	monthly	monthly	monthly (Jan-Feb. only)		
1972		monthly	monthly	monthly		monthly	
1973		monthly	monthly	monthly		monthly	monthly
1974		monthly	monthly	monthly		monthly	monthly
1975		monthly	monthly	monthly	monthly	monthly	monthly
1976		monthly	monthly	monthly	monthly	monthly	monthly
1977		monthly	monthly	monthly	monthly	monthly	monthly
1978		monthly	monthly	monthly	monthly	monthly	monthly
1979		monthly	monthly	monthly	monthly	monthly	monthly
1980		monthly	monthly	monthly	monthly	monthly	monthly
1981		monthly	monthly	monthly	monthly	monthly	monthly
1982		monthly	monthly	monthly	monthly	monthly	monthly

Table 4.1-2. (Continued)

1983		monthly	monthly	monthly	monthly	monthly	monthly
1984		monthly	monthly	monthly	monthly	monthly	monthly
1985		monthly	monthly	monthly	monthly	monthly	monthly
1986		monthly	monthly	monthly		100 areas total	
1987		monthly	monthly	monthly			
1988		monthly	monthly	monthly			
1989		monthly	monthly	monthly		monthly	monthly
1990		monthly	monthly	monthly			
1991		monthly	monthly	monthly		monthly	monthly
1992		tritium facilities total (monthly)	tritium facilities total (monthly)	tritium facilities total (monthly)	tritium facilities total (monthly)	monthly (except Nov.)	monthly (except Nov. and Apr.)

Uncertainty of Source Term Estimates

The accuracy of the atmospheric source term estimates is largely dependent on the type of monitor used to measure the releases. A detailed description of the equipment and the data used in our uncertainty analysis appears in Appendix E. It has not been possible to determine the exact times that each type of equipment was installed and operated in each of the facilities. However, in reviewing the documentation, it is possible to find dates on equipment operation procedures or reports of plans or prototype tests.

For the purposes of this work, general use of the various tritium monitoring systems that were used onsite since the 1950s are documented as

Dehumidifier	1954–1986 possibly longer (reactors)
Silica gel	1954–1958 (reactors and disassembly areas)
Kanne ionization chambers	1954–present (Tritium Facilities)
Stack tritium monitor	1970–1988 (reactors)
Berthold tritium monitor	1988–present (reactors)
Stack monitor integrator	1974–present (Tritium Facilities)
FORMS	1985–present (Tritium Facilities and reactors)

It was not possible to determine exact start and stop dates because systems were tested and then kept running in parallel to provide some redundancy if the main system failed. It is not clearly stated when the measurement results of one system became the primary monitoring device in replacement of an older version. For example, in early years, it is not clear when silica gel was no longer used for stack monitoring (replaced by dehumidifiers) and shifted to general area monitoring for health physics protection purposes. The documentation we found refers to both the silica gel and dehumidifier monitors being used for stack monitoring.

The uncertainty assessments for each of the monitoring systems have taken into account the representativeness of the sample; the frequency of sampling; efficiency of sampling; the effects of environmental conditions; the accuracy of the measurements; and finally, errors in reporting results. Using a Monte Carlo sampling technique, we developed uncertainty estimates, which are reported in Table 4.1-3. Details of the analysis and further explanations can be found in Appendix E.

Table 4.1-3. Uncertainty Estimates for Tritium Releases Reported by Monitors on SRS

Monitor	For results in years	Facilities	Distribution	Range[a]
Silica gel	1954–1958	Reactors	Lognormal	GM = 1.15 GSD = 2.16
Dehumidifier	1954–1970	Reactors	Lognormal	GM = 0.99 GSD = 2.3
Stack tritium monitor	1970–1988	Reactors	Lognormal	GM = 0.91 GSD = 2.19
Berthold tritium monitor	1988–present	Reactors	Lognormal	GM = 0.99 GSD = 1.16
Kanne chamber	1954–1974	Tritium Facilities	Lognormal	GM = 0.9 GSD = 1.8
Stack monitor integrator	1974–present	Tritium Facilities	Lognormal	GM = 0.9 GSD = 1.8

[a] GM = geometric mean, GSD = geometric standard deviation, SD = standard deviation.

To apply these results to a monthly tritium atmospheric source term, use the monthly value for a given facility and select the appropriate uncertainty distribution (based on year and facility). To obtain the median value on the distribution, multiply the monthly best tritium release estimate by the geometric mean (GM). To obtain the 5th percentile value, divide the median value by the square of the geometric standard deviation (GSD). The 95th percentile is calculated by multiplying the median value by the square of the geometric standard deviation. The uncertainty for annual tritium release estimates has been calculated, and is compiled in a reactor release file and a tritium facility release file. These tables contain annual release estimates along with 5th and 95th percentiles and the calculated GM and GSD.

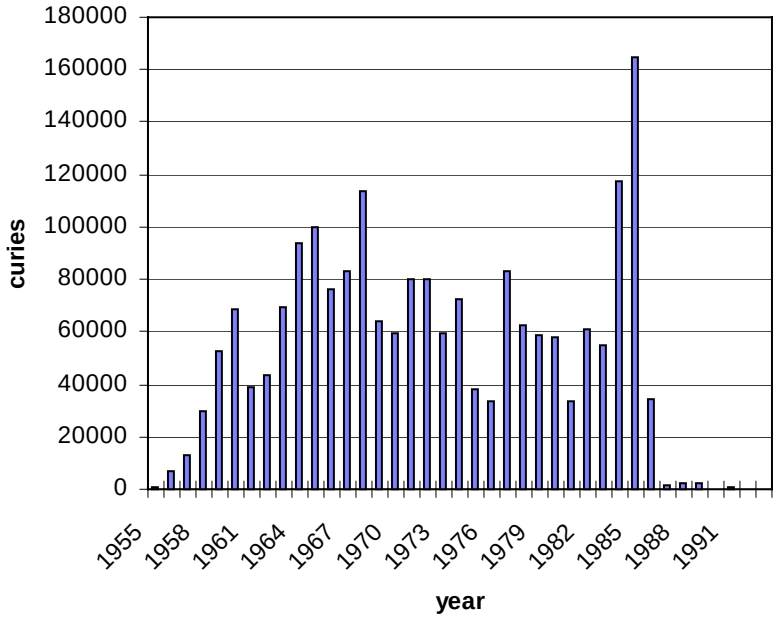


Figure 4.1-5. Annual atmospheric tritium releases from reactor 105-C (Ci). Link to tabulated data.

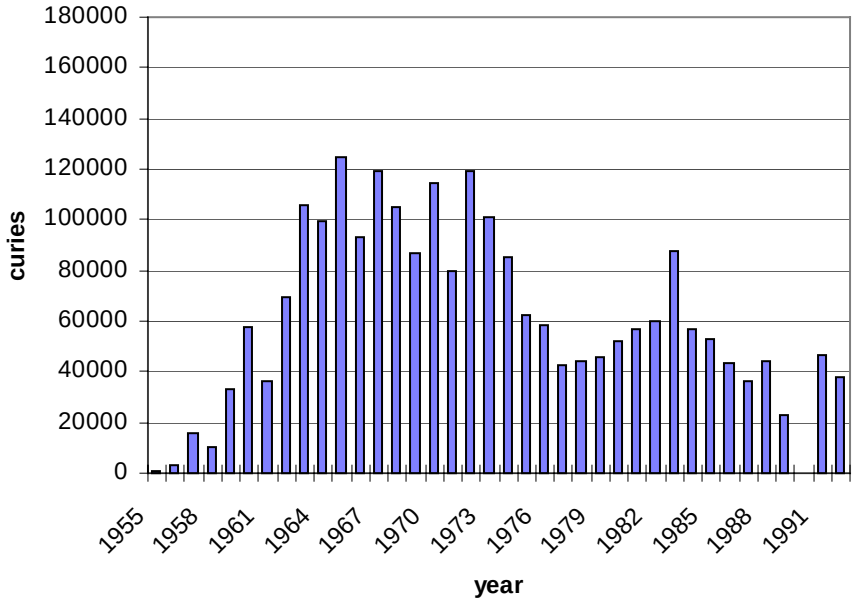


Figure 4.1-6. Annual atmospheric tritium releases from reactor 105-K (Ci). Link to tabulated data.

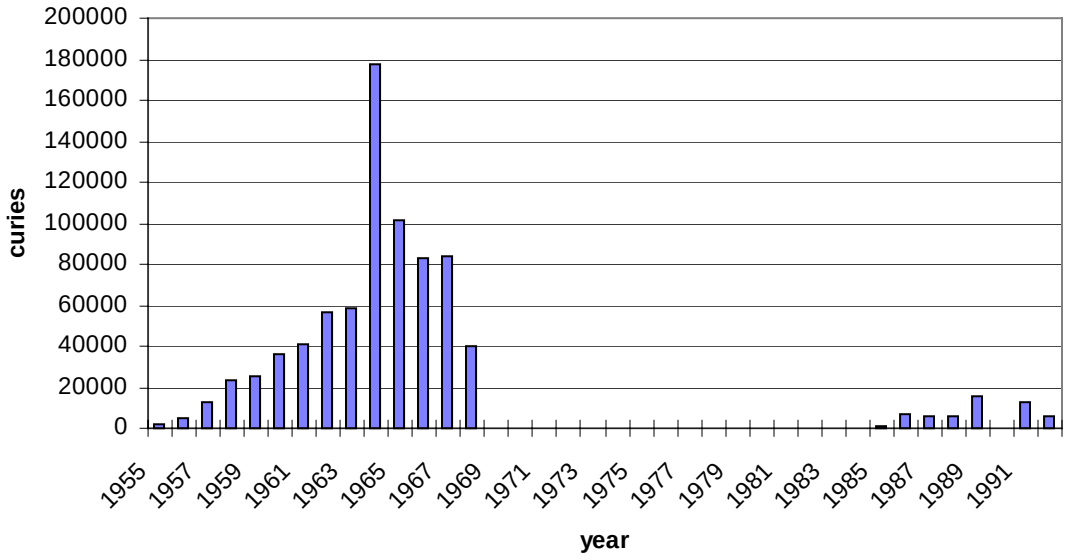


Figure 4.1-7. Annual atmospheric tritium releases from reactor 105-L. Link to tabulated data.

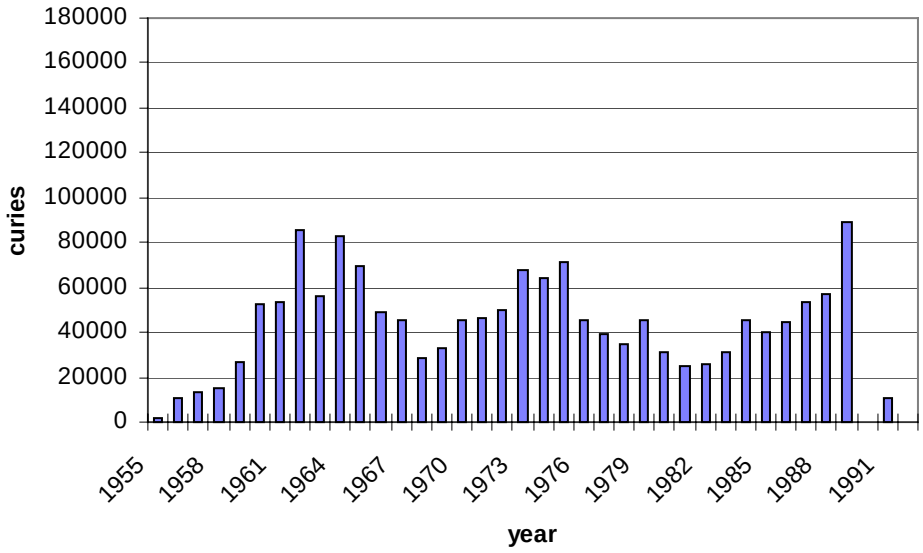


Figure 4.1-8. Annual atmospheric tritium releases from reactor 105-P (Ci). Link to tabulated data.

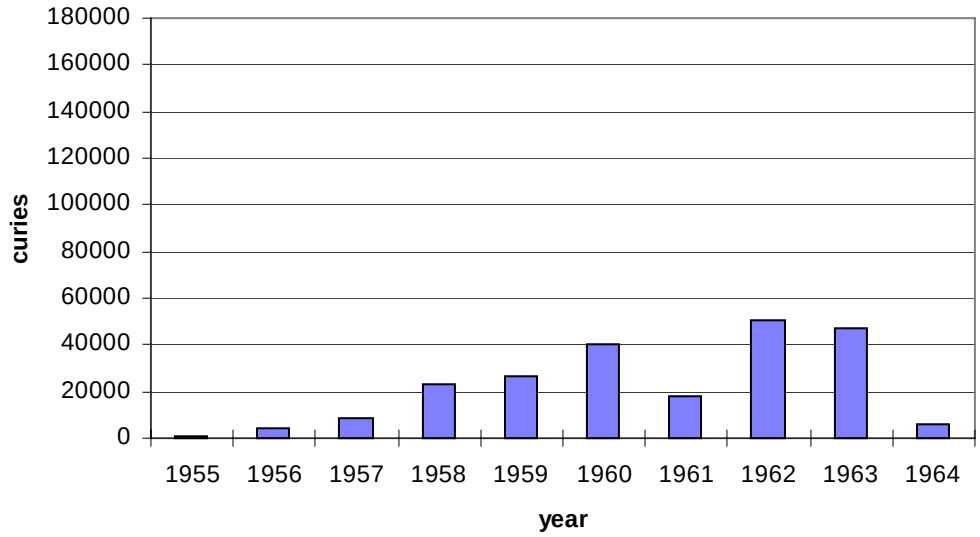


Figure 4.1-9. Annual atmospheric tritium releases from reactor 105-R (Ci). Link to tabulated data.

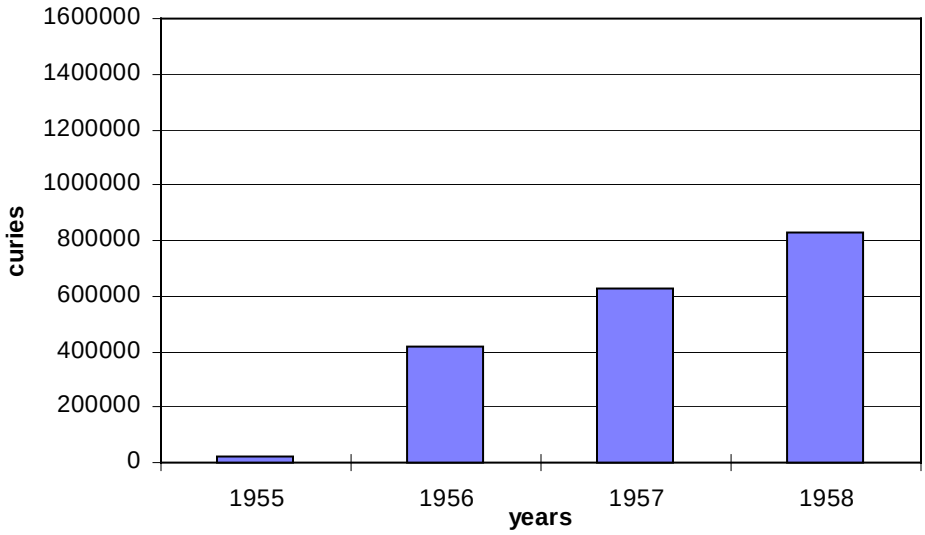


Figure 4.1-10. Total annual tritium releases (Ci) from Building 232-F. Link to tabulated data.

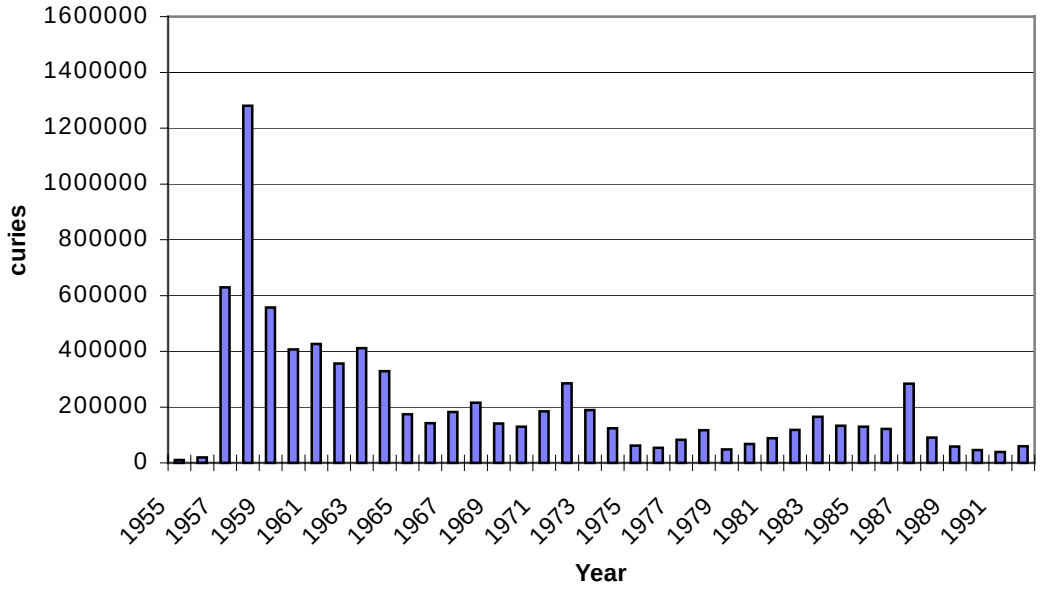


Figure 4.1-11. Total annual tritium releases (Ci) from Building 232-H. Link to tabulated data.

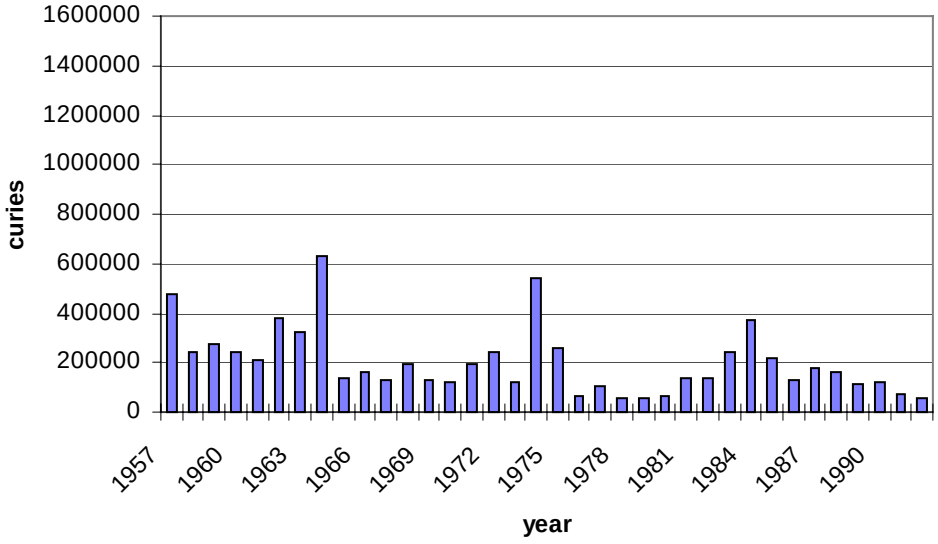


Figure 4.1-12. Total annual tritium releases (Ci) from Building 234-H. Link to tabulated data.

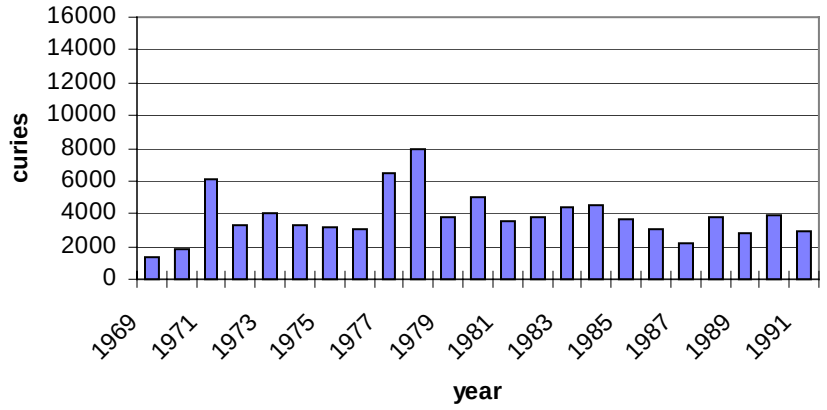


Figure 4.1-13. Total annual tritium releases (Ci) from Building 238-H. Link to tabulated data.

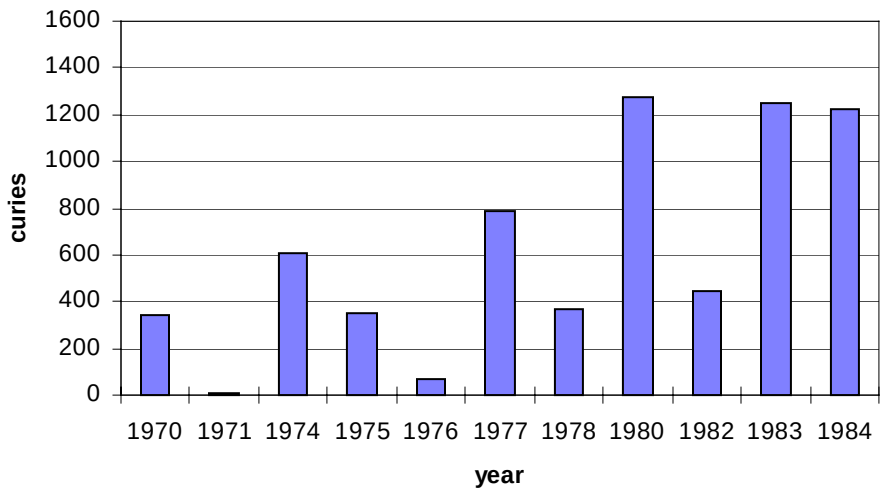


Figure 4.1-14. Available total annual tritium releases (Ci) from Building 244-H. Link to tabulated data.

We believe that the monthly data presented here include all the releases, both routine and inadvertent. However, some of the inadvertent releases were large and occurred over a short period of time, so it is likely that future dose reconstruction work will address these separately. To support this, we have included information we found on the details of inadvertent releases, which are discussed in the following section.

INADVERTENT RELEASES OR INCIDENTS

Introduction

Over the years at the SRS, inadvertent releases were defined as those releases that were not planned and resulted from an unexpected sequence of events or failures in a process. Many incidents occurred, resulting in inadvertent releases of tritium. These were documented in Incident Reports and an incidents report database. Incident Reports were found for releases as early as 1956 (Miller 1956c). All evidence from our record search leads to the conclusion that the inadvertent releases were included in the total monthly and yearly tritium values reported because no large release that were identified seem unaccounted for in the monthly data reported. Starting in the 1970s, any large releases with public health, worker safety, or economic implications, were assessed in special reports written by SRS staff.

We have researched and reported routine and inadvertent releases separately to (1) ensure that the releases (which are expected to be larger than usual) have been included in the release totals published by SRS, and (2) identify these events for possible special treatment during the future phases of the SRS Dose Reconstruction Project.

A report summarizing the results of Phase I of the SRS dose reconstruction project discussed unusual events and accidents that caused environmental releases considered significant to the dose reconstruction process (Meyer et al. 1995). In developing a list of the most important events or accidents, it became clear that the definition of important releases would greatly influence the number of occurrences that would be included as distinct analyses in the future dose reconstruction.

In summaries of the tritium releases from SRS, Murphy et al. (1991) used the term “inadvertent tritium releases” to the environment and designated recorded releases of over 700 Ci to the atmosphere as “significant” releases. This level was chosen because it is roughly 0.1% of the average annual tritium atmospheric releases. In this report, we used the 700-Ci level designated in Murphy et al. (1991) to define significant releases.

Because inadvertent releases were defined as unplanned, the relatively large releases that occurred during the early days of operation as a result of developing new facilities and procedures were not flagged as unusual events. They were included in the routine reporting of releases (Murphy et al. 1991; Du Pont 1955). During later times, much smaller releases that occurred as a result of some malfunction were considered and documented as inadvertent releases. We believe that the comparison of specific incident release with the published monthly release values shows that the tritium release has been included in routine reporting. For example, if you look at the release from B234-H over the year of 1974, you will see the monthly release ranged from 3580 Ci to 8247 Ci, excluding the month of May, during which a large inadvertent release occurred. On May 2, 1974, a release of 479,000 Ci occurred. The May 1974 release was reported to have been 481,954 Ci. This value allows for a “routine” monthly release of about 3000 Ci (which is a bit low but close to the range seen for the rest of the year) plus the inadvertent large release.

Another main source of data for inadvertent releases (as they were defined at the time) was the Tritium Area Facilities Fault Tree Data Storage and Retrieval System, or TAFFTDSRS, (TAFFTDSRS 1994) beginning with the year 1956. Inadvertent releases between 1959 and 1974 are indicated to have been a result of reactor operations and from reactor moderator losses (Murphy et al. 1991), but the few unusual events resulting in increased atmospheric releases of

tritium from the reactors were found in reports or logbooks, not the database. The releases from the Tritium Facilities, which are reported in detail, have been attributed to aging equipment (Murphy et al. 1991).

Search profiles were prepared for the four unclassified data banks during Phase I of the SRS dose reconstruction project (Meyer et al. 1995), and a separate search profile was prepared for the TAFFTDSRS (TAFFTDSRS 1994). The entries range from procedures violations and minor equipment malfunctions to incidents that released radioactivity or chemicals or resulted in injury to workers. The data banks are used primarily for probabilistic risk assessment, training, trend analysis, safety analysis reports, and management and administrative controls analysis. Most of the detail in the incidents listed in the database pertains to the workers, the SRS facilities, and the immediate onsite environment and equipment.

The search profile for the TAFFDSRS (TAFFTDSRS 1994) produced a printout listing 2994 incidents for just the five different curie levels (1 to 100 Ci; 100 to 1000 Ci; 1000 to 10,000 Ci; 10,000 to 100,000 Ci; and greater than 100,000 Ci). For each incident, the database lists the unique incident identification number, source of information describing the incident, date of occurrence, and description of incident. Another list of approximately 3000 incidents was printed out under the remainder of the tritium search. The actual incidents listed in these two printouts are approximately the same (e.g., a release under airborne also would be listed in one of the curie categories). Incidents of tritium releases over 700 Ci were used as the basis for documenting inadvertent releases for this report (link to tabulated data). The data from this printout were augmented by information found in period incident reports and limited information found in logbooks.

Documented Inadvertent Releases

Reactors

The investigation of unplanned and accidental releases of tritium from reactors has uncovered very little specific information. Most of the details that were found are for the Tritium Facilities, where larger amounts of tritium were at potential for release. A list of “Inadvertent Tritium Releases to the Environment from SRS Operations,” found among C. Zeigler’s personal files, notes three specific cases of atmospheric releases from reactors: (1) November to December 1961 (20,000 Ci HTO) from P-reactor stack, (2) March to June 1977 (83,000 Ci HTO) from C-reactor, and (3) early in 1978 (62,810 Ci HTO) from C-Reactor stack (Zeigler 1994). (The notes are varied and are not all dated. The year 1994 depicts the year the notes were reviewed.) However, we found no other specific mention of these releases as unplanned incidents or accidents on specific dates or times in any other report.

The P-Reactor release in 1961 (no date indicated) could have been included in the 6-month releases reported for 1961, which take a jump of 18,500 Ci from the first 6 months to the second 6 months. These biannual values were published and are also shown in the logbook data (Table 4.1-4). Therefore, in this case we cannot draw a clear conclusion regarding the release indicated in the logbook.

The two C-Reactor release values mentioned in Zeigler’s logbook are the same values as were reported for the yearly total atmospheric release. No further information is provided and no

mention of this is indicated in any other source, which would probably have occurred during the 1970s. In this case, it is not possible to interpret this information.

Table 4.1-4. P-Reactor Tritium Releases Noted in Logbook (Zeigler 1994)

Year	Releases (Ci)	
	January–June	July–December
1960	28100	24400
1961	17600	36090
1962	37300	48100

Tritium Facilities

For the Tritium Facilities (Buildings 232-H, 234-H, and 238-H), it became clear upon reviewing the results of the database search, logbooks, and incident reports that there were hundreds of incidents resulting in a wide range of releases. These releases had been individually recorded and included in the monthly release reports. For this report, we have used the level that was defined as significant by Murphy et al. (1991) and only report releases of over 700 Ci to the atmosphere. This is roughly 0.1% of the average annual release to the atmosphere.

With this criterion, we identified 289 events in the time period from July 1956 to June 1990. Figure 4.1-15 plots these incidents and limited details of these events can be found in the next section of this chapter.

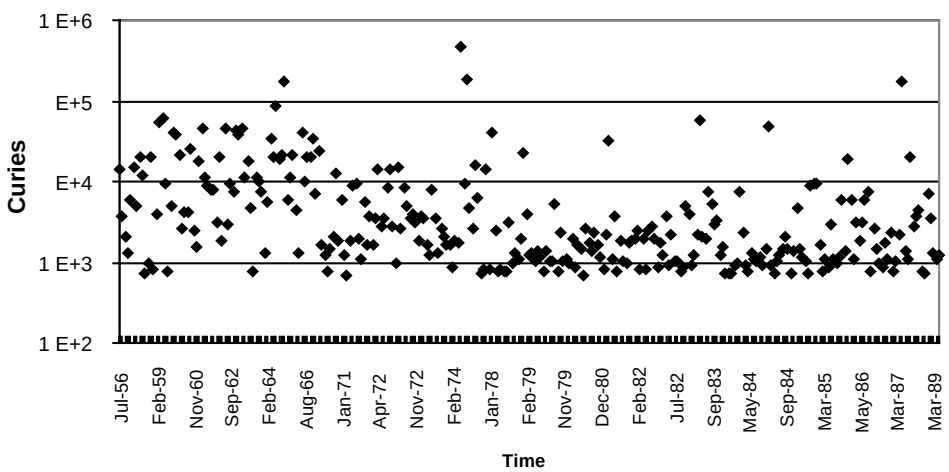


Figure 4.1-15. Summary of incidents of inadvertent or accidental releases of tritium to the atmosphere (over 700 Ci). The y-axis of this figure has values shown in scientific notation. A value such as 1 E+6 is equivalent to 1 x 10^6 or 1,000,000. Link to tabulated data.

To have a better idea of the importance of these releases, Table 4.1-5 groups the data by 5000-Ci increments (above 4999 Ci) and Figure 4.1-16 plots these releases.

Table 4.1-5. Inadvertent Tritium Releases Over 700 Ci from the Tritium Facilities from 1956 to 1990

Curies	Number of events	Curies	Number of events
700–4999	2	55,000–59,999	1
5000–9999	202	60,000– 64,999	1
10,000–14,999	34	65,000–69,999	1
15,000–19,999	12	70,000–74,999	0
20,000–24,999	11	75,000–79,999	0
25,000–29,999	6	80,000–84,999	0
30,000–34,999	1	85,000– 89,999	0
35,000–39,999	3	90,000–94,999	1
40,000–44,999	4	100,000–149,999	0
45,000–49,999	3	150,000–199,999	0
50,000–54,999	3	200,000	3
		479,000	1

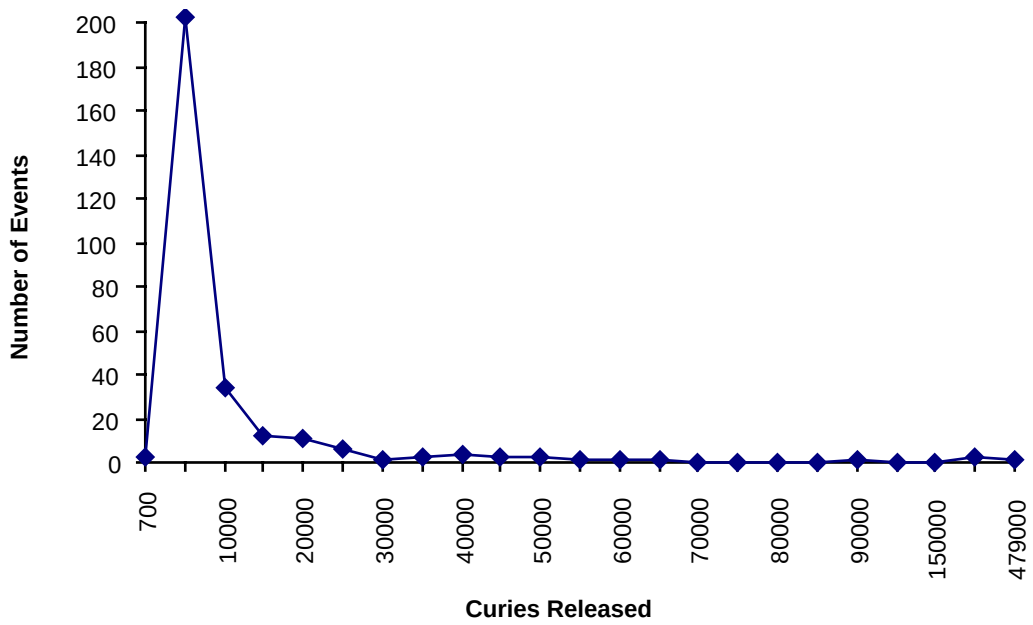


Figure 4.1-16. Activity releases from unplanned incidents and accidents (over 700 Ci releases) (based on data in Table 4.1-5). Link to tabulated data.

Special Incidents

Because of the magnitude of the release, composition of the release, or environmental conditions, more detailed studies were implemented by SRS to investigate the potential impact on

the environment and the public. In the 13 cases where more detailed information was available from SRS reports, the information is summarized below[a].

June–July 1964. Incidents resulting in around a total of 100,000 Ci loss to the atmosphere can be seen in Figure 4.1-15 during mid-1964. Although no detailed description of the incidents were found, the monthly reports describe operational incidents that were not considered to be unusual, such as routine purging of the stack system or during letdown of argon after loading room check operations (Du Pont 1964a, 1964b).

May 2, 1974. In Building 234-H, at 8:01 a.m., tritium monitors for the 200-ft exhaust stack and for the enclosure cabinet exhaust system to the process room alarmed. Over the next 4 minutes, over 99% of the 479,000 Ci of tritium gas (T_2) was released, as indicated by the stack monitor integrator (exhaust monitor) and consistent with the material balances within the measurement accuracy. Approximately 50 g of tritium (about 7 ft^3, STP), mixed with the building ventilation exhaust air, discharged from the stack at rate of 135,000 ft^3 min^{-1}. The total volume released during the 4 minutes was 540,000 ft^3 of air. It exited the 8-ft diameter stack top at a velocity of 2700 ft min^{-1}. Averaged over the 4 minutes, the release was estimated to be a concentration of 13 ppm (approximately 0.25 Ci ft^{-3}) (Morris and Scaggs 1974; Haywood 1974a). Measurements offplant indicated that less than 1% of the tritium was in oxide form (Morris and Scaggs 1974). A Kanne monitor chart is presented in the reports (Haywood 1974a, 1974b; Marter 1974; Morris and Scaggs 1974). The two peaks of the release were estimated to be at 600,000 Ci min^{-1} and 170,000 Ci min^{-1} (the tops were over the top of the chart). A much smaller peak a bit later was estimated to be 9900 Ci min^{-1} (Haywood 1974b).

It appears that the release was estimated using a manual integration technique because the electronic integrator was out of service during the release period (Patterson 1975). Another account written in 1991 indicated that the release started at 07:55 a.m. (Murphy et al. 1991). A summary of the environmental effects of the release was summarized in Marter (1974).

December 31, 1975. At 10:00 p.m. on December 31, 1975, a process failure (vacuum gauge failed releasing about 20 g of tritium) in Building 234-H resulted in a release of 182,000 Ci of tritium via the 200-ft stack. The automatic integrator was out of service because of a malfunction, so the health physics shift supervisor and inspector manually integrated the stack Kanne recorder chart. The graphic integration method they used involves making a calculation for each one-third of a minute during the time the release rate was greater than 100 Ci m^{-1} (Cofer 1976; Webb 1975; Jacobsen 1976).

The extended range stack monitor recorder peaked at a release rate of about 290,000 Ci m^{-1} and about 90 seconds later indicated a release rate of about 600 Ci m^{-1}. About 90% of the release occurred during a 1.5-minute period. The chart is attached to the report (Cofer 1976).

"Tritium absorber samples" from the 234-H stack indicate that approximately 99.4% of the release was in elemental form and 0.6% in oxide form (Webb 1975). Tritium FORMS samplers, which determine the amounts of elemental tritium gas and tritium oxide, had been installed in August 1974 (Jacobsen 1976).

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[a] We received information during the review phase of this project indicating that we should have included a tritium release from K Reactor in December of 1991 in this section. We found no detailed information about such a release, and believe that it was likely included in the monthly source term. We do, however, want to draw attention to the possibility that such a release did exist and needs to be studied in more detail in a later phase of the dose reconstruction project.

March 27, 1981. On March 27, 1981, 3.42 g (33,000 Ci) of tritium in the form of water vapor was released via the 234-H stack from 09:45 a.m. to 12:00 noon. Maintenance personnel were attempting to change a plugged inline filter (Anonymous 1981). The stack monitor indicated a loss of 308 Ci min^{-1} (0.03 g min^{-1}) as work to remove an inlet filter was being done (no water was seen). As the work continued, the loss dropped to 12 Ci min^{-1} (0.001 g m^{-1}). However, during the installation of the new filter, water dropped into the hood and the stack monitor activity increased to (2200 Ci min^{-1} [0.23 g min^{-1}]) for about 2 minutes (Green 1981). The tritium FORMS samplers indicated that the release was over 99% tritium oxide (Anonymous 1981). Tritiated water vapor was 99.7% of the release (Murphy et al. 1991).

July 16, 1983. On July 16, 1983, about 56,392 Ci (5.8 g) (Epting 1983) of elemental tritium was released from Building 234-H at 11:16 p.m. (Du Pont 1983). The release took place over a period of about 3 minutes, and only 1% of the release was in oxide form (Murphy et al. 1991; Garrett et al. 1983). The release was monitored by the Kanne chambers.

March 23, 1984. On March 23, 1984, 7673 Ci of tritium was released from Building 234H during a product transfer (Du Pont 1984a). A process line leak occurred in a process hood that began at 5:40 a.m. and continued at decreasing rate until about 9:00 a.m. From 5:40 a.m. to 8:40 a.m., 7400 Ci was released and an additional 100 Ci was released from 8:00 a.m. to 9:00 a.m. as monitored by the Kanne chambers. The tritium FORMS monitor installed on the sampling line indicated that the average composition of the release was 70% HTO/30% HT (Evans et al. 1985).

September 2, 1984. A total of 46,690 Ci of tritium was released from 234-H starting at 10:00 p.m. on September 2, 1984, until 10:00 am on September 3, 1984. The release occurred during preparation for maintenance work on a process pump. The activity was in the form of spilled HTO. Over the next several days, an additional 11,160 Ci was released for a total of 57,850 Ci by the time the cleanup efforts were completed on September 7, 1984. The tritium FORMS monitors confirmed that all the release was in oxide form (Du Pont 1984b; Anonymous 1984). Another report stated that 99% of the release was in oxide form (Murphy et al. 1991).

Another account (Hoel et al. 1990) indicated that 43,8000 Ci was released between 10:00 p.m. September 2 to 3:00 a.m. September 3. The additional 14,000 Ci was released during cleanup operations between September 3 and late in the day on September 7.

January 31, 1985. A total of 9285 Ci of tritium was released from the Tritium Separations Area from 2:00 p.m. until 5:00 p.m. The majority of the tritium (7400 Ci) was released during the first 15 minutes. About 54% of the release was in oxide form and the remaining 46% in hydrogen form (Murphy et al. 1991).

March 27, 1985. An estimated 19,422 Ci of tritium oxide was released from H-Area on March 27, 1985. The release began at 1:53 p.m. and averaged about 90 Ci min^{-1} until 5:30 p.m. (Addis et al. 1985). The rate of release was 76 Ci min^{-1} at 2:14 p.m. and 96 Ci min^{-1} at 2:31 p.m. The tritium FORMS analysis showed that the release was 99.99% oxide.

May 29, 1986. Approximately 5900 Ci of tritium was released from Building 232-H during repairs of process equipment on May 29, 1986. Approximately 95% of the tritium was in oxide form (Lawrimore 1986).

July 31, 1987. A release between 8:17 a.m. and 8:55 a.m. occurred on July 31, 1987, from the separations area stack. The total releases estimate (172,000 Ci) was based on the tritium FORMS monitor, where 2.7% was in oxide form and the remaining 97.8% was in elemental form (Murphy et al. 1991).

March 1, 1988. At 6:18 a.m., a total of 19,980 Ci was released from Building 234-H. The chart recorder peaked at approximately 90,000 Ci min^{-1}. By 6:19 a.m., the stack monitors had decreased to approximately 300 Ci min^{-1}; by 6:25 a.m., the rate was down to about 4 Ci min^{-1}. The tritium FORMS monitor showed that approximately 86% of the release (17,120 Ci) was in elemental form, and the remaining (2860 Ci) was in tritium oxide form (Lott and Mason 1988).

June 7, 1988. About 3650 Ci of 96% tritiated hydrogen gas was released on June 7, 1997, from the separations area tritium facility (Murphy et al. 1991).

October 6, 1988. Approximately 7000 Ci of tritium was released from a separations area tritium facility stack. About 11% was in the form of tritiated water vapor (Murphy et al. 1991).

December 7, 1988. Approximately 3500 Ci of tritium was released from a separations area tritium facility stack. About 99.5% was in the form of tritiated water vapor (Murphy et al. 1991).

The relative importance of the incident releases from the Tritium Facilities is shown in Figure 4.1-17. The larger releases that occurred during the early days of operation were the result of developing new facilities and procedures. They were not documented as unusual events. In later years, these incidents were considered unusual incidents or at least unplanned. Figure 4.1-17 illustrates the percentage of the total annual release attributed to accidental or unusual releases.

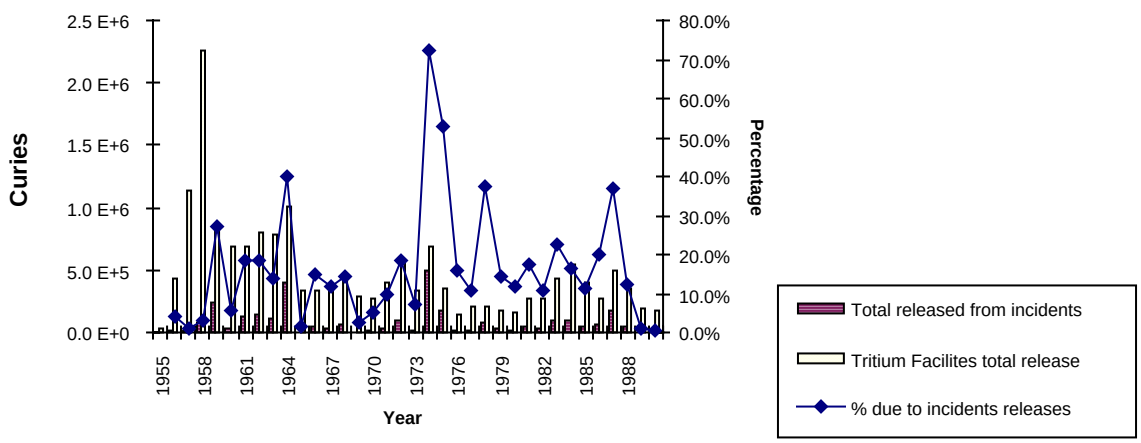

Figure 4.1-17. Annual releases (Ci) attributed to unusual incidents as a percentage of the total release from the Tritium Facilities. Link to tabulated data.

As with the source term releases, several documents that discussed inadvertent releases were identified but not explicitly cited in the text of this chapter. These document citations are included in the "Reference – Inadvertent Releases" section.

SUMMARY

We conclude that the releases of tritium from the SRS are generally well documented in published internal reports. The main sources of information were SRS weekly and monthly reports, published since near-commencement of site operations. We used daily or shift data to check the monthly published data where feasible. In general, agreement was found to be quite good. We have found no indication that there were serious or consistent errors made in SRS tritium release monitoring.

It is not possible to provide a brief summary of the key release data in this summary section. The number of figures provided to summarize the key data is too large to incorporate here. The reader is instead referred to the groups of figures that summarize releases from specific SRS sources. The total annual releases, both routine and inadvertent, from each reactor are plotted with time in Figures 4.1-5, 4.1-6, 4.1-7, 4.1-8, and 4.1-9. Figures 4.1-10, 4.1-11, 4.1-12, 4.1-13, and 4.1-14 provide plots of the annual releases from the Tritium Facilities. Two spreadsheets contain the detailed information, including uncertainty estimates: Annual Reactor Releases, and Annual Tritium Facility Releases. While we believe that these data incorporate the records of inadvertent tritium releases over the years, we have included in this chapter a detailed discussion of documented inadvertent releases to provide the reader with a more complete discussion of this important topic.

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CHAPTER 4.2

RELEASES OF RADIOIODINES AND BETA-GAMMA-EMITTING PARTICLES TO THE ATMOSPHERE

ABSTRACT

This chapter first discusses the releases of the most important radioiodine isotopes (^{131}I and ^{129}I) to the atmosphere from facilities at the Savannah River Site (SRS). The historic measurements at the facilities are interpreted in the context of current knowledge of radioiodine chemistry and the effectiveness of various sampling techniques. The analysis leads to revised estimates of radioiodine releases. These revised estimates, or radioiodine source terms, can be used to estimate thyroid doses to persons residing near the SRS. The fuel processing facilities in F-Area and H-Area were sources of the highest releases of radioiodines, which were discharged through tall stacks. The largest estimated releases occurred in 1956. Releases during the years 1955–1961 were significantly higher than the releases that occurred between 1962 and 1971. Those latter releases were, in turn, markedly greater than releases in the later 1970s and 1980s.

The latter part of the chapter addresses atmospheric releases of particles that contain beta-gamma-emitting radionuclides. These non-volatile fission and activation isotopes are generally associated with solid particles, in contrast to the radioiodines that are most likely to be released in gaseous forms. The principal radionuclides released were isotopes of ruthenium, cerium, cesium, strontium, zirconium, and niobium. As was the case for radioiodines, the most important release points were the stacks at the fuel processing facilities. Overall, releases of ^{103}Ru plus ^{106}Ru were largest. (Separate analyses were not performed for many years.) However, in some years, the pair of radionuclides, ^{95}Zr plus ^{95}Nb, was discharged in greater quantities. In one year, the highest release was of ^{137}Cs. Releases of ^{141}Ce plus ^{144}Ce and ^{89}Sr plus ^{90}Sr were generally lower than those of ^{103}Ru plus ^{106}Ru and ^{95}Zr plus ^{95}Nb.

INTRODUCTION

Radioactive isotopes of iodine and other beta-gamma-emitting radionuclides are produced by fission of uranium and plutonium in reactor fuels and by neutron activation of both radionuclides and stable isotopes in the fuel. As is discussed in the following sections, many such radioisotopes are produced, but only a small number are important for environmental dose assessment. Because most of the releases of radioiodines and beta-gamma-emitting particles occurred when the fuel was processed (usually many days after removal from the reactor), the radionuclides of greatest interest in this context are those that are produced in relatively large quantities and have half-lives that exceed 1 day. This is illustrated in the discussion of the radioiodine releases.

RELEASES OF RADIOIODINES

As a general term, “radioiodines” refers to the entire set of radioactive isotopes of the element iodine. Many such isotopes, with mass numbers between 128 and 142, are produced in

nuclear reactor fuel by fission and activation. However, in this report we focus primarily upon those isotopes whose half-lives are long enough that significant amounts can be present at the time of fuel processing. The first of these is ^{131}I; its half-life is 8.04 days and it is produced in large quantities (tens of millions of curies) in a load of reactor fuel. Other radioiodine isotopes, with half-lives ranging from about 1 hour to 1 day (^{132}I, ^{133}I, ^{134}I, and ^{135}I), are produced in similarly large amounts during reactor operation. However, they decay in the fuel quickly following reactor shutdown and much smaller quantities are available for release when the fuel is processed. A number of radioiodines have even shorter half-lives and their decay in the fuel is correspondingly more rapid. The second isotope of interest here is ^{129}I, which has a half-life of about 16 million years. Although only a small quantity (less than 1 curie) of ^{129}I is produced in the normal life of a reactor fuel core, the amount produced has not been diminished by decay since the start of SRS operations. The ^{129}I that was released from SRS facilities remains as part of the pool of environmental ^{129}I, but the ^{131}I that was released decayed to low levels within about two months.

The following section contains a brief introduction to some aspects of the chemistry of radioiodine. This information is important to understanding the importance of radioiodine releases and their assessment. Then, the methods and problems of sampling radioiodine releases are discussed. Historic estimates of radioiodine releases are presented and the rationale for revising those estimates is described. Finally, revised estimates of the amounts released are presented; these are the so-called source terms for radioiodines.

Sources of Radioiodine at the Savannah River Site

Radioiodines are produced by nuclear fission and activation processes in nuclear reactor fuel. Thus, the several reactors at the Savannah River Site (SRS) were sources of radioiodine releases. Because reactor fuel rods are surrounded by a cladding designed to contain the fission and activation products and the rods are stored in cooling water, atmospheric releases from those facilities were limited. The reprocessing facilities, where the cladding and the reactor fuel itself were dissolved, were more important sources of gaseous fission product releases. Research facilities of the Savannah River Laboratory (SRL) were a third source of radioiodine releases. These facilities were used for pilot studies and research; some work involved irradiated fuel or target elements containing radioiodines.

All these facilities are shown in Chapter 2, Figure 2-3. The reprocessing facilities are located in the 200 Areas (F-Area and H-Area) near the center of the SRS. The P, L, K, R, and C Reactors are also fairly centrally located; the areas were named 100-P, 100-L, etc. The SRL facilities are located in the Administrative Area (A-Area) near the northwestern boundary of the SRS.

This chapter focuses on the reprocessing facilities, which were the sources of the highest releases of radioiodines identified by Kantelo et al. (1993). The release point at each of these plants was a tall ~61-m (200-ft) stack, designated 291-F and 291-H, respectively. Gases that were carried from the dissolver vessels were treated to reduce the radioiodine concentrations; however, radioiodines were evolved from liquids in other parts of the facilities as well, and those gases were also carried to the tall stacks. Sand filters used to reduce releases of particulate radionuclides did not have a large effect on the quantities of ^{131}I and ^{129}I released. For the reactors, the main atmospheric discharge points were also tall stacks. Several stacks of differing heights served SRL facilities.

Radioiodine Chemistry

The chemistry of iodine in nuclear facilities and in the environment is complex and not fully understood. We discuss here only basic features that are important to estimating radioiodine releases from nuclear facilities and to understanding the principal human exposure pathway. Iodine is one of the halogens, a series of reactive elements that also includes fluorine, chlorine, and bromine. Iodine is an essential element in human nutrition and is primarily located in the thyroid gland. Radioactive iodine, such as ^{131}I, is chemically the same as stable iodine (^{127}I) and, when ingested or inhaled, also is concentrated in the thyroid.

Relationship between Chemical Form and Deposition on Surfaces

The reactive nature of elemental iodine gas, which is composed of two iodine atoms and is symbolized by I_2, makes it likely to deposit on surfaces. Examples are (a) walls of vessels in a nuclear facility and (b) vegetation in the environment. Deposition on vegetation is the first step in the pathway that leads from iodine in air to iodine in cows' milk and subsequently to irradiation of the thyroid glands of consumers of contaminated milk. That sequence of transfers is often referred to as the air-grass-cow-milk exposure pathway. (For the small fraction of the population that consumes goats' milk, the pathway is basically the same, but the milk source differs.) Deposition of elemental iodine on the interior wall of a sampling line can lead to underestimating the amount of radioiodine that was released from the stack being sampled. Loss of the elemental iodine from the sample air stream means that the concentration measured in the sample will be lower that the concentration that was present in the stack exhaust air.

When iodine combines with other elements, the resultant compounds are generally less reactive than elemental iodine. Hydrogen iodide (HI) is the most reactive of such compounds. Iodine may be a component of a radioactive particle (firmly bound within it) or sorbed on the surface of a non-radioactive particle. Iodine may combine with other gases, such as oxygen, nitrogen, and organic compounds, to form less reactive gases. Least reactive of the variety of iodine compounds are the organic iodides. Methyl iodide, symbolized by CH_3I, is the most common example of this group. These compounds are formed in nuclear facilities and comprise part of the radioiodine releases from them. Organic iodides are much less likely to deposit on surfaces in the environment; as a result, they contribute much less to human thyroid doses from the air-grass-cow-milk transport pathway. Their lack of reactivity also means that techniques that use chemical reactions to trap elemental iodine in an air sample will not successfully collect organic iodides in the same sample.

There are forms of iodine whose reactivity is intermediate between those of elemental iodine and organic iodides. Iodine associated with particles is one; others are gaseous compounds. A number of inorganic iodine compounds, including HOI, have been postulated; however, firm definition of the intermediate reactivity fraction has proved elusive. Knowledge of these compounds is limited by sample collection methods that have been devised to isolate them and by a lack of understanding of the reactions that produce them. Voillequé (1979) provided a summary and discussion of the sampling techniques that had been used for radioiodines and for stable iodine and the relationship between iodine species and environmental transport. More recently, the question of radioiodine species was revisited as part of a workshop conducted for the Centers

for Disease Control and Prevention to address issues related to results of the dose reconstruction for Hanford releases (Grogan 1998).

The relationship between chemical form and deposition velocity, a measure of the air to surface transfer process, is summarized in Table 4.2-1. Because deposition of airborne iodine onto pasture grass is the first step in the air-grass-cow-milk pathway, the relative importance of a species for that exposure pathway is indicated by the magnitude of its deposition velocity. For that reason, the table is a useful guide to the relative doses received by the most highly exposed groups in a population. It should be noted that the table contains only representative values to show the relative ranking and that not all are based upon the same amount of experimental data. For example, elemental iodine has been studied more than any of the other compounds (e.g., Chamberlain and Chadwick 1953, 1966; Chamberlain 1960; Heinemann and Vogt 1980). The estimate for HOI is based upon a single laboratory experiment (Voillequé and Keller 1981). Deposition of radioiodine onto pasture grass in any particular circumstance depends upon several other variables in addition to the chemical form of the radioiodine.

Table 4.2-1. Approximate Relationships Between Chemical Forms and Deposition Velocities for Radioiodine

Chemical form	Deposition velocity for pasture grass (cm s^{-1})
Elemental iodine (I_2)	1
Other inorganic forms (such as HOI)	< 0.1
Iodine associated with particles	0.1–0.2
Organic iodides	0.0001–0.005

Although doses are generally much lower than those due to milk consumption, humans are also exposed to radioiodine by inhalation of contaminated air. For inhalation exposures, there are no great differences in the dose contributions from the various species. Doses from external exposure are also much lower than those following consumption of contaminated milk. Because external exposure depends in part upon deposition of radioiodine, elemental iodine makes the most important contribution to that pathway.

In summary, elemental iodine is the most important chemical form from the point of view of the air-grass-cow-milk (or air-grass-goat-milk) exposure pathway that leads to the largest thyroid doses in human populations. Because of the large differences in human exposure associated with the release of different radioiodine species, it is important to distinguish among them if possible. Information that can be used to make inferences about iodine species released at the SRS is discussed in the next section and in the later discussion of early sampling techniques at the SRS.

Measurements of the Forms of Radioiodine in Facility Effluents

The chemical forms of radioiodine in facility effluents were not and are not measured routinely at the SRS or at other Atomic Energy Commission (AEC)/U.S. Department of Energy (DOE) or commercial facilities. However, special studies at various locations have provided data on the chemical forms of radioiodine in releases from operating power reactors and reprocessing facilities.

In an early evaluation of a caustic and thiosulfate scrubber for radioiodine monitoring, Sill and Flygare (1960) found that 70% of the ^{131}I in effluents from the Idaho Chemical Processing Plant (ICPP) was not trapped by their system. Because the scrubber had been shown to be efficient for I_2, it was concluded that the effluent must contain chemical forms other than elemental iodine. Much later, Hetzer et al. (1980) found that the principal gaseous component of the ^{129}I discharge from the ICPP was organic iodides, which accounted for 60–80% of the total. The elemental iodine fraction ranged from 2–20% of the total, and a species of intermediate reactivity (identified as HOI by their species sampler) consistently accounted for about 20% of the radioiodine activity.

Measurements at two European reprocessing plants have been reported (IAEA 1980). At the Dounreay reprocessing plant in the United Kingdom, measurements indicated that about 70% of the radioiodine release was in organic form. At the Karlsruhe reprocessing facility in Germany, the organic iodide fraction of the ^{129}I discharge was found to be about 25%. The reasons why effluents from that facility had a smaller organic iodide fraction are not known, but they could relate to differing processing schemes or other factors.

Special investigations in similar facilities at the SRS and Hanford identified several organic iodides in atmospheric discharges using gas chromatography (Smith and West 1967; Haller and Perkins 1967). Methyl iodide, ethyl iodide, propyl iodide, and butyl iodide were identified, and it is possible that other longer chain compounds were also present. These studies did not quantify the fractions of various species in the total ^{131}I releases. Other investigations by Perkins (1964) at Hanford showed that the fraction not present as I_2 or HI (both of which were effectively captured by their scrubber) ranged from 50–70% of the total. Subsequent studies at the SRS found that the organic iodide fraction at the 200-H facility comprised 80–90% of the total ^{131}I. The predominant airborne organic forms were found to be methyl iodide and butyl iodide (Kantelo et al. 1993).

A series of investigations of the sources of ^{131}I in commercial power plant effluents yielded the bulk of the available information on radioiodine species for reactors. Data from nine boiling water reactors (BWRs) and ten pressurized water reactors (PWRs) were examined to develop composite species distributions for these two reactor types. The PWR species distribution is expected to be most applicable to the SRS production reactors, but there is not a great difference between those results and the composite distribution for BWRs (Keller et al. 1982). A small fraction (2%) of the released ^{131}I was associated with particles, 27% was elemental iodine, 31% was in organic form, and 40% was present as inorganic forms of intermediate reactivity (identified as HOI in the studies).

Sampling of Airborne Radioiodine

There are several aspects of successful sampling for contaminants in airborne effluents. An enumeration of these principles was published as an American National Standards Institute (ANSI) guide (ANSI 1969). This section briefly discusses issues that are relevant to understanding the effectiveness of the sampling for airborne radioiodines at the SRS.

A primary requirement of sampling is that a representative sample be withdrawn from the air being discharged from the stack. That is, the sample should faithfully show “the quality and characteristics of the entire volume” of the exhaust. If the discharge is not well mixed, this is difficult to accomplish. Because the airborne radioiodine is primarily in gaseous form, difficulties that affect extraction of samples of particles from the exhaust air stream are not highly relevant.

Following its extraction from the air being discharged, the sample is transported to the point of radioiodine collection from the sample stream. Ideally, the distance between the point of sample withdrawal and the point of collection will be short to minimize any losses due to deposition of radioiodine in the sampling line. The next steps are the capture of the radioiodine in the sampled air and the measurement of the collected radioiodine activity. These important issues are discussed in reverse order in the following sections.

Collection and Measurement of Radioiodine Samples

The earlier discussion of radioiodine species suggests that different chemical forms require particular attention when attempting to collect a sample of all airborne radioiodines or of the various forms individually. These sampling problems were not widely understood during the 1940s and 1950s and were not solved until the 1960s.

Based upon the Hanford experience, releases of radioiodine from the reprocessing facilities at the SRS were expected to be important and a stack monitoring system for ^{131}I was designed (Anonymous 1953a, 1953b; Gould 1953; Moison 1953; Carmichael, and Karraker 1955). The system employed a caustic scrubber to collect the ^{131}I in the sampled air. Its success depended upon a chemical reaction between the airborne radioiodine, presumed to be present as I_2, and the NaOH solution through which the air was bubbled. This sampler would not collect organic iodides; its efficiency for collection of inorganic forms other than I_2 and HI is uncertain. The NaOH solution was transported through a coiled line, which was monitored by a scintillation detector and analyzer focused on the main ^{131}I gamma ray. The output of the system, proportional to the ^{131}I release rate, was recorded in the control room. This real-time monitor could detect a release rate of about 1 Ci of $^{131}I_2$ per day. The caustic scrubber solution was also collected over a 24-hour period. Sampling and laboratory analysis of the solution provided a more sensitive, but not real-time, measure of ^{131}I releases. A release rate of about 2 mCi of $^{131}I_2$ per day could be detected in that way (Carmichael and Karraker 1955; Marter 1962).

Beginning in 1956, a second sampling system that employed particulate filters impregnated with silver nitrate ($AgNO_3$) was operated in parallel with the caustic scrubber stack monitor. These filters were replaced daily and counted in the laboratory to determine the ^{131}I content and obtain an estimate of the daily discharge of ^{131}I. The filter was tested in the laboratory using I_2; measured collection efficiencies routinely ranged from 85–95%, but they were as high as 100% under carefully controlled experimental conditions. Field tests in 1956 yielded filter collection efficiencies of 76–100% relative to a caustic scrubber (Du Pont 1956i, 1956l). Because collection of ^{131}I by these $AgNO_3$-impregnated filters also relied on a chemical reaction and binding of the ^{131}I to the silver in the filter, these filters also would not trap organic iodides. The collection efficiency of these filters for other inorganic forms is not known.

The effluent monitoring systems were thoroughly reviewed following the first widely discussed ^{131}I release from the SRS reprocessing facilities in May and June 1961 (Reinig 1961; Marter 1963). The investigators concluded that neither the caustic scrubber and monitor nor the $AgNO_3$-impregnated filters used to collect ^{131}I had performed well. Initial indications from the monitor that there was a high ^{131}I release rate were not considered credible because of previous errant indications by that system. However, later analysis of the amount of ^{131}I collected in the scrubber solution showed that the indications had been correct. Analysis of the data led to the conclusion that the collection efficiency of the $AgNO_3$-impregnated filters, relative to the caustic

scrubber, had dropped to 32% and was well below the nominal value of 80% that had been assumed to provide cautious estimates of the releases (Reinig 1961; Marter 1963).

The releases at the end of May and in early June led to readily detectable contamination in milk in the environs of the SRS. See Chapter 10 of this report for a more detailed discussion regarding radioiodine concentrations in milk. There was a formal investigation of the event, which was reported to AEC Headquarters (Blair 1961).

A detailed investigation of the sampling and monitoring systems that were in use at the reprocessing facilities was conducted. As part of this effort, plant personnel evaluated alternative collection media for ^{131}I in the 200-F and 200-H exhaust stack sampling and monitoring systems. As a result of comparative testing, it was decided that charcoal cartridges would be used to collect ^{131}I in reprocessing facility effluent samples. Although not considered entirely satisfactory because of the heating produced by reactions of undefined oxides of nitrogen (NO_x) with the charcoal, this practice was started in the fall of 1961 (Marter 1962).

The investigations into effluent sampling produced a number of significant findings, some of which are summarized below. Others are discussed in the following two sections.

- Laboratory testing of the NaOH scrubber indicated an efficiency of 92% for I_2; however, daily comparisons against charcoal sampling of 200-H plant effluents yielded a 20-day average scrubber efficiency of only 20%. Because charcoal is a good collector for all iodine species, this comparison indicates that much (nearly 80%) of the ^{131}I in the effluent was not I_2.
- Two laboratory tests indicated a collection efficiency for I_2 of 85% for the $AgNO_3$-impregnated filters (compared with a caustic scrubber) that was not affected by a change in sampling flow rate from 472 to 944 $cm^3\ s^{-1}$ (1 to 2 $ft^3\ min^{-1}$). Field testing, also compared with a caustic scrubber, showed efficiencies of between 34 and 64%. In those tests, the collection efficiency was found to be inversely proportional to the sampling flow rate. Plant staff concluded that an efficiency of 32% was appropriate for the sampling flow rate during period of abnormal release in May–June 1961.
- Further field testing of $AgNO_3$-impregnated filters, compared against charcoal, showed filter collection efficiencies for ^{131}I that were much lower. For 28 days of sampling the 200-H exhaust and 25 days of sampling the 200-F exhaust, collection efficiencies averaged 1.3 and 4.9%, respectively. These results are again consistent with an effluent that is primarily composed of organic iodides and contains relatively little I_2.

Follow-up studies and measurement were reported by Smith and Jolley (1963). At the inlet to the stack, the $AgNO_3$-impregnated filters were only 8 percent efficient when compared against charcoal collectors. This finding supports the previous conclusion that most (~90% in this case) of the ^{131}I was not in elemental form. It is consistent with the earlier field efficiency measurements that employed those filters and with later measurements, at the SRS and elsewhere, that showed a high organic iodide fraction in reprocessing plant effluents. Although the indicated organic fractions for the SRS reprocessing facilities are higher than seen elsewhere, this might be explained by the fact that at SRS the effluent passed through a sand filter. That filter provided a large surface area for deposition of I_2 and subsequent chemical conversion to less reactive forms.

Jacobsen and Jolly (1963) reported efficiencies of 24 and 30 percent for two $AgNO_3$-impregnated filter papers exposed to process ^{131}I, but the sampling times were only 15 minutes. Efficiencies of two NaOH scrubber solutions for the same test conditions were 56 and 59 percent. For short-term samples the efficiency of activated charcoal exceeded 99 percent, but for 24-h

samples in the presence of NO_2, or a combination of NO_2, NH_3, and NH_4NO_3, the average efficiency was about 88 percent (Jacobsen and Jolly 1963).

Sampling Line Losses for Radioiodines

The effluent sampling lines for the reprocessing facilities at the SRS were quite long. Samples were extracted at the ~59-m (195-ft) level of the stack. The line extended to near ground level and then an additional 30.5 m (100 ft) horizontally to the point of sample collection. The total line length, including the in-stack portion, is estimated to be about 93 m (~305 ft).

Table 4.2-1 also serves as a guide to the relative probability of losses due to deposition of gaseous iodine species in sampling lines. Although the numerical values of deposition velocity differ, the general relationships are the same. Thus, the large organic iodide fraction of the ^{131}I in the sample would be carried through the sampling line effectively, with little loss due to deposition on the walls of the line. The ratio of the organic iodide concentration leaving the line to that entering it (called the transmission factor for organic iodide) would be very close to the desired value of 1. However, based upon current knowledge, we expect that there would be significant deposition of the reactive iodine fraction (I_2) during passage through the line. A model of the behavior of radioiodine in sampling lines is discussed below.

Measurements at the SRS. Sampling line losses for I_2 were investigated by SRS staff following the elevated releases of ^{131}I in reprocessing facility effluents in May–June 1961. Testing was conducted in both the laboratory and in the actual sampling line in the field. Because the laboratory test employed only a very short (~1-m) piece of tubing, the field test is most relevant. Elemental iodine was released into the horizontal 30.5-m (100-ft) section of the sampling line and measured at the normal sample collection point. About 32% of the I_2 was initially lost because of deposition in the line. However, during the first day after the injection period, 7% of the deposited I_2 was resuspended from the walls of the line and collected at the sampling point. This amounted to an additional 2% of the amount injected, resulting in a net loss of ~30% in that section of line (Marter 1962).

The observed transmission factor for the test (0.7) can be extrapolated to the full length of the sampling line. The entire line is nominally three times longer than the section used for the test. The transmission factor for samples of I_2 carried through the entire line is estimated to be approximately 0.7^3 or ~0.34 for conditions comparable to those used for testing.

Two later tests involving injection of 1 and 6 mCi of $^{131}I_2$ into the stack did not yield detectable amounts of ^{131}I at the sample collection point. These unusual results do not seem to have been investigated further. It may be, as was speculated, that a combination of deposition in the stack and in the sampling line reduced the concentrations of ^{131}I below the level that was detectable.

In a later presentation, Jacobsen and Jolly (1963) reported rather different results for line loss. They performed experiments using a separate line running from the ~15-m (50-foot) level of the stack to the sample collection point in the fan building. Simultaneous measurements were made at both end of the line, which was about half as long as the normal sampling line, to estimate deposition losses. They found that NO_2 in the air stream greatly decreased the deposition loss, but that losses were higher when ammonia was also present to react with the NO_2. On the basis of their results, they estimated that typical losses would average only 10–15 percent.

Unfortunately, the document prepared for the oral presentation contains few details about the experiments. Information about the flow rate through the line, the concentrations of ^{131}I, NO_2, NH_3, and NH_4NO_3 measured during the tests, and the relationship of the levels of the non-radioactive constituents to routine operational conditions would be very useful, but has not been found. The results provided are difficult to interpret without the supporting data. They imply a transmission factor for the main sampling line in the range 0.7–0.8, which is quite different from the estimate of ~0.34 based on a test using a section of that line (discussed above).

Model of Radioiodine Transmission Sampling Lines. This section describes a model that has been developed over many years and is believed to reflect the principal processes that affect transmission of radioiodine through sampling lines. An early version of the model considered only deposition and resuspension and was consistent with experimental data that showed there was a delay in the movement of radioiodine through lines and a change in the activity balance from reactive species (I_2) to nonreactive forms like organic iodides (Pelletier et al. 1978a). Figure 4.2-1 shows a diagram of one segment of the current model. It includes a third process, fixation of some of the deposited iodine on the surface.

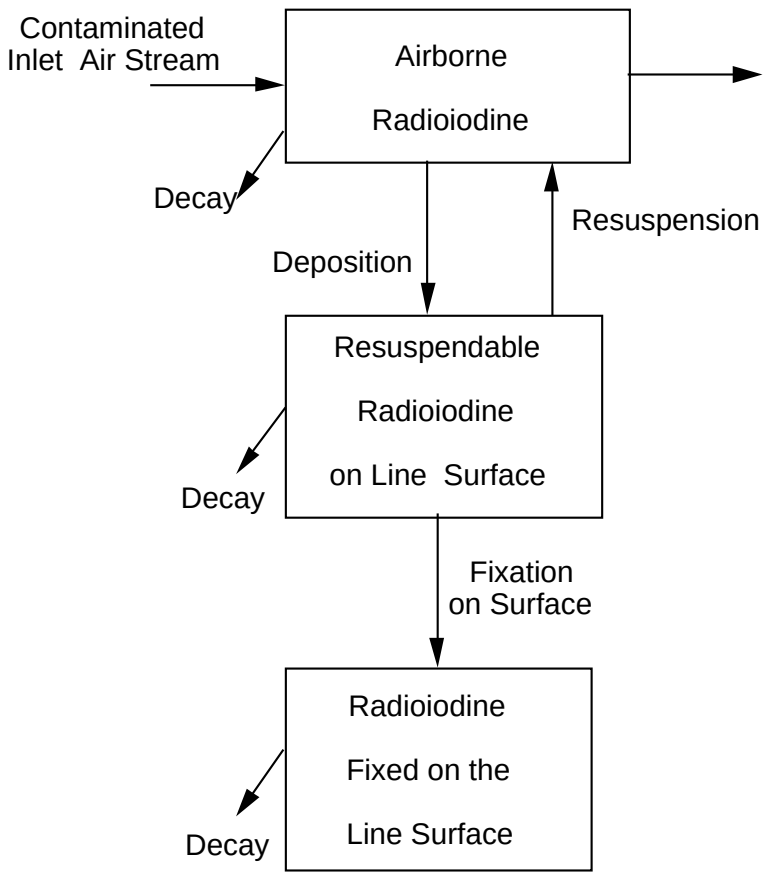

Figure 4.2-1. Model of radioiodine transport in a sampling line.

Typically, a sampling line is modeled as a sequence of 20 segments like the one shown in Figure 4.2-1. The airborne activity leaving one line segment becomes the input for the next

segment of the model. Radioactive decay is shown as a removal process for all three compartments; however, it is important only for the radioiodine on surfaces.

Laboratory measurements of radioiodine deposition and resuspension in mock-ups of steel sampling lines (Unrein et al. 1985; Widner et al. 1985; Edson et al. 1987; Glissmeyer and Sehmel 1991) are the source of estimates of the deposition and resuspension rate constants. Kabat (1983) provides information for other surfaces. In addition to the SRS measurement described above, experimental studies of installed sampling lines were also conducted and documented in Ström and Hesböl (1978) and Curtis and Guest (1986). It was analysis of experimental measurements that led to the inclusion of the fixation process in the revised model, which greatly reduced the variability of the resuspension rate constants computed from the data. Voillequé (2001) describes the model and derivation of parameter values in more detail.

Three differential equations describe the transport of radioiodine activity in a line segment as functions of time (t, s). Each equation addresses the amount of radioiodine in one of the three compartments shown in Figure 4.2-1. The three equations are

$$\frac{dq_a}{dt} = I + rq_s - \left(\lambda_v + \Delta + \lambda\right)q_a \tag{4.2-1}$$

$$\frac{dq_s}{dt} = \Delta q_a - \left(r + \lambda + \phi\right)q_s \tag{4.2-2}$$

$$\frac{dq_f}{dt} = \phi q_s - \lambda q_f \tag{4.2-3}$$

where

q_a = activity (Bq or µCi) of the airborne radioiodine in the segment
I = rate (Bq s^{-1} or µCi s^{-1}) at which activity enters the line segment
q_s = activity (Bq or µCi) on line surfaces that is available for resuspension
r = resuspension rate constant (s^{-1})
λ_v = air removal rate constant (s^{-1}); $\lambda_v = Q\ V_s^{-1}$
Q = sampling line flow rate ($cm^3\ s^{-1}$)
V_s = volume of the line segment (cm^3)
Δ = deposition rate constant (s^{-1}); $\Delta = V_d\ A_s\ Vs^{-1}$
V_d = radioiodine deposition velocity (cm s^{-1})
A_s = surface area (cm^2) of the line segment
λ = radiological decay rate constant (s^{-1}) for the radioiodine isotope
ϕ = rate constant (s^{-1}) for the surface fixation process
q_f = activity (Bq of µCi) fixed on the surface.

The three equations are solved as functions of time for each of the line segments, usually 20, used to model the sampling line. The transient behavior following either an increase or decrease of radioiodine input to the line can be computed. Because the focus of this discussion is on long term average releases, the behavior at equilibrium is addressed here.

During a period of relatively constant input of elemental iodine into the sampling line, the surface activity in each segment will gradually increase to an equilibrium value. The equilibrium

is reached when deposition of I_2 onto the surface of the segment is balanced by the removal due to resuspension from the surface and fixation to the surface. The air concentration in each segment also reaches an equilibrium value as this balance is achieved.

The equilibrium transmission factor for elemental iodine (TF_{ee}) is defined as the following ratio:

$$TF_{ee} = \frac{\lambda_v q_{ae20}}{I} \tag{4.2-4}$$

The numerator, which is the product of the equilibrium ^{131}I activity in the last (20th) line segment and the exhaust rate constant, is the rate (Bq s^{-1} or µCi s^{-1}) at which activity leaves the line under equilibrium conditions. The denominator is the rate (Bq s^{-1} or µCi s^{-1}) at which $^{131}I_2$ activity enters the sampling line. The transmission factor depends upon the properties of the sampling line as well as the behavior of elemental iodine.

In general, transmission factors for various iodine species depend upon the properties of the sampling line (length, diameter, flow rate) and the properties of the particular chemical form. While the dimensions of the sampling line are usually known and the equations for the model can be solved, knowledge of the parameters needed to use them is not exact. Uncertainty is associated with the estimates of the deposition, resuspension, and fixation rate constants. Monte Carlo techniques were used to estimate the transmission factors for elemental iodine in the sampling lines.

The Monte Carlo procedure uses information about the expected values and distributions of possible deposition, resuspension, and fixation parameter values to make a series of estimates of the transmission factor. The result of these calculations is a distribution of values of TF_{ee}. That distribution of estimates is typically lognormal, or approximately so, and is characterized by a median (50th percentile value) and a geometric standard deviation (GSD). It reflects the uncertainties in the input parameters and their effect on the estimates of TF_{ee}.

The model was used to compute sampling line losses for elemental iodine under various operating conditions that are discussed below. The whole range of estimates of sampling bias due to line losses was then used in the reevaluation of the radioiodine releases.

Extraction of a Representative Sample

Following the May–June 1961 releases, plant staff also investigated the overall sampling process. Tritium gas was used as a tracer to check whether the sample extraction system and sampling line were functioning properly. Known amounts of tritium gas (~190 Ci) were injected into the stack and measured with a portable Kanne chamber at the sample collection point. The measured tritium concentrations in two tests were an average of 96% of the expected tritium concentrations. These results provide confidence that a representative sample of stack gases was being collected and that the sampling system was functioning properly (Marter 1962).

It was noted that the linear velocity of air in the stack exhaust was about double the linear velocity in the sampling system. This difference in velocities means that sampling was not isokinetic (Marter 1962). Anisokinetic sampling is not an important factor for gaseous iodine, but it is considered in the evaluation of discharges of radioactive particles.

Estimates of Airborne Releases of Radioiodine at the SRS

Revised estimates of radioiodine releases to the atmosphere from the reprocessing facilities, production reactors, and research facilities are presented in the following sections. Original weekly and monthly reports have been used to prepare a history of reported releases. The data have been compared with summaries of releases that have been compiled in recent years (Cummins et al. 1991; Kantelo et al. 1993). Occasional small differences or inconsistencies were found when comparing the documents. In spite of them, the summaries are considered to be accurate reflections of the ^{131}I releases that were reported historically; however, the summaries do not address issues related to the reliability of the historic data.

Reprocessing facilities, which were the largest sources of airborne radioiodine releases and for which monitoring records are most complete, are discussed first and given the greatest attention. Overall, it is estimated that the F-Area and H-Area stacks discharged more than 300 times more radioiodine than all the reactor and research facility stacks combined. The most important discharges, from the point of view of human health, are those of elemental iodine. As was discussed earlier, this is because of its role in exposure via the air-grass-cow-milk pathway. It is estimated below that historic discharges of I_2 from the F-Area and H-Area stacks exceed those from other locations by at least a factor of 50. Those releases, which mostly occurred during early years of operation, are the most significant radioiodine releases at the SRS.

Estimates of Radioiodine Releases from Reprocessing Facilities

Reassessment of the radioiodine releases from the reprocessing facilities must consider the history of sampling techniques and the information on radioiodine species that were described above. The early monitoring and sampling systems were designed for collection of I_2. At that time, there was little or no knowledge of other less reactive chemical forms. After charcoal cartridges were employed (late 1961), more complete collection of radioiodine was accomplished and more attention was given to problems of incomplete sample collection. Subsequently, a new monitoring system was installed (Jolly et al. 1968). Table 4.2-2 shows the time periods considered in this reassessment.

Table 4.2-2 illustrates a gradual improvement of radioiodine monitoring at the reprocessing facilities. The most important changes were made following the 1961 releases, which led to a serious investigation of radioiodine monitoring practices. The only factor important to estimation of releases that was consistently not considered is loss of sample due to deposition in the sampling line. In the more recent summary of reported SRS radioiodine releases (Kantelo et al. 1993), line loss was not considered to be an important factor because most of the releases have been shown to consist of organic iodides. That view overlooks the fact that during the periods when the highest releases were reported only the reactive species were being measured.

The reported amounts of radioiodine released from the separations areas are discussed in the next section. That is followed by the reevaluation of those releases, which takes into account the factors identified in Table 4.2-2.

Table 4.2-2. Overview of Radioiodine Sampling and Monitoring Methods

Period	Method of sample collection	Comments on reported results
Dec 1954 to Oct 1956	NaOH scrubber	Only reactive forms were measured; system was not considered reliable; no corrections for line loss were made
Oct 1956 to Sept 1961	$AgNO_3$-impregnated filters	Only reactive forms were measured; unknown variability in collection efficiency; no corrections for line loss were made
Sept 1961 to Sept 1965	Charcoal cartridges	All chemical forms were measured; attention paid to collection efficiency variations; no corrections for line loss were made
After Sept 1965	Charcoal cartridges	New monitoring system with higher flow rate; all chemical forms were measured; attention paid to performance; no corrections for line loss were made

Reported Releases of ^{131}I. Values of the reported releases of ^{131}I from the F and H stacks during early years of operation (late 1954 to the end of 1961) are plotted in Figure 4.2-2 (Du Pont 1954a-l, 1955a-l, 1956a-l, 1957a-l, 1958a-l, 1959a-l, 1960a-l, 1961a-l). The year of largest releases was 1956, when several of the reported monthly releases exceed 100 Ci. The reported total for that year was slightly under 1600 Ci. Reported releases during 1957, 1959, and 1961 were ~290, ~160, and ~160 Ci, respectively.

Annual releases reported for years following 1961 did not exceed 40 Ci and those reported for 1972–1989 were all less than 3 Ci. Comparison of release estimates from monthly reports from 1954 through December 1971 (Du Pont 1954a-l, 1955a-l, 1956a-l, 1957a-l, 1958a-l, 1959a-l, 1960a-l, 1961a-l, 1962a-l, 1963a-l, 1964a-l, 1965a-l, 1966a-l, 1967a-l, 1968a-l, 1969a-l, 1970a-l, 1971a-l) with the summary of annual releases by Kantelo et al. (1993) and all the annual releases reported by those authors are contained in an Excel® spreadsheet. Clicking on the following hyperlink will provide automatic access to these data: Reported_I-131_Releases_(F,H).xls. Agreement between the sums of values in the monthly reports and the annual totals in the recent summary report is generally very good.

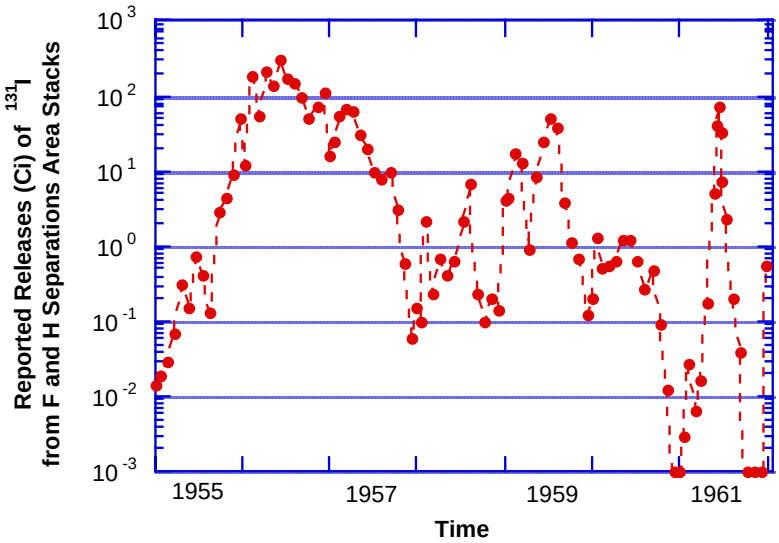

Figure 4.2-2. Reported releases of ^{131}I from the stacks of the F and H separations facilities during the earliest years of operation.

Revised ^{131}I Release Estimates. The revised estimates of ^{131}I releases from the reprocessing facilities reflect estimates of bias and uncertainty that were not addressed in previous reports of those releases. Uncertainties that are addressed include those associated with sample collection and with radioactivity measurements. The biases in the reported results stem from the fact that some sampling media did not collect organic forms of radioiodine and because there were losses of I_2 due to deposition in the sampling line.

As indicated in Table 4.2-2, several time periods must be considered because the methods of radioiodine monitoring changed. Those same time periods are listed in Table 4.2-3, which contains information related to several questions about radioiodine monitoring for elemental iodine and organic iodides. The less reactive inorganic fraction that may have been present is not addressed explicitly because of lack of information; however, identification of that fraction is much less important than the distinction between I_2 and organic iodides. In the two earliest periods, the organic iodide releases were not monitored. They are estimated from the releases of elemental iodine using observations that organic forms comprised 70–90% of the total release from U.S. reprocessing facilities. The calculational procedure that was used is described later.

Estimates of the efficiencies of the collection media (NaOH scrubber solutions and $AgNO_3$-impregnated filters) for I_2 are based primarily upon reported SRS experience. However, the selected ranges are somewhat broader than that to reflect a greater variety of field sampling conditions. For the scrubber efficiency, a range of about ±10% is considered. A broader range is considered for the $AgNO_3$-impregnated filter papers. This range is based upon the results of field tests that were conducted in 1961. A collection efficiency as low as 34% was measured for a sampling flow rate that was lower than that normally used for the filter paper. It is not certain why the field tests did not include sampling at the normal flow rate, but it may have been related to the requirements for the caustic scrubber used for comparison. The lower bound of 0.2 is

believed to cover the range of possible efficiencies. Relatively high efficiencies had been measured in field tests in 1956. The factors that changed during the roughly 5-year period and led to lower collection efficiency for I_2 and when the change occurred are not known.

Table 4.2-3. Factors Considered in Preparing Revised Estimates of ^{131}I Releases

Period	Chemical Form of ^{131}I released	Release Measured?	Sample collection efficiency	Estimated measurement uncertainty	Estimated transmission factor
Dec-54 to Oct-56	Elemental	Yes	0.75–0.95	40%	0.28[c]
	Organic	No[a]	NA[b]	NA	NA
Oct-56 to Sept-61	Elemental	Yes	0.2–0.9[e]	30%	0.42[f]
	Organic	No[d]	NA	NA	NA
Sept-61 to Sept-65	Elemental	Yes		20%	0.28[h]
	Organic	Yes	[g]	20%	1
After Sept-65	Elemental	Yes		15%	0.59[i]
	Organic	Yes	[g]	15%	1

[a] Organic iodides were not collected by NaOH scrubber; releases estimated using observations that organic iodides constituted 70–90% of the total radioiodine discharge.
[b] Not applicable.
[c] Median estimate based upon the design flow rate of 944 $cm^3\ s^{-1}$ (2 $ft^3\ min^{-1}$) of the NaOH scrubber system; estimated GSD = 1.94 (see Figure 4.2-3).
[d] Organic iodides were not collected by AgNO3-impregnated filters; releases estimated using observations that organic iodides constituted 70–90% of the total radioiodine discharge.
[e] Estimates based upon reported results of 1956 field tests and measurements following the 1961 releases.
[f] Median estimate based upon the reported total flow rate of 1316 $cm^3\ s^{-1}$ (3 $ft^3\ min^{-1}$); 944 $cm^3\ s^{-1}$ (2 $ft^3\ min^{-1}$) to the $AgNO_3$-impregnated filter and 472 $cm^3\ s^{-1}$ (1 $ft^3\ min^{-1}$) to the NaOH scrubber system; estimated GSD = 1.65 (see Figure 4.2-3).
[g] Although variable, collection efficiencies were checked routinely and corrections made; efficiencies of the charcoal cartridges are estimated to be within 10% of the value estimated.
[h] Estimate based upon a flow rate of 944 $cm^3\ s^{-1}$ (2 $ft^3\ min^{-1}$) through the charcoal cartridge.
[i] Median estimate based upon the design flow rate of 2360 $cm^3\ s^{-1}$ (5 $ft^3\ min^{-1}$) of the stack monitoring system; estimated GSD = 1.34 (see Figure 4.2-3).

Equilibrium transmission factors for organic iodides and elemental iodine through the long sampling lines have been computed using the line loss model discussed earlier. Transmission factors for organic iodides through sampling lines, even for short times after an increase in concentration, are practically identical to 1 and no correction has been made here for line losses of organic iodides.

The transmission factor for elemental iodine depends upon the sampling flow rate, which varied with time. Values of the deposition, resuspension, and fixation parameters of the model are based upon experimental results for lines that are similar and the distributions reflect a range of results. The distribution of deposition velocities was taken to be lognormal with a median value of 0.02 cm s^{-1} and a GSD of 1.4. The distributions of resuspension and fixation rate constants were logtriangular, with the mode equal to the upper bound. For resuspension, the range of values was from 1.1×10^{-7} to $6.8 \times 10^{-6}\ s^{-1}$. For the fixation rate constant, the range considered was 9.2×10^{-8} to $1.0 \times 10^{-5}\ s^{-1}$. The calculations of the elemental iodine transmission factors reflect the observed variability of these parameters as well as a potential 15% variability in the nominal flow rates.

The distributions of estimates of the equilibrium transmission factors for elemental iodine are shown in Figure 4.2-3. The distributions are contained in an Excel® spreadsheet. Clicking on the following hyperlink will provide automatic access to the information: Transmission Factor Estimates.xls. Sampling line losses decrease as the sampling flow rate increases. The median TF_{ee} was estimated to be 0.28 for a flow rate of 944 $cm^3\ s^{-1}$ (1 $ft^3\ min^{-1}$), but for a flow rate of 2360 $cm^3\ s^{-1}$ (5 $ft^3\ min^{-1}$) it was 0.59. Dispersion of the estimates decreased with increasing flow rate. Approximate GSDs are 1.94, 1.65, and 1.34 for flow rates of 944, 1416, and 2360 $cm^3\ s^{-1}$ (1, 2, and 5 $ft^3\ min^{-1}$), respectively. The median transmission factors for the two lower flow rates, 0.28 and 0.42, are approximately the same as the estimate made above by extrapolation of the 1961 SRS result to the entire line.

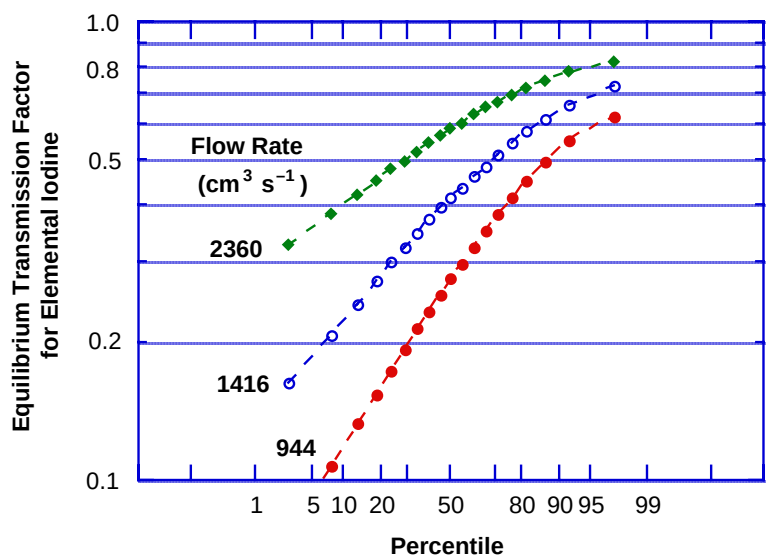

Figure 4.2-3. Equilibrium transmission factor estimates for elemental iodines for three sampling flow rates through a 93-m sampling line.

The method used here to reevaluate releases of ^{131}I at various times depends upon the measurement technique that was used. During the two earliest time periods defined in Table 4.2-3, the measurement procedure only gave an estimate of the elemental iodine release. After the fall of 1961, when routine use of charcoal cartridges was begun, that technique yielded an estimate of the total radioiodine release. The reanalysis of these results relies upon our knowledge of the fraction of the total radioiodine in organic form (f_o), which we estimate to be 70–90%. Designating the total amount of radioiodine released by Q_t and the two fractions by Q_e and Q_o, respectively, we may write:

$$Q_t = Q_o + Q_e$$
$$Q_o = f_o Q_t \qquad (4.2\text{-}5, -6)$$

Substitution and rearrangement yields:

$$\frac{Q_o}{f_o} = Q_o + Q_e \tag{4.2-7}$$

$$Q_o = \frac{f_o Q_e}{(1 - f_o)} \tag{4.2-8}$$

It should be noted that the quantity Q_e in these and subsequent equations refers to the corrected estimate of elemental radioiodine release. That is, it reflects corrections of the measured amount of elemental iodine (Q_{me}) for incomplete sample collection and for sample transmission losses discussed above. If the sample collection efficiency for elemental iodine is ε_e, and the equilibrium transmission factor for that species is TF_{ee}, then:

$$Q_e = \frac{Q_{me}}{\varepsilon_e TF_{ee}} \tag{4.2-9}$$

Corrections for sample collection efficiency were made historically. However, we consider a range of possible corrections as opposed to point estimates made previously. Corrections for sample transmission losses, not made previously, were discussed above. When the measurement technique could only detect elemental iodine, we first estimate the release of elemental iodine and then estimate the release of organic iodides.

A different approach is necessary to reevaluate radioiodine releases after September 1961, when the measurement technique collected both elemental iodine and organic iodides. The sample collection efficiency (called ε_o) was primarily determined by collection of organic iodides, and probably overcorrected somewhat for the elemental iodine fraction. The initial estimate of the total radioiodine release given in previous reports is:

$$Q_t = \frac{Q_{mt}}{\varepsilon_o} \tag{4.2-10}$$

However, this estimate neglects sample transmission losses. The relationship between Q_t and the amounts of organic and elemental radioiodine released is:

$$Q_t = Q_o + Q_e TF_{ee} \tag{4.2-11}$$

Rearrangement and use of the previous relationship (equation 4.2-8) between Q_e. and Q_o yields:

$$Q_o = Q_t - \frac{Q_o (1 - f_o) TF_{ee}}{f_o} \tag{4.2-12}$$

Solving for the organic iodide release, we have:

$$Q_o = \frac{f_o Q_t}{\left[f_o + (1 - f_o) TF_{ee} \right]} \tag{4.2-13}$$

and then the elemental iodine release is calculated using:

$$Q_e = \frac{Q_o(1 - f_o)}{f_o} \tag{4.2-14}$$

The two procedures just discussed were applied, in appropriate time periods, to previous estimates of radioiodine releases from the separations stacks in the F-Area and H-Area. The median estimates of the revised ^{131}I releases of elemental iodine are shown in Figure 4.2-4. The estimates are highest for 1955–1961 and are much lower for all subsequent years. It should be noted that the vertical scale is logarithmic and that each of the major divisions is ten times greater than the previous one. Thus, the median estimate for 1962 is more than 1000 times lower than the median estimate for 1956.

Figure 4.2-5 contains the release estimates for ^{131}I in the form of organic iodides from the same stacks. The pattern is similar to that in the previous figure and the vertical scale is also logarithmic. Because organic iodides are believed to comprise a large fraction of the total release, estimated organic iodide releases exceed those for elemental iodine. However, as noted earlier, releases of organic iodides are much less important for exposure of the thyroid via the air-grass-cow-milk pathway.

The main (5th, 25th, 50th, 75th, and 95th) percentiles of the distributions of release estimates for both elemental iodine and organic iodides are tabulated in an Excel® spreadsheet. In that listing, the median (50th percentile) value is highlighted to guide the reader to the central estimate of the amount released. The 5th, 50th, and 95th percentile values from this table are the basis for the release estimates plotted in Figures 4.2-4 and 4.2-5. The Excel® spreadsheet can be directly accessed by clicking on the following hyperlink: Revised_I-131_Releases_(F,H).xls.

The revised estimates of ^{131}I releases have been criticized by SRS staff (Heffner 1999) because they found good agreement between environmental concentrations predicted using the originally reported release estimates for 1956 and contemporary environmental measurements. They also cited similar comparisons made in an earlier plant report (Reinig 1959). As was discussed above, in the 1950s, both the effluent and environmental concentrations of ^{131}I were estimated using sampling media that were not highly reliable. Uncertainties in meteorological modeling, even for flat terrain, are on the order of a factor of 3–4 without consideration of plume depletion. In view of the quality of both sets of measurements and the uncertainties involved, the comparisons between predicted and measured concentrations in 1956 are not conclusive. When Reinig made similar comparisons in 1959, he had no knowledge of the unreliability of the sampling systems.

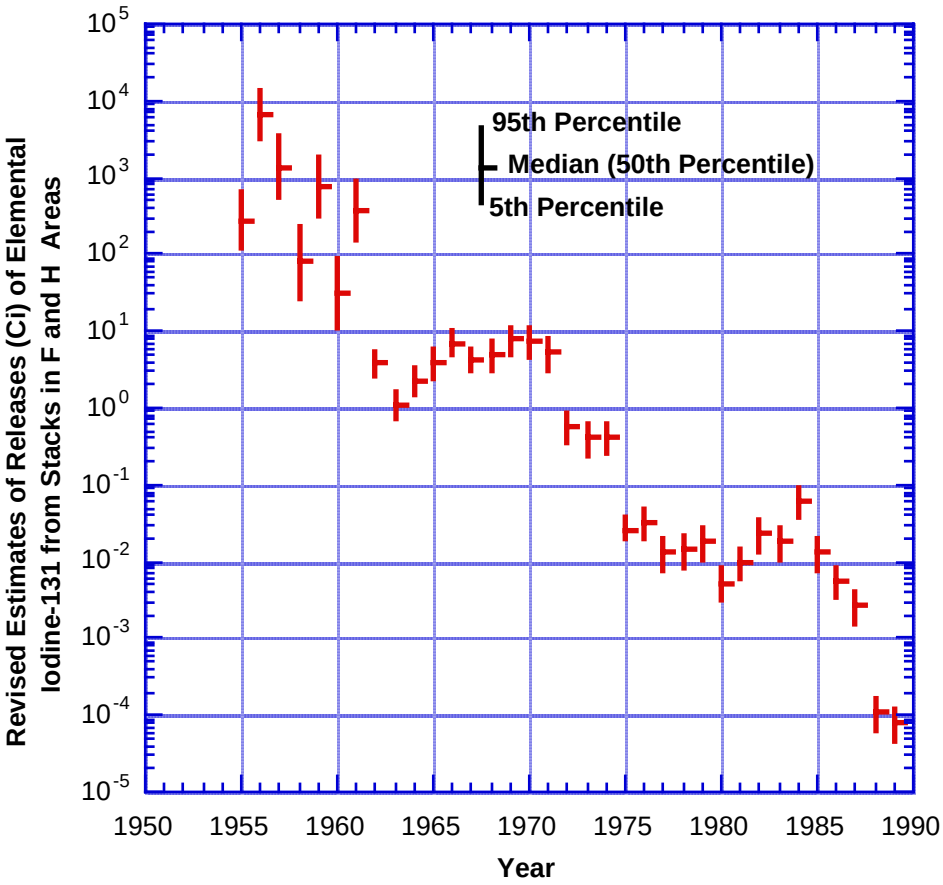

Figure 4.2-4. Revised estimates of releases of ^{131}I in elemental form (I_2) from stacks in the F and H separations areas. For each year, the length of the vertical line shows the range (5th to 95th percentiles) of the distribution of release estimates and the median estimate is indicated by the horizontal bar.

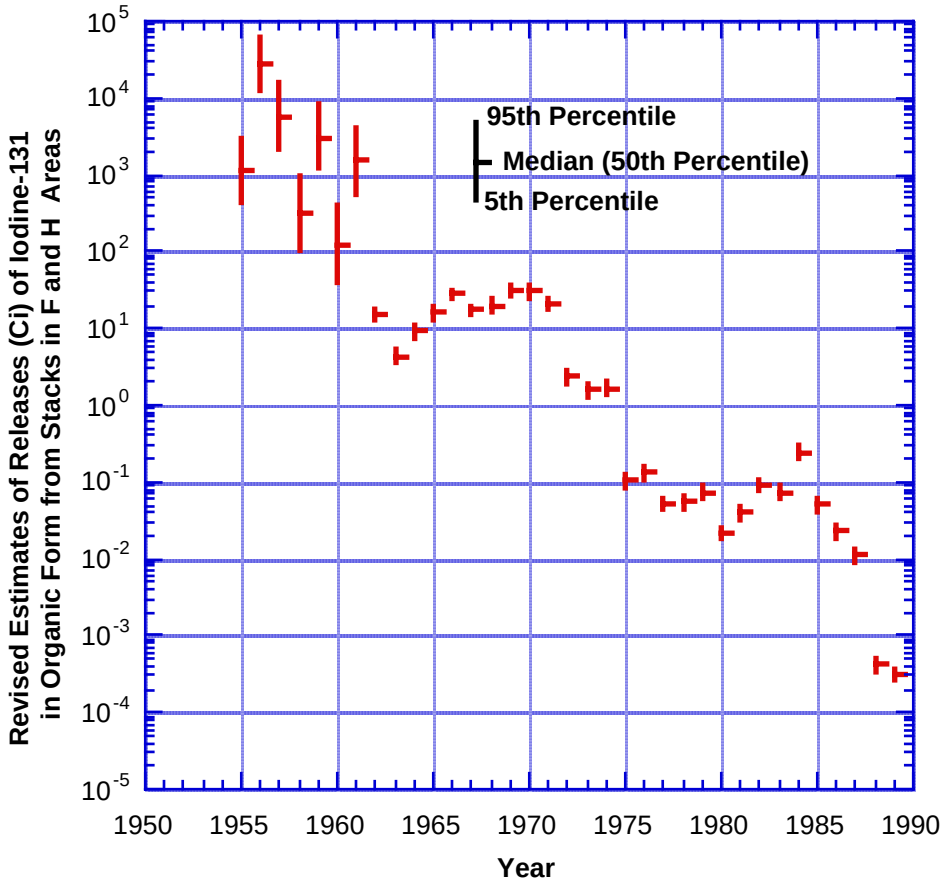

Figure 4.2-5. Revised estimates of releases of ^{131}I as organic iodides (e.g., CH_3I) from stacks in the F and H separations areas. For each year, the length of the vertical line shows the range (5th to 95th percentiles) of the distribution of release estimates and the median estimate is indicated by the horizontal bar.

Releases of ^{129}I. Iodine-129 is a fission product produced in reactor fuel that has a very long (~16 million-year) half-life. Most releases of ^{129}I occur when the fuel is being processed because the fuel cladding is dissolved and then the fuel itself is dissolved. These processes first liberate fission gases that have diffused into the gap between the fuel and the cladding and subsequently provide an opportunity for all volatile elements to be released from the solutions in the dissolver or in other vessels to which solutions are transferred.

The amount of ^{129}I produced in a particular batch of fuel depends upon the time that the fuel is irradiated in the reactor core. Figure 4.2-6, which shows the total power production of the SRS reactors during the years 1954–1989, is a guide to the amount of ^{129}I that was produced at the SRS. Note that the highest production occurred during 1959–1962, when the total power levels were at or above 3000 GW-d per year. During the years 1958, 1963, 1964, 1966, and 1967 the total power production was above 2500 GW-d. In years prior to 1958 and after 1968 reactor power levels were less than 2000 GW-d.

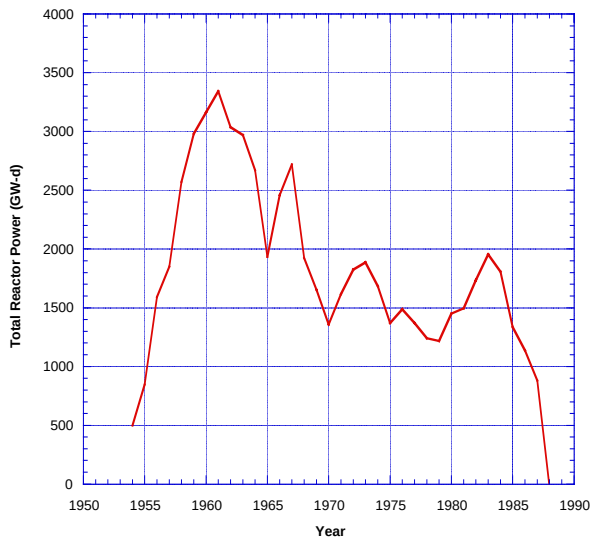

Figure 4.2-6. Total reactor power levels in operating SRS reactors. Data are given in Chapter 2.

The annual production of plutonium at the SRS (DOE 1996) is shown in Figure 4.2-7. That figure provides a guide to the time of releases of ^{129}I as the result of fuel processing. The delays (a) between removal of fuel from the reactor and start of fuel processing and (b) between the start of processing and the shipment of the product are reflected in the plutonium production plot. The peak releases of ^{129}I likely fell between the peak in power production and the peak in plutonium production. The plutonium production data from DOE (1996) are contained in an Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: SRS_Plutonium_Production.xls.

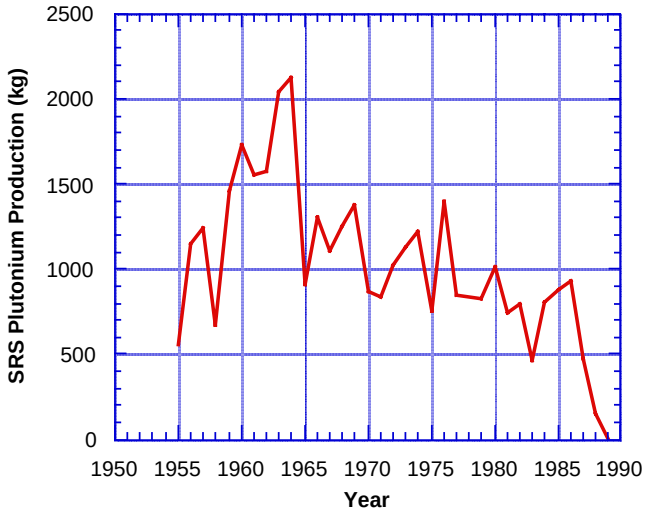

Figure 4.2-7. Amounts of plutonium produced at the SRS (DOE 1996).

Two different ^{129}I release histories have been reported for the SRS. Both are primarily based upon estimates of fuel inventories, release fractions, and cleanup system performance. Cummins et al. (1991) and Kantelo et al. (1993) present an average release of 0.21 Ci y^{-1} of ^{129}I between 1955 and 1973. Although Kantelo et al. (1993) identify a set of estimates based more upon plant operations (Boone et al. 1985), those estimates were not used for dose calculations contained in their report. The estimates made by Boone et al. are more consistent with the data on SRS reactor power levels and processing facility operations illustrated above. Measurements of ^{129}I releases did not begin until 1981. Measurements made in 1984 and later years are reported by Kantelo et al. (1993).

Estimated releases of ^{129}I that reflect historic operations are shown in Figure 4.2-8. The estimated releases are tabulated in an Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: I-129_Releases_(F,H).xls. For the years 1955–1960, we estimate the uncertainty in the plotted ^{129}I releases to be ± 40% because of variations in scrubber performance during the earliest years of operations. We estimate that uncertainties in release estimates are ± 25% for years between 1961 and 1980 and that those for estimates in years after 1980 are ±20%.

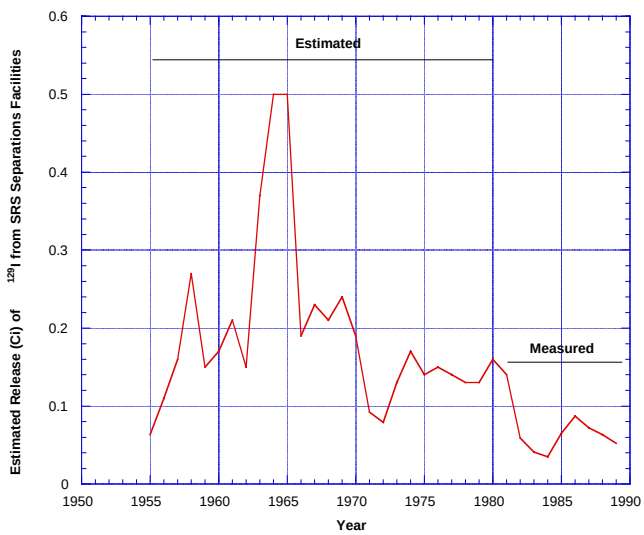

Figure 4.2-8. Estimated releases of ^{129}I from separations facilities in F-Area and H-Area.

Releases from SRS Reactors

This section focuses upon the release of ^{131}I to the atmosphere from the SRS production reactors. Releases of ^{129}I from reactors are expected to be small because of the low level of production of ^{129}I compared with that for ^{131}I. There are no quantitative estimates of the releases of ^{131}I to the atmosphere from SRS reactors for years prior to 1972. The summary document for radioiodine (Kantelo et al. 1993) contains only the releases that were reported after that time. There are some historic reports of releases of nonvolatile beta activity from the reactors. There are no atmospheric release estimates for R reactor, which was shut down in 1964, in Kantelo et al. (1993). The only release estimates given for L reactor are those based upon measurements during the latter years of operation (1986-1988). For the other reactors (P, K, and C), the report contains results of measurements of airborne releases that were performed after 1972.

Although no releases of ^{131}I to the atmosphere are reported for the early years of SRS reactor operations, such releases surely occurred. Considering the nature of the iodine sampling systems at that time, failure to observe airborne ^{131}I releases is not an adequate basis for suggesting that they did not occur. Silver nitrate-impregnated filters were used for collection of ^{131}I in samples of airborne effluent until the early 1960s. As noted above, this technique was inadequate. At about the same time that charcoal cartridges were introduced for sample collection, particle filtration and iodine absorption systems were installed in the SRS reactors to protect against large releases in case of accidents. During power operation, the building exhaust air was discharged through the filtration systems. During outages, part of the building air bypassed the filters. Tests of the activated charcoal from these absorbers were reported by Durant (1967) to show that the medium was quite efficient (> 99.9%) for removal of elemental iodine. The charcoal was not tested for collection of organic iodides but it is believed that the efficiency was not as high for that species.

Estimates of early atmospheric releases of ^{131}I from the SRS reactors are needed to assess the importance of those releases relative to those from the SRS reprocessing facilities. A number of factors affect ^{131}I releases from reactors. The reactor power level determines the maximum inventory of ^{131}I in the fuel. After a month of power operation, the ^{131}I content of the fuel is near its maximum value. Thus, duration of operation is not a major determinant of the release potential. After the inventory of ^{131}I is near the equilibrium level, the potential for release depends primarily upon the integrity of the fuel cladding. If the cladding is intact, radioiodine gas that has diffused into the gap between the fuel and the cladding will be contained. If not, radioiodines can leak from the fuel elements into the reactor moderator or fuel storage pool water and will be available for release in liquid or gaseous effluents.

The following rationale forms the basis for a method to estimate releases of ^{131}I to the atmosphere from the reactors. Because releases of ^{131}I to the atmosphere are a consequence of releases of ^{131}I from fuel elements to moderator or pool water, discharges of ^{131}I in liquid waste indicate that releases from the fuel have occurred and indicate that releases to the atmosphere were likely. The summary of Cummins et al. (1991) contains 26 pairs of estimates of annual releases of ^{131}I to the air and water from three reactors after 1971. The data were used to compute the ratio of airborne release (Ci) to the liquid release (Ci) for a particular year. The reactors and years with data available are: P Reactor (1973), K Reactor (14 years between 1972 and 1987), and C Reactor (11 years between 1973 and 1985). Because the releases were filtered, the radioiodine collected was most likely present as organic iodides. No correction for line losses was made when computing the ratios. Figure 4.2-9 shows the distribution of ratios of releases to the atmosphere to those in liquid wastes. The figure reflects 25 of the 26 ratios; one value of 45 (K Reactor in 1981) was excluded because it was clearly different from the other results and was considered an outlier.

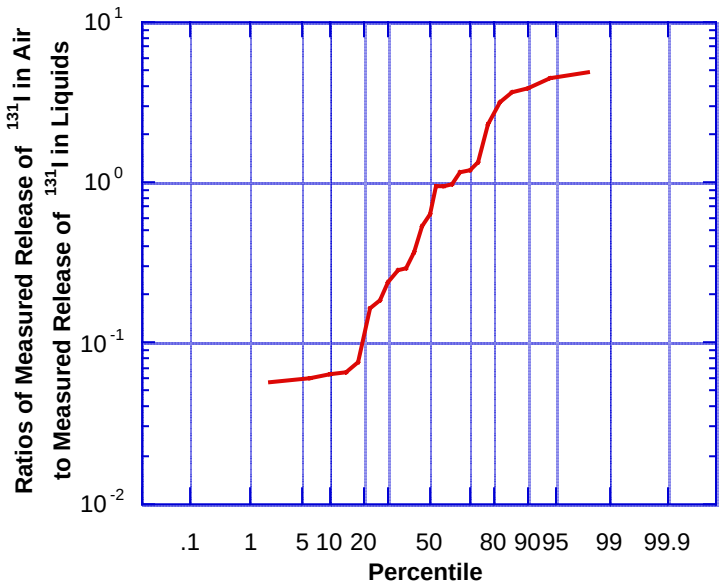

Figure 4.2-9. Distribution of ratios of the annual ^{131}I release (Ci) to the atmosphere to the annual ^{131}I release (Ci) in liquid wastes in years between 1972 and 1987. Gaseous effluents were filtered during power operation.

The distribution of release ratios has three components. About 20% of the ratios are relatively low (0.05–0.08) and a comparable fraction are relatively high (3–5). The remaining 60% of the ratios are roughly lognormally distributed between 0.2 and 2.3, with a median of about 0.6 and a GSD of about 2.3. The release data and computed ratios are contained in an Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: Ratios_of_Releases_(A-LW).xls. These ratios can be used together with previously reported and estimated releases of ^{131}I in liquid wastes to estimate the releases of ^{131}I to the atmosphere during years when no definitive measurements were made.

Figure 4.2-10 shows the reported and estimated releases of ^{131}I in liquid wastes from all the SRS reactors for the years between 1953 and 1975. For the years 1958 and 1959, the release was estimated to be 50 Ci, which is the average of the releases reported for five nearby years (1957, 1960–1964). Releases for 1953–1956 were also estimated. Releases were interpolated between a low release (0.1 Ci) estimated for 1953 to 65 Ci reported for 1957. The release data are contained in an Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: Reactor_Liquid_Waste_Releases.xls.

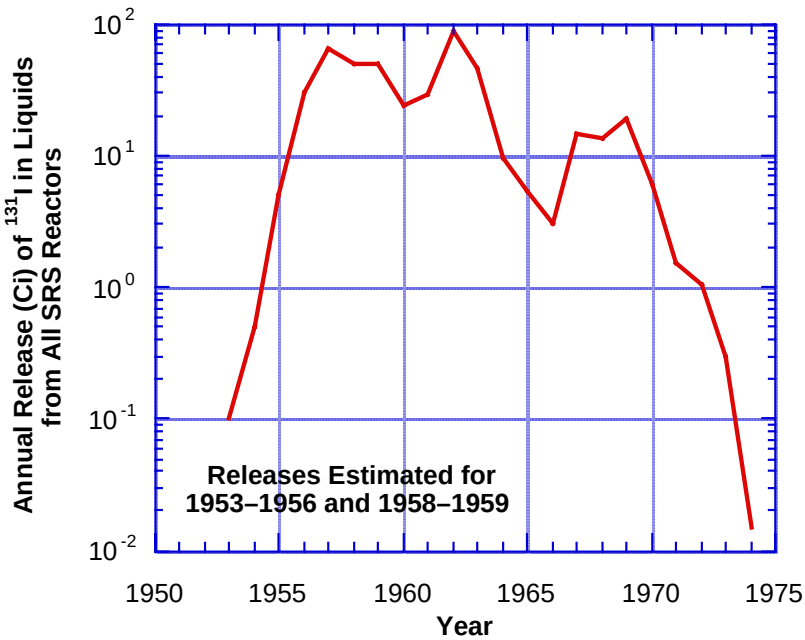

Figure 4.2-10. Reported and estimated reactor liquid waste releases of ^{131}I for all SRS reactors combined. See text for information about the estimates made for 1953–1956 and 1958–1959.

The measured releases of airborne ^{131}I during the years for which ratios were computed were lower than the amounts that would have been released had the charcoal filters not been present. Air purged from lower levels of the reactor buildings was not filtered. Containment purges during shutdown were found to contribute about 55% of the total release from commercial PWRs (Pelletier et al. 1978b) but that result offers only a rough guide to the appropriate fraction for the SRS reactors. A range of 30–70% is assumed for the fraction of the ^{131}I release that was unfiltered. The charcoal filter units were assumed to remove 0.999–0.9995 of the inorganic iodine and 0.8–0.95 of the organic iodides in air discharged through them. It is estimated, based on measurements reported by Pelletier et al. (1978b), that about 40% of the ^{131}I in routine PWR discharges was present as organic iodides. Using these assumptions, it is estimated that the sampling program estimated 31–74% of the release of airborne ^{131}I that would have occurred had the filters not been in place. All of the reactors had filter systems at some time in 1964 (Durant 1967). For the earlier years (1953–1963) the release ratios shown in Figure 4.2-9 must be adjusted for the effects of the charcoal absorbers on the amounts released.

The liquid waste release amounts shown in Figure 4.2-10 and the distribution of release ratios shown in Figure 4.2-9 were used in Monte Carlo calculations of estimated releases of ^{131}I from the SRS reactors. The liquid waste releases were represented by triangular distributions with a mode equal to the reported release. Minimum and maximum values for these distributions were ±25% of the mode for times before 1971 and ±20% of the mode for later years. For years prior to

1964, the effect of filtration on the release ratios was included. A uniform distribution over the range of values discussed above (0.31–0.74) was used for the calculations.

Results of the calculations are shown in Figure 4.2-11 for all years of operation. The plot provides three percentiles (5th, 50th or median, and 95th) of the distributions that were estimated. The release estimates shown are those for all forms of ^{131}I. Based upon measurements at commercial PWRs (Pelletier et al. 1978b), it is estimated that about 30% of the total would be elemental iodine and that the remainder would be divided between less reactive inorganic forms (~40%) and organic iodides (~30%).

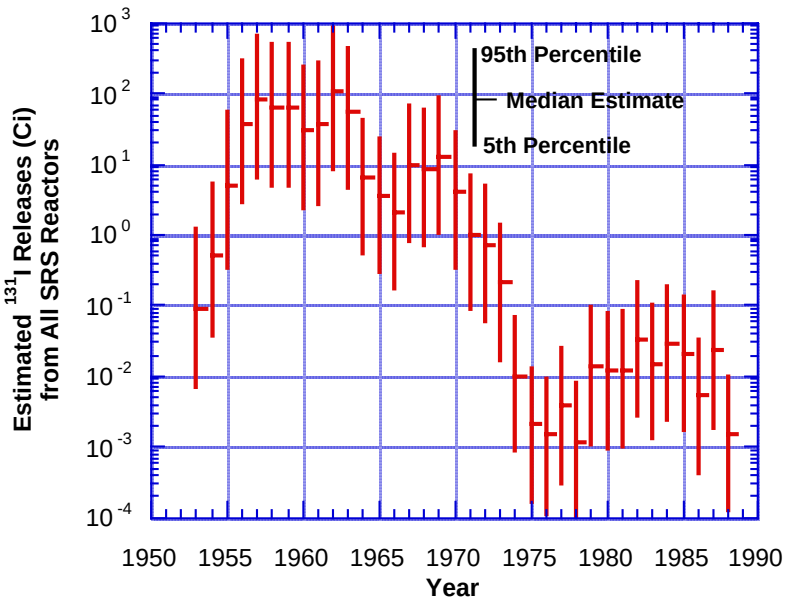

Figure 4.2-11. Estimated releases of ^{131}I from all SRS reactors to the atmosphere. For each year, the length of the vertical line shows the range (5th to 95th percentiles) of the distribution of release estimates and the median estimate is indicated by the horizontal bar.

The main (5th, 25th, 50th, 75th, and 95th) percentiles of the distributions of release estimates for all SRS reactors are tabulated in an Excel® spreadsheet. In that listing, the median (50th percentile) value is highlighted to guide the reader to the central estimate of the amount released. The 5th, 50th, and 95th percentile values from this table are the basis for the release estimates plotted in Figures 4.2-11 and 4.2-12. The spreadsheet can be directly accessed by clicking on the following hyperlink: Est_I-131_Releases_(Reactors).xls.

Figure 4.2-12 shows results of the calculations for only those years when releases were reported. It provides a comparison between the estimates resulting from the estimation procedure and those that were reported. Correspondence between the estimated and reported values is not surprising because the release ratios used in the calculations were taken from this period. However, the comparison does indicate that the estimates are in the expected range.

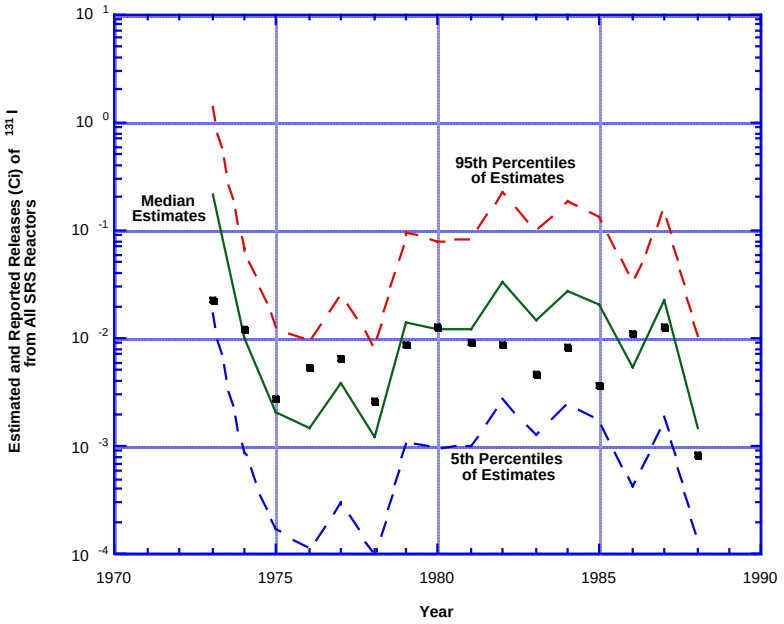

Figure 4.2-12. Comparison of estimated radioiodine releases from all SRS reactors to the atmosphere with measured values (Kantelo et al. 1993). The median and range (5th to 95th percentiles) of the distribution of release estimates are shown by solid and dashed lines, respectively. The reported annual releases are indicated by the solid squares.

Releases from Other SRS Facilities

The only other facility that released measurable amounts of ^{131}I at the SRS was the Savannah River Laboratory (SRL) research facility. Laboratory research in support of processing activities was conducted by the SRL. Some of the research employed short-cooled irradiated fuel that contained ^{131}I and some of the radioiodine was released. Kantelo et al. (1993) indicate that the total measured release was 6.4 Ci of ^{131}I, about half of which was reported to have occurred in 1958 (Cummins et al. (1991). The next largest release, about 1.6 Ci, was reported to have occurred in 1964 (Cummins et al. 1991).

Based upon the previous discussion of radioiodine monitoring at the SRS, it is unlikely that releases occurring prior to 1962 were measured reliably. Recorded releases between 1958 and 1961 total about 3.4 Ci of ^{131}I; no estimates are given for prior years (Cummins et al. (1991). It is believed, based on other experience (Sill and Flygare 1960), that most of the radioiodine released from the shielded cells was probably in elemental form. It seems unlikely, considering previous discussions of corrections of estimated releases, that more than 20 Ci of ^{131}I were actually released from the SRL stacks. The SRL releases are small compared to those from reactors and much smaller than those from reprocessing plants.

RELEASES OF BETA-GAMMA-EMITTING PARTICLES

The term "beta-gamma-emitting particles" refers to a large group of radioactive fission and activation products of many non-volatile elements. These radionuclides are produced in reactors when fuel and reactor components are irradiated. When released, they are generally associated with solid particles and are thus distinguished from the radioactive noble gases and the radioiodines, which are always or much more likely to be in gaseous form. Some radionuclides in this category are very short-lived and decay prior to fuel processing, which (for reasons discussed earlier) provides the principal opportunity for release to the atmosphere. Those that are released generally have half-lives on the order of tens of days (like ^{89}Sr, ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{141}Ce), hundreds of days (like ^{106}Ru and ^{144}Ce), or many years (like ^{90}Sr and ^{137}Cs).

This section also focuses on the F and H fuel processing facilities because fuel dissolution and processing activities occurred there. Measurement records, summarized by Cummins et al. (1991), show that the F and H fuel processing plants were the largest sources of release of beta-gamma-emitting particles at the SRS. Some releases were measured from the reactor facilities and from the SRL stacks, but the quantities were much smaller.

Original weekly and monthly reports have been reviewed and the data on atmospheric releases contained in them has been compiled to prepare a history of reported releases of beta-gamma-emitting particles from the SRS fuel processing facilities. Initial measurements did not quantify releases of specific isotopes, but were estimates of the amounts of "non-volatile beta" activity released from the stacks (Du Pont 1954a-l, 1955a-l, 1956a-l, 1957a-l, 1958a-l, 1959a-l, 1960a). Early in 1960, results of more specific analyses were first reported. During most of the 1960s, release estimates were given for the sums of: two ruthenium isotopes (^{103}Ru plus ^{106}Ru); two strontium isotopes (^{89}Sr plus ^{90}Sr); two cerium isotopes (^{141}Ce plus ^{144}Ce); the zirconium-niobium pair (^{95}Zr plus ^{95}Nb); and for the long-lived cesium isotope, ^{137}Cs (Du Pont 1960b-l, 1961a-l, 1962a-l, 1963a-l, 1964a-l, 1965a-l, 1966a-l, 1967a-l, 1968a-l). Beginning in 1969, routine monthly reports contained separate release estimates for the two ruthenium isotopes and that practice was continued in later years (Du Pont 1969a-l, 1970a-l, 1971a-l). At the start of 1971, the listing was further segregated to provide estimates for ^{95}Zr and ^{95}Nb as well as for ^{141}Ce and ^{144}Ce (Du Pont 1971a-l).

Some additional data are provided in the SRS summary release report (Cummins et al. 1991). Included there are estimates of releases of the two strontium isotopes (^{89}Sr plus ^{90}Sr), the two ruthenium isotopes (^{103}Ru plus ^{106}Ru), and ^{137}Cs from the F and H processing plants during the early years of operation. The following illustrations are based upon the report of Cummins et al. (1991). Figures 4.2-13 and 4.2-14 show, for F-Area and H-Area respectively, the sums of the reported releases of the beta-gamma-emitting particles discussed here. Those results generally agree with the sums of releases of beta-gamma-emitting particles that were derived from the monthly reports. Two significant differences can been seen in the spreadsheet containing the reported releases. For F-Area in 1959, the sum of releases reported by Cummins et al. (1991) is about double that derived from the monthly reports (~8 Ci versus ~4 Ci). For H-Area in 1955, the sum of releases reported by Cummins et al. (1991) is about 19 Ci. That total is about 5 Ci smaller that the total of about 24 Ci derived from the monthly reports.

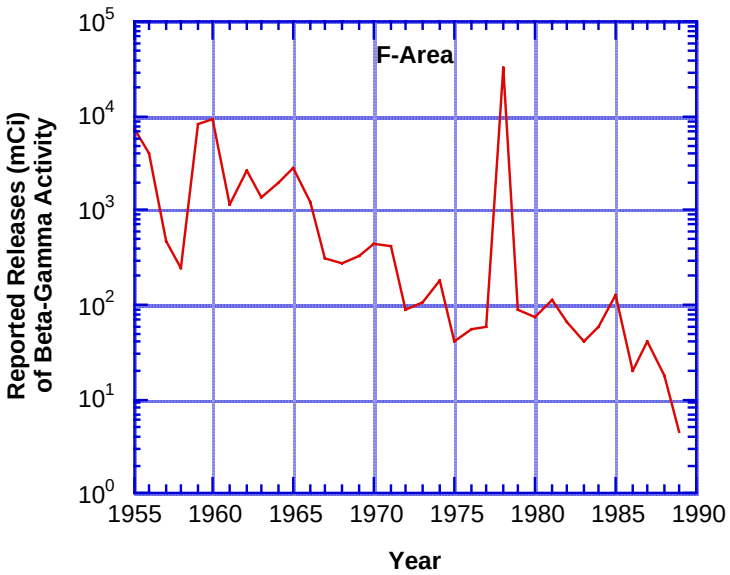

Figure 4.2-13. Reported releases of beta-gamma-emitting particles to the atmosphere from F-Area (Cummins et al. 1991). Estimates for ^{89,90}Sr, ^{95}Zr-Nb, ^{103,106}Ru, ^{137}Cs, and ^{141,144}Ce have been summed.

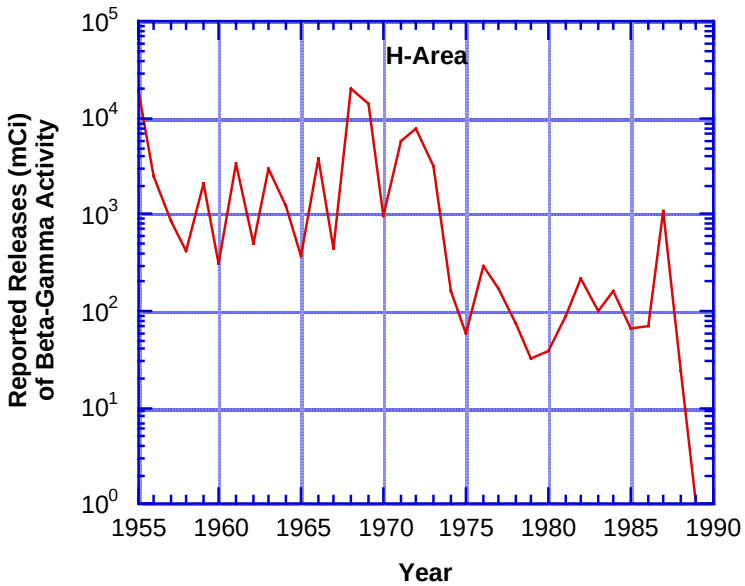

Figure 4.2-14. Reported releases of beta-gamma-emitting particles to the atmosphere from H-Area (Cummins et al. 1991). Estimates for ^{89,90}Sr, ^{95}Zr-Nb, ^{103,106}Ru, ^{137}Cs, and ^{141,144}Ce have been summed.

Figure 4.2-15 shows the releases of ruthenium and cerium isotopes from F-Area. There are no estimates for the latter nuclides prior to 1960. Comparison of this figure with Figure 4.2-13 shows that releases of ^{103}Ru and ^{106}Ru comprise the largest fraction of the total releases of beta-gamma-emitting particles from F-Area. Ruthenium isotopes are identified in some monthly reports as being major components of stack releases; for example, see Du Pont (1956i, 1958j). The large release in 1978 was also of ruthenium (Cummins et al. 1991). Figure 4.2-16 shows the releases of ^{89,90}Sr, ^{95}Zr-Nb, and ^{137}Cs from F-Area. Releases of zirconium and niobium, which are only reported for years after 1959, exceed those of the longer lived isotopes in later years. In some years the releases of ^{95}Zr and ^{95}Nb exceed those of ruthenium isotopes.

Figure 4.2-17 shows the releases of ^{103,106}Ru and ^{141,144}Ce from H-Area and Figure 4.2-18 shows the releases of the isotopes of strontium, zirconium, niobium, and cesium from H-Area. Again, the ruthenium isotopes account for large fractions of the releases for most years, although a release of about 1 Ci of ^{137}Cs is prominent in 1987. Separate reviews of the releases of cesium and strontium, based upon the summary report, have been prepared (Carlton et al. 1992a, 1992b).

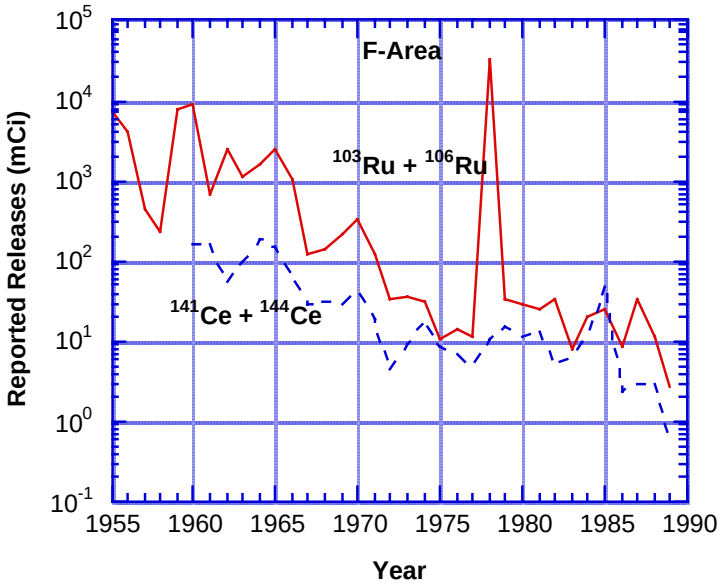

Figure 4.2-15. Reported releases of ^{103,106}Ru and ^{141,144}Ce to the atmosphere from F-Area at the SRS (Cummins et al. 1991).

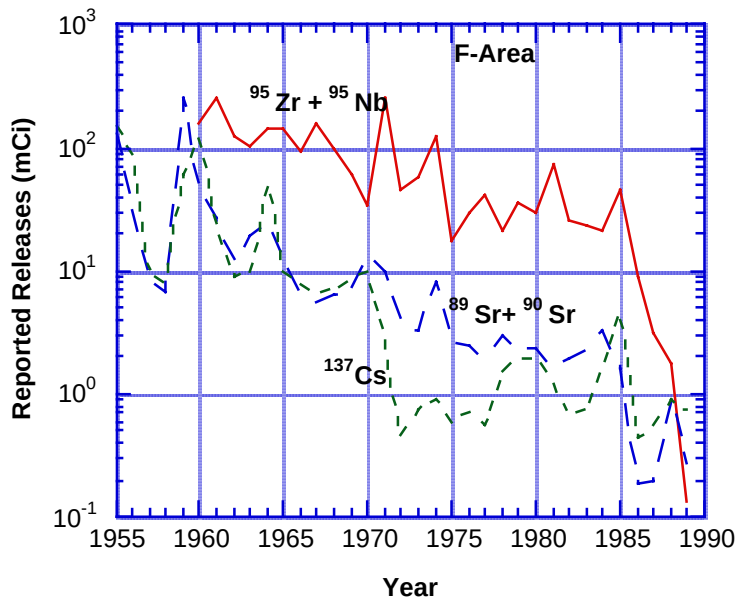

Figure 4.2-16. Reported releases of ^{89,90}Sr, ^{95}Zr-Nb, and ^{137}Cs to the atmosphere from F-Area at the SRS (Cummins et al. 1991).

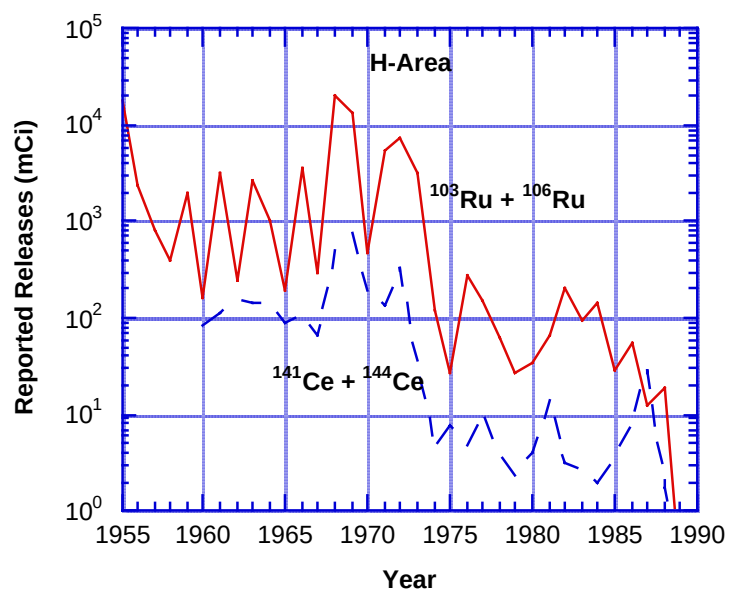

Figure 4.2-17. Reported releases of ^{103,106}Ru and ^{141,144}Ce to the atmosphere from H-Area at the SRS (Cummins et al. 1991).

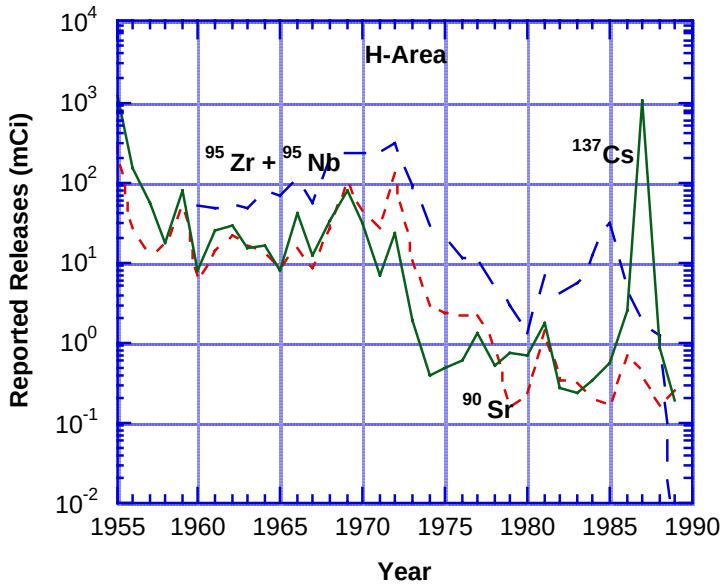

Figure 4.2-18. Reported releases of ^{89,90}Sr, ^{95}Zr-Nb, and ^{137}Cs to the atmosphere from H-Area at the SRS (Cummins et al. 1991).

The release estimates that are shown in these figures do not reflect any corrections for sampling line losses. As discussed previously, the sampling line for the F- and H-Area stacks were long and some deposition of particles would be expected. However, evaluation of such losses is problematic. Information on the sizes of particles in the stack effluents is limited and largely confined to results for plutonium. Those results are presented and discussed in Chapter 4.4. Whether the sizes of particles containing beta-gamma-emitters are comparable to those containing plutonium (from a different area in the facility) is an open question.

Some estimates of sampling line losses are also presented in Chapter 4.4. Losses of 20–30% were estimated for particles with geometric mean diameters less than 1.5 µm. That size range is consistent with normal operation of the effluent filtration systems, which would efficiently remove larger particles. However, sampling line losses during periods when there were filter failures would probably be greater because larger particles could pass through the defective filters. Filter failure events occurred during startup of both fuel processing plants and in H-Area in 1969. Calculations presented in Chapter 4.4 suggest that, for plutonium particles, sampling line losses of 70-80% may have occurred at those times.

Estimates of releases of beta-gamma-emitting particles are tabulated in an Excel® spreadsheet. The spreadsheet can be directly accessed by clicking on the following hyperlink: Ann_B-G_Part_Releases.xls. Because of the great uncertainty about the applicability of the available plutonium particle size data, these estimates (like those shown in the figures) have not been corrected for sampling line losses. For screening calculations, a cautious procedure would be to multiply releases of beta-gamma emitting particles in the spreadsheet by a factor of 4. It is unlikely that all actual releases were more than four times those that were reported; thus estimates of potential doses would likely not be underestimated by such a procedure.

SUMMARY OF RELEASE ESTIMATES

Releases of Radioiodines to the Atmosphere

The largest releases of radioiodines from SRS facilities to the atmosphere were from the tall stacks of the fuel processing facilities located in F-Area and H-Area. The main focus of the analysis was estimation of the releases from these facilities. Estimates of the annual releases of ^{131}I and ^{129}I from the F and H fuel processing plants are contained in Excel® spreadsheets, which can be accessed by clicking on the following hyperlinks: Revised_I-131_Releases_(F,H).xls and I-129_Releases_(F,H).xls. Radioiodine releases from the SRS production reactors were estimated to be more than 80 times smaller than those from the fuel processing facilities. The estimated annual releases of ^{131}I from all reactors combined are compiled in the Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: Est_I-131_Releases_(Reactors).xls. Releases from research facilities at the Savannah River Laboratory were estimated to have been more than 20 times smaller than those from reactors and were not evaluated in detail. We estimated that an upper bound total for those releases is 20 Ci.

Radioiodines released in elemental form (I_2) are more likely to enter the air-grass-cow-milk food chain. For that reason, such releases have greater significance to the health of the public living near the SRS. Releases of elemental iodine were estimated separately for the reprocessing facilities and are shown in the spreadsheet referenced above. Measurements at commercial PWRs suggest that about 30% of the ^{131}I releases from the reactors may have been in elemental form. Experience at other facilities suggests that all of the release from the SRL hot cells was likely to be I_2.

Releases of Beta-Gamma-Emitting Particles to the Atmosphere

The largest releases of particles containing beta-gamma-emitting fission and activation products to the atmosphere also occurred at the fuel processing areas in F-Area and H-Area at the SRS. The particles, which are not volatile like the radioiodines, contained a variety of radionuclides. Releases of the following radionuclides were the most important: ^{89}Sr, ^{90}Sr, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{137}Cs, ^{141}Ce, and ^{144}Ce. Initial estimates were of the composite activity and individual isotopes were not measured separately for many years. Estimates of releases of these radionuclides are contained in an Excel spreadsheet, which can be accessed by clicking on the following hyperlink: Ann_B-G_Part_Releases.xls.

Release estimates for these radionuclides have not been revised to account for deposition in the sampling lines. Adequate information about particle sizes is not available to permit estimation of the correction factors. For screening purposes, a cautious approach of multiplying the release estimates for beta-gamma-emitting particles by 4 is recommended.

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CHAPTER 4.3

PRODUCTION AND ATMOSPHERIC RELEASE OF ACTIVATION PRODUCTS

ABSTRACT

The primary activation product of interest in terms of airborne release and potential offsite dose is ^{41}Ar. Even though it is a short-lived radionuclide, ^{41}Ar is a noble gas readily released from the reactor stacks, and most has not decayed by the time it moves offsite with normal wind speeds. SRS reactor operations produced and released relatively large quantities of ^{41}Ar, and its production rate in the air blanket surrounding a reactor should have been roughly proportional to the reactor power level. Cummins et al. (1991) provides an SRS-developed estimate of ^{41}Ar releases, which we compare to other measurements, check against reactor power levels, and accept as a generally reasonable estimate of SRS ^{41}Ar releases.

While these values represent the best available estimates for ^{41}Ar releases from the SRS reactors, the values presented for the later years (1974–1988) are quite low when compared to reactor power levels and overall average ^{41}Ar production levels. We also observe that the ^{41}Ar release values presented for certain of the early years (1955–1967) are quite high when compared to reactor power levels for the same period. The reason for these apparent discrepancies is not clear, and adds to the uncertainty in our estimates of ^{41}Ar releases.

INTRODUCTION

Most of the radioactivity produced by the five SRS production reactors involved fission products, created when ^{235}U, ^{239}Pu, or ^{233}U split into two or more smaller atoms. In addition, neutrons captured by some materials inside the reactor created radioactive isotopes called activation products. Some activation products were created outside the fuel or target protective cladding, and thus were more readily released to the outside environment than fission products produced inside intact fuel elements. Of these, ^{41}Ar was quite readily released to the environment, and must be evaluated in a dose reconstruction for its potential to expose people living nearby.

The Health Physics and Radiological Health Handbook lists some 300 target/product pairs in its neutron activation database (Shleien 1992). It is neither feasible nor necessary to consider all possible activation products in this study; we limit this research to consideration of those materials with any significant potential for dose to individuals living offsite. *RAC*'s approach to identifying these key radionuclides involves screening for such characteristics as quantity. Chapter 3 of this report provides details concerning our screening of radioactive materials encountered at the SRS, to identify those of greatest potential significance. We reproduce some of that screening logic below, to demonstrate the approach used to focus our activation product research.

THE SCREENING PROCESS

We began the screening process during Phase I of the study, and published a report in June 1995 identifying key radionuclides in the context of dose reconstruction (Meyer et al. 1995).

For air, the more important contributors emerging from the Phase I screening were ^{241}Am, ^{14}C, ^{137}Cs, tritium, ^{129}I, ^{131}I, ^{238}Pu, ^{239,240}Pu, ^{103}Ru, ^{106}Ru, ^{89}Sr, ^{90}Sr, uranium, and the noble gases including ^{41}Ar. For surface water, the more important dose contributors were ^{241}Am, ^{137}Cs, ^{60}Co, tritium, ^{131}I, ^{238}Pu, ^{239,240}Pu, ^{89}Sr, ^{90}Sr, ^{35}S, ^{99}Tc, ^{65}Zn, and Zr,^{95}Nb.

During Phase II of the project, we performed a more detailed assessment of release estimates to provide a more realistic ranking of the radionuclides that were identified in the Phase I, cautious screening methodology. The assessment was based on a methodology recently published by the National Council on Radiation Protection and Measurements (NCRP 1996). We looked at the relative importance of each radionuclide with respect to pathway of exposure. Chapter 3 of this report summarizes the results of that assessment.

For surface water pathways, ^{137}Cs dominates all pathways except the ingestion of drinking water, where tritium is most important. Strontium-90 is also relatively important as a contributor to dose via the surface water pathways when considering the ingestion of vegetables and meat as the major exposure pathways. Phosphorus-32 is important with respect to the ingestion of fish, while ^{131}I should be considered when the ingestion of drinking water and meat are evaluated.

For releases of radionuclides to air from the SRS, Chapter 3 shows that tritium and iodine are the major contributors to radiation dose from all airborne pathways except plume immersion. Argon-41 contributes almost 100% of the dose to a person who may be standing in the plume when it is released from the facility. Plutonium-238 and uranium are also relatively important contributors to dose via the inhalation pathway, contributing about 20% and 5% of the dose, respectively. Plutonium-239,240, a minor contributor to dose, is responsible for less than 5% of the dose from eating vegetables. Nevertheless, because it does meet our criteria of contributing greater than 1% to the dose, a more detailed source term reconstruction was undertaken.

We therefore focus our attention on activation products with potential for significant offsite human exposure: ^{32}P, ^{35}S, ^{41}Ar, ^{51}Cr, ^{60}Co and ^{65}Zn. We exclude radionuclides with an estimated total dose contribution of less than 1% via either the water or air pathway.

ACTIVATION PRODUCT CHARACTERISTICS AND RELEASE POTENTIAL

The radionuclides considered here were created in the SRS reactors by neutron capture. Their half-lives range from 1.8 hours (^{41}Ar) to 5.3 years (^{60}Co). Table 4.3-1 lists characteristics of these radionuclides. The radionuclide ^{41}Ar is a noble (nonreactive) gas produced in the air surrounding the reactor. Because this air volume communicated with the air in the reactor building proper, much of the ^{41}Ar produced in a reactor was released with ventilation air exiting the reactor stack.

Potential for Activation Product Releases at the Production Reactors

The largest releases of activation products occurred during the 1960s, and consisted primarily of ^{41}Ar from the reactors. The five reactors primarily responsible for ^{41}Ar gaseous releases were located on a rough circle, about halfway between the Site center and the boundary. As discussed in Chapter 2 of this report, the primary role of the production reactors was to produce plutonium and tritium for weapons development. Neutrons produced during fission interacted with fuel, targets and structural materials, and with chemicals in the moderator, creating activation products but releasing relatively small quantities of most to the environment.

This was because the products were largely contained within the materials in which they were created. Such activation products, if contained in solid materials including the fuel or metal reactor structure, did have the potential for fractional release later when fuel rods and targets were dissolved in the reprocessing canyons. Activation products produced within the liquid moderator chemicals, or within fuel elements which subsequently developed leaks, had a higher release potential, especially during the early years when the reactor fuel cooling basins were routinely purged as a maintenance procedure. Chapter 5 of this report considers activation product releases to surface waters, and the reader is referred to that chapter for a detailed discussion of such releases.

Table 4.3-1. Characteristics of Activation Products

Radionuclide	Primarily produced from	Half-life	Beta energy (MeV) max	Gamma energy (MeV)
^{32}P	^{31}P	14.3 d	1.7	None
^{35}S	^{35}Cl	87.3 d	0.17 (100%)	None
^{41}Ar	^{40}Ar	1.83 h	1.2 (99%)	1.29 (99%)
^{51}Cr	^{50}Cr	27.7 d	Electron capture	0.32 (9%)
^{60}Co	^{59}Co	5.27 y	0.31	1.17 (100%) 1.33 (100%) 0.51 (3.4%)
^{65}Zn	^{64}Zn	244 d	Electron capture	1.12 (49%)

[a] Source: Health Physics Handbook (Shleien 1992).

At least one activation product was produced in such a way that it was rapidly released to the outside environment. Neutrons interacting with the natural noble gas ^{40}Ar in the air around the reactor vessel created ^{41}Ar, which quickly mixed with reactor building air and was released via the airflow through the stack. Once out the stack, reactor gases moved with local air flows offsite, and individuals living nearby were potentially exposed to beta and gamma radiation emitted during ^{41}Ar decay. Because the half-life of ^{41}Ar is relatively short, radiation exposures calculated for this radionuclide were highest near the site boundary, and decreased more rapidly than for most other radionuclides with distance from the site. Argon-41 is the principal activation product of interest in terms of atmospheric releases, and when considering offsite human health risk.

The reactors began operating in the years 1953–1954, and were all operating by 1955. They were routinely shut down to replace fuel and targets and occasionally when unplanned events occurred. Test reactors, spontaneous fission, and some materials delivered from outside the SRS also contributed small quantities of activation products to the SRS inventory.

Potential for Release of Activation Products in Areas Other Than the Reactors

As mentioned above, under ideal conditions, most activation products produced within reactor fuel and targets would remain contained within the cladding until reprocessing occurred. Some leakage inevitably occurred, however, allowing activation products to move into the moderator circulating around the fuel, or to leak into the fuel cooling and disassembly basins. In addition, some materials circulating in the moderator were activated during neutron exposure. Under normal conditions, a significant fraction of the activation products in the moderator adhered to the fuel and targets; these materials were transported to the disassembly basin when

the reactor fuel was unloaded. During cooling time in the basin, dissolution and corrosion allowed a fraction of the activation products to escape into the basin water. Air and water at the reactors were monitored for such possible releases.

Starting in the 1960s, basin water was passed through ion exchange resins to remove most radionuclides. Resins were either reworked to capture and concentrate the radionuclides for disposal or buried in the Burial Grounds (renamed the Solid Waste Disposal Facility in 1990). Chapter 5 considers these processes in more detail. During cooling in the basins, short-lived radionuclides decayed away.

After cooling, fuel and targets were treated in the chemical separations areas (the canyons). During these chemical separations, small quantities of some activation products volatilized during the evaporation of aqueous wastes; some material was discharged to seepage basins. In addition, small quantities of activation products moved through H-Area process cooling water because of heating and cooling coil leakage. Most of this activity was collected in retention basins and sent to seepage basins, but some leakage did occur into Four Mile Branch (Four Mile Creek) (Carlton and Denham 1996).

Chemical separations process aqueous wastes were evaporated and sent to the large waste storage tanks in F-Area and H-Area. Condensate from the evaporators was sent to seepage basins until 1988, when the Effluent Treatment Facility began to be used to remove radionuclides and chemicals from such wastes. Treated wastes from the new facility were discharged into Upper Three Runs Creek. High-level liquid wastes were stored for decay in temporary tanks, then shipped to F-Area for processing through the waste system. Solid wastes were buried in the Solid Waste Disposal Facility.

According to Carlton and Denham (1996), “Most of the atmospheric and aqueous effluents in the chemical separations and SRTC have been monitored for possible activation product releases...Activation product activity released through stacks was small enough that offsite transport through the atmosphere has not been detected.”

Activation Products Release Data

The SRS has in recent years published a number of documents summarizing radioactive releases for the primary operating history of the Site (1954–1989). The Carlton and Denham (1996) report discussed above presents annual data by release point (reactors, reprocessing areas, and certain other areas onsite) for the activation products ^{32}P, ^{51}Cr, ^{60}Co and ^{65}Zn. While these summaries are of interest in the examination of releases by location onsite, an earlier report prepared, Cummins et al. (1991), summarizes releases for the same period by pathway and year and for all six activation products of interest here. For the purposes of dose reconstruction, we choose to extract annual release data from Cummins et al. (1991).

Table 4.3-2 and Figure 4.3-1 present annual data for the six activation products we considered. To display the data more effectively, the ^{41}Ar values were divided by 100 before presentation in Figure 4.3-1.

Table 4.3-2. Activation Products Released from the SRS Per Year, All Sources, by Pathway (Ci)[a]

Year	^{32}P – Seepage basins	^{32}P – Streams	^{35}S – Seepage basins	^{35}S – Streams	^{41}Ar – air[b]	^{51}Cr – Seepage basins	^{51}Cr – Streams	^{60}Co – Seepage basins	^{60}Co – Streams	^{60}Co – Air	^{65}Zn – Seepage basins	^{65}Zn – Streams
55					5.0×10^{4}				6.0×10^{-2}			
55					1.4×10^{5}				2.2×10^{-1}			
56					2.5×10^{5}				6.1×10^{-1}			
57					2.9×10^{5}			2.4	1.6			
58					3.4×10^{5}			9.1×10^{-1}	1.1			
59					4.4×10^{5}			4.7×10^{-1}	2.7			
60					4.0×10^{5}		1.1×10^{1}	2.7×10^{-1}	7.8			4.3
61				6.6×10^{1}	4.2×10^{5}		6.2×10^{1}	1.6×10^{-1}	6.3			2.1×10^{1}
62				5.1×10^{2}	4.3×10^{5}		2.1×10^{2}	8.1×10^{-2}	1.3×10^{1}			3.2
63				1.6×10^{2}	4.5×10^{5}		1.3×10^{3}	7.5×10^{-1}	5.1			3.3×10^{1}
64		3.1		9.1×10^{1}	3.7×10^{5}	6.1×10^{1}	1.1×10^{3}	2.7	3.1		4.0×10^{-2}	2.0×10^{1}
65	3.7×10^{1}	1.4×10^{1}		8.6×10^{1}	2.7×10^{5}	1.0×10^{2}	7.8×10^{2}	1.6	1.1×10^{1}		3.3×10^{-1}	8.9
66	1.1×10^{2}	1.0×10^{1}		1.2×10^{2}	2.8×10^{5}	7.6	6.3×10^{2}	2.7×10^{-1}	5.1		1.0×10^{-2}	7.5
67	9.0	4.1		1.5×10^{2}	3.2×10^{5}	2.5	2.4×10^{2}	2.5×10^{-1}	1.7		1.2×10^{-1}	7.3
68		2.4	1.9×10^{2}	1.7×10^{2}	2.2×10^{5}	1.2×10^{2}	8.6×10^{1}	1.9	1.8	3.9×10^{-2}	3.8	4.5
69		7.3×10^{-1}	7.7×10^{1}	6.4×10^{1}	1.4×10^{5}	3.0×10^{2}	6.9×10^{1}	1.4	2.1×10^{-1}	1.1×10^{-2}	5.6×10^{-1}	1.5
70		7.7×10^{-1}	1.8×10^{1}	2.6×10^{1}	1.1×10^{5}	8.6	1.6×10^{1}	4.9×10^{-1}	1.7×10^{-1}	2.8×10^{-3}		5.1×10^{-1}
71		9.2×10^{-1}	3.1	1.1×10^{1}	1.4×10^{5}	2.5×10^{1}	7.3	4.7×10^{-1}	4.6	2.2×10^{-2}	3.0×10^{-2}	1.8
72		3.0×10^{-1}	9.7×10^{-1}	1.5	1.7×10^{5}	3.1×10^{1}	1.4	4.0×10^{-2}	1.8×10^{-1}	7.2×10^{-3}		
73	1.8×10^{-2}	1.3×10^{-1}	1.7×10^{-1}	1.7	1.8×10^{5}	1.4×10^{1}	4.5×10^{-1}	4.3×10^{-2}	2.7×10^{-2}	1.8×10^{-3}		
74	2.2×10^{-2}	1.4×10^{-2}	7.3×10^{-1}	5.0	1.1×10^{5}	7.9	6.0×10^{-1}	4.9×10^{-1}	1.0×10^{-3}	3.9×10^{-3}	7.6×10^{-1}	1.4×10^{-1}
75	8.0×10^{-3}	2.0×10^{-3}	5.1×10^{-2}	2.9×10^{-1}	6.5×10^{4}	4.6	1.2×10^{-1}	4.2×10^{-1}	9.0×10^{-3}	1.1×10^{-3}	5.7×10^{-1}	2.0×10^{-3}
76	9.0×10^{-3}	1.7×10^{-2}	3.1×10^{-1}	3.4×10^{-1}	8.3×10^{4}	6.4	3.6×10^{-1}	4.7×10^{-1}	2.0×10^{-3}	1.3×10^{-4}	4.4×10^{-1}	
77	2.2×10^{-2}	6.4×10^{-3}	5.1×10^{-2}	6.1×10^{-2}	6.5×10^{4}	6.1	6.6×10^{-1}	4.0×10^{-1}	1.0×10^{-1}	3.8×10^{-4}	5.3×10^{-1}	
78	3.6×10^{-3}	2.7×10^{-5}	1.3×10^{-2}	1.5×10^{-2}	5.3×10^{4}	1.5	4.0×10^{-2}	1.7×10^{-1}	3.9×10^{-4}	3.8×10^{-4}	5.4×10^{-2}	
79	2.0×10^{-3}	4.0×10^{-3}	1.9×10^{-2}	1.1	5.3×10^{4}	1.3	5.4×10^{-1}	1.0×10^{-1}	4.1×10^{-1}	4.0×10^{-3}	8.0×10^{-2}	
80	1.9×10^{-3}		1.1×10^{-2}		7.0×10^{4}	1.5		2.2×10^{-1}	1.6×10^{-3}	6.2×10^{-4}	1.4×10^{-1}	
81	3.7×10^{-3}		7.4×10^{-2}		6.2×10^{4}	2.4		1.6×10^{-1}	2.5×10^{-4}	8.9×10^{-5}	7.4×10^{-2}	
82	6.9×10^{-3}		1.8×10^{-1}		6.0×10^{4}	2.9		2.6×10^{-1}	1.1×10^{-4}	4.4×10^{-5}	9.2×10^{-2}	
83	1.6×10^{-2}		1.0×10^{-1}		4.1×10^{4}	1.2		2.2×10^{-1}	1.9×10^{-3}	1.7×10^{-4}	9.5×10^{-2}	
84	3.0×10^{-3}		1.5×10^{-2}		3.6×10^{4}	5.9		2.7×10^{-1}	3.1×10^{-4}	5.4×10^{-5}	1.0×10^{-2}	
85	6.1×10^{-4}		4.3×10^{-2}		5.2×10^{4}	3.8		2.9×10^{-1}			3.0×10^{-3}	
86	8.5×10^{-5}		2.7×10^{-2}		8.3×10^{4}	5.5×10^{-1}		1.3×10^{-1}		8.0×10^{-6}	5.2×10^{-2}	
87	1.3×10^{-5}		2.3×10^{-2}		8.8×10^{4}	9.8×10^{-2}		1.5×10^{-2}			2.1×10^{-2}	
88	9.8×10^{-6}		2.4×10^{-3}		3.0×10^{4}	2.7×10^{-3}		8.1×10^{-4}		3.0×10^{-6}	4.7×10^{-3}	
89	2.1×10^{-4}		2.1×10^{-4}									

[a] Source: Cummins et al. (1991).
[b] ^{41}Ar values before 1971 are estimates per Cummins et al (1991).

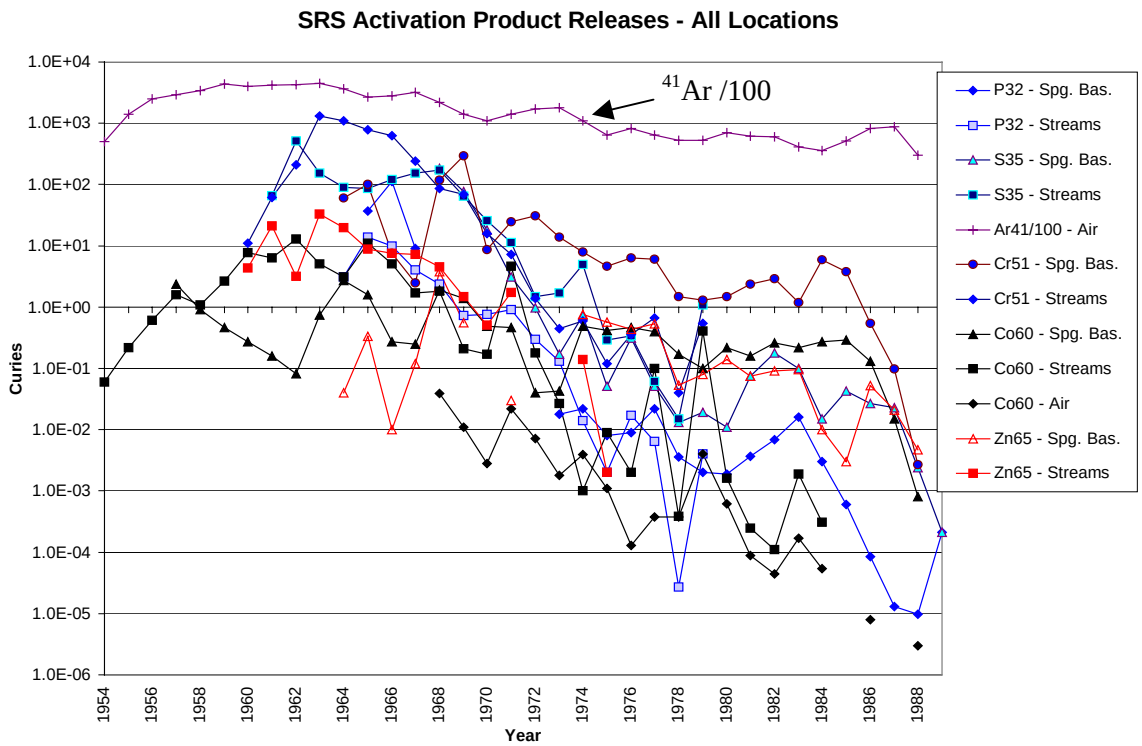

Figure 4.3-1. Activation products released from the SRS per year, all sources, by pathway (Cummins et al. 1991).

SUMMARIES OF OTHER AVAILABLE DATA

We searched the *Radiological Assessments Corporation* (*RAC)* SRS Dose Reconstruction Phase I document database for records with information related to the activation products. Most of the documents recovered are brief reports of specific events and do not lend themselves to developing a clear pattern related to activation product releases over the entire history of the Site. They primarily record offnormal incidents at the SRS, when specific activation products were noticed or anticipated in an effluent and a study was undertaken to determine the radionuclide's source or actual release rate. Because ^{32}P, ^{35}S, and ^{51}Cr have relatively short half-lives, there are few observations of these nuclides in effluents. The nuclides ^{65}Zn and ^{60}Co, with longer half-lives, were more often measured in effluent and in offsite media. Although ^{41}Ar has a very short half-life, it was created in relatively large quantities routinely during reactor operation. As a noble (nonreactive) gas, it was released much more readily to the environment with the stack air stream. Data and estimates discovered during the Phase I document review and summarized here are more complete than for the other activation products.

The Phase I records concerning these nuclides make up a rather sparse set and do not support detailed, comprehensive estimates of SRS releases or dose calculations. However, they do focus on important events associated with activation product releases dating back to early 1956. These events coincide with peak releases seen in recent radionuclide release summaries published by the SRS (Cummins et al. 1991; Carlton and Denham 1996) and, thus, may be used to corroborate key

releases and release periods. We reviewed these records and summarized them below, listed by primary document date (some of the records are actually record groups, containing memos written on a specific topic over several years). Where data are presented, the summaries below contain excerpted tables of those data and graphics where useful. These tables represent the primary data sets for these data fragments in this report -- no spreadsheets were prepared to support them. Similarly, the data graphs presented below are the primary data sets for these data fragments -- no spreadsheet analyses were performed or are available to support them.

Early Discussions Concerning Potential Increases in ^{41}Ar Releases

A note, Hall and Coombs (1956), states that the stack discharges of "A^{41}" were on the order of 1500 Ci per month per reactor area. The monthly stack discharge ranged from 570 Ci in C-Area to 1840 Ci in R-Area. The note states that, "Assuming the present stack activity to be the average activity, the stack discharge ranges from 860 to 2300 Ci per month, with the exception of L area which is shut down." Hall and Coombs note that the ^{41}Ar activity of the stack effluent is almost insignificant while the reactor is shut down and increases greatly during and following startup. The note references an attached table summarizing the results, but the table is missing from the copy recovered by *RAC*.

A memo, Hoy (1962), notes that design modifications being considered for the reactors may cause increased ^{41}Ar releases to the atmosphere. The Health Physics group estimated that 1500 Ci d^{-1} released per reactor would cause dose rates as high as 2.4 mrem h^{-1} at 0.8 mi (well inside the Site boundary). Maximum offsite dose, with overlapping plumes from all five reactors, would not exceed 0.0022 mrem h^{-1}. Hoy (1962) noted that the stack Kanne chambers, low-level beta-gamma counters, and area background detectors might be affected. The memo noted that "the normal gamma background level in the vicinity of SRP is 0.35 mr/24 hr . . .."

A note to file, Coombs (1964), indicates that an anticipated 50-fold increase in the ^{41}Ar release rate, due to ventilation of the K-Reactor with dry air vs. CO_2 beginning in June 1964 did not occur. Kanne chamber readings indicated an ^{41}Ar release of 85 Ci d^{-1}, three times higher than the previous K-Area discharge rate. Exposure rates at the K-Area fence (using a Geiger-Mueller tube and scaler) ranged from 0.34 to 0.49 mrem d^{-1}, "within the normal background range" but highest at the predicted ^{41}Ar maximum concentration. Coombs (1964) speculates that a high percent of air may have already been in the reactor cavities from leakage. Calculations, based on the 50-fold predicted ^{41}Ar increase, had indicated a potential for 2.4 mrem h^{-1} at 0.8 mi downwind. An increase in ^{14}C emission was also predicted. Coombs (1964) notes an estimated discharge of ^{14}C of 0.84 Ci y^{-1} at 30–65 ft^3 min^{-1} purge of K-Area reactor cavities at 10% reactor down time.

Stream Releases

A two-page document fragment, titled "Reactor Area Release Summary" (Anonymous ca. 1964), summarizes amounts of radioactive material released to streams from 1954 through 1964. From 1954 through 1960, radionuclides were characterized as "primarily short-lived materials," associated with the discharge from the reactors of failed fuel elements. The fragment notes "Discontinued routine purge of deionized water," and "75% Np," in association with the 1959 data. The data related to (or likely related to) activation products are summarized below in Table

4.3-3. Table 4.3-4 provides an indication as to the specific radionuclides probably making up the "primarily short-lived materials" source term in the first table. No further information was available to support this speculation.

Table 4.3-3. Releases to Streams

Year	Releases to streams (Ci)
1954	30
1955	112
1956	303
1957	875
1958	537
1959	1323
1960	836

For 1960, Anonymous ca. 1964 notes that "Specific radioanalysis (was) initiated last half of year," but it does not provide the results of the analyses. For 1961–1964, five to six radionuclides are considered individually in author (year). Only activation products are listed in Table 4.3-3.

Table 4.3-4. Activation Products Released to SRS Streams

Year	Total curies to streams	Specific radionuclides analyzed	Curies to streams for the specific radionuclides analyzed
1961	360	^{51}Cr	62
1962	1142	^{51}Cr	199
		^{35}S	512
1963	2594	^{51}Cr	1280
		^{35}S	156
1964	1725	^{51}Cr	1085
		^{35}S	91

The document contains a remarks section, noting changes in processes leading to changes in release rates. Better chemistry control and less corrosion, plus discontinued purging of the thermal shield system, reduced releases in 1960.

Johnson (1962) states that ^{35}S, the main contributor of the long-lived materials, was likely due to degradation of the cation (sulfonated) resin as a result of radiation damage. Johnson (1962) also states that "Cation resin beds were replaced with mixed beds in the moderator purification system to facilitate better Pd control; ^{35}S releases decreased as a result." A 1964 note states that, "The failure to sorb the ^{35}S was attributed to saturation of the anion resin." Also noted was a six-fold increase in the release of ^{51}Cr at that time because of a change in chemistry increasing the retention of chromium by fuel assemblies.

A table titled, "Savannah River Plant. Radioactivity in Reactor Effluent Waters–CY 1968," (Anonymous 1969) tabulates combined reactor effluents for 17 radionuclides including the activation products listed in Table 4.3-5.

Table 4.3-5. Radioactivity in SRS Effluent Waters, All Reactors Combined

Activation product	Combined reactor effluents (Ci released)	Contribution in Savannah River downstream at Highway 301 bridge (pCi L^{-1})
^{51}Cr	85.8	<8
^{32}P	2.4	NA
^{65}Zn	4.5	<1.1
^{60}Co	1.8	<1.4
^{35}S	172	26

Other Data

A memo (Patterson 1969) that discusses ^{60}Co releases notes that a ^{60}Co release was "first" reported in September 1967 and that, "To date only a gaseous form of ^{60}Co has been released from the High Level Caves short stacks, from the 'B' stack, and from the 776-A short stack." Particulate cobalt activity had not penetrated the high efficiency filters. The High Level Caves Off Gas Exhaust system was the source of ^{60}Co in "B" stack." The memo notes particulate ^{60}Co problems in ducts and filters; with the ducts of Cell 12 radiating 1 to 2 R h^{-1} due to internally deposited ^{60}Co. High-efficiency filters in the Cell 12 system had shown dose rates up to 10 R h^{-1} through the filter housing after ^{60}Co manipulations in the cell. Table 4.3-6, and Figure 4.3-2 summarize onsite and offsite ^{60}Co measurements in air for a 7-month period in 1968 and 1969 when activities involving cutting materials containing ^{60}Co were occurring. A partial summary of these same data is also presented in a memo, (Taylor 1969a).

According to Patterson (1969), since September 1967 there had been three periods of significant ^{60}Co release:

1. September–November 1967: 8.8 mCi
2. February 8–March 15 1968: 33 mCi
3. January 9–March 20, 1969: 3.2 mCi.

Patterson (1969) notes that the Environmental Monitoring group collected vegetation samples in the Technical Division area after the first two release periods, with very low concentrations detected. The report notes that ^{60}Co was released from the 776-A short stack when cutting solution was discharged to the high-level drain after ^{60}Co specimens were cut in the metallurgy cell. Several tables, including several handwritten pages, are attached to the report. *RAC* has compared the handwritten (Patterson 1969) and summarized values (above) for the three periods noted and found them to be essentially identical. A note indicates that the SRS ultralow level counting laboratory had just been completed, enabling the detection via coincidence counting of very low levels of ^{60}Co (to 0.5 disintegrations min^{-1}, which equals 227 fCi or 227 × 10^{-15} Ci).

Table 4.3-6. Onsite and Offsite SRS Monitoring for ^{60}Co (Ci/m^3 × 10^{-15})[a]

Month	Hi Vol.[b]	Uncrt. [b]	735-S[b]	Uncrt.	200F&H	Uncrt.	OP[b]	Uncrt.	25 mile[b]	Uncrt.	100 mile[b]	Uncrt.
Dec-68			0.29	0.25	0.11	0.06	0.04	0.02	0.071	0.01	0.062	.010
Jan-69	0.09	0.008	1.34	0.65	0.11	0.05	0.1	0.02	0.05	0.01	0.048	.010
Feb-69	0.038	0.01	9.53	1.46	0.34	0.06	0.116	0.01	0.052	0.01	0.166	.020
Mar-69			7.88	1.45	0.19	0.06	0.133	0.02	0.08	0.01	0.042	.010
Apr-69			1.93	0.52	0.11	0.03	0.178	0.02	0.075	0.01	0.064	.020
May-69			4.81	0.87	0.144	0.1	0.095	0.02	0.135	0.03	0.049	.020
Jun-69			13.06	0.51	0.512		0.529	0.04				

[a] Source: Patterson (1969).

[b] HiVol - high volume air sampler. Uncrt. - Uncertainty. Building 735-S. Buildings 200-F and H. OP - Outer SRS Perimeter. 25 Mile radius from SRS. 100 Mile radius from SRS.

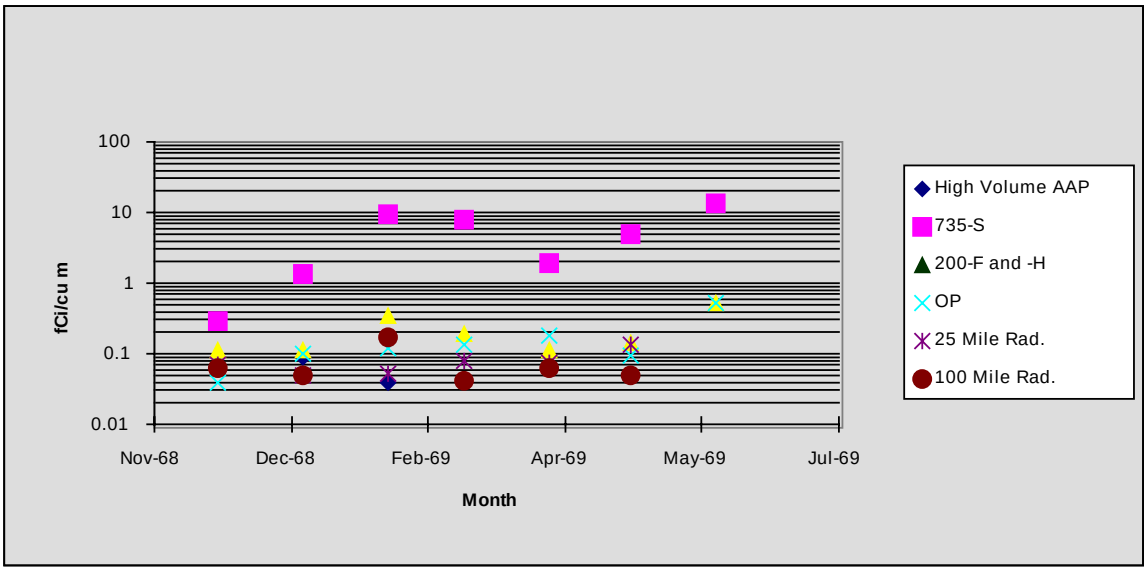

Figure 4.3-2. Onsite and offsite SRS monitoring for ^{60}Co (Patterson 1969).

The following data were compiled from two memos (Taylor 1969a, 1969b) attached to Patterson (1969) after *RAC* evaluation and correction of an apparent error in Taylor (1969b). The correction was based on comparisons to contemporary SRS data, data overlap between the two memos, and a note in Taylor (1969b) indicating a detection limit, using ^{60}Co coincidence counting that was a factor of 1000 lower than the lowest ^{60}Co values presented in the subject memo. The data in Table 4.3-7 and Figure 4.3-3 reflect the factor-of-1000 correction where indicated.

Table 4.3-7. Cobalt-60 in Air—Aiken Airport and Onsite

Date	Sample (m^3)	^{60}Co Ci m^{-3} [× 10^{-15}])	+/–
1/30/68	36,833	.017[b]	.001
10/17/68	178,715	.035[b]	.003
1/3/69	106,040	.038[b]	.004
1/9/69	36,589	.065[b]	.009
1/14/69	35,768	.140[b]	.012
1/15/69	35,149	.103[b]	.010
1/17/69	106,030	<.003[b]	
1/24/69	104,309	.097[b]	.006
1/31/69	109,679	.189[b]	.008
2/7/69	106,073	.073[b]	.005
2/11/69		.034	.010
2/13/69		.065	.010
2/15/69		.030	.010
2/18/69		.042	.020
2/22/69		.012	.003
2/25/69		<.010	
Onsite:			
1/1–31/69[c]	45,851	.590[b]	.021

[a] Source: Extracted from Taylor (1969a and 1969b).
[b] Data corrected for apparent error in the SRS source memo; these values have been reduced by a factor of 1000.
[c] Onsite air sampler, Administration Area (SRS 700 Area).

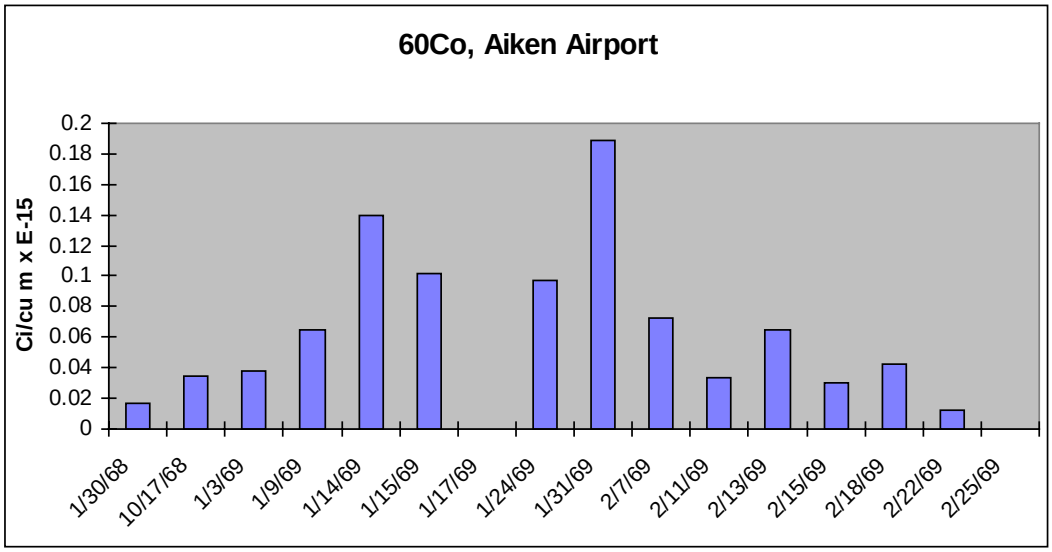

Figure 4.3-3. Cobalt-60 in air—Aiken Airport (extracted from Taylor 1969a, 1969b).

Jacobsen and Marter (1972) provides an interesting perspective on anticipated reactor releases. L-Reactor shut down in February 1968 and was restarted in October 1985. The reactor ran through December 1987, but with two extended shutdowns during the period. While this 1972 report is not of value in estimating L-Reactor activation product releases after the actual 1985 restart, it does base many of its conclusions on the functioning 1972-era reactors, stating that ". . . all facility improvements made in the three operating production reactors at SRP for minimizing environmental effects of operation will be incorporated (in L-Reactor)." The most significant of these modifications was to be construction of a closed-loop heat exchanger-filter-deionizer for cooling, clarifying and decontaminating disassembly basin water. The disassembly basin had been the primary source of release of radioactive materials to liquid effluent during prior operation of L-Reactor. The new system was said to be "comparable to those now used in other operating reactors and will greatly reduce the potential for release of radioactive materials." A program of seal welding was also proposed to reduce leakage from other areas in the reactor system, including the heavy water recirculation system.

Jacobsen and Marter (1972) considers the potential for release of all types of radionuclides; this section extracts data related to the activation products. Jacobsen and Marter anticipated that ^{41}Ar releases would be relatively constant from year to year, with reactor power ranging from about 2000–2300 MW(t). Actual maximum average power after the 1985 L-Reactor restart was reached in April 1986 at about 1900 MW(t), but average power over the 25-month period was just under 700 MW(t), including 8 months of extended shutdowns, or 1040 MW(t) not including the extended shutdowns. This latter value was about one-half the average value developed by the other reactors over their full power lives.

Jacobsen and Marter (1972) notes that some 55,000 gal of heavy water, 99.7% isotopic purity, would be used as moderator. The moderator would initially contain very little tritium, which would build in over the years after startup. Table 4.3-8, extracted from the startup report, shows the anticipated offsite effect of the tritium buildup and consequent increase in routine tritium leakage from the reactor. It is reproduced here to indicate the baseline dose predicted by the SRS for the release of other radionuclides, including the activation products.

Table 4.3-8. Predicted Population Dose from Operation of L-Reactor, for a Potential Mid-1970s Startup[a]

Operation (years following L-Reactor restart)	Population dose (man-rem), from:		
	Atmospheric releases	Aqueous releases	Total
1	26	1.3	27.3
2	28.8	3.9	32.7
3	31.5	5.9	37.4
4	33.7	7.8	41.5
5	36.3	9.8	46.1

[a] Source: Jacobsen and Marter (1972).

More specifically, the annular cavity (the space between the reactor tank and the circumferential shield systems) was to be filled with dehumidified air, with an assumed exchange rate to the reactor building of 100 cfm. This space had been designed to be filled with CO_2, but leakage had made this approach impractical. Natural (1%) ^{40}Ar in the annular cavity air would be

converted to ^{41}Ar by neutron activation and would then be released to the environment via the main stack. Jacobsen and Marter calculate a production rate, decay period, and equilibrium concentration for ^{41}Ar in the operating reactor, but they also compare the value to the then-current value for P- and K-Reactors with similar neutron fluxes and reactor system designs and annual releases of about 50,000 Ci ^{41}Ar. They discuss the possible design of a delay tank to further reduce ^{41}Ar concentration prior to stack exhaust, but they indicate that no such design had been developed for L-Reactor or the other reactors as of 1972. Leakage of air through the moderator system was anticipated to result in another 1000 to 2000 Ci ^{41}Ar released up the stack per year. Fission product gases, concentrating in the blanket gas helium over the moderator, were predicted to be approximately 1000 Ci y^{-1} of ^{85m}Kr, 300 Ci y^{-1} of ^{88}Kr, 800 Ci y^{-1} of ^{135}Xe and 4,000 Ci y^{-1} of ^{133}Xe.

Based on past history in the L-Reactor fuel and target storage basin, estimated first year aqueous releases of activation products to Steel Creek were calculated in Jacobsen and Marter (1972) and are presented in Table 4.3-9.

Table 4.3-9. Estimated First Year Aqueous Releases, Based on 1971 K-Area Data[a]

Activation products	Release (Ci)
^{51}Cr	0.6
^{32}P	<0.1
^{65}Zn	<0.1
^{60}Co	<0.1
^{35}S	3.1

[a] Source: Jacobsen and Marter (1972).

The Jacobsen and Marter report projects the L-Reactor exhaust air rate from the 200-ft stack to be 120,000 ft^3 min^{-1} (3400 m^3 min^{-1}). On the basis of the expected ^{41}Ar release rate, the exhaust stack concentration of ^{41}Ar during L-Reactor operation (presumably at 2000–2300 MW[t]) was estimated to be $2.8 \times 10^{-5} \times$ Ci cc^{-1}, or $28 \times$ Ci m^{-3}. This is equivalent to 95,000 × Ci min^{-1}, or 50,000 Ci y^{-1} ^{41}Ar released , as previously noted in the report.

The Jacobsen and Marter report also estimates annual atmospheric SRS releases associated with L-Reactor operations to be 7×10^{-3} Ci y^{-1} for ^{60}Co. The report estimates maximum atmospheric plant perimeter concentrations associated with L-Reactor operations for ^{41}Ar as $9.9 \times 10^{-12} \times$ Ci cc^{-1}, and for ^{60}Co as $5.6 \times 10^{-19} \times$ Ci cc^{-1}.

Wood and Randolph (1974) describes the development, calibration, and in-stack testing of a continuously operating monitor for the noble gases at SRS. No release data of value are presented, however.

A memo, Mirshak (1983), notes the results of a comparison between SRS estimated ^{3}H and ^{41}Ar concentrations using the WIND system and concentrations measured by the U.S. Environmental Protection Agency. The average ^{41}Ar emission rate for P-Reactor was 1×10^{-3} Ci s^{-1} (a rate of 32,000 Ci y^{-1}) on December 15. A memo from Garrett to Corey on March 28, 1983 notes the same study. *RAC*'s reactor power study (see Chapter 2 of this report) indicates that P-Reactor was in stable operation at the time, at a power level of approximately 1700 MW(t).

A memo, (Anonymous 1986) tabulates SRS annual releases of ^{41}Ar as shown in Table 4.3-10.

Table 4.3-10. SRS Annual Releases of ^{41}Ar (all reactors)[a]

Year	^{41}Ar released (Ci)
1982	60,000
1983	41,000
1984	36,000
1985 (preliminary)	52,000

[a] Source: Anonymous (1986).

Anonymous (1986) estimates the offsite maximum individual dose as 1.54×10^{-6} mrem Ci^{-1} released. *RAC* notes that during this period P-, K-, and C-Reactors were all operating at high average power levels (except that C-Reactor shutdown in mid-1985 and L-Reactor restarted in late 1985).

Merz and Marter (1986) states that the annual guide of 30,000 Ci for ^{41}Ar released to the atmosphere from P-Area was exceeded during November 1985. A release of 2250 Ci in November increased the 1985 total to 30,460 Ci. It is noted that 90% of the ^{41}Ar released originates in the annular cavity, leaks into the process room, is mixed with air and released out through the stack. Because the K-Area and P-Area reactor systems are similar with respect to the conditions that produce ^{41}Ar, one or both of the monitors was believed to be inaccurate. Releases of ^{41}Ar for both areas are tabulated for 1985 (Table 4.3-11). Reactor power levels were similar over this period, although power data for March and April were not recovered during Phase II of this SRS study. To date, *RAC* has been unable to resolve the apparent discrepancy in ^{41}Ar release levels.

Table 4.3-11. Argon-41 Releases from SRS P- and K-Reactors, January–November 1985[a]

1985 Month	P-Reactor release (Ci ^{41}Ar)	K-Reactor release (Ci ^{41}Ar)
January	4200	758
February	5340	950
March	5340	600
April	-	1070
May	1360	1210
June	3100	1142
July	3100	1173
August	3080	1050
September	1750	19
October	940	940
November	2250	826

[a] Source: Merz and Marter (1986).

Sigg and Huang (1986) describes the development, calibration, testing, and validation of a mobile ^{41}Ar plume monitor, the TRAC. The device is sensitive to normal ^{41}Ar releases and is said to be capable of detecting a plume at 70 km downwind from a reactor, under typical transport conditions. The document provides no ^{41}Ar release data, although it does note, that since ^{41}Ar is formed by neutron activation of stable argon in air, its formation rate should be proportional to reactor power for a given reactor. Differences in reactor fluxes and annular cavities may give different source terms from different reactors at the same power level. Although SRP reactor power levels would be a better gauge of source terms, for simplicity in this report it is assumed the average of TRAC's observations represent average operating conditions.

SUMMARY: BEST ESTIMATES OF ^{41}AR RELEASES DURING SRS OPERATIONS, 1954–1988

Figure 4.3-1 earlier in this chapter presents airborne release rate data for the activation products in general. Of these, the primary radionuclide of interest in terms of airborne release and potential offsite dose is ^{41}Ar. SRS reactor operations produced and released relatively large quantities of ^{41}Ar, and its production rate in the air blanket surrounding a reactor should have been roughly proportional to the reactor power level. Cummins (1991) provides an SRS-developed estimate of ^{41}Ar releases. The data were presented in Table 4.3-2, and are summarized in Table 4.3-12 and Figure 4.3-4, below.

Table 4.3-12. Best Estimate of ^{41}Ar Releases to Air, All Reactors

Year	^{41}Ar to air (Ci)	Year	^{41}Ar to air (Ci)	Year	^{41}Ar to air (Ci)
55	5.0×10^{4} [a]	66	2.8×10^{5} [a]	78	5.3×10^{4}
55	1.4×10^{5} [a]	67	3.2×10^{5} [a]	79	5.3×10^{4}
56	2.5×10^{5} [a]	68	2.2×10^{5} [a]	80	7.0×10^{4}
57	2.9×10^{5} [a]	69	1.4×10^{5} [a]	81	6.2×10^{4}
58	3.4×10^{5} [a]	70	1.1×10^{5} [a]	82	6.0×10^{4}
59	4.4×10^{5} [a]	71	1.4×10^{5}	83	4.1×10^{4}
60	4.0×10^{5} [a]	72	1.7×10^{5}	84	3.6×10^{4}
61	4.2×10^{5} [a]	73	1.8×10^{5}	85	5.2×10^{4}
62	4.3×10^{5} [a]	74	1.1×10^{5}	86	8.3×10^{4}
63	4.5×10^{5} [a]	75	6.5×10^{4}	87	8.8×10^{4}
64	3.7×10^{5} [a]	76	8.3×10^{4}	88	3.0×10^{4}
65	2.7×10^{5} [a]	77	6.5×10^{4}	89	

[a] Values from Cummins (1991). Values before 1971 were estimated; data after 1970 were based on measurements.

While these values represent the best available estimates for ^{41}Ar releases from the SRS reactors, we note that the values presented for the later years (1974–1988) are quite low when compared to reactor power levels and overall average ^{41}Ar production levels. We also observe that the ^{41}Ar release values presented for certain of the early years (1955–1967) are quite high when compared to reactor power levels for the same period. The reason for these apparent discrepancies, demonstrated in the following graphs, is not clear.

The following graph (Figure 4.3-4) plots the data from Cummins (1991) data on a linear scale, for comparison to reactor power levels as seen in Figure 4.3-5.

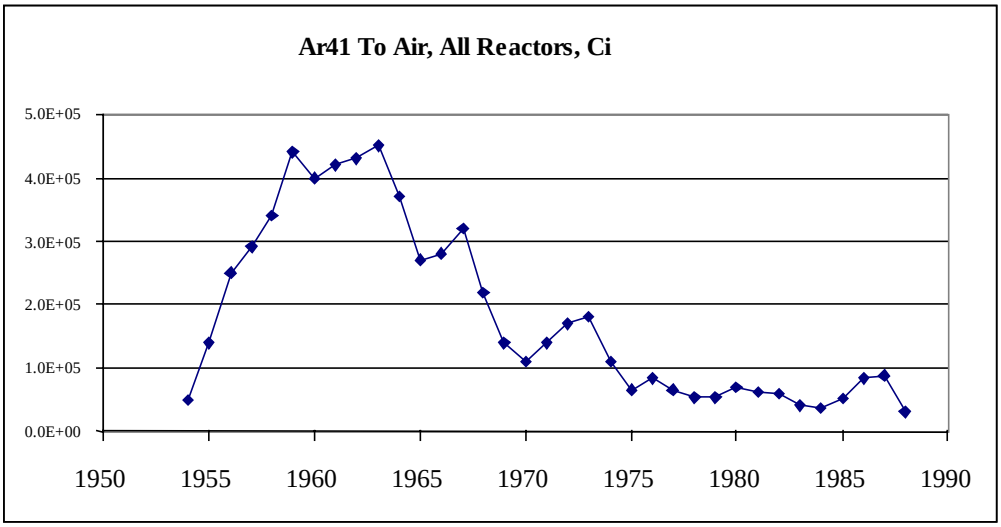

Figure 4.3-4. SRS Estimate of ^{41}Ar releases, 1954–1989 (from Cummins et al. 1991).

Chapter 2 of this report presents data extracted by *RAC* concerning SRS reactor power levels for the period 1955–1988. Those data are summarized in Figure 4.3-5.

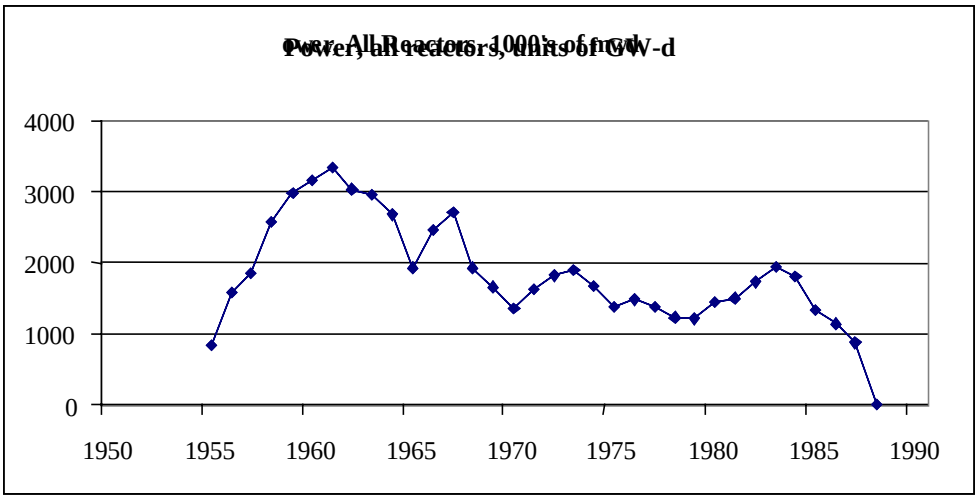

Figure 4.3-5. Combined SRS reactor power output, 1955–1988.

Figure 4.3-6 allows examination of annual reactor power level data by individual reactor.

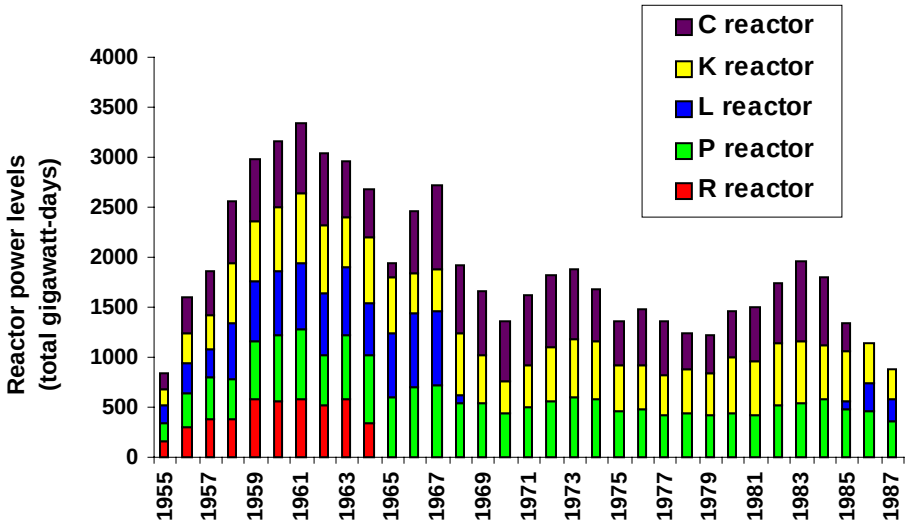

Figure 4.3-6. SRS reactor power output, 1955–1988. (Note: for black and white copies: the vertical order within the data bars is the same as the order of reactors in the key.) From Chapter 2 of this report.

Figure 4.3-7 presents a comparison of the ^{41}Ar and reactor power data sets, with the ^{41}Ar values having been divided by 100 to allow direct comparison on the same scale. Here, the discrepancy between the two sets of information becomes apparent, with relatively high estimated ^{41}Ar release values apparent in the early years, and relatively low values apparent during the later years.

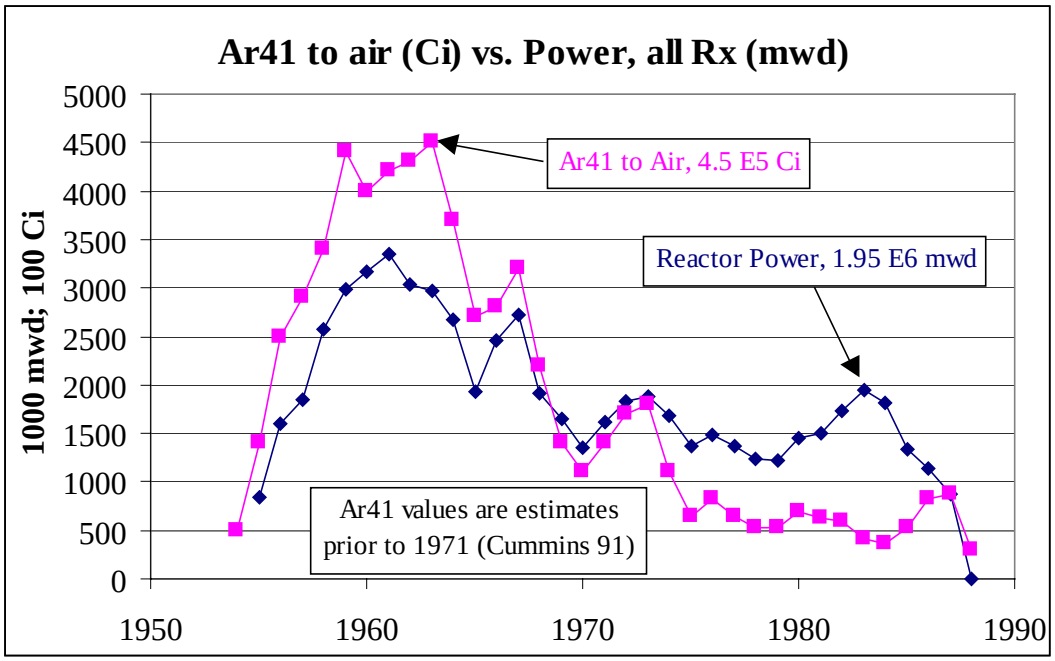

Figure 4.3-7. Comparison of SRS ^{41}Ar release estimates versus reactor power levels.

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CHAPTER 4.4

RELEASES OF ALPHA-EMITTING RADIONUCLIDES TO THE ATMOSPHERE

ABSTRACT

This chapter discusses releases of alpha-emitting radionuclides, primarily plutonium and uranium to the atmosphere from Savannah River Site (SRS) facilities. The primary objective of our efforts is to develop release estimates, or a source term, for alpha-emitting radionuclides that can be used to estimate potential exposure and risks to surrounding populations. We have attempted to validate reported releases by comparing data from as many separate SRS sources as possible. In general, reported release values are consistent among the various reports that we have reviewed. Additionally, we have estimated the uncertainty associated with the reported releases, based on sample collection and counting procedures, and estimated possible sample losses during transmission through sampling probes and lines. Annual atmospheric release values for plutonium and uranium and associated uncertainty have been compiled for both F-Area and H-Area stacks.

The data indicate that the majority of plutonium emissions from both F-Area and H-Area stacks occurred during 1955 and 1969. The data also indicate that the majority of uranium emissions from H-Area stacks occurred during 1955, 1968, and 1969, and that the majority of uranium emissions from F-Area stacks occurred during 1955, 1956, and throughout the 1960s. Evaluation of potential impacts resulting from alpha-emitting radionuclide releases should be focused on these years because the relative magnitude of total emissions during other years (including all years since 1970) appears to be quite small.

POTENTIAL RELEASE SOURCES

To assess atmospheric releases of alpha-emitting radionuclides and their potential impacts, it is important to understand the general processes that have historically been carried out at the SRS. This enables release data compilation and analysis to focus on those facilities with the greatest potential for releases as well as those time periods during which the largest releases occurred.

Carlton et al. (1993) and Evans et al. (1992) provide detailed description of the processes that may have led to historical plutonium and uranium emissions. The information in this section was taken primarily from these two references. Most SRS operations focused on the production of plutonium and tritium for national defense purposes. The production of plutonium initially involves the fabrication of fuel and target assemblies (in the M-Area facilities) for use in the five onsite production reactors (R-Reactor, C-Reactor, K-Reactor, P-Reactor, and L-Reactor). Plutonium and other radionuclides are formed in the fuel and target elements, which are reprocessed in one of two chemical separation facilities located in the F-Area and H-Area canyon buildings (221-F and 221-H). Various experiments and analyses with the potential to release alpha-emitting radionuclides have also been conducted in the Administrative Area (A-Area).

Administrative Area

Organizations that supply direct support for SRS operations, including the Department of Energy (DOE) office for the Site, the Savannah River Ecology Laboratory (SREL), administrative offices, the Savannah River Laboratory (SRL), which is now called the Savannah River Technology Center (SRTC), are all located in the Administrative Area, or A-Area (Cummins et al. 1990). The SRTC has historically been involved with analyses of fuel and target material and has dealt with various alpha-emitting radionuclides, including ^{238}Pu and isotopes of americium, curium, californium, and various other transuranics with high specific alpha activities (Evans et al. 1992). Releases from the SRTC are routed through a sand filter, and some of the ventilation streams pass through some combination of HEPA filters, charcoal filters, or caustic scrubbers before reaching the sand filter.

Fuel Fabrication

Most production of fuel and target assemblies took place onsite at the M-Area facilities and involved cladding of uranium with aluminum. Additionally, enriched uranium fuel rods were manufactured by first alloying the uranium with aluminum and were then machined and extruded. These processes have been associated with generating uranium metal filings and dust as well as dissolved and suspended uranium in solution.

Potential releases of uranium from M-Area facilities have included both atmospheric stack releases and liquid effluent releases to Tim's Branch and M-Area seepage basins. The majority of uranium releases from M-Area, however, occurred through liquid effluent releases to Tim's Branch. Chapter 5 addresses these releases in detail.

Reactor Operations

Fuel and target assemblies, heavy water moderator, and control rods comprise the major components of the reactor core. Following fabrication, fuel and target assemblies are transferred to the production reactors. Irradiated target and spent fuel elements are then removed from the reactor core and placed in the vertical tube storage basin of the reactor building. After a delay to reduce radioactivity levels, the elements are relocated to the disassembly basins and prepared for transfer to the chemical separation facilities. Under ideal conditions, all radionuclides would remain contained within the aluminum cladding of the fuel and target elements. However, during normal reactor operations, fuel and target elements can develop defects or ruptures in the aluminum cladding. Uranium, plutonium, other neutron capture products, and fission products have been released to the reactor moderator and basin water during such element failures. The reactor was shut down following a fuel or target element failure, and the failed elements were transferred to a harp container for storage in the reactor basin.

Potential releases of plutonium, uranium, and other alpha-emitting radionuclides were primarily through the discharge of liquid effluent. Atmospheric releases have likely been minimal because all fuel and target handling activities were accomplished under water. The primary mechanism for atmospheric release of radionuclides from the reactor building was resuspension of particulate material from dried liquids. Furthermore, atmospheric releases were minimized through the use of a confinement system that did not allow for air exchange with the gases

present inside the reactor tank, vertical tube storage basin, or disassembly basins. All ventilated air was passed through a demister filter bank, a high-efficiency particulate air (HEPA) filter bank, and a carbon filter bank.

Fuel Processing

From the reactors, irradiated target and spent fuel elements were transferred to the chemical separation facilities located in the F-Area and H-Area canyon buildings for reprocessing. Exhaust stacks from these facilities have historically had the highest measured atmospheric emissions of alpha-emitting radionuclides, primarily plutonium (^{238}Pu and ^{239,240}Pu) and uranium. Therefore, the majority of release data analyses focuses on these facilities.

Beginning in November 1954, target elements were treated by the Purex process to recover ^{239}Pu, ^{238}U, and ^{237}Np from irradiated ^{238}U in the F-Area canyon buildings. As part of the Purex process, target slugs were stripped of their aluminum cladding and dissolved in nitric acid. The ^{239}Pu, ^{238}U, and ^{237}Np were segregated into separate aqueous streams by solvent extraction and several stages of contactors and centrifugal separators. The plutonium, uranium, and neptunium were present in the process streams as nitrate salts in strong nitric acid solutions. During the precipitation and reduction processes carried out in the B-Line facilities, some particles containing metal oxides were formed. The primary routes for atmospheric releases were through the process ventilation vents and the aqueous stripping streams. All process vessels are vented through HEPA filters and large sand filters before being released through the main process 61-m stack. Air from other areas is directly vented to the sand filter only before discharge or sent through HEPA filters only before discharge. Offgas from the A-Line passes through bag filters before being discharged to the atmosphere via the 291-F stack (Evans et al. 1992 and Sanders 1977). In the early years of operation, exhaust gases from the B-Line facilities were vented only through HEPA filters before release through short stacks on the roof of the canyon buildings. However, the ventilation systems were modified to provide additional filtration following a fire at the Rocky Flats Plant (Carlton et al. 1993).

The Purex process was also used in the H-Area canyon building from July 1955 through 1959, after which the facility was modified to recover ^{235}U and ^{237}Np from enriched fuel elements. The H Modified (HM) process is different from the Purex process because it used mercuric nitrate as a catalyst to control dissolution of the uranium-aluminum alloy used to fabricate enriched fuel elements. The Frames process, which employs ion exchange instead of solvent extraction to separate and purify ^{238}Pu and ^{237}Np from waste products, was also used in the H-Area to recover ^{238}Pu from ^{237}Np targets (Carlton et al. 1993). Additionally, special campaigns for the production of radionuclides, such as ^{252}Cf and ^{244}Cm, have occasionally occurred. The atmospheric ventilation system in the H-Area facilities is essentially the same as that described for the F-Area facilities.

Other Sources of Alpha-Emitting Radionuclides

Much of the electric power distributed at the SRS was generated onsite at the power plants. These plants also provided steam for the Site. Historically, both electricity and steam at the SRS have been generated by burning coal, which in turn has resulted in the atmospheric release of signficant quantities of fly ash. Coal, and consequently the ash created by burning it, contains a

number of radionuclides, which originate from trace amounts of naturally occurring ^{238}U and ^{232}Th. Additionally, a number of other radionuclides result from decay of ^{238}U and ^{232}U, including a number of radioactive isotopes of radium, uranium, thorium, bismuth, polonium, and lead.

To make comparisons of releases associated with fly ash to releases associated with other Site operations, we can estimate the amounts of certain radionuclides that may have been released as a result of coal burning, based on the measured concentrations of radionuclides in fly ash and the amount of fly ash released (see Chapter 17 for additional details regarding coal burning and estimated airborne releases of fly ash). Eisenbud and Petrow (1964) reported concentrations of several radionuclides associated with coal burning, including ^{226}Ra, ^{228}Ra, ^{238}U, ^{228}Th, and ^{232}Th. Table 4.4-1 lists the concentrations of these radionuclides in ash reported by Eisenbud and Petrow (1964), along with estimated potential releases, based on the maximum estimated annual release of fly ash from the SRS (see Chapter 17).

Table 4.4-1. Radionuclide Concentrations in Fly Ash and Associated Maximum Airborne Emission Potential

Radionuclide	Concentration in Fly Ash[a] (pCi g^{-1})	Estimated Fly Ash Release[b] (g)	Calculated Radionuclide Release (Ci)
^{226}Ra	3.8	2.9×10^{10}	0.11
^{228}Ra	2.4	2.9×10^{10}	0.07
^{228}Th	2.6	2.9×10^{10}	0.08
^{232}Th	2.2	2.9×10^{10}	0.06
^{238}U	4.4	2.9×10^{10}	0.13

[a] Source: Eisenbud and Petrow (1964)
[b] Maximum annual fly ash release estimate (for 1988) from Chapter 17

These potential maximum release amounts suggest that coal burning may have released sufficient quantities of radionuclides to become important by comparison to F-Area and H-Area releases of plutonium and uranium, after approximately 1960, excluding 1969. If annual releases of plutonium and uranium on the order of 0.1 to 1 Ci are determined to be important contributors to potential risk to members of the public during future phases of this dose reconstruction, it may be necessary to further evaluate the potential importance of radionuclide releases associated with the burning of coal.

RELEASES AND RELEASE MONITORING

Administrative Area

Releases of alpha-emitting radionuclides have occurred at A-Area, but the releases are relatively small by comparison to the larger releases that of occurred at the Chemical Separation Area Facilities. The most notable releases occurred in the mid to late 1960s. Cummins et al. (1991) report 0.032 and 0.021 Ci releases of ^{244}Cm in 1964 and 1969, respectively. These releases are comparable to and generally less than total releases of alpha-emitting radionuclides associated

with fly ash generated by burning coal and are significantly less than the highest releases associated with the Chemical Separation Area Facilities.

M-Area Facilities

Isotopes of uranium were the only alpha-emitting radionuclides that had the potential to be released in significant quantities from M-Area facilities. From the first days of operation through 1974, activity release alarms were in place on building exhausts, but chronic atmospheric releases through stacks have only been measured since 1975. From 1975 through 1989, the highest recorded annual atmospheric release of uranium from M-Area facilities was 0.0001 Ci, or about 143 g (Evans et al. 1992), which is more than an order of magnitude less than the lowest recorded annual uranium release at the chemical separation facilities. Total reactor power levels during this time period (1975 through 1987) were approximately one-half the total power levels during peak production years (late 1950s and early 1960s). Fabrication of a larger number of fuel elements would have accompanied the higher power levels, so it is likely that atmospheric releases of uranium during peak production years would have been greater than releases measured since 1975. It is unlikely, however, that M-Area releases would have ever been great enough to appreciably add to chemical separation area releases.

Reactor Buildings

Because of the low potential for atmospheric radionuclide releases of any kind from the reactors, only gross beta and gamma activity was measured on air filters. Air filters were submitted for alpha analysis only if activity above background was detected. After the early 1970s, all filters were submitted for gross alpha analysis, but specific radiochemical analyses for plutonium or uranium were not made. Maximum annual alpha releases to the atmosphere, when they were quantified, were generally 2 or 3 orders of magnitude less than the lowest recorded annual plutonium and uranium releases from the chemical separation facilities. As with A-Area and M-Area releases, it is unlikely that reactor releases to the atmosphere would ever have been great enough to appreciably add to the chemical separation area releases.

Chemical Separation Area Facilities

Atmospheric releases of alpha-emitting radionuclides (including plutonium and uranium) from the separation facilities have been quantified since shortly after startup in November 1954. The primary release points at the F-Area and H-Area consisted of two tall ~61-m (200-ft) stacks, designated 291-F and 291-H, respectively. The first sampling period for plutonium and uranium in atmospheric effluents was December 14, 1954, to January 7, 1955 (Carlton et al. 1993). Continuous sampling was accomplished by passing a portion of stack effluent through a filter designed to trap particles and associated radionuclides. The filter was changed weekly and submitted for specific (including plutonium and uranium) radiochemical analysis. Carlton et al. (1993) indicated that plutonium emissions data for both F-Area and H-Area stacks were based on specific radiochemical analyses. Evans et al. (1992) indicated that uranium emissions data from F-Area were based on specific radiochemical analyses, and that emissions data from H-Area were based on gross alpha analyses. However, the original monthly reports through 1971 provide gross

alpha release estimates only, and there is no indication of routine specific isotopic analyses. It is possible that specific isotopic analyses were carried out only for quarterly or semi-annual filter composites and were therefore not reported on a monthly basis. However, the routine semi-annual and annual monitoring reports from 1955 through 1971 also reported only gross alpha release estimates.

The vast majority of alpha-emitting radionuclides released from the separation facilities likely consisted of plutonium and uranium. However, in 1967, approximately 28 mCi of ^{244}Cm were released between November 11 and 25, accounting for the majority of H-Area atmospheric releases during that month (Du Pont 1967k).

F-Area and H-Area Stacks

Total alpha emissions were reported monthly in 1955 and semiannually from 1956 through 1963 in the Health Physics Regional Monitoring report series and were reported semiannually from 1964 through 1966 and annually from 1967 through 1971 in the Environmental Monitoring at the Savannah River Plant report series. Annual plutonium (both ^{238}Pu and ^{239,240}Pu) and uranium emissions data were reported by Cummins et al. (1991). Carlton et al. (1993) and Evans et al. (1992) also provided emissions data for plutonium and uranium, respectively. However, a few of their release estimates were inconsistent with the data provided by Cummins et al. (1991), and the tabulated plutonium data reported by Carlton et al. (1993) appeared to represent only ^{239,240}Pu. All emissions data depicted in the following figures have been taken from the Health Physics Regional Monitoring report series (total alpha), the Environmental Monitoring at the Savannah River Plant report series (total alpha), and Cummins et al. (1991) (specific radionuclides).

Figures 4.4-1 and 4.4-2 (note the logarithmic scale) show the total alpha and plutonium plus uranium emissions data for 1955 through 1971 for F-Area and H-Area stacks, respectively. It appears that plutonium and uranium have comprised the majority of measured total alpha emissions from F-Area stacks during most years, particularly those years during which the most significant releases occurred (1955, 1956, and 1960). It also appears that plutonium and uranium have comprised the majority of measured total alpha emissions from H-Area stacks for the years during which the most significant releases occurred (1955 and 1969). For two years (1960 and 1967), however, total alpha releases from H-Area stacks were greater than the sum of plutonium and uranium releases. It is likely that other radionuclides were emitted during those years in sufficient quantities to account for a significant portion of the total alpha activity. For instance, as mentioned previously in this section, the majority of H-Area atmospheric releases during November 1967 consisted of ^{244}Cm, and other alpha-emitting radionuclides, such as ^{252}Cf, have occasionally been processed in the chemical separation areas.

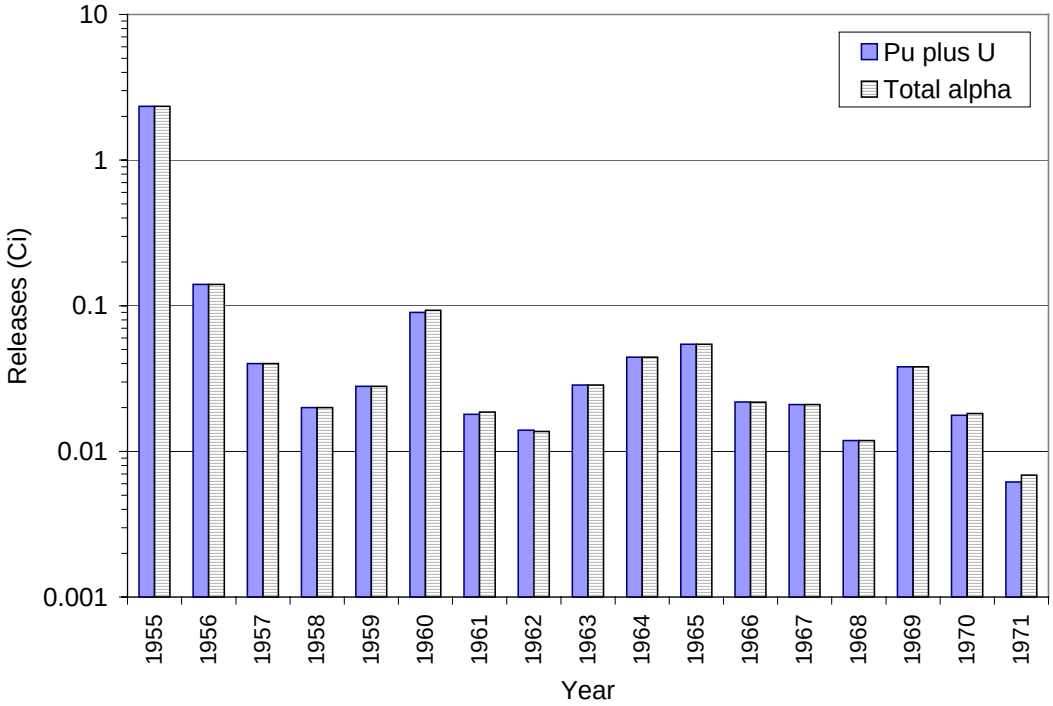

Figure 4.4-1. Total alpha and plutonium plus uranium emissions from F-Area stacks. Link to tabulated figure data.

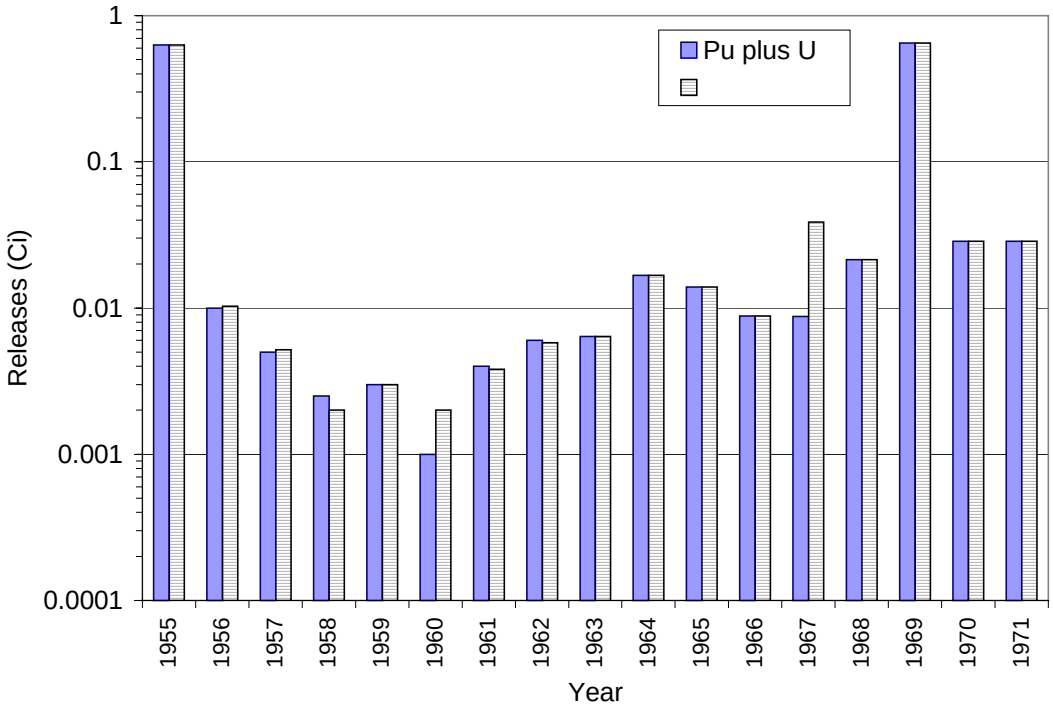

Figure 4.4-2. Total alpha and plutonium plus uranium emissions from H-Area stacks. Link to tabulated figure data.

Figure 4.4-3 shows the percent of total alpha releases by year from 1955 through 1971 for F-Area and H-Area stacks. It is clear that the majority (about 83%) of releases occurred during 1955 and 1969. More than 70% of the releases (predominantly ^{239,240}Pu) in 1955 occurred between August and December during a number of B-Line exhaust filter failures (Horton and Mealing 1956a). Backup filters were installed in both areas during December, reducing measured emissions significantly. In April 1969, breakage of supporting tile in the H-Area exhaust stack sand filter caused local depletion of sand and resulted in a significant alpha release, primarily ^{238}Pu (Ashley 1970a).

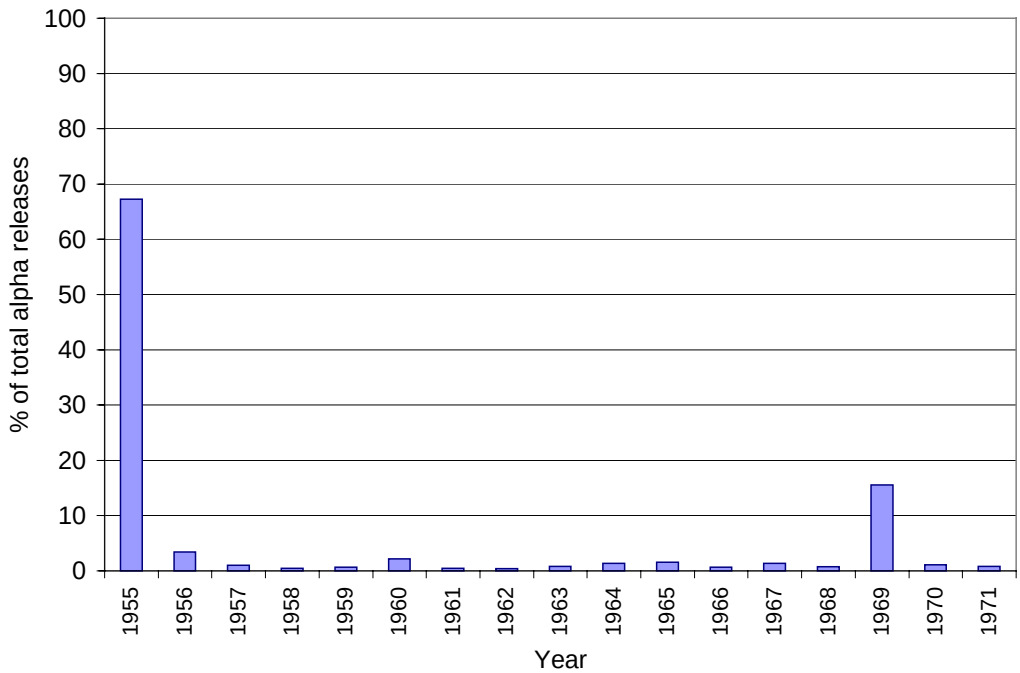

Figure 4.4-3. Percent of total alpha releases by year from F-Area and H-Area stacks from 1955 through 1971. Link to tabulated figure data.

Figures 4.4-4 (note the logarithmic scale) and 4.4-5 show the annual plutonium emissions and percent of total emissions by year, respectively, for F-Area and H-Area stacks (Cummins et al. 1991). Greater than 90% of the total plutonium emissions from separation area stacks between 1955 and 1989 occurred in 1955 and 1969 (specific incidents discussed previously).

Based on data provided by Cummins et al. (1991), distinctions were not made between ^{238}Pu and ^{239,240}Pu, and all plutonium emissions data were reported as ^{239,240}Pu before 1967. Beginning in 1967, ^{238}Pu emissions data were provided. Figure 4.4-6 shows the percent ^{239,240}Pu of total plutonium emissions (^{238}Pu plus ^{239,240}Pu) for F-Area and H-Area stacks from 1967 through 1989. It is clear that ^{238}Pu comprised a significant portion of total plutonium emissions during this time period, particularly from H-Area facilities, which were involved with ^{238}Pu recovery through the Frames process. In fact, reported plutonium emissions from H-Area stacks in 1977 and 1978 consisted entirely of ^{238}Pu. Totals of 2.46 and 1.26 Ci of plutonium were released from F-Area and H-Area stacks, respectively, between 1955 and 1989.

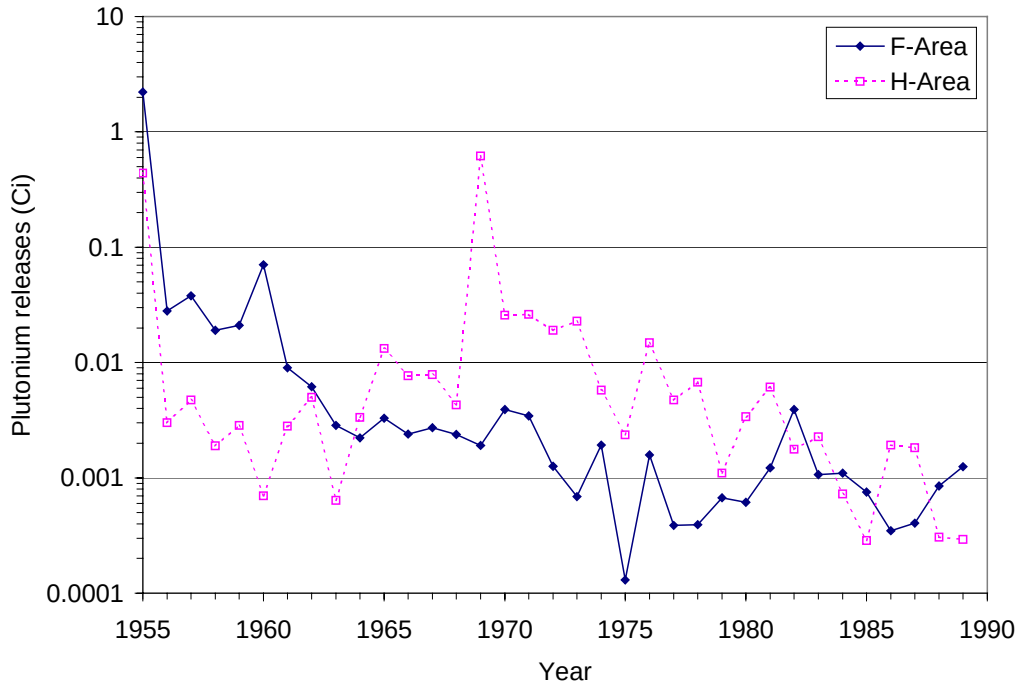

Figure 4.4-4. Annual plutonium releases from F-Area and H-Area stacks from 1955 through 1989. Link to tabulated figure data.

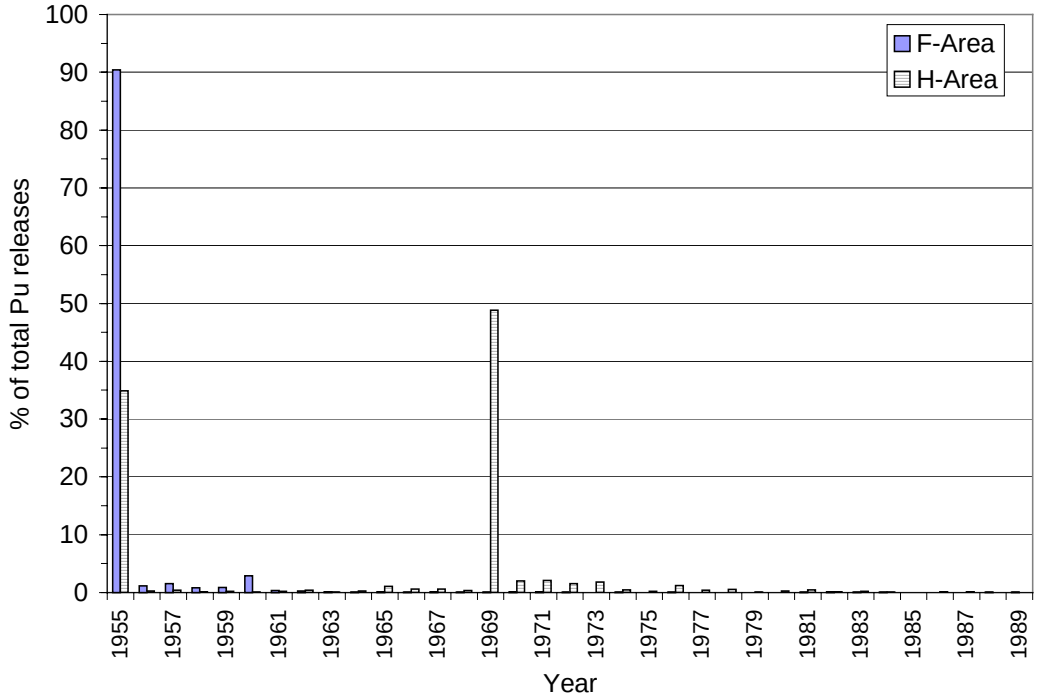

Figure 4.4-5. Percent of total plutonium released by year from F-Area and H-Area stacks from 1955 through 1989. Link to tabulated figure data.

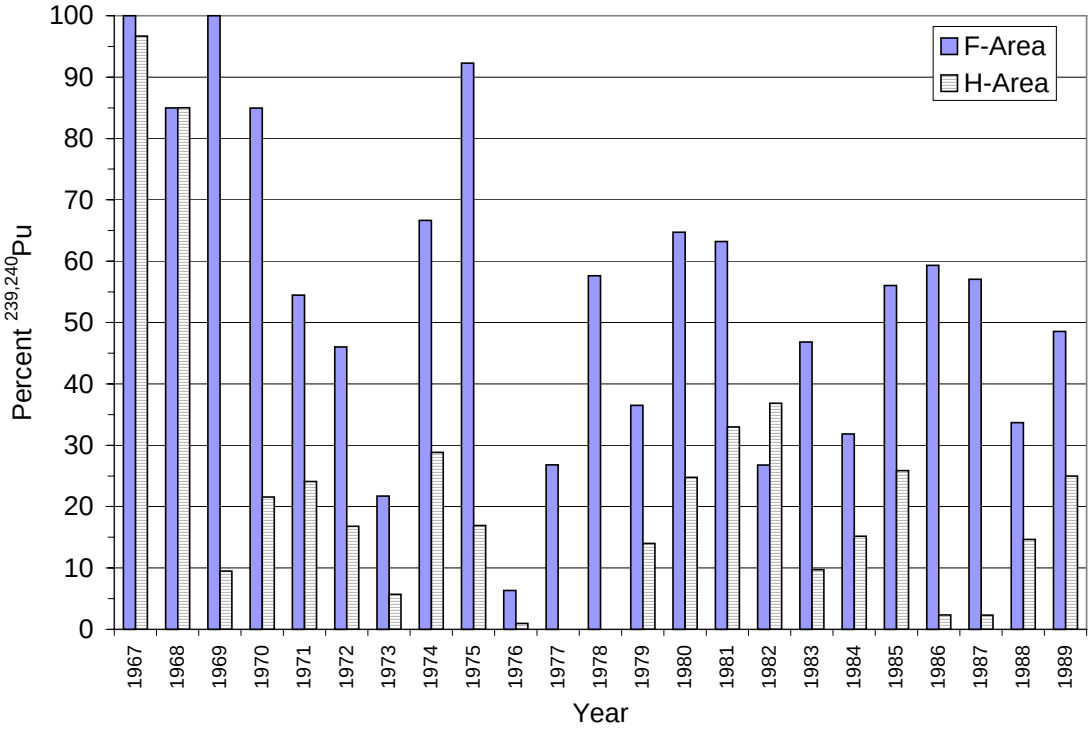

Figure 4.4-6. Percent of ^{239,240}Pu out of total plutonium emissions (^{238}Pu plus ^{239,240}Pu) for F-Area and H-Area stacks from 1967 through 1989. Link to tabulated figure data.

Figures 4.4-7 (note the logarithmic scale) and 4.4-8 show the annual uranium emissions and percent of total emissions by year, respectively, for F-Area and H-Area stacks (Cummins et al. 1991). The uranium emissions provided by Cummins et al. (1991) reportedly consisted of natural uranium, and include ^{238}U, ^{235}U, and ^{234}U. It is not clear why the emissions are indicated to consist only of natural uranium since both enriched and depleted uranium have been processed and recovered at SRS. If uranium emissions are determined to be important contributors to dose to members of the public, it may be necessary to more closely examine the specific isotopic composition of uranium emissions.

The majority of uranium emissions occurred during the first 2 years of operation (1955 and 1956) and during several years in the 1960s. Evans et al. (1992) indicated that greater emphasis was placed on reducing emissions since 1970; this is supported by both the plutonium and uranium emissions data. Totals of 0.57 and 0.29 Ci of uranium were released from F-Area and H-Area stacks, respectively, between 1955 and 1989.

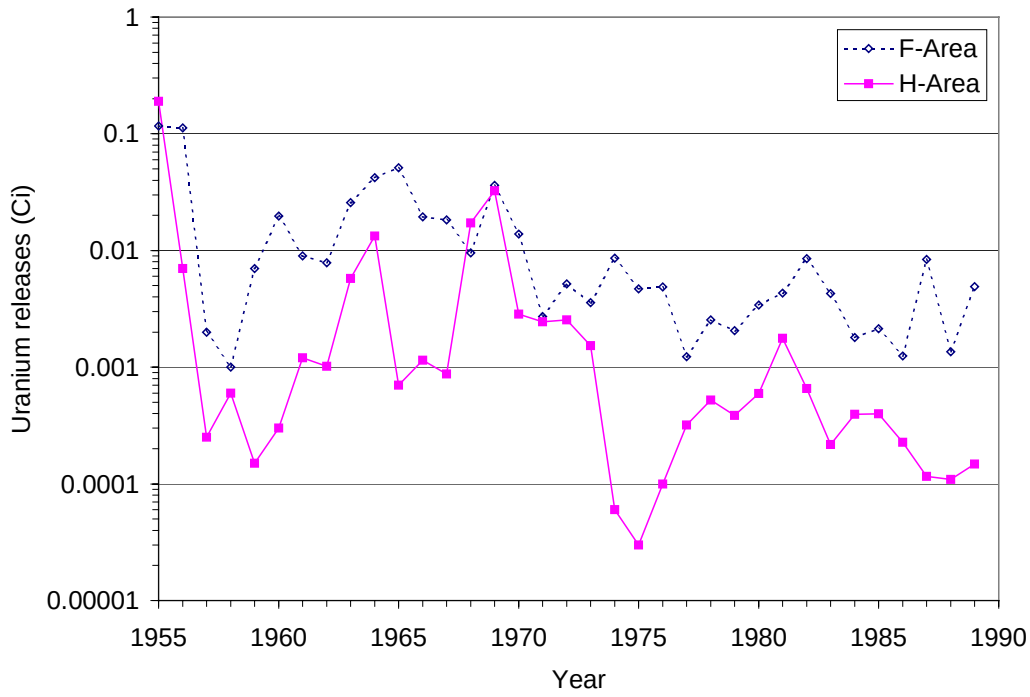

Figure 4.4-7. Annual uranium releases from F-Area and H-Area stacks from 1955 through 1989. Link to tabulated figure data.

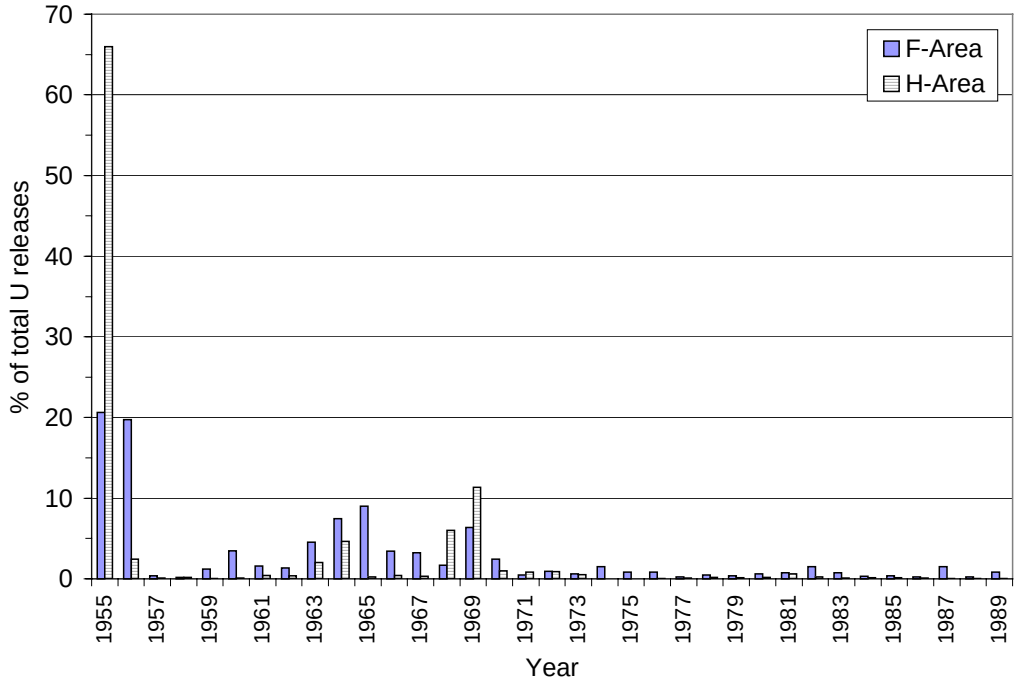

Figure 4.4-8. Percent of total uranium released by year from F-Area and H-Area stacks from 1955 through 1989. Link to tabulated figure data.

ACCOUNTING FOR SAMPLE LINE LOSSES

It is not apparent that Site-reported release quantities were modified to account for potential line loss or particle plateout along the inner walls of the sampling line, which would result in an underestimate of the amounts released. The ratio of the contaminant concentration leaving the sampling line to the concentration entering the line is often referred to as a transmission factor. We used a model developed by Texas A&M University to estimate transmission factors for alpha bearing particles sampled from the F-Area and H-Area effluent stacks (ATL 1993). These transmission factors can be used to modify reported releases to account for potential sample line losses.

A number of factors can influence the degree to which particles are transmitted through a sampling line. In general, larger and denser particles are most prone to deposition and loss along the length of the sampling line, particularly along horizontal sample line sections and by impaction in bends. Sample flow rate, effluent free stream velocity, and probe type also affect transmission through the sampling lines.

The effluent sampling lines for the reprocessing facilities at the SRS were quite long. Samples were extracted at the ~59-m (195-ft) level of the stack. The line extended to near ground level and then an additional 30.5 m (100 ft) horizontally to the point of sample collection. The total line length, including the in-stack portion, is estimated to be about 93 m (305 ft). This estimate is based on the following description: the sampling system consisted of a probe, a 0.15-m (6-in.) vertical tube, a 4.57-m (15-ft) horizontal tube, a 57.9-m (190-ft) vertical tube, and a 30.5-m (100-ft) horizontal tube. Each section of tubing was assumed to be connected by a 90-degree bend. It is possible that the actual sampling system was somewhat more complex and consisted of a greater number of bends and individual sections of tubing as described by Zippler (1979). However, the greatest amount of loss generally occurs in the probe and in the horizontal sections of tubing, the distances of which are known, so assuming an overly simple design does not significantly affect the calculation.

The flow rate through the sampling lines has varied somewhat and is assumed to be 2 cubic feet per minute (cfm) from December 1954 through October 1956, 3 cfm from November 1956 through September 1961, 2 cfm from October 1961 through September 1965, and 5 cfm after September 1965. See Chapter 4.2, Table 4.2-3 for more details regarding the sample flow rate during different time periods.

The free stream velocity refers to the rate or speed at which the effluent is moving through the stack and, therefore, the rate at which the sample enters the probe. Zippler (1979) measured the free stream velocity of the effluent in the 291-H stack and reported an average value of 2574 feet per minute (fpm) or approximately 13 m s^{-1}. This is consistent with the average flow for the 291 F stack (226,000 cfm) reported in Du Pont (1968a). Assuming a 10-ft stack diameter (Zippler 1979), this flow rate results in a free stream velocity of approximately 15 m s^{-1}. A flow rate of 13 m s^{-1} was assumed for the transmission factor calculations.

Several researchers have attempted to characterize the particle size distribution for the material released from the separations area stacks. Croley (1973) reported an average particle size of 1.5 microns (μm) (ranging from 0.5 to 4.5 μm) for samples collected during July 1973 from the 291 F and H stacks at the 195-ft level. Average particle sizes were determined by the light microscope technique, which does not account for the fact that alpha-emitting radionuclides are generally not uniformly distributed among different particle sizes.

Activity-based particle sizes are more appropriate for determining transmission factors. Such distributions are estimated by collecting effluent samples with a cascade impactor using several fractionation stages. The activity that is collected at each stage is determined, and an activity-based particle size distribution can be estimated.

Sanders (1978) collected samples from the 50-ft level of the 291-F stack in October 1975 and reported a geometric mean particle size of 5.43 µm with a geometric standard deviation of 2.69. Sanders (1977) described the procedures that were used to determine the particle sizes reported by Sanders (1978). Particles were selected for analysis based on the number of observed fission-fragment tracks, with those particles surrounded by many tracks being selected in favor of those surrounded by few tracks. This method of particle selection would appear to characterize the size distribution of particles carrying the majority of the plutonium.

Particle sizes as low as 0.4 µm were reported, but submicron particles accounted for only 7% of the total number of particles. The relatively large size of these particles was ascribed to coagulation of submicron particles by thermal and turbulent mechanisms to form larger particle agglomerates. The elemental composition of these larger particles showed them to be comprised primarily of crustal elements or dust in combination with very small amounts of plutonium. Sanders (1978) hypothesized that the larger dust particles did not pass through the HEPA filters, but instead they entered the exhaust system through leaks in the ducts, which would be undetected as long as the exhaust system remained under negative pressure compared to the atmosphere.

Carlson et al. (1983) reported activity-based particles sizes, or activity median aerodynamic diameters, for samples collected from the 50-ft level of the 291-F stack in 1982. Table 4.4-2 shows the average percentage of total activity measured on particles of several size intervals during four separate sampling tests. About two-thirds of the total activity is associated with submicron particles. This size distribution is consistent with distributions reported by Croley (1978) and by Gay and Watts (1981) for environmental samples collected from the vicinity of the 291-H stack in 1975, 1976, and 1977. On the other hand, this size distribution is not consistent with the data reported by Sanders (1978). The fact that the particle sizes determined by Sanders (1978) were not activity-based may help account for this inconsistency. It is also possible that leaks in the exhaust system had been repaired since the study by Sanders (1978), and less dust was entering the system in 1982, when Carlson et al. (1983) collected their samples.

Table 4.4-2. Percentage of Total Plutonium Activity Measured on Different Particle Sizes in Samples Collected from the 291 F Stack in 1982[a]

Particle diameter (microns)					
>8.2	8.2–3.5	3.5–2.1	2.1–1.0	1.0–0.5	<0.5
19.1%	6.6%	2.9%	4.2%	13.6%	53.4%

[a] Data from Carlson et al. (1983)

Selecting an appropriate particle size distribution is an important factor in establishing a credible transmission factor estimate. Based on the data *RAC* has been able to locate, the most appropriate particle size distribution is somewhat uncertain. The particle size distribution reported by Sanders (1978) is significantly larger than the distributions reported by Croley (1973) and Carlson et al. (1983). For this reason, we have calculated several transmission factors based on various particle size distributions and sample flow rates (Table 4.4-3). During the time period of

highest release amounts (i.e., before 1970), samples were collected using anisokinetic probes. For comparison, transmission factors for isokinetic probes, which collect samples with the same velocity in the sample line as in the effluent stream being sampled, are also shown. A particle density of 2.0 g cm^{-3} was assumed for the distributions reported by Sanders (1978) and Croley (1973), which is characteristic of the crustal elements that typically comprise dirt and dust. A particle density of 1.0 g cm^{-3} was assumed for the distribution reported by Carlson et al. (1983) because a unit density is inherent for an activity median aerodynamic diameter.

Table 4-4.3. Transmission Factors Calculated for Several Particle Size Distributions

Particle diameter	Sample flow rate (cfm)	Transmission factors (%) Anisokinetic	Transmission factors (%) Isokinetic
Sanders (1978)	2	23	26
GM[a] = 5.43 µm	3	26	30
GSD[b] = 2.69	5	29	37
Croley (1973)	2	71	76
GM = 1.5 µm	3	74	80
GSD = 1.5	5	78	86
Carlson et al. (1983)	2	69	71
(see Table 4.4-2 for	3	71	74
particle sizes)	5	73	77

[a] GM = geometric mean.
[b] GSD = geometric standard deviation.

Transmission factors calculated based on the particle sizes reported by Sanders (1978) are likely conservative estimates because the large particle size distribution results in a greater amount of deposition and particle loss as the effluent passes through the sampling line. Transmission factors calculated based on the particle sizes reported by Carlson et al. (1983) may be more accurate because the distributions were based on measured activity. Additionally, the particle sizes reported by Carlson et al. (1983) are generally consistent with the particle sizes reported by Croley (1973) and Gay and Watts (1981). However, the particle sizes reported by Sanders (1978) may be appropriate for estimating line losses for uranium-bearing particles. Voillequé et al. (1995) reported uranium to be associated with larger-sized particles, similar to the distribution reported by Sanders (1978). Additionally, assuming a larger particle size may be appropriate for estimating releases associated with filter breaks (such as those that occurred in 1955 and 1969), events which would have led to releases consisting of larger particles. Anonymous (date unknown) estimated a transmission factor of 65% using the PLATEOUT computer model and an assumed particle size of 1.5 µm, which is generally consistent with the transmission factors we calculated using the model developed by Texas A&M University and similar assumed particle sizes. In addition, Zippler (1979) states “Due to the complexity of our sample lines theoretically less than half of the particles with median diameters greater than two microns would ever reach the filter paper”, which is consistent with the transmission factors we calculated based on the larger particle size distribution reported by Sanders (1978).

Effluent samples from the 291-F and H stacks were extracted with anisokinetic probes through at least the 1970s, which may result in collecting samples that do not represent the stack effluent particle composition. Isokinetic probes, on the other hand, collect samples with the same

velocity in the sample line as in the effluent stream being sampled, and they are considered more appropriate for obtaining representative samples. Based on results obtained by running the model developed by Texas A&M University, anisokinetic probes result in a slightly lower total transmission factor than isokinetic probes as a result of lower transmission through the probe itself. Transmission through the sampling lines and bends is slightly higher, however, when an anisokinetic probe is assumed in the model, perhaps because of decreased turbulence.

The SRS became concerned about the possible difficulties associated with anisokinetic sampling and attempted to draw conclusions about the differences between anisokinetic and isokinetic sampling. Zippler (1979) compared the two sampling probe types and concluded that no significant advantage could be shown for the isokinetic sampling over the routine anisokinetic sampling. However, low levels (i.e., near the detection limit) of activity were collected for many of the samples, and higher measured concentrations of ^{103,106}Ru collected with the isokinetic probe indicated greater sampling by nearly a factor of 2 compared to the anisokinetic probe. For the purpose of estimating transmission factors, it is assumed that the Site collected samples using anisokinetic probes.

UNCERTAINTIES IN THE REPORTED RELEASES

Uncertainty analysis is important because it provides an estimate of the possible range of releases that is consistent with our knowledge of the individual parameters in the release estimate calculation. The uncertainty analysis combines the best scientific knowledge available about each parameter in a single calculation to provide a distribution of results that is realistic given what we know about each parameter.

At the chemical separation areas (F-Area and H-Area), the majority of alpha releases were confined to the 61-m (195-ft) stacks. Aliquots of effluent were taken from these stacks daily, analyzed by health physics area personnel for gross alpha activity measurements, and sent to the Environmental Monitoring Group for final analysis and determination of release quantities. Release quantities were calculated as described in Du Pont (1968a), Equation (4.4-1).

$$Q = \frac{(cpm)(CF)(F_{stack})}{(F_{sample})(E)(2.22x10^{12}\,dpm/Ci)}$$

where

Q	=	stack release (Ci)
cpm	=	counts per minute
CF	=	conversion factor to convert cpm to dpm (disintegrations per minute)
F_{stack}	=	stack effluent flow rate ($ft^3\ min^{-1}$)
F_{sample}	=	sampler flow rate ($ft^3\ min^{-1}$)
E	=	collection efficiency of particulate air filter.

The total uncertainty in the release quantities (Q) that were determined is dependent upon the uncertainty associated with each of these factors. Inherent in the measured counts per minute are additional factors including counter error and percent recovery of extracted radionuclide. Total uncertainty resulting from counter error is likely small (5% or less), particularly during periods of the highest releases, because the filters would have accumulated significant activity

during a continuously sampled 24-hour period, and counter error decreases as a function of increasing count rate. This is assumed to be a negligible source of uncertainty. The uncertainty in the various extraction processes that were used, including ether and ethyl acetate and tri-butyl phosphate extractions, is likely around 20% based on average recovery values provided by Geiger (1954). A triangular distribution is assumed.

Uncertainty in the conversion factor that was used to convert counts per minute to disintegrations per minute is dependent upon the efficiency of the detector. These conversion factors were based on an alpha standard with a ¼-in. diameter active area (Johnson 1977). There may be some error associated with the different geometries of the standard and sample planchets. However, the 2-in. diameter zinc-sulfide crystal used for alpha counting was slightly larger than the 47-mm filters that were counted, and the samples were placed less than 5 mm from the active surface of the crystal (Johnson 1998). It is expected that this would contribute a negligible source of error.

Stack flow rates for the F-Area and H-Area 61-m stacks were determined from monthly Power Department flow measurements of the sand filter discharge plus fan capacities of contributing streams that do not go through the sand filter (Zeigler 1986). The uncertainty for reactor stack flow rates was reported as ±20% (Du Pont 1965a). The uncertainty in F-Area and H-Area stack flow rates is assumed to be the same, and a triangular distribution is assumed for this source of error.

Uncertainty in the sampler flow rate is dependent upon the type of particulate filter that was used. Inherent uncertainty in the calibration of the moto-air sampler flow rate was reported as ±10% (Du Pont 1968b). However, from 1955 through at least February 1957, Whatman #41 particulate filters were used for effluent sampling. Flow rates for these filters were found to vary as much as 50% when operating at a fixed pressure differential (Hoy 1957). In 1957, a change was made to MSA 1106-B filter papers, which are more uniform and do not show significant flow rate variations. Therefore, from 1955 through 1957, the uncertainty in the sampler flow rate is assumed to be ±60%. In subsequent years, uncertainty is assumed to be ±10%. A triangular distribution is assumed.

Uncertainty in the collection efficiency of the filter paper is dependent upon the type of filter paper that is used. An assumed collection efficiency of 80% was used for all calculations (Du Pont 1968c). However, MSA 1106-B filter paper efficiency is essentially 100%, and assuming 80% efficiency would have overestimated releases. On the other hand, when Whatman #41 filters were used, the efficiency of collection varied with the face velocity across the filter, and the efficiency was found to vary as much as 25% when operating at a fixed pressure differential (Hoy 1957). Therefore, from 1955 through 1957, the uncertainty in the collection efficiency is assumed to be ±25%. In subsequent years, uncertainty is assumed to be a negligible source of error because the efficiency of MSA 1106-B filters appears to have been underestimated by assuming an efficiency of 80%. It is not entirely clear what types of filters may have been used after 1957. Therefore, we are not making an adjustment to correct for this potential underestimation. A triangular distribution is assumed for collection efficiency uncertainty between 1955 and 1957.

We used the Crystal Ball® uncertainty analysis software package Version 4.0c (Decisioneering 1996). The Crystal Ball package is used within Microsoft Excel and allows the user to define the distribution of possible values for an input parameter. The possible range of release estimates is then calculated using a Monte Carlo analysis, which involves multiple trial calculations, using randomly selected values from the possible range of parameter values. In a

single trial, each of the parameter distributions is sampled, and the selected values are used to compute an estimate of the release. This procedure is repeated many times in a Monte Carlo analysis, and all the release estimates are saved and displayed as a probability histogram. The more Monte Carlo trials run, the more continuous this histogram appears. This histogram can then be fit to a more conventional distribution, and the statistics describing that distribution are given by the Crystal Ball software, accounting for the uncertainty in the input variables.

Many distributions of environmental data uncertainty are lognormal and are best represented by a geometric mean and a geometric standard deviation. The parameters calculated for the alpha release uncertainty are generic uncertainty parameters that can be applied to any release value during the appropriate time period. To determine the range of possible values for a given release value, the median value would be represented by the release estimate multiplied by the geometric mean. The range of values within 1 standard deviation of the mean is defined by multiplying and dividing the mean value by the geometric standard deviation. The 5th and 95th percentile values are obtained by dividing and multiplying the mean value by the square of the geometric standard deviation.

For alpha releases from 1955–1957, the geometric mean is 1.08 and the geometric standard deviation is 1.64. For alpha releases after 1957, the geometric mean is 0.98 and the geometric standard deviation is 1.21.

CONSISTENCY IN REPORTED DATA

Total plutonium and uranium emissions reported by Cummins et al. (1991), Carlton et al. (1993), and Evans et al. (1992) are consistent with total alpha emissions reported in the Health Physics Regional Monitoring report series (Alexander and Horton 1956; Horton and Mealing 1956a, 1956b; Mealing 1957; Mealing and Horton 1957; Mealing et al. 1958; Harvey et al. 1959a, 1959b; Du Pont 1959a, 1960a, 1960b, 1961a, 1962a, 1962b, 1963a, 1963b, 1964a) and the Environmental Monitoring at the Savannah River Plant report series (Ashley 1965b, 1966b, 1967b, 1968, 1969b, 1970a, 1971, 1972). Data provided by Cummins et al. (1991) were compiled from air monitoring results gathered from both routine and special monitoring at effluent sampling locations by the Environmental Monitoring Section of the Environmental Protection Department at the SRS. The reported alpha emissions are also consistent with data provided in annual SRP Waste Audit reports (Ashley 1960, 1962a, 1962b, 1963, 1965a, 1966a, 1967a, 1969a, and 1970b), and specific radionuclide emissions are consistent with data provided by Ashley et al. (1982). Total alpha release data compiled from original monthly monitoring reports from 1954 through 1956, 1959 through 1965, and 1967 through 1970 (Du Pont 1954a–l, 1955a–l, 1956a–l, 1959b–m, 1960c–n, 1961b–m, 1962c–n, 1963c–n, 1964b–m, 1965b–m, 1967a–l, 1968d–o, 1969a–l, 1970a–l) are also consistent with the data provided by the above sources. We have attempted to locate original hand-written release data compilations, but no original data have been located that could be used to further verify the reported emissions.

SUMMARY OF ALPHA RELEASES

Plutonium and uranium annual release values for F-Area and H-Area between 1955 and 1989 reported by Cummins et al. (1991) are compiled in a Microsoft Excel® workbook (Estimated_source_term.xls). Median values and 5th and 95th percentile values are calculated for

both F-Area and H-Area, based on the uncertainties discussed above. We have assumed that line-loss corrections were not made to reported release values and that only anisokinetic probes have been used. Therefore, we have also applied calculated transmission factors (Table 4.4-3) to all reported plutonium and uranium releases.

For plutonium releases occuring in 1955 (F-Area and H-Area) and 1969 (H-Area only) and all uranium releases, we have used the transmission factors calculated assuming the particle size distribution provided by Sanders (1978). This results in estimated releases approximately a factor of four greater than Site-reported releases values. Zippler (1979) states "Due to the complexity of our sample lines theoretically less than half of the particles with median diameters greater than two microns would ever reach the filter paper", which is generally consistent with the transmission factors we calculated based on the larger particle size distribution reported by Sanders (1978).

For plutonium releases during all years except 1955 and 1969 (H-Area only), we have used the transmission factors calculated assuming the particle size distribution provided by Carlson et al. (1983). This results in values approximately 35% to 45% greater than the Site-reported release values. Anonymous (date unknown) estimated a transmission factor of 65% using the PLATEOUT computer model and an assumed particle size of 1.5 µm, which is generally consistent with the transmission factors we calculated using the model developed by Texas A&M University and similar assumed particle sizes.

These estimates may exaggerate actual releases if the released material is not appropriately characterized by the assumed size distribution data provided by Sanders (1978) and Carlson et al. (1983). However, we feel it is important to conservatively estimate potential releases, particularly because filter breaks, which have resulted in the majority of alpha-emitting radionuclide releases, may have resulted in releases consisting of larger-sized particles. Additionally, Voillequé et al. (1995) reported uranium releases to be associated with larger-sized particles, similar to the distribution reported by Sanders (1978).

Figures 4.4-9 and 4.4-10 show the annual median release estimates we calculated for plutonium and uranium, respectively. The error bars represent the 5th and 95th percentile values, which were calculated based on the uncertainties and transmission factors discussed above.

The data indicate that the majority of plutonium emissions from both F-Area and H-Area stacks occurred during 1955 and 1969, primarily as a result of known filter break events. The data also indicate that the majority of uranium emissions from H-Area stacks occurred during 1955, 1968, and 1969, and that the majority of uranium emissions from F-Area stacks occurred during 1955, 1956, and throughout the 1960s. Evaluation of potential impacts resulting from alpha-emitting radionuclide releases should be focused on these years because the relative magnitude of total emissions during other years (including all years since 1970) appears to be quite small.

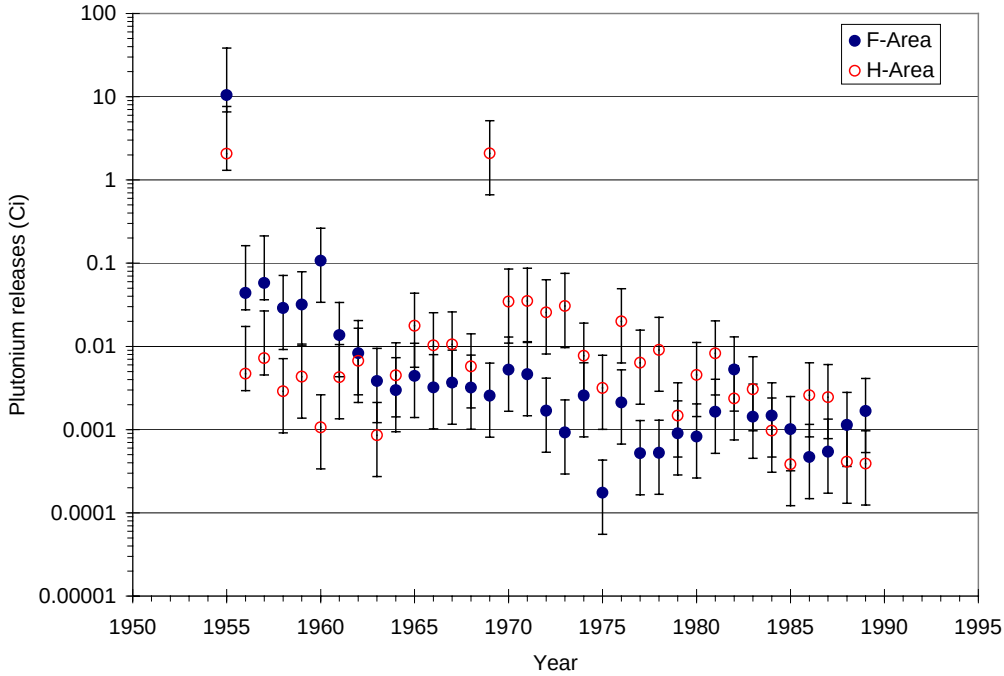

Figure 4.4-9. Annual median plutonium, including ^{238}Pu and ^{239,240}Pu, release estimates for F-Area and H-Area. The upper and lower error bars represent the 95th and 5th percentile values, respectively. Link to tabulated figure data.

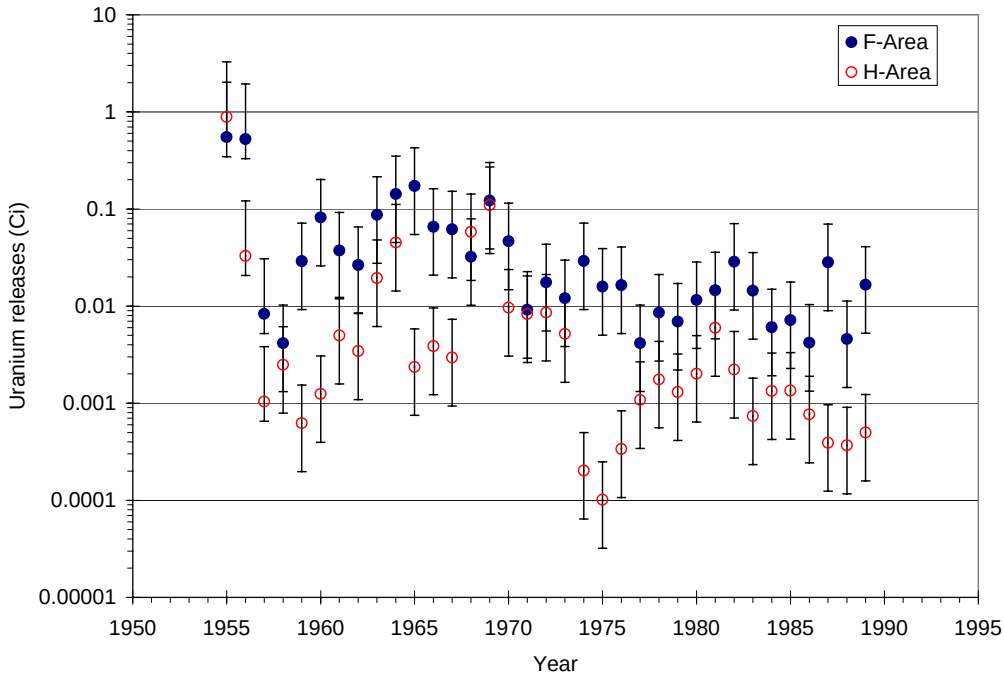

Figure 4.4-10. Annual median uranium release estimates for F-Area and H-Area. The upper and lower error bars represent the 95th and 5th percentile values, respectively. Link to tabulated figure data.

It should also be noted that emissions, including a number of alpha-emitting radionuclides, associated with the burning of coal may approach and even exceed the F-Area and H-Area plutonium and uranium emissions for many years, particularly after 1960 (see Table 4.4-1). Based on the relative importance of plutonium releases (to be determined during future phases of this project), particularly those occurring during 1955 and 1969, it may be necessary to more closely evaluate the potential health consequences associated with radionuclides present in airborne fly ash.

VALIDATION OF RELEASE ESTIMATES

A detailed validation of release estimates goes beyond the scope of work for this phase of the dose reconstruction project. However, because of several reviewer comments related to the appropriateness of sampling line deposition corrections made to Site-reported release estimates, we have included this discussion to further address the adequacy of our release estimates. Heffner (1999) offered lines of reasoning to support Site-reported release values by suggesting that modeled concentrations of Site-reported release estimates for 1955, the year of the highest plutonium release, show good correlation with measured gross alpha air concentrations during that year. It is asserted that either sampling line losses were insignificant or that some other process, such as large particle size, reduced the offsite impact of the release. It is not indicated what particle size was assumed for the modeling estimates made by the Site, but the larger particle size distribution we assumed for the transmission factor calculated for plutonium releases in 1955 certainly would result in increased deposition and, consequently, could similarly limit impact at the plant perimeter locations.

Based on soil (Chapter 12.2), air (Chapter 8), and vegetation (Chapter 9) monitoring data, it is not apparent that releases of alpha-emitting radionuclides have significantly impacted concentrations in media at plant perimeter or offsite locations. However, it is clear that these releases have impacted concentrations measured at onsite locations, near the F-Area and H-Area. We maintain that our estimated releases of plutonium and uranium, which take into account potential line losses, are conservative and appropriate, based on the events that led to the releases and on reported particle sizes. It is quite possible that modeling of our higher release estimate for 1955, taking into account the assumed particle sizes, will produce results that still correlate with measured air concentrations at the plant perimeter because of increased deposition of larger particles at onsite locations. At any rate, additional modeling of alpha-emitting radionuclide release estimates should necessarily include some amount of calibration to achieve reasonable correlation with gross alpha concentrations measured in air and vegetation samples.

ELECTRONICALLY COMPILED ALPHA RELEASE DATA

The data summarized in this section are electronically compiled in two Microsoft Excel® workbooks. One workbook (Ch4-4-Figure_data.xls) contains the figures provided in this chapter as well as the tabulated data that were used to produce the figures. In this workbook, there is a separate worksheet for each figure and one worksheet that contains the tabulated data for all of the figures.

The second workbook (Ch4-4-All_data.xls) contains the data that have been tabulated from various environmental monitoring reports and release summary documents. The workbook

contains several named worksheets, each containing a brief summary of the data compiled in the worksheet.

Table 4.4-4 summarizes the data that have been electronically compiled for alpha releases from the SRS. Additionally, the names of the individual spreadsheets in which these data are compiled (including a brief description of the data) are provided.

Table 4.4-4. Description of Data Electronically Compiled for Alpha Releases

Workbook name	Worksheet name	Brief description of data
Ch4-4-Figure_data.xls	Figures 4.4.1 through 4.4.10	Each worksheet contains a separate figure depicted in this chapter
	Data for figures	This worksheet contains the tabulated data for each of the figures
Ch4-4-All_data.xls	Total alpha	Total alpha releases reported in Health Physics Regional Monitoring report series
	Total alpha (2)	Comparison of total alpha releases to plutonium releases reported by Carlton et al. (1993) and uranium releases reported by Evans et al. (1992)
	SRP audits	Data compiled from annual Audit of SRP Radioactive Waste reports
	Plutonium releases	Plutonium release data compiled from Cummins et al. (1991) and Carlton et al. (1993)
	Uranium releases	Uranium release data compiled from Cummins et al. (1991) and Evans et al. (1992)
	Monthly reports	Total alpha monthly release data compiled from monthly monitoring reports
	Comparison of data	Comparison of data reported by different sources

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CHAPTER 5

RELEASES OF RADIONUCLIDES TO SURFACE WATER

ABSTRACT

The five production reactors were the source of the majority of radionuclide releases to surface water from the Savannah River Site (SRS), primarily because most surface water releases came from the disassembly basins in the reactors areas. Releases of liquid effluents from the separations areas were discharged into storage tanks and seepage basins and not directly to the streams. Tritium and ^{137}Cs are the main radionuclides of concern from past releases to surface streams that eventually reached the Savannah River, and detailed source terms have been calculated for those radionuclides. Other radionuclides that are important depending upon the exposure pathways include ^{90}Sr, ^{131}I, the activation products, ^{60}Co, ^{32}P, and ^{65}Zn, and uranium releases from the M-Area. Surface water releases of radionuclides were highest in the early to middle 1960s and decreased into the 1980s. Our median estimate of release of tritium for all years (1952 through 1992) is 1.8 million Ci, with the 5th and 95th percentiles of the distribution of 1.3 million and 2.5 million, respectively. The median estimate of the total ^{137}Cs for all years is about 250 Ci with the 5th and 95th percentiles of the distribution of 100 and 600 Ci ^{137}Cs. The median estimate of the total ^{90}Sr for all years is about 100 Ci with the 5th and 95th percentiles of the distribution of 45 and 250 Ci ^{90}Sr. There is overall general agreement between our reconstructed release estimates to the Site streams based on original measurement data, supported by weekly and monthly reports from the 1960s and 1970s, and the annual total reported by SRS. There is more uncertainty associated with release estimates in the 1950s and 1960s than in later years because of improvements in monitoring capabilities and techniques and in methods to prevent the release of materials from the reactors and processing facilities.

INTRODUCTION

A general overview of the key operational areas at the SRS, and important liquid effluent release points may be helpful in understanding the sources of information and methods used for reconstructing the releases of key radionuclides released in liquid effluents from the SRS in the past. A detailed history of the SRS Site and processes can be found in a report produced during Phase I (Meyer et al. 1995), and a summary of that history in Chapter 2 of this current report. Some of the areas onsite that are important contributors to radionuclides in liquid effluents are reactor areas, separations areas, fuel fabrication area (M-Area), heavy water reprocessing (D-Area), and administration area (A-Area).

Reactor Areas

Five heavy water production reactors, called R, P, K, L, and C Reactors, operated at the Site. They were all constructed in the early 1950s and are located near the center of the Site in separate areas designated R-Area, P-Area, K-Area, L-Area, and C-Area and are separated by at least 2.5 mi. The operation periods for the reactors were: C Reactor 1954-1987, K Reactor 1954-1988, L Reactor 1954-1968 and 1985-1988, P Reactor 1954-1988, and R Reactor 1954-1964. A restart of

the K Reactor began in 1991. Plutonium and tritium—the primary products of the SRS reactors—are created by uranium and lithium absorption of neutrons. Tritium is also by ternary fission of transuranic elements in the reactor fuels and targets. The controlled fission process within the reactors produced enormous amounts of energy. These are heavy-water moderated reactors, which means that heavy water is circulated in a closed system through heat exchangers to moderate and cool the reactors. Water from the Savannah River and Par Pond provided the cooling water for the heat exchangers. The heavy water in the heat exchangers was cooled by ordinary water from the Savannah River. The secondary cooling water from the Savannah River was returned to the river through the Site streams. The river water did not come into direct contact with the reactor core but was used to cool the heavy water that served as a moderator and primary coolant. This secondary cooling water was stored in holding basins at each reactor area, passed through the heat exchangers as needed and then discharged as wastewater to the Site streams. In 1958, Par Pond, a 2,700-acre human-made lake, began providing cooling water from the P and R Reactors. The pond water was pumped to the P and R Reactors and returned to Par Pond, which allowed more river water to be pumped to L, K, and C Reactors. In 1985, L Lake was formed by damming Steel Creek to provide cooling water for the L Reactor to comply with the permit to restart the L Reactor. Before Par Pond and L-Lake, the water was discharged directly to Lower Three Runs Creek (from P and R Reactors), to Steel Creek (from L Reactor), to Pen Branch (from K Reactor), or to Four Mile Creek (from C Reactor).

The Reactor Support Facility for each reactor consisted of an Assembly Area, Disassembly Area and Purification Area (Figure 5-1) (Bauer 1986). In the Disassembly Area, depleted fuel and irradiated materials were stored for a time to allow short-lived fission products to decay and then were disassembled and loaded into casks. All disassembly operations were performed remotely under water. Materials were transferred sequentially through the four sections of the basin through narrow vertical gateways between sections. Each basin contained four main sections:

1. Vertical tube storage (VTS) assemblies suspended by an overhead monorail system for cooling.
2. Disassembly basin, where slugs from target assemblies containing plutonium were placed in buckets for shipment to F Canyon, and fuel assemblies were bundled for shipment to H Canyon.
3. Horizontal and bucket storage, where buckets and bundles were stored before transfer.
4. Transfer bay, where materials were loaded into shipping casks for transport to the separations areas.

The main source of radionuclide releases to surface water from the reactors was from the VTS and disassembly basins. Spent fuel and irradiated target elements were removed from the reactor and stored in these large water-filled basins adjacent to the reactor building. The water in the basins cooled the components and provided shielding for the radioactive components. During the years of peak operations (1950s and 1960s), materials remained in the disassembly basins for less than nine months, but the storage period was longer than nine months in later years. The 17 to 30 foot-deep basins had 3-ft thick concrete walls and held 3.5 million gal of water in C-Area, L-Area, and K-Area and about 4.8 million gal in the P-Area and R-Area.

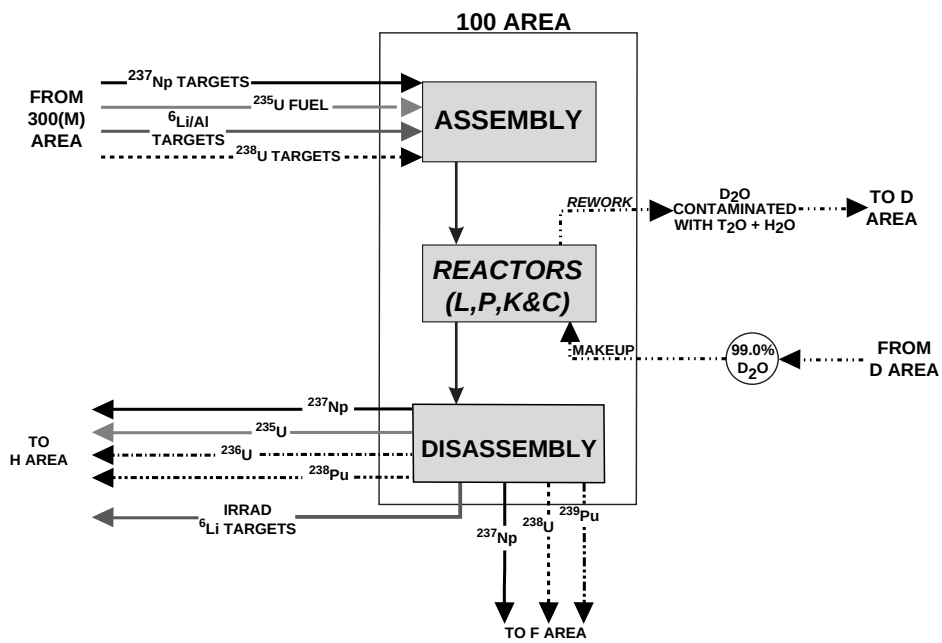

Figure 5-1. Materials flow diagram for the reactor (100) areas at the SRS. The area is comprised of the reactors as well as an assembly area, which receives fuel and target materials from the 300-M Area, and the disassembly area, which contains the vertical tube storage and disassembly basins that sent products, irradiated fuel, and irradiated targets to the F-Area and H-Area. The moderator for the reactor, deuterium oxide (D_2O), comes from the Heavy Water Plant in D-Area, and was returned to the D-Area for processing when the heavy water became degraded with tritium oxide (T_2O) and light or natural (H_2O) water. (Redrawn from Bauer 1986).

From startup to the mid-1960s, visual clarity was maintained in the basins by continuously purging them with fresh, filtered river water. The basin purge water was discharged to the Site streams along with the secondary cooling water. In the mid-1960s filters were added to the VTS basins. Sand filters were added to the disassembly basins in the early 1970s and continuous basin purging to the Site streams was discontinued (Murphy et al. 1991). However, periodic purges continued directly to the Site streams from 1970 to 1977 to reduce the tritium exposures to operating personnel because the sand filters did not remove tritium. Finally, in 1978, the purge water from the fuel storage basins was discharged to the seepage basins (Reinig et al. 1973, Towler 1980).

Separations Areas

Products produced in the reactors were separated chemically in the F-Area and H-Area separations area, located near the center of the SRS. Complex chemical and physical processes in the F-Canyon and H-Canyon Buildings, 200-F and 200-H, separated uranium, plutonium, and fission products. Major operational milestones for both the F-Area and H-Area were increases in the processing capacities throughout the 1950s and 1960s. Both the F-Area and H-Area were operated throughout the 1980s and 1990s. The separated plutonium and uranium were then

transferred to other facilities in the F-Area and H-Area and processed into solid forms. Fission products were stored in high-level waste tanks in the separations areas. Originally, the tritium received in the separations areas was a by-product of plutonium production. By 1955, greater production of tritium was needed, and a second tritium production line became operational in 1957. Cooling water for portions of the F-Area separations process line was pumped from deep wells and discharged to Four Mile Creek after use. At times, this water contained measurable amounts of radioactivity because of the occurrence of cooling coil leaks. Cooling coils are used in the head end dissolvers, which contain a mixture of many fission and activation products. Uranium is a predominant chemical constituent of this mixture (Evans et al. 1992). The F-Area liquid effluent consists of process cooling water, sanitary wastewater treatment effluents, and spill runoff, similar to the H-Area effluent.

The H and F facilities each had their own waste tanks, seepage basins and retention basins, but they shared a common burial ground (643-G). For processes in both 221-H and 221-F, liquid high activity wastes were evaporated to recover nitric acid and reduce volume, neutralized with sodium hydroxide and sent to the Building 241 tanks for storage. Radioactive wastes were transferred from both buildings in four stainless steel waste headers along the east side of the buildings in pipes that ran at a gradual slope within a hollow concrete encasement to the Building 241 waste tanks. If actinides were present, the evaporated high activity concentrate may have been run through the ion exchange primary recovery column and frames in the separations area to recover plutonium and neptunium before being sent to the waste tanks.

Most of the liquid low activity waste was concentrated by evaporation, neutralized with sodium hydroxide and stored in the Building 241 tanks. Equipment removed from the canyon was decontaminated by jetted detergent and chemicals before repair. Equipment to be discarded was often cleaned first, in a special cell called the swimming pool, to be classified as low activity waste. Solid waste from most of the facilities was trucked to the burial ground (Building 643-G) near the F-Area and H-Area (Figure 5-2). Solid waste was shipped from the hot canyon by rail.

Some of the tritium releases from the separations area originated as tritium-contaminated air. In 1973, a special study was conducted in 200-H during November and December to determine the source of the apparent tritium contamination in the 221-H cooling water systems. Results suggested that more than 90% of the tritium released to the streams from the 221-H cooling water systems could originate as tritium-contaminated air near 285-H, which was “cleaned” as it passed (or was drawn) through the cascading water in the cooling tower (Epting 1974). Sampling stations, set up near the 285-H cooling water tower, were designed to measure the influence of airborne tritium near the tower on a covered container of clean, deionized water. The station also collected tritium-contaminated rainwater. Airborne tritium concentrations in the vicinity of the cooling water tower were determined using the silica gel moisture collection method (see Chapter 4.1). Results showed that there was sufficient ambient airborne tritium to account for an average of 3 Ci mo^{-1} (and higher during periods of high stack releases from the tritium facilities) of releases to the stream via cooling water releases. There was an apparent direct relationship between tritium air activity near the 285-H tower and tritium concentrations in the tower cooling water. It was noted that less than 10% of the tritium releases via 221-H cooling water were thought to be due to canyon building processes (Epting 1974).

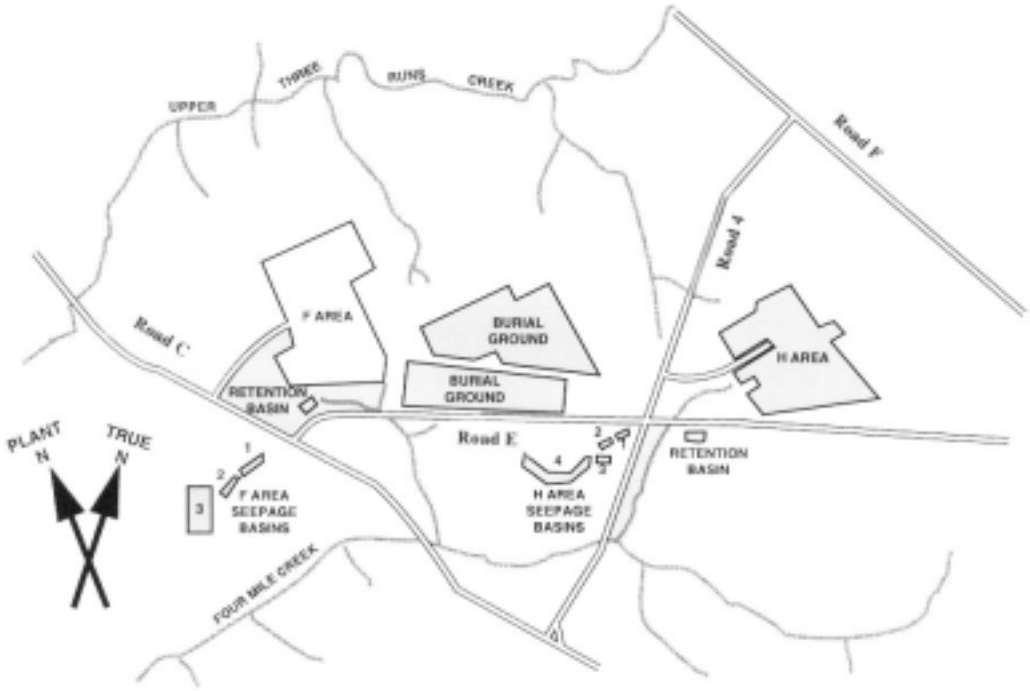

Figure 5-2. A diagram showing one location of waste areas at the SRS—the radioactive burial grounds, and seepage and retention basins in the separations area at the SRS. (Redrawn from Ice 1971).

Fuel Fabrication Area, M-Area

The fuel fabrication facilities produced fuel and target elements from aluminum and uranium that were then sent to the 100-Area production reactors. There was also a treatment facility for radioactive liquid wastes in the M-Area. See Meyer et al (1995) and Chapter 2 for a thorough description of the fuel fabrication area. Over time, changes in the fuel and targets were made, new and different fuels were tested, and more efficient and productive fuels were designed (Pelfrey 1987; Pickett 1997). The reactor assemblies became increasingly complex as different products were desired and more efficient cooling means were developed.

Of the radioactivity released from this area, uranium dominated other radionuclides in liquid effluents (Horton 1955b). The largest releases of uranium onsite occurred from the M-Area to Tim's Branch and to the seepage basins after the seepage basins were put into service in 1973. The main sources of uranium in liquid effluents from this area were from:

- Uranium in etching solutions—The uranium metal was electrolytically plated with nickel and then clad, or covered, to protect it from contact with the reactor coolant because uranium corrodes rapidly in water. The moderator and coolant used at the SRS was heavy water. The cladding also served as a barrier to keep fission products from contacting the water. For the aluminum cover or cladding to bond to the

uranium, the uranium target slugs had to be etched with acid. As a result, the etching solution contained up to 300 g L^{-1} of uranium (Evans et al. 1992).

- Ruptured aluminum seals on uranium slugs—After cladding, the aluminum seals on the uranium slugs were tested by heating the slugs under pressure in an autoclave. If there were leaks in the cladding, slug rupture occurred and those slugs could then be rejected for use. After rupture, the autoclave had to be washed out, and particulate uranium oxide was present in this wash-down water of the autoclave.
- Chemical removal of the aluminum and nickel cladding from the rejected slugs, using nitric acid—When a uranium target slug was rejected, the aluminum and nickel cladding was removed using nitric acid. Even though some of this dissolved material was precipitated and filtered to remove the suspended solids, much of it was carried along in the waste liquids (Evans et al. 1992).

Heavy Water Reprocessing, or D-Area

A heavy water production plant, in D-Area, began operation in 1953 to concentrate heavy water from Savannah River water to moderate and cool the Site's reactors. Heavy water was produced in the Heavy Water Plant, 400-D, from October 1952 to 1982. The Heavy Water Plant extracted deuterium oxide, D_2O from Savannah River water by dual temperature exchange using hydrogen sulfide (H_2S). Natural river water contains D_2O at a concentration of about 150 ppm, or 0.015%. The Dana Plant, a facility in Dana, Indiana, was a precursor to the SRS facility and produced heavy water by the GS process for the SRS (Bebbington 1990). The facility stopped production in 1981 because there was a sufficient supply of heavy water. A heavy water reprocessing facility, a coal-fired power plant, and a laboratory that analyzes process effluent samples are also located in the D-Area (See Meyer et al. 1995 and Chapter 2).

Administration Area, or A-Area

The A (Administration) Area contained organizations that provided direct support for SRS operations. Generally, the 700-numbered buildings were offices, laundries, fire stations, maintenance buildings, research laboratories, with a lower potential for offsite release of hazardous materials than the Reactor Areas or separations areas. The Savannah River Laboratory (SRL), or Savannah River Technology Center (SRTC) as it is now called, are located here as well as the University of Georgia's Savannah River Ecology Laboratory (SREL).

Although not located in the A-Area, the TNX and CMX Semiworks were considered to be a part of the SRL and were some of the first facilities to operate at the SRS. CMX and TNX were code designations and had no logical derivation according to Bebbington (1990). CMX investigated problems associated with using Savannah River water for cooling, and it housed the river water pumps (water treatment took place at the reactors), a pressure facility for testing of reactor elements, and a hydraulic test facility. CMX was shut down in 1984. The TNX facility, one of the first facilities to operate at the Site, provided technical support, pilot data, and personnel training. In later years, the facility was involved in waste processing research and development. By the end of the first 5 years, all of the basic production facilities were in operation and the products, plutonium metal and tritium gas, were being delivered.

GENERAL HYDROLOGY

The Savannah River is the principal river in the area with an average velocity of about 3 to 5 mi hr^{-1} (MPH) or 4.4–7.3 ft s^{-1} and average flow in 11,000 cubic feet per second (cfs) (Ice 1971). The water table at the Site varies from ground surface to slightly more than 100 ft deep. Groundwater in the area occurs as perched water tables, normal water tables and artesian aquifers (Ice 1971). The principal aquifer is the Tuscaloosa formation, with a potential water yield ranging from 30,000 to 400,000 gal per day per foot. In the chemical separations areas (F-Area and H-Area), the groundwater velocity ranges from a few hundredths of a foot per day to nearly 1 ft d^{-1} near flowing streams. Both solids and liquid wastes have been stored at or just beneath ground level during much of its operational history. Thus, the local hydrology and geology play an important role in assessing the releases via surface water.

Onsite Streams

Both chemicals and radionuclides in liquid effluents from the SRS facilities are released either directly into onsite streams or first into seepage basins and then into onsite streams. In the early years of operations, most liquid waste was discharged to streams or swampy areas. With time, seepage basins, settling ponds, tanks, and retention basins were used. Contaminants from these often entered groundwater then migrated to the nearest downgradient surface water.

Of the radionuclides that reach the water table, tritium is the most important because of its high mobility and abundance. Low concentrations of tritium are present in liquid waste streams and burial ground leachates as tritiated water. The tritiated water moves with the groundwater, some of which outcrops to surface streams. A study in 1983 (Christensen and Gordon 1983) reported that the waste sites that are the principal contributors of tritium from shallow groundwater to surface water at the SRS are

- K-Area containment basin—about 10,000 Ci y^{-1} outcrops to a tributary of Pen Branch
- H-Area seepage basins—about 7,000 Ci y^{-1} outcrops to Four Mile Creek and tributaries
- F-Area seepage basins—about 2,000 Ci y^{-1} outcrops to Four Mile Creek
- Radioactive burial grounds—an estimated 200 Ci y^{-1} outcrops to a tributary of Four Mile Creek.

Almost all of the SRS is drained by five main streams: Upper Three Runs Creek, Four Mile Creek, Pen Branch, Steel Creek and Lower Three Runs Creek. The onsite streams meander for 8–13 km (5–8 mi) onsite, through the Savannah River swamp system before eventually discharging offsite into the Savannah River (Westinghouse 1996). One smaller stream in the northeast area of the Site drains to the Salkehatchie River rather than the Savannah River.

The following section describes each of the major onsite streams and close-up maps for the five Site streams show the drainage patterns of various facilities to specific streams. These diagrams show that Upper Three Runs received runoff from the storm sewer from the north side of F-Area and seepage from the F-Area ash basin. At Road C, Tim's Branch discharges industrial, laboratory, water treatment, and sanitary wastewater treatment effluents from A-Area and M-

Area to Upper Three Runs. Steed's Pond, located on Tim's Branch just north of Road 2, has served as a mixing and settling pond for A-Area and M-Area wastes. The creek merges with the C-Reactor cooling water about 0.5 mi south of Road 3. The operating reactor cooling water flow is much greater than that in the receiving stream, and as a consequence, materials already in Four Mile Creek are greatly diluted. This may be advantageous from the standpoint of release concentration limits, but it makes analyses more difficult.

The onsite streams of particular importance for carrying reactor effluents are Steel Creek and Lower Three Runs Creek. Steel Creek received discharges from the L and P Reactor Areas in the early years before Par Pond was constructed and filled in late 1958. After that, the P Reactor cooling water was released to a canal for recirculation in Par Pond. The K Reactor effluent from Pen Branch was discharged to Steel Creek via the Savannah River swamp through one major and one or more minor streams about 0.25 mi above Steel Creek mouth. Lower Three Runs Creek received some discharge from the R Reactor area before 1958 when Par Pond opened. The overflow from Par Pond to Lower Three Runs carried runoff from the northeast portion of the plant, sewage treatment effluent, water treatment chemicals added to P Reactor cooling water, and any materials leaking to the cooling water.

Significant modifications to creek drainage patterns, channel depth, and surrounding vegetation configurations have occurred over the years as a result of SRS discharges. The streams most heavily affected were Steel Creek, Pen Branch, and Four Mile Creek where small deltas have been deposited at their creek mouths where they enter the Savannah River swamp system or flood plain (Ruby et al. 1981). These sedimentation deltas have resulted from erosion of stream banks because the stream channels have carried many times their natural flows. For example, the natural flow in Steel Creek of about 35 cfs increased roughly ten-fold (250-400 cfs) when effluents from the L and P Reactors were discharged into Steel Creek in the late 1950s and early 1960s.

The Savannah River swamp system borders the Savannah River for about 16 km (10 mi) and averages about 2.4 km (1.5 mi) wide. About 10,000 acres of the SRS forest lie on the Site from Upper Three Runs Creek to Steel Creek. The Savannah River flow records its highest levels in the winter and spring, and lowest levels in the summer and fall. The river historically overflows its channel and floods the swamps bordering the Site when its elevation rises higher than 27 m above mean sea level at the SRS Boat Dock. This level corresponds to flows equal to or greater than about 438 $m^3\ s^{-1}$. Studies have indicated that the swamp was flooded approximately 20% (74 days per year on the average) during the period 1958 to 1967 (Westinghouse 1996). Clearly, the onsite streams play an important role in receiving, transporting, removing and diluting materials before they are released offsite in the Savannah River.

Upper Three Runs Creek

This creek is the only SRS stream with headwaters arising offsite, north of the Savannah River Plant (Figure 5-3). This stream and its major tributaries, Tinker Creek and Tim's Branch, drain an area of about 545 km^2 (Evans et al. 1992). Upper Three Runs has the largest natural flow of any plant stream. The average flow of 257 $ft^3\ s^{-1}$ is slightly increased by a Site contribution of less than 10 $ft^3\ s^{-1}$. Before the point where the creeks converge, the streams received no plant process discharges prior to the construction of the Effluent Treatment Facility (ETF) in 1988 but

are subject to runoff of herbicides from activities at the U.S. Forest Service-Savannah River Environmental Laboratory facilities, and part of the H-Area storm sewer system.

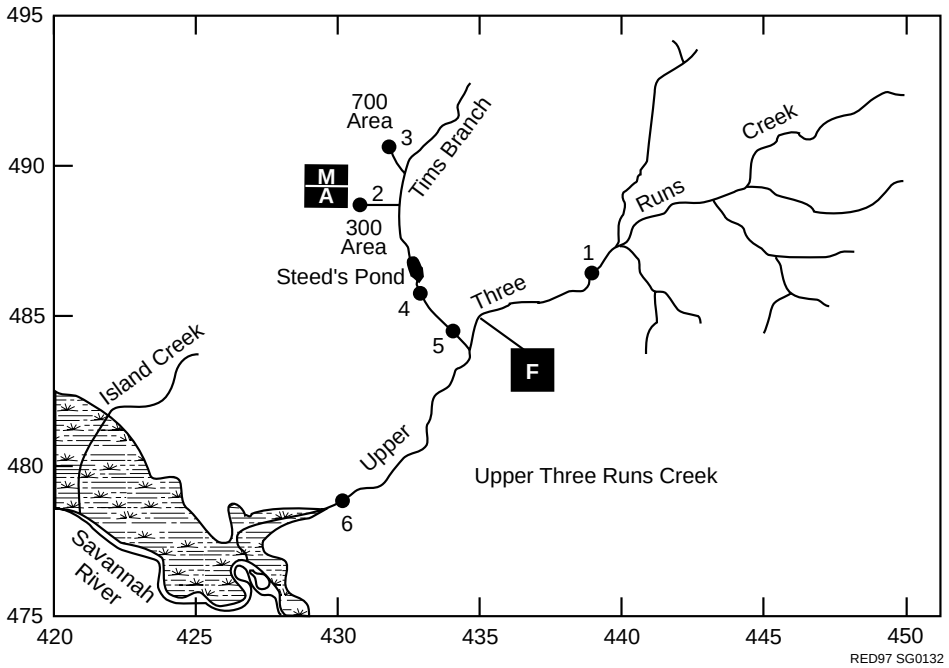

Figure 5-3. Map of the Upper Three Runs Creek and its tributaries at the SRS with water sampling locations in the early years. Sampling location 6 represents the point where Upper Three Runs Creek crosses Road A, the final sampling location onsite before the creek flowed into the Savannah River. The numbers on the axes are from an SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

At Road C, Tim's Branch discharges industrial, laboratory, water treatment, and sanitary wastewater treatment effluents from A-Area and M-Area to Upper Three Runs. The M-Area fuel fabrication facility discharges liquid effluents to Tim's Branch, which contains large quantities of acids, bases, and salts with widely varying pH values. Starting in early 1973, these wastes were diverted to a settling basin. Other discharges to the A-Area and M-Area effluents include drainage from such activities as power facilities, automobile maintenance, and equipment overhaul and can contain contaminants such as oils, greases, detergents, corrosion inhibitors, boiler water chemicals and other trace chemicals, and pesticides. Steed's Pond, located on Tim's Branch just north of Road 2, has served as a mixing and settling pond for A-Area and M-Area wastes, and the Site routinely sampled the outfall from the pond to determine deposition or holdup in the pond.

From Road C to Road A, the only source of releases should be runoff from power line right-of-way herbicide treatment, sanitary wastewater treatment effluent, and, possibly, corrosion inhibitors. Between Road A and the Savannah River, the stream is again subject to drainage from offsite activities, particularly from the land area recently released to the U.S. Forest Service.

Four Mile Creek

This creek rises on SRS property, about 1.5 mi east of its first confluence with an SRS discharge, the effluent from H-Area separations at a point a short distance east of Road C (Figure 5-4). The H-Area discharge consists of process cooling water, which contains water treatment chemicals, possibly corrosion inhibitors, sanitary wastewater treatment effluent, and, potentially, runoff from spills. The effluent also contains sulfuric acid and caustic solutions from ion exchange resin regeneration in the water treatment plant. An ash pit is located just south of the area, below Road E. Leaching from this pit would reach Four Mile Creek by way of a small stream that joins the H-Area effluent. Between Road 4 and Road C, the creek receives water from the H-Area seepage basins that has permeated the soil to the water table and moved with the ground water to the outcrop location at the stream (Ashley 1964).

Discharges from the F-Area reached Four Mile Creek just before Road C. Below Road C, outcropping of water permeating from the F-Area seepage basins added to the further discharge of materials to the creek. Below this point, Four Mile Creek received sanitary wastewater effluent; traces of oil, chemicals, and solvents from the Central Shop Area; runoff from nonpoint sources; and possible leaching from the C-Area ash pit. The creek merges with the C Reactor cooling water about 0.5 mi south of Road 3. The operating reactor cooling water flow was much greater than that in the receiving stream; as a consequence, materials already in Four Mile Creek were greatly diluted. This was considered advantageous from the standpoint of release concentration limits, but it is a disadvantage in that analyses become more difficult.

From the junction with the C-Area effluent, Four Mile Creek flows for about 7 mi before reaching the swamp. This section of the stream received no point discharges, only runoff from highway and power line right-of-way. The water was dispersed in the swamp and part of the flow reached the river through Beaver Dam Creek, part at the mouth of Four Mile Creek, and part, after flow in the swamp parallel to the river, through Steel Creek. At the mouth of Four Mile Creek, the temperature of the discharge to the river was usually several degrees higher than the ambient river temperature. In fact, this discharge had the most significant thermal effect on the river of any SRS discharge.

Beaver Dam Creek

This creek receives the effluent from the D-Area facilities, including the heavy water production plant and a coal-burning powerhouse (Figure 5-4). Conventional water treatment chemicals to condition the water for various uses are present in the discharge. Water used for process and powerhouse condenser cooling is elevated in temperature when discharged. Leaching from the ash pits at the 400-D power house is source of releases of chemicals to this stream. The drainage systems from the 400-D power house consisted of a 300-m channel that connected the ash settling basin and the SRS swamp. The water flowed through the swamp to Beaver Dam Creek. Water from Beaver Dam Creek (1.3 km in distance from the confluence) entered the swamp and followed the edge of the swamp to the entrance of Four Mile Creek (Newman et al. 1985). The creek follows a meandering route through the swamp enroute to the river, including mixing with a portion of the Four Mile Creek flow.

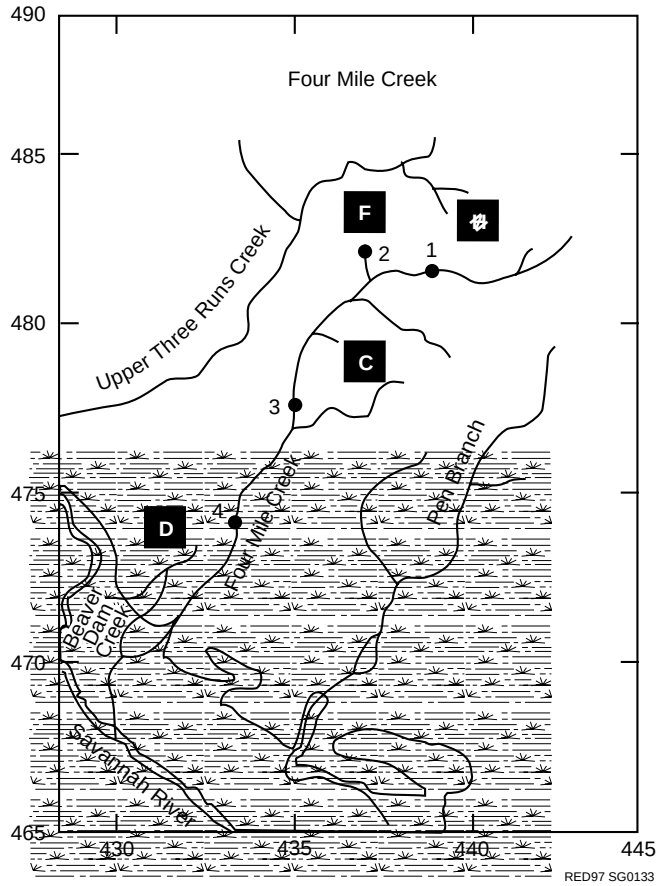

Figure 5-4. Map of Four Mile Creek and its tributaries at the SRS with water sampling locations in the early years. Sampling location 4 represents the point where Four Mile Creek crosses Road A, the final sampling location onsite before the creek flowed into the Savannah River. The numbers on the axes reflect an internal SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

Pen Branch

Pen Branch, above its confluence with the K Reactor effluent, receives only drainage water from sources such as Central Shops, the L-Area ash pit, and herbicide applications (Figure 5-5). The K-Area cooling water discharged into Indian Grave Branch, a tributary of Pen Branch. The materials released in the cooling water were similar to those described for the C Area. Leachate from the K-Area ash pit drained into Indian Grave Branch above its confluence with Pen Branch about 0.5 mi north of Road A. Pen Branch discharges to the swamp about 4 to 5 mi below Road A and is subject only to runoff along this distance. Pen Branch water then flowed through the swamp, dissipating heat from K Reactor cooling water, and joined Steel Creek about 0.5 mi from its mouth.

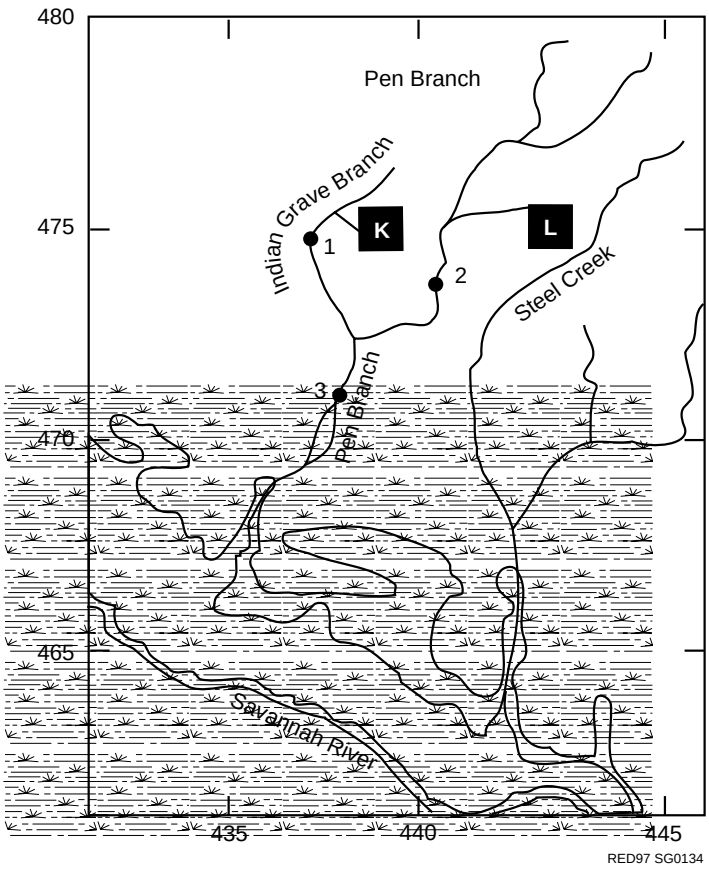

Figure 5-5. Map of Pen Branch and its tributaries at the SRS and water sampling locations in the early years. Sampling location 3 represents the point where Pen Branch crosses Road A, the final sampling location onsite before the creek flowed into the Savannah River. The numbers on the axes are reflect an SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

Steel Creek

After SRS operations began, Steel Creek headwaters were diverted to receive miscellaneous discharges from the 105-P Reactor building (Figure 5-6). The P Reactor cooling water was not released directly to Steel Creek but to a canal system for recirculation in Par Pond after Par Pond was constructed in 1958. The P Reactor supply water came from the river pumphouses and, to a greater extent, from the Par Pond Pumphouse 681-6G. From P Reactor to its confluence with Meyers Branch, just north of Road A, Steel Creek was subject only to runoff, including possible drainage from waste disposal and storage facilities in the L Area. No point discharges were made to Steel Creek below Meyers Branch. The K Reactor effluent from Pen Branch was discharged to Steel Creek via the swamp through one major and one or more minor streams about 0.5 mi above Steel Creek mouth. The water in Pen Branch cooled as it moved through the swamp, and the thermal effects of Steel Creek discharges to the river were somewhat dissipated.

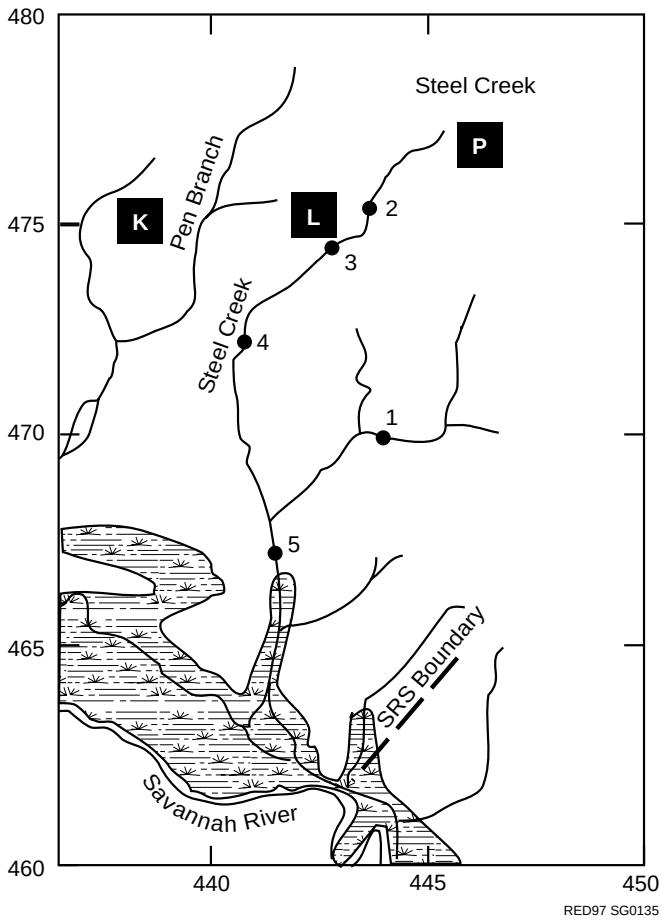

Figure 5-6. Map of Steel Creek and its tributaries at the SRS and water sampling locations in the early years. Sampling location 5 represents the point Steel Creek crosses Road A, the final sampling location onsite before the creek flowed into the Savannah River. The numbers on the axes reflect an internal SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

Lower Three Runs Creek

Lower Three Runs (LTR) Creek has the second largest drainage area of streams on the SRS. Both P- and R-Reactors have discharge effluent to the LTR system. The overflow from Par Pond to Lower Three Runs Creek carried runoff from the northeast portion of the plant, sewage treatment effluent, water treatment chemicals added to P Reactor cooling water, and any materials leaking to the cooling water such as sodium borate from the emergency cooling water system (Figure 5-7). Monitoring at this point (where Par Pond overflowed to the Lower Three Runs Creek) provided the major source of discharge inventory data from SRS to this creek. It also proved necessary to monitor the P-Area cooling water discharge to the canal system. Just below the Par Pond dam overflow, the Barnwell Nuclear Fuel Plant effluent was discharged to Lower Three Runs, and still further downstream, the effluent from other offsite facilities, as well as SRS runoff entered this creek. Below Patterson's Mill Bridge, the stream was still within the SRS property boundary, but the corridor was so narrow that it afforded little protection for runoff from

surrounding agricultural activities. In addition, the J. P. Stevens Co. textile facility effluent was discharged to the stream between Tabernacle Church Road and S. C. Highway 125. The Site considered that monitoring at locations along the corridor and at the mouth of the Lower Three Runs provided data to contest alleged discharge violations (by comparison with Par Pond overflow data). Because of the presence of these other non-SRS discharge points, the Site felt that monitoring stations along the corridor and at the mouth of the Lower Three Runs Creek would not be useful to determine discharge inventories.

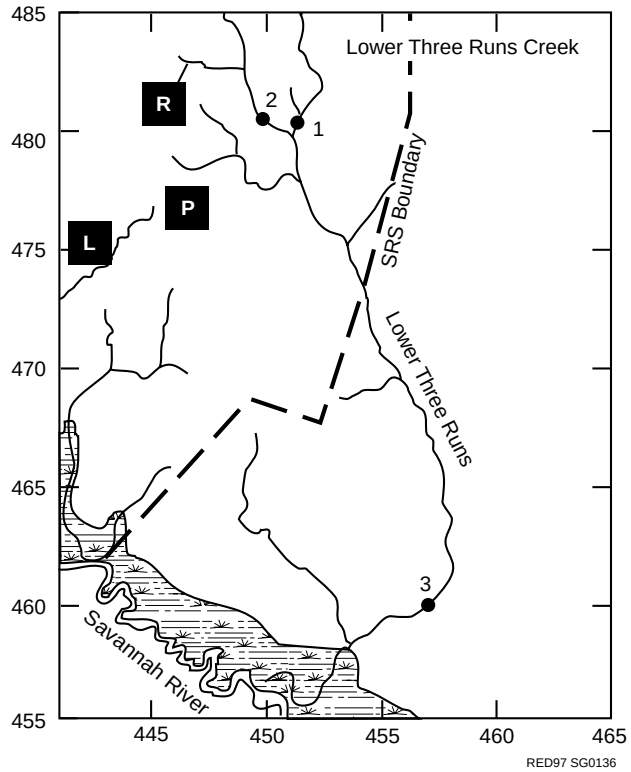

Figure 5-7. Map of Lower Three Runs Creek and its tributaries at the SRS with water sampling locations in the early years. The numbers on the axes reflect an internal SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

Site Stream Flow

Site stream flow data can provide an indication of processing activities onsite and are important for calculating total levels of activity released when radionuclide concentration is measured. Data like these play an important role in reconstructing releases from the Site streams to the Savannah River because the flow rate directly affects the dilution factor.

The U.S. Geological Survey (USGS) measured stream flow on four of the five Site streams beginning in 1959. The flow for Four Mile Creek and Steel Creek were influenced to a large extent by SRS operations. Four Mile Creek received effluents from the separations areas and cooling water from the C Reactor. In addition, shallow groundwater outcrops to Four Mile Creek from the H-Area and F-Area seepage basins and from the radioactive burial grounds. Because the C Reactor cooling water contributed a large fraction of water to the Four Mile Creek flow, the

general flow in Four Mile Creek decreased dramatically when the C Reactor was not operational. Figure 5-8 shows the flow rate in Four Mile Creek and how greatly it was influenced by C Reactor operations (Ashley 1959). Figure 5-9 shows the flow rates of Steel Creek and Lower Three Runs Creek from 1959 through 1967.

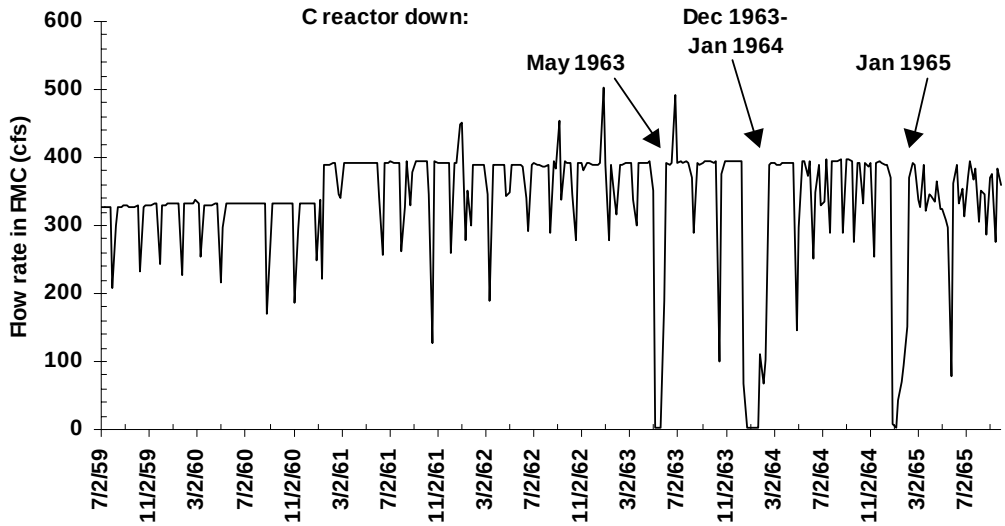

Figure 5-8. The flow rate in cfs for Four Mile Creek from July 1957 through June 1965. Four Mile Creek received cooling water and liquid effluent discharges from the C Reactor and the separations areas. The flow rate in Four Mile Creek more than doubled from that measured upstream when the C Reactor was operating. The dates indicate some of the times when the C Reactor was not operating and, therefore, was not discharging cooling water and other liquid effluents into Four Mile Creek.

Statistical characteristics of the flow rates in each of the streams discharging liquid wastes from SRS are given in Table 5-1. Steel Creek had the highest average flow rate of almost 500 cfs and Upper three Runs had the lowest. These data, along with concentration measurements made at the same locations, were used to calculate the total quantities of key radionuclides discharged from the Site to the Savannah River.

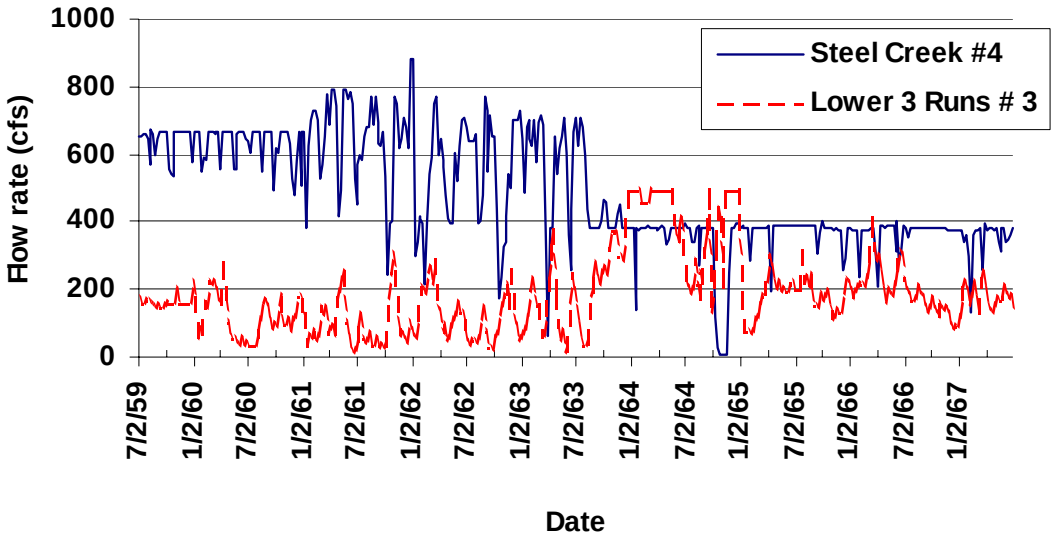

Figure 5-9. Flow rate in Steel Creek and Lower Three Runs Creeks from July 1959 through June 1967 measured at Road A on each stream by the USGS. The noticeable decrease in flow rate in Steel Creek and increase in Lower Three Runs in the fall of 1963 coincided with fuel failures and other operational problems in the R Reactor. The R Reactor was permanently shut down in 1964, at which time the flow of cooling water from the R Reactor to Steel Creek was stopped, and some cooling water from the P Reactor was diverted to Lower Three Runs. Cooling water flow from the reactors at SRS was a major contributor to stream water.

SRS WASTEWATER CONTROL

The SRS implemented various measures to control and limit the volume of liquid effluents that entered the Site streams directly and would eventually drain into the Savannah River. The liquid wastes from the Reactor Areas consisted primarily of disassembly basin water purged to maintain clarity in the basins. Each reactor area also had a 50 million gallon basin for emergency conditions to retain water that contained short lived radionuclides. In the separation areas, liquid effluents originated as cooling water from the process buildings in the F-Area and H-Area, from runoff to storm sewers, and from the seepage basins (Du Pont 1973).

The cooling water was classified as either circulated cooling water (circulated in the single-purpose coils of the canyon vessels) or segregated cooling water (circulated in the dual-purpose coils for heating and cooling). It was more likely to have radionuclide releases to the streams from the segregated system (Ice 1971). In both the circulated and segregated water system, the water was monitored and armed with alarms that would indicate high levels of either alpha or beta-gamma activity. If this occurred, the water was diverted to a retention basin or to the seepage basins. Storm sewer water from the separation areas was normally discharged to the Four Mile Creek. When spills or other sources of contamination threatened to drain into the storm sewers, emergency measures were implemented. These measures involved damning the streams into which the sewer discharged and then pumping the impounded water into the seepage basins.

Table 5-1. Flow Rate Characteristics of SRS Site Streams in the 1960s[a]

Flow rate parameter	Upper Three Runs[b]	Four Mile Creek #6	Pen Branch #3	Steel Creek #5	Lower Three Runs #2	Par Pond	Savannah River
	Flow Rate (cfs)						
Median		374	387	395	162	337	
Average	270	340	349	486	184	305	11,000[c]
Standard deviation		87	87	164	119	118	
Maximum	550	502	518	880	496	486	
Minimum	190	0	3	6	12	3	

[a] Based on reported weekly flow measurements made by the USGS from July 1959 through June 1967 (Ashley 1959). These values were converted into cfs for this table.

[b] Upper Three Runs has the largest natural flow rate (about 257 cfs) of the onsite streams at SRS; plant contributions raise the natural flow rate by only about 10 cfs. This stream and its major tributaries, Tinker Creek and Tim's Branch, drain an area of about 545 km^3 (Evans et al. 1992).

[c] This is equal to about 300 $m^3\ s^{-1}$; the average velocity of the Savannah River is 5–7 feet per second (3–5 mi per hour) (Zeigler et al. 1985)

Seepage Basins

Seepage basins were constructed in each of the two chemical separations areas before startup in 1955. They were originally designed and arranged so that liquid discharged to basin 1 overflowed to basin 2, and finally overflowed to basin 3 (Ice 1971). Tables 5-2 and 5-3 provide statistics on the history, size, and seepage characteristics of the basins. There is an underground seepline that roughly parallels Four Mile Creek that carries some contaminants from the seepage basins into FMC. The distance and flow time between the seepline and FMC varies depending on location (Table 5-3). Christensen and Gordon (1983) gave estimates of about 9 years travel time from the F-Area SB and from 1-4 years for contaminants from the H-Area. Thus, the location of the outcropping from groundwater to FMC is not a single point. The outcrop from F-Area occurs between sampling locations 2 and 3 (Figure 5-4). For H-Area, the outcrop to FMC occurs between sampling locations 1 and 2; thus, the distance is shorter but still not a single point location. This entire area is wet and swampy and the seep line actually comes quite close to FMC in several places.

Immobilization plus radioactive decay greatly reduce the amount of radioactive material eventually leaving control of the SRS (Marter 1969). Marter and others calculated the fraction of each radionuclide that would eventually move through the soil by a groundwater/soil transport equation that takes into account:

Table 5-2. Characteristics and General Information about the Seepage Basins in the Separations Areas

Location	Basin No.	Bldg. No.	Used	Area (acres)	Acre (ft^2)	Volume (gal)
200-F	-	904-49	1954–1955	1.3	56,550	na - NW of area
	1	904-41	1955–1988 [a]	0.34	14,800	1,000,000
	2	904-42	1955–1988 [a]	0.69	30,000	1,900,000
	3	904-43	1955–1988 [a]	4.44	193,000	14,000,000
Total F-Area seepage basins					238,000	
200-H	1	904-44	1955–1988[a]	0.29	12,615	1,000,000
	2	904-45	1955–1988 [a]	0.79	34,365	2,800,000
	3	904-46	1955–1962	3.3	143,550	2,300,000
	4	904-56	1962–1988 [a]	9.3	404,550	31,000,000 (crescent-shaped)
Total H-Area seepage basins					595,000	

a From Ice 1971; the F-Area and H-Area seepage basins were not used after 1988 when the Effluent Treatment Facility for the separations areas began operation.

Table 5-3. Seepage Characteristics of F-Area and H-Area Basins[a]

Parameter	F-Area	H-Area
Mean seepage rate (gal ft^{-2} d^{-1})	0.37	0.36
Distance to outcrop (ft)	1600	400–1400
Groundwater flow rate (ft d^{-1})	0.5	1.0
Travel time (y)	9	1–4
Basin evaporation (%)	25	27

[a] From Christensen and Gordon (1983).

The advantages of seepage basins over direct disposal to surface streams are that some ionic species are immobilized by the soil while others are greatly delayed in movement.

Using the known values for size and volume of the basins, it is possible to calculate an estimate of volume seeping from the basins over time:

Total area x mean seepage rate = volume releases per time
For F area:
238,000 ft^2 x 0.36 gal ft^{-2} d^{-1} = 88,060 gal per day from F-area seepage basins
= ~ 3.2 x 10^7 gallons per year from F-area seepage basins
For H-Area :
595,000 ft^2 x 0.36 gal ft^{-2} d^{-1} = 220,150 gal per day from H-area seepage basins
= ~ 8.0 x 10^7 gal year from the H-area seepage basins.

- Decay constant of the radionuclides
- Holdup time in the seepage basin
- Distance between the seepage basin and the nearest plant stream
- Adsorption distribution coefficient for the radionuclide in soil
- Weight of soil per unit volume of soil
- Volume of water per unit volume of soil
- Velocity of movement of groundwater (Evans et al. 1968).

Seepage basins were constructed at all reactor areas during the second half of 1957. The primary purpose was designed for disposing of waste discharged by underwater vacuum cleaning of the disassembly basins (Mealing et al. 1958). Interestingly, during the first year, at least, the radioactivity discharged to the basins was from sources other than vacuum cleaner discharges. The first R-Area seepage basin was placed in service on June 26, 1957 and received 9 Ci of nonvolatile beta activity from July through October. Then on November 8, 1957 a fuel failure in the R-Area lead to the discharge of over 1900 Ci of nonvolatile beta activity during the next 2 months. To handle the contaminated effluents, three additional seepage basins were placed in service in the R-Area on November 16, 21, and 29, respectively (Mealing et al. 1958). Special analyses of the seepage basin water collected on November 13 showed about 54% of the nonvolatile beta activity was due to radiostrontium, 14% to radiocesium and about 25% to rare earth elements. The maximum nonvolatile beta concentration observed in samples collected from these basins during this time was 8.5×10^8 pCi L^{-1}(Mealing et al 1958). The average nonvolatile beta activity measured in Lower Three Runs Creek ranged from about 200 to 300 pCi L^{-1}, and from 30 to 200 pCi L^{-1} in the Savannah River (Harvey et al. 1959). During the subsequent 6 months the concentration averaged about 25 pCi L^{-1} downstream of where Lower Three Runs enters the river (Harvey et al. 1959).

Sanitary Wastewater Treatment

When the plant was constructed in the early 1950s, primary treatment was provided for sanitary wastewater (sewage), which met South Carolina regulations. By the 1960s, regulations changed, and secondary wastewater treatment was required (Marter 1965). Du Pont Engineering Department Design developed secondary treatment facilities over time to comply with the state regulations.

Prefabricated extended aeration plants for the required secondary treatment were designed for 100-P, 200-F, 200-H, 700-A, and 400-D in the 1970s. Central Shops, with its variable work force, had a dual stabilization pond system. The remaining septic tank facilities were provided with individual tile fields, which were designed to retain liquid effluents in the soil. Table 5-4 lists the SRS wastewater treatment facilities in place by 1970.

Table 5-4. Sanitary Wastewater Facilities Present at the SRS by 1970[a]

Bldg. No.	Area	Flow (gpm)	Type of Plant in 1970	Discharge watershed
607-A	3/700	74	primary plant	Tim's Branch-Upper Three Runs
607-F	200-F	20	primary plant	Four Mile
607-H	200-H	20	septic tank	Four Mile
607-P	100-P	8	septic tank	Canal to Par Pond
607-D	400-D	8	septic tank	Beaver Dam Creek
607-K	100-K	8	septic tank	Pen Branch
607-C	100-C	10	septic tank	Four Mile
CMX-TNX		1	septic tank	Savannah River
607-U	TC-1	1	Imhoff Tank	Upper Three Runs
N. Lagoon	Central Shops	6	Lagoon	Four Mile
S. Lagoon	Central Shops	6	Lagoon	Pen Branch

[a] Adapted from Marter 1969.

High-Level Liquid Waste Storage

From the outset of operations at the SRS, special waste storage tanks were designed and installed to handle the huge quantities of liquid effluents that had activity levels too high for discharge to the seepage basins. Most high level radioactivity in liquid wastes was generated in the separations areas as a result of chemically processing the irradiated fuel from the reactor areas. The original storage tanks, eight in the F-Area and 4 in the H-Area, relied on gravity or steam-jet siphons for the transfer of radioactive liquids. Valves were not used so waste flow was directed to the appropriate tank by a change of pipe jumpers in diversion boxes located near the tanks. All piping was stainless steel and concrete trenches, which provided secondary containment, were equipped with leak collection and detection facilities. Some of the key highlights of the early waste handling history of the Site were described in the monthly and semi annual reports (Alexander and Horton 1956, Ashley 1967b, Du Pont 1958, Du Pont 1961-1962, Du Pont 1971). Some of the important events include:

- 1956 Four additional cooled, million-gallon waste storage tanks completed in the H-Area
- 1957 Four 1.3 million gal , uncooled tanks completed in F-Area
- 1958 Construction of four 1.3 million-gallon uncooled tanks started in H-Area; some leakage through steel tank wall in 3 H-Area tanks
- 1960 Operation of F-Area tank farm evaporator started
- 1962 Continuous inventory of fission products and alpha emitters in tanks begun
- 1963 Operation of H-Area tank farm evaporator started
- 1964 Total fission products in storage increased to 865 million Ci
- 1965 Total fission products in storage decreased to 745 million Ci; a fire in a tank ventilation filter due to an accumulation of vapor phase inhibitor; cesium removal ion exchange columns limits ^{137}Cs releases to seepage basins
- 1966 Total fission products in storage increased to 885 million Ci

- 1967 Total fission products in storage decreased to 851 million Ci; construction of four additional 1.3 million-gallon cooled waste tanks started in H-Area
- 1968 Total fission products in storage increased to 937 million Ci
- 1969 Total fission products in storage decreased to 798 million Ci; construction of 2 1.3 million-gallon cooled waste tanks started in F-Area
- 1970 Total fission products in storage decreased to 790 million Ci; several tiny explosions occurred at the H-Area tank farm evaporator; 7 of 36 cooling coils in one tank started leaking bringing he number of leaking coils to 17 and leaving 19 coils available for cooling.

Throughout this period, there was continual effort to reduce the volume of high-level effluent, usually by evaporation. From startup through about 1970, about 39 million gal of high and low level waste were sent to the tank farms in the F-Area and H–Area. Of that volume, 18 million gal remained in 1971, the other volume had been lost by evaporation and concentration of wastes (Ice 1971).

EFFLUENT AND ONSITE SAMPLING

Liquid effluents from the major release points at SRS were monitored from startup of operations at the SRS. This onsite effluent monitoring served to alert the Site personnel of problems that might be occurring and of the release of contaminants to the Site streams. A routine onsite monitoring program was established by 1952 and reports were issued routinely in the form of weekly, monthly and semiannual reports (For example, Du Pont 1953, Du Pont 1958).

Liquid Effluent Sampling Procedures

Water samples from the various reactor building effluent systems were monitored extensively to track levels of radioactivity leaving the facilities. Historically, the disassembly basins were the main source of radioactivity from the reactor areas to the Site streams, with some effluent contributed from the heat exchanger cooling water and the shield cooling water. To obtain gamma measurements from the disassembly basin flow to the Site streams in the early years, continuously operating ionization chambers were suspended in the liquid effluent stream of the weirs of the disassembly basin overflows and the gamma readings recorded. Water samples of the overflow from the disassembly basins to the streams were collected by six weir samplers located along the disassembly basins and canal during each shift at a designated time and recorded at approximately the same time as the gamma measurements were done (Du Pont circa1960).

In the 1950s, the heat exchanger cooling water was discharged by pipeline to an effluent ditch that drained to the Site streams. There was no continual monitoring of this discharged water; rather, periodic samples were taken through a hatch in the grating over the discharge tank in the 107 buildings at a designated time during each shift. The shield cooling water held in an underground tank and designated the 109 building, was located adjacent to the 105 building, the reactor building. The deionized water purged from the shield cooling systems was piped to the tank, The purged water contained corrosion products and fission and activation products from the reactors. The shield cooling water tank held up the water to permit radioactive decay of some

products and to allow settling of the heavier radioactive materials. Sampling was done during each shift through a hatch in the top of the overflow weir in the tank to allow for radioactive decay and to allow settling of materials.

In general, the Site stream and river water were analyzed for the same radionuclides as reactor effluent samples. Before the 1970s, suspended and dissolved solids were not separated for routine analyses. River flow was estimated from information provided by the USGS, while plant effluent stream flow measurements were supplied by the SRS Power Department. Table 5-5 provides a summary of the radionuclides in liquid effluents that were routinely monitored by the early 1960s.

In 1973, a major survey of the effluent and environmental monitoring program at the SRS found that overall improvements could be made and that the Site needed to make "substantial improvements in its capability to monitor nonradioactive releases because of new pollution control legislation" (Reinig et al 1973). Special attention was directed to ensure that representative samples were collected at the point of release. In general, effluents were continuously monitored at a major release point where releases may have been in excess of 10% of an operating guideline or have a potential for large unplanned releases. If the anticipated annual release from a specific emission point was less than 1% of the operating guideline then only grab sampling was the normal operating procedure. For some emission points, quantities released were calculated where emission were accurately established; for example the release of noble gases in irradiated fuels.

By the 1980s, all purges, or batch releases, from the disassembly basins were discharged to the reactor area seepage basins (from the P, L, and C Reactors) or to a containment basin (K Reactor). An in-line calibrated water meter measured the flow to the seepage basin during a purge. A sample was collected automatically every 3 minutes during a purge from the disassembly basin. The area survey health physicist removed the samples sent them to the Environmental Monitoring Laboratory for analyses at the end of each month. For process control, the Operating Department collected a sample every 4 hours during a purge and sent the sample to the 772-D Laboratory for gross alpha and beta, tritium and gamma pulse height analysis (Zeigler 1986). Meanwhile, continuous sampling of the effluent going to the process sewers was done. Every 2 minutes, a predetermined quantity, which was proportional to the expected average flow, was collected and a composite of the daily collection was analyzed. The Environmental Monitoring Laboratory analyzed the composite samples monthly for a number of radionuclides, including tritium, ^{137}Cs and ^{90}Sr.

During this same time, weekly grab samples were collected from the reactor heat exchanger cooling water and sent to the Environmental Monitoring Group for tritium analysis. The "high activity" cooling water gamma monitors had alarms in the central control rooms, but there were no alarms or monitors for tritium. The adequacy of weekly grab sampling was questioned although changes in tritium concentrations in such large volumes of water (approximately 180,000 gallons per minute) were said to occur very slowly (Zeigler 1986).

Table 5-5. Radionuclides Routinely Analyzed in Reactor and Separations Areas Effluents in 1962[a]

		Liquid Effluent	
Type of analyses	Radionuclide	Reactor to streams[b]	Separations to seepage basins[c]
Chemical separations	^{147}Pm	x	x
	^{91}Y	x	
	^{90}Sr	x	x
	$^{89,90}Sr$	x	x
	^{35}S	x	
	^{32}P	x	
	(Uranium and plutonium)		x
Gamma spectrometry	^{239}Np	x	
	^{233}Pa		x
	$^{141,144}Ce$	x	x
	$^{140}BaLa$	x	
	$^{134,137}Cs$	x	x
	^{131}I	x	x
	$^{124.125}Sb$		x
	$^{103.106}Ru$	x	x
	$^{95}ZrNb$	x	x
	^{65}Zn	x	
	$^{58,60}Co$	x	
	^{54}Mn	x	
	^{51}Cr	x	
Liquid Scintillation	^{3}H	x	x
Alpha Spectrometry	Alpha emitters[a]	x	x

[a] Adapted from Du Pont 1965; alpha releases from reactor areas were considered negligible; samples were counted for only gross alpha activity.
[b] For reactors, sampling frequency was varied according to fuel discharge and concentration of radionuclides in the disassembly basin. Flow estimates were made according to purge rates and the Ci release inventory based on flow and radionuclide concentration.
[c] In the separations facilities, liquid wastes were discharged to open seepage basins. Sampling volume was proportional to flow and was done continuously. Routine analyses were done on weekly samples.

Liquid effluent releases from the separations areas to streams were continuously sampled with a high-activity alarm in place in the 1980s. A paddlewheel sampler with a Brailsford battery operated pump was used at the H-Area and F-Area effluent discharge points. Samples were collected and analyzed weekly. The sample concentrations were combined with USGS flow data to determine release quantities. The volume in the H-Area effluent stream varied widely at times, causing sampler failure to occur. Releases from the M-Area to Tim's Branch were also continuously sampled with a flow-proportional (trebler) sampler in the effluent line and analyzed weekly. Only periodic calibrations of flow measurements were performed with the trebler and flow measurement calibration was noted as an area of uncertainty at that time (Ziegler 1986).

The Operations Department collected samples of the wastewaters that had been stored in tanks from the 420-D and 421-2D areas. The samples were analyzed for tritium and beta-gamma emitters. If the analyses were within the established limits, the water was released to Beaver Dam Creek (Figure 5-4). The quantity of water in each tank was determined from a sight glass reading before release to the creek. The area survey health physics team collected daily samples from the Brailsford pump sampler on effluents from 420-D and the process sewer downstream of all tritium rework facilities. There was no sampling point downstream from the Heavy Works facilities onsite so release estimates were based on measurements from the facilities. Because it was possible for radioactivity to be released to the effluent from routes other than the batch holding tanks, the uncertainties associated with tritium releases from the D-Area were much broader than those on Site streams where multiple sampling points were located (Marter 1969). On the other hand, the tritium releases from the D-Area were quite small when compared to releases from the reactor and separations facilities (See Figure 5-12).

Stream Sampling

Effluents from the processing and reactor areas onsite flowed into the five major Site streams, each described at the beginning of this chapter (Figures 5-3, 5-4, 5-5, 5-6, and 5-7). Water samples were collected at regular intervals from several locations on all Site streams, and at least one sample was taken above the location of the effluent discharge into each stream so increases in radioactivity could be detected (Horton 1954). The number of weekly water samples collected in this program increased from 384 samples during the first six months of 1953 (Albenesius 1954) to almost 1200 water samples by the end of 1954 (Horton 1955a). During the late 1950s about 950 water samples were collected in a typical six-month period from the onsite streams.

SRS initially took grab samples of streams and the Savannah River during the preoperational surveys and early monitoring programs. A 2 bottle siphon action apparatus was later used for continuous stream sampling. In 1958, a water wheel sampler was developed when line plugging and maintenance problems continued to occur with the siphon action apparatus (Johnson 1964). The water wheel continuously pumped a small volume through one system to a collection container. The second system was geared to pump at a rate of about 40–80 L wk^{-1} through the ion exchange column. The exact volume through the ion column was determined by multiplying the small volume collected in the first system by a known gear ratio of the pumps. Prefilters were placed before the ion column to prevent plugging with sediments.

The water sampling and monitoring methods for each stream depended upon the particular effluents they received. For example, paddlewheel samplers collected continuous samples of the effluents from the F-Area and H-Area that flowed into the Four Mile Creek. The anchored paddlewheel sampler floated in the stream, and collected samples in a small cup attached to the paddlewheel. As the stream flowed, the paddlewheel water was collected in the cup and emptied into a collection trough. The water flowed down the trough and into a collection bottle. The preferred paddlewheel samplers were also used on the streams at Road A, the final onsite sampling location for all streams before flowing into the Savannah River. On the other hand, effluents from the A and TNX areas were grab sampled and those from the M-Area were monitored with a trebler sampler. These latter areas did not contribute the levels of radioactivity that other facilities did (see Figures 5-12, and 5-13). Nevertheless, all Site streams were monitored routinely and fairly extensively from plant start up through the present day. Although

the actual sampling locations remained approximately the same over time, the designations for the sampling locations changed. Figure 5-10 shows the SRS site with major facilities, site streams, and roadways. While there were several sampling locations along each of the Site streams, the final sampling point onsite before the streams flowed into the Savannah River was at Road A. Table 5-6 summarizes the sampling location identification numbers from 1954 through 1980 on the five major Site streams (Upper Three Runs, Four Mile Creek, Pen Branch, Steel Creek, and Lower Three Runs), and their smaller tributaries. The table shows how the identification of sampling locations changed over the years, and, more interesting, is the fact that the number of sampling locations on Site streams at the SRS was greatest in the 1950s (24 locations), and decreased to less than half that number (10) by the mid-1970s.

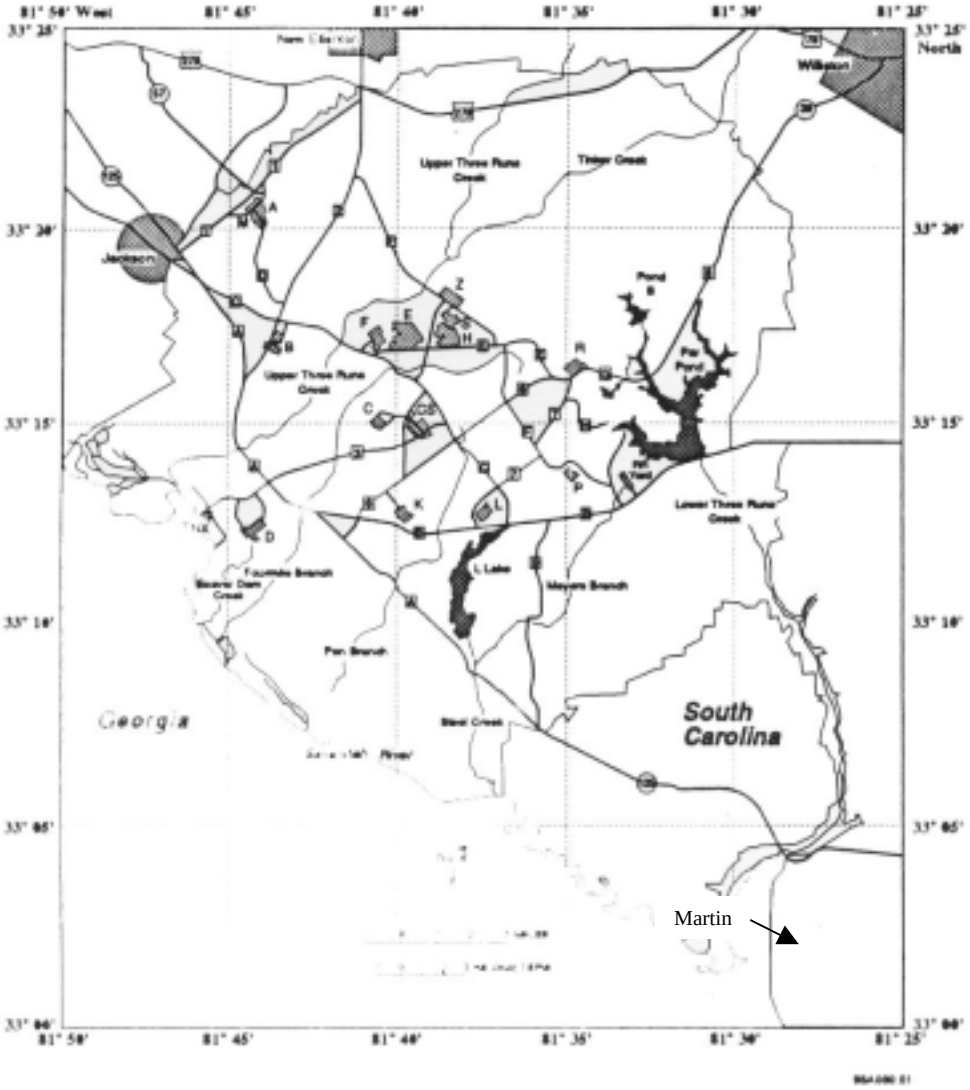

Figure 5-10. The SRS showing major facilities and roadways. The final sampling locations for Upper Three Runs, Four Mile Creek, Pen Branch and Steel Creek, before entering the Savannah River, were taken at Road A which becomes Highway 125 to the southeast.

Table 5-6. Identification Numbers for Stream Sampling Locations at the SRS[a]

		Sample location ID #							
Site stream	Physical Description	1954-1958	1959-1960	1961-1963	1964-1968	1969-1970	1971	1972	1973-1980
Upper Three Runs	**UTR at Road A**	**4**	**4**	**4**	**4**	**7**	**7**	**7**	**ns**
(UTR)	UTR at rail line	3	3	5	5	5	5	5	5
	Discharge stream -- F to UTR	2	2	2	2	2	2	2	2
	UTR at Tinker Crk	1	1	1	1	1	1	ns	ns
Tim's Branch	TB at UTR	5	5	ns	ns	Ns	ns	ns	ns
(TB)	Discharge stream - M/A to TB	3	3	4	4	4	4	4	ns
	N. offsite stream to TB	2	2	3	3	3	3	3	ns
Four Mile Creek	FMC at Fischer Pond Rd	7	ns	ns	ns	ns	ns	ns	ns
(FMC)	**FMC at Road A**	**6**	**6**	**10**	**11**	**12**	**12**	**12**	**6**
	FMC S. of C Reactor	5	5	ns	ns	ns	ns	ns	ns
	FMC NW of C Reactor	4	4	9	10	10	10	ns	ns
	FMC SW of F-Area	ns	ns	ns	ns	1	1	1	ns
	FMC at Road C (S of H-Area)	2	2	8	9	10	10	10	ns
	Discharge stream-F to FMC	3	3	7	8	8	8	8	ns
	Discharge stream-H to FMC	ns	ns	ns	ns	9	9	9	ns
	FMC at Tinker Crk	1	ns	ns	ns	ns	ns	ns	ns
Pen Branch	PB at SR swamp	3	3	11	12	13	13	ns	ns
(PB)	**PB at Road A**	**ns**	**ns**	**ns**	**ns**	**ns**	**ns**	**13**	**3**
	PB between K & L Reactors	7	ns	ns	ns	ns	ns	ns	ns
Indian Grave	Indian Grave Branch NW of K Reactor	2	2	ns	ns	ns	ns	ns	ns
Steel Creek	SC at PB confluence	ns	ns	ns	ns	ns	ns	ns	5
(SC)	**SC at Road A**	**4**	**4**	**12**	**13**	**14**	**14**	**14**	**4**
	Discharge stream-L to SC	3	3	ns	ns	ns	ns	ns	ns
	SC between L and P	2	2	ns	ns	ns	ns	ns	ns
Meyer's Branch	Meyer's Branch	1	ns	ns	ns	ns	ns	ns	ns
Lower Three Runs	LTR at Martin	ns	6	17	18	18	18	18	3
(LTR)	LTR at Stinson Bridge	3	ns	ns	ns	ns	ns	ns	ns
	LTR at Patterson Mill Bridge	ns	5	16	17	17	17	17	2
	LTR at upper E. branch	1	ns	ns	ns	ns	ns	ns	ns
	LTR at upper W. branch	2	ns	ns	ns	ns	ns	ns	ns
	Par Pond into LTR	na	4	ns	ns	ns	ns	ns	ns
	Par Pond into LTR-P branch	na	3	14	15	15	15	15	2
	Par Pond into LTR-R branch	na	2	15	16	16	16	16	3
Total Locations		24	22	16	16	18	18	16	10

[a] Adapted from Du Pont 1965 and Du Pont 1973.
ns = not sampled
na = not applicable, Par Pond was constructed later

DOCUMENTATION OF RELEASES

For surface water source term development, release estimates are calculated for the key radionuclides that are released offsite. For surface water, offsite means the point that the effluents drain into the Savannah River. Unlike atmospheric source term development where releases are estimated from the point of release from a particular facility into air, the liquid effluents are released from facilities to seepage basins or to onsite streams.

To reconstruct the releases to surface water, we have used the most original and basic information available. The most basic data for surface water releases are the original weekly results handwritten from the laboratory ledger books and retained at the SRS on aperture cards (which are similar to microfiche). *Radiological Assessments Corporation (RAC)* copied the measurement data for air, water, and environmental monitoring that were needed to complete this study. These references are called aperture card printouts throughout this report. Another useful information source for surface water source term development was the experimental notebooks of Clarice Ashley (Ashley 1959), in which she compiled analytical results of the surface water releases. Ashley tracked the results of the weekly stream flow and concentration measurements for nonvolatile beta, tritium, ^{131}I, strontium, and cesium at key sampling locations on Site streams. We corroborated the experimental notebooks of C. Ashley with the handwritten entries in the aperture card printouts.

These handwritten entries provided radionuclide concentration, flow rate data and some quality control information for the major onsite streams. The Site used these data to calculate and report release estimates as total activity in curies (Ci) in the appropriate monthly report. In the early years, the radioactivity in surface streams was usually reported as nonvolatile beta, and tritium. Beginning in the late 1950s specific radionuclides, like ^{137}Cs and ^{90}Sr, were measured at the facility discharge points, in the Site streams and in the Savannah River. For our work, we calculated monthly averages from these weekly data and compared the results to the values reported in monthly reports to check the validity and credibility of monthly or other summary types of reports. The monitoring data of radionuclides in the Site streams for the key radionuclides have been compiled in the Water Releases Excel workbook. In the electronic version of this document, double-clicking on the following hyperlink provides access to this workbook: Water Releases.xls.

Figure 5-11, which shows this type of comparison for 1962, supports quite good agreement between the two approaches. The calculated monthly totals from the weekly values reported in the ledger sheets give a better view of when the releases actually occurred. With the routine monthly reports, the releases were reported in the month after they occurred (e.g. February and May 1962) (Du Pont 1962). This most likely happened because the monthly reporting period was from the middle of one month to the middle of the month in which the report was issued. For the annual totals however, the values are very close. In some cases, we used these summary reports when more detailed daily or weekly data were not located.

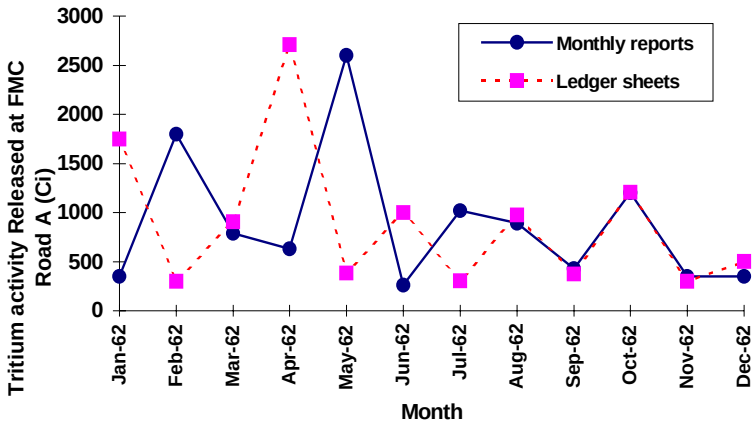

Figure 5-11. Comparison of monthly totals of tritium measured in Four Mile Creek at Road A in 1962. The solid line represents Site totals reported in monthly reports, and the dotted line shows the monthly totals calculated from weekly values in the handwritten ledger sheets. This sampling location at Road A was the final monitoring point on the Four Mile Creek before it flowed into the Savannah River.

Several complete series of monthly reports, as well as some semi-annual and annual reports for this period provided additional information for our source term reconstruction work. The monthly reports of most value were the: *Environmental Monitoring Monthly Report. Radiological Control and Methods* series (For example, Du Pont 1962, Du Pont 1964a, Du Pont 1964b, Du Pont 1967). For 1962 to 1964, these reports were each 25 to 30 pages in length, and provided quite thorough summaries of effluent and environmental monitoring for both airborne and waterborne releases. Related to surface water releases, these summaries report the liquid waste releases to separations area seepage basins for the nonvolatile beta emitters: ^{103,106}Ru, ^{89,90}Sr, ^{95}Zr-Nb, ^{137}Cs and ^{141,144}Ce; gross alpha, ^{3}H, and radioiodine. Furthermore, the average levels of activity of nonvolatile beta, radiostrontium, radiocesium, radioiodine, and tritium were reported for the onsite streams: Four Mile Creek, Pen Branch, Steel Creek, Lower Three Runs, and Par Pond. Radioactivity levels in these streams reflected discharge operations in the separations areas and reactor areas. Periodically, measurements of ^{35}S released to Four Mile Creek were reported. These documents also provided estimates of radioactivity measured in the Savannah River upstream and downstream of the discharge points from the Site. Estimates of total levels of nonvolatile beta, tritium ^{90}Sr, and ^{35}S were routinely reported.

RANKING THE RADIONUCLIDES RELEASED TO SURFACE WATER

Chapter 3 of this report describes the rationale and process used to determine the radionuclides released to water and air from the SRS that historically were the most important contributors to offsite dose. Some radionuclides were relatively more important to offsite residents than others; therefore it was important to focus our efforts on more important radionuclides so resources of the study were used most effectively. The relative importance of releases of these radionuclides to the environment depends upon the quantities released, differences in the potential for nuclide concentration in the environment, and the relative toxicity

of the radionuclides (as measured by their dose conversion factors). An updated methodology developed by the National Council on Radiation Protection and Measurements (NCRP) (NCRP 1996) was used to assess the relative importance of the identified radionuclides as potential contributors to offsite radiation dose.

For surface water pathways, the radionuclides that contribute 0.1% of the total screening radiation dose (our screening criterion) are ^{137}Cs, ^{60}Co, ^{3}H, ^{129}I, ^{131}I, ^{32}P, ^{238}Pu, ^{239,240}Pu, ^{89}Sr, ^{90}Sr, ^{35}S, ^{99}Tc, uranium, ^{91}Y, ^{65}Zn, and ^{95}Zr,Nb. The results of the NCRP screening for radionuclides in surface water showed that ^{137}Cs was the major contributor to offsite dose (~75% of the dose), with ^{60}Co, tritium, ^{131}I, ^{32}P, and ^{90}Sr jointly contributing about another 20%). Sulfur-35 and ^{65}Zn together contributed less than 4% to the screening dose via the surface water pathway, and all other radionuclides meeting our screening criteria (^{129}I, ^{239,240}Pu, ^{99}Tc, uranium, and ^{95}Zr,Nb) together contribute less than 2% to the screening dose. Refer to Chapter 3 for full details of the screening analysis.

In addition to ranking individual radionuclides for their relative importance to dose, we also evaluated the relative importance of the facilities at the SRS for their contribution to radioactivity in the Site streams. The monitoring data clearly indicate the facilities that historically contributed the highest levels of radioactivity in liquid effluents and to Site streams.

To focus our efforts on the key radionuclides and facilities, we compared the relative contributions of the major facilities to the total levels of tritium, total alpha and beta activity discharged to Site streams. Figure 5-12 summarizes results of measured tritium and beta activity discharged to Site streams from 1965 to 1969 and indicates clearly that the reactors and reactor areas are the largest contributors to total tritium and beta activity released to Site streams (Marter 1970). Beta activity represents a number of radionuclides that can be categorized as fission products and activation products. Important fission products from reactor operations at the SRS are ^{137}Cs, ^{90}Sr, and ^{131}I. Roughly 40% to 60% of the tritium and beta activity in liquid waste came from the reactor effluents. Another 30 to 40% originated from the seepage basins in the reprocessing and reactor areas. The Heavy Water Facility in D-Area contributed about 10% of the tritium activity in the liquid effluent waste stream. Figure 5-13 shows the results for alpha activity discharged in liquid effluents (Du Pont 1966, Du Pont 1967, Du Pont 1968, Marter 1970).

SOURCES OF UNCERTAINTY FOR RELEASES OF RADIONUCLIDES TO THE SAVANNAH RIVER

A number of factors and processes can affect the quantity of radionuclides that are released in liquid effluents from the reactor, separations and miscellaneous areas and eventually are released offsite into the Savannah River. As a result, a number of factors must be considered in developing the surface water source term with uncertainty estimates. Some of the factors include:

- Sampling and analytical procedures
- Properties of the radionuclide being evaluated
- Volume of liquid effluent released from the facility
- Holdup time in seepage basin, if applicable
- Distance between the seepage basin and the closest stream
- Velocity of groundwater movement where groundwater outcrops to surface streams
- Distance between Road A (the final sampling point onsite)and the river

- Adsorption distribution coefficient for radionuclides in soil (for seepage basins and the SRS swamp)
- Hold-up time in the Savannah River swamp.

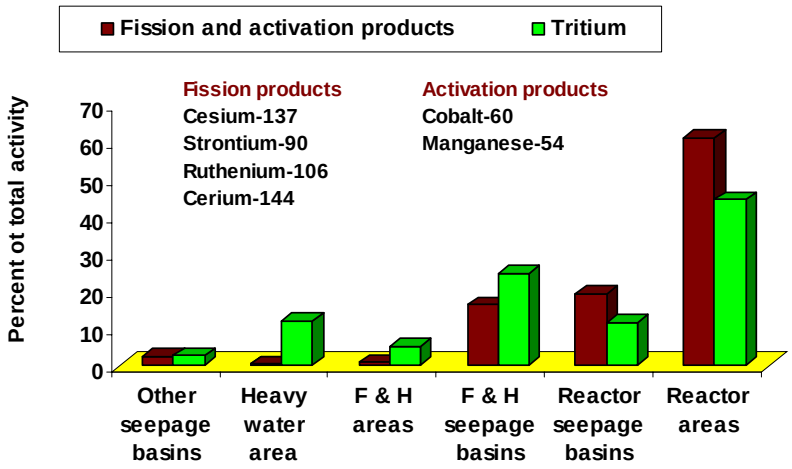

Figure 5-12. Distribution of tritium and beta activity (fission and activation products) in liquid wastes from six sources onsite. These percentages were based on operations in 1965–1969 (Du Pont 1966, Du Pont 1967, Du Pont 1968, Marter 1970).

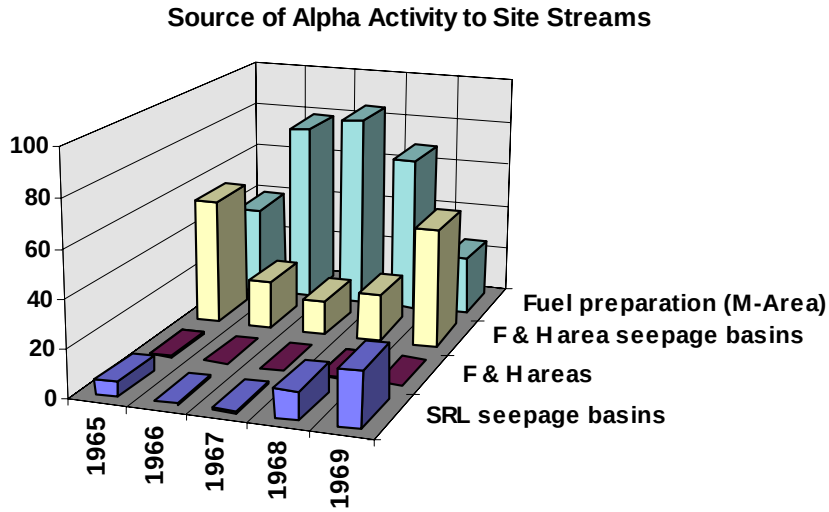

Figure 5-13. Sources of gross alpha activity in liquid wastes from six areas onsite. These percentages were based on operations in 1965–1969; however, in general, they reflect the pattern at other times as well. Roughly 60% of the alpha activity in liquid waste came from the M-Area. Another 30% originated from the F-Area and H-Area seepage basins and less than 10% came directly from the F-Area and H-Area and from the Savannah River Laboratory seepage basins.

For surface water source terms, we provided release estimates for the key radionuclides that were released offsite, which means the point that the effluents drain into the river. Unlike atmospheric source term development where releases are estimated from the point of release from a particular facility into air, the liquid effluents were released from facilities to seepage basins or to onsite streams, and still well within the site boundaries. Using effluent data measured directly at the point of release from the facilities is not appropriate if the goal is to provide estimates of release with uncertainty to the offsite environment where the public would have access to the surface water. This point of pubic access is the Savannah River and along the Lower Three Runs Creek to the southeast. Using measurements of radionuclides at points of release from the facilities is not suitable because the onsite streams flowed several miles before reaching the river.

An example of an ideal situation for surface water source term development might be having liquid effluents discharged from the facilities through a pipe directly into the Savannah River. In that case, measurements taken at the effluent release point to the river would be well suited for estimating releases from the site before any dilution occurred in the river. In the actual situation at the SRS, radionuclides were routinely measured onsite at several locations along each stream and then at several locations in the river. For the source term work at SRS, we reasoned that the last measurement point onsite would be the best data to use as the starting point. This location was at Road A where it intersected each stream. At these locations, a large quantity of data was available historically for all site streams. Measurements at several locations along Lower Three Runs had also been reported over the years. We used the data obtained at the Road A locations (final sampling point before discharge from the Site) on the five Site streams as the basis for our source term development. However, it was clear that some of the factors that might affect the release estimates that are listed above had not been evaluated quantitatively. The hold-up time in the swamp, and sampling and analytical procedures were the major components of the uncertainty analysis. The Water Releases workbook contains the details of the uncertainty calculations.

Effect of the SRS Swamp

The fact that the final measurement points of radionuclides in surface streams were located east of the Savannah River Swamp has important implications for the computation of the surface water source terms. Several studies help to understand the effect the swamp has on the discharge of certain radionuclides to the river. Beyond Road A, the onsite streams can meander for 4 to 5 mi or more onsite before discharging into the Savannah River. Most importantly, it is necessary to evaluate the impact of the Savannah River Swamp on retaining certain radionuclides, at least for a period of time, and thereby, changing the rate they move offsite into the Savannah River. On the other hand, periodic flooding of the swamp also affects the levels of radionuclides that are flushed from the swamp into the river.

Some of the key elements we considered and incorporated into the uncertainty estimates for releases to surface water are:

- The SRS swamp borders the Savannah River for about 16 km (10 mi) and averages about 2.4 km (~1.4 mi) wide
- The SRS swamp flooded about 20% of the time
- Elements are absorbed or retained by soils differently; a measure of this retention in soils is called the K_d value (See Table 5-7).

Clearly, the onsite streams and swamp play an important role in receiving, transporting, removing and diluting materials before they are released offsite in the Savannah River. Immobilization in soils plus radioactive decay reduce the amount of some contaminants entering the river. The Site conducted various studies on the manner in which the SRS swamp might act to remove and/or hold certain radionuclides for a period of time, or to prevent them from entering the Savannah River soon after being released from processing areas onsite. For example, the absorption distribution coefficient, or K_d value, is a measure of how materials are retained in soils. In one study at the SRS, K_d values varied from 50 mL g^{-1} for ^{90}Sr to 1000 mL g^{-1} for ^{137}Cs to 100,000 mL g^{-1} for ^{239}Pu (Marter 1969). Table 5-7 summarizes the key elements of that study. For soluble radionuclides, like tritium, the SRS swamp does not have a great impact on the levels released offsite; however, for more insoluble materials, like ^{137}Cs, the swamp influences the calculations of release estimates a great deal.

Table 5-7. Fraction of Radionuclides Released to Fuel Reprocessing Area Seepage Basins Reaching Four Mile Creek[a]

Isotope	Half life (y)	K_d at pH 1[b] mL g^{-1}	Fraction reaching creek from F-Area	Fraction reaching creek from H-Area
^{3}H	12.3	0	0.25[c]	0.4[c]
^{60}Co	5.2	100	0.1	0.5
^{90}Sr	27.7	50	0.7	0.9
^{106}Ru	1	800	1×10^{-16}	1×10^{-9}
^{134}Cs	2.1	1000	1×10^{-5}	0.01
^{137}Cs	26.6[e]	1000	0.2	0.7
^{147}Pm	2.5	100	0.01	0.3
^{239}Pu	24,000	10,000	0.9	0.9

[a] Selected from Marter (1969); partial listing of most significant radionuclides.
[b] Adsorption distribution coefficients reported in Marter (1969).
[c] Assuming 50% loss by evaporation.
[d] Through 1969, the Site reported that the only radionuclides reaching surface streams from the Fuel Reprocessing Area Seepage Basins were ^{3}H and ^{90}Sr. All other radionuclides shown, with the exception of ^{239}Pu, have been detected in ground water in the vicinity of the basins.
[e] The half-life of ^{137}Cs is 30.0 years (Schleien 1992).

Researchers at the Site carried out a number of studies to estimate the impact of the swamp, sediment retention and decay on the quantities of radionuclides that actually reached the river. They inferred that the advantages of seepage basin disposal over direct disposal to surface streams were that some ionic species were immobilized by the soil while others were greatly delayed in movement. They indicated that immobilization plus radioactive decay greatly reduced the amount of radioactive material eventually leaving control of the SRS (Evans et al 1968).

Retention in the streams and swamp

Radionuclide monitoring was quite extensive in the Site streams above the Road A location, but there were sparse measurements made between the Road A location and the Savannah River. Furthermore, the flow of each stream through the swamp is unique and not well understood. The onsite streams meander for 3–12 km (2-8 mi.) onsite before reaching the Savannah River Swamp System, which borders the Savannah River for about 16 km (10 mi) and averages about 2.4 km (1.5 mi) wide. Effluents from the Site flow through the swamp before discharging offsite into the Savannah River and are affected by seasonal variations water levels (Westinghouse 1996). Flows in the Savannah River are highest in the winter and spring, and lowest in the summer and fall. For these reasons, a simpler model was a more appropriate alternative than a complex model with more unknown parameters.

We chose a simple model to calculate the concentration of dissolved radionuclides and radionuclides attached to suspended and bottom sediments in the swamp (Jirka et al. 1983) because of the limited measurements of radionuclide in the surface water and sediments of the SRS swamp available historically. The model uses an unsteady, one-dimensional, liquid pathway submodel to calculate temporal and longitudinal distribution of dissolved radionuclide concentration as well as the concentration of radionuclides attached to suspended and bottom sediments of various sizes. The dissolved radionuclide concentration at a given location is found by applying the mass conservation equation with radioactive decay:

$$C_{x,t} = \frac{1}{Q_{x,t}} Q_{(x-\Delta x,\, t-\Delta t)} C_{(x-\Delta x,\, t-\Delta t)} e^{-\lambda \Delta t} \qquad (5\text{-}1)$$

where

$C_{x,t}$	=	dissolved radionuclide concentration at location x and time t,
$Q_{x,t}$	=	flow rate at location x and time t,
λ	=	radionuclide decay
$\Delta_{x,t}$	=	travel time in stream (s)
Δx	=	change in location (m)
x	=	distance (m)
t	=	time (s)

The travel time in the stream ($\Delta_{x,t}$) is the time it takes the radionuclide to move from its last point of measurement (e.g., Road A) to its entry into the Savannah River. For example, for Four Mile Creek this distance is about 12 km or 12,000 m. The travel time depends upon the flow rate and velocity of the particular stream. The final activity of the radionuclide entering the Savannah River is affected by the radioactive decay that might occur during this travel time.

The concentration of radionuclides attached to sediment (C_p) is calculated from the known dissolved radionuclide concentration and the distribution coefficient (K_d). The distribution coefficient, K_d is a measure of the amount of radionuclide sorbed on sediment.

$$K_d = \frac{\text{amount of radionuclide sorbed on sediment}}{\text{amount of radionuclide left in solution}} \qquad (5\text{-}2)$$

The K_d value for each radionuclide depends on various parameters, including sediment type and concentration, the flow characteristics, and water quality of the surface water and the contact time (Whicker and Schultz 1982). Table 5-8 gives the range and median values that have been reported in the literature for the elements of interest for releases from the SRS. In our source term calculations we assumed a lognormal distribution of K_d values for each element with the given median estimate and the range as the 5th and 95th percentiles of that distribution. We applied these equations to our source term calculations for releases of cesium and strontium in surface water releases from the SRS.

Table 5-8. K_d Values for Elements in Freshwater[a]

Element	Range (mL g^{-1})	Median (mL g^{-1})
Cesium	50–80,000	10,000[b]
Cobalt	1,000–71,000	5000
Iodine	0–75	10
Plutonium	100–10,000,000	100,000
Strontium	8–4000	100
Tritium	0	0

[a] From Jirka et al. 1983.
[b] Studies with cesium show an increase in K_d values with contact time; we assume a median value of 10,000 because of the longer contact period in the SRS swamp than in a flowing stream (Whicker and Schultz 1982).

In the following section, we illustrate the method using an example from 1961 for ^{137}Cs releases in four Mile Creek. This example uses the following assumptions and measurement data:

- Average concentration of ^{137}Cs at Road A in Four Mile Creek was about 10 pCi L^{-1} (see Table 5-11).
- The distance from the Road A location on Four Mile Creek to the Savannah River is about 12 km (12,000 m).
- At the Road A location, the average flow rates ranged from 270-490 cfs, with an average of 325 cfs or 9.2 m^3 s^{-1}. The SRS facilities contributed significant flow to the Site streams, as noted previously. For Four Mile Creek, the average flow rate upstream of the SRS effluent discharge point is approximately 0.35 m^3 s^{-1}. During the periods of intense operations at the Site, the flow rate in Four Mile Creek was increased to 10 m^3 s^{-1} ± 3 m^3 s^{-1} (340 ±90 cfs) at the Road A location. By about 1987 the flow rate in Four Mile Creek at Road A had returned to its historic flow rate of 0.5 m^3 s^{-1}.
- The average velocities varied from 0.03 to 0.08 m s^{-1} from the late 1950s through the mid 1960s (see Hydrology section). The average velocity in Four Mile Creek in the late 1950s through the mid 1960s was about 0.06 m s^{-1}.
- The half-life of ^{137}Cs is 30.0 y, so the decay rate (λ) is 7.3 x 10^{-10} per second (equal to (ln 2)/ half-life).
- For cesium, we assume a median K_d value of 10,000 mL per g^{-1} (Table 5-8) (Jirka et al. 1983).

Thus, in the early operational years, the travel time in Four Mile Creek,

$$\Delta t = x/t$$
$$= 12{,}000 \text{ m}/0.06 \text{ m s}^{-1} = 200{,}000 \text{ s (or ~55 hr)} \quad (5\text{-}3)$$

So the dissolved ^{137}Cs concentration 12 km downstream from Road A is calculated using equation (5-4):

$$C_{12\ km,\ FMC} = \frac{1}{(9 \text{ m}^3 \text{ s}^{-1}) + (0.35 \text{ m}^3 \text{ s}^{-1})} (9 \text{ m}^3 \text{ s}^{-1})(10 \text{ pCi L}^{-1}) e^{-7.3 \times 10^{-10} \text{ s}^{-1}\ 200{,}000 \text{ s}} \quad (5\text{-}4)$$

$$C_{12\ km,\ FMC\ water} = 9.6 \text{ pCi L}^{-1}$$

The results of this calculation show that the radioactive decay of ^{137}Cs as it travels the 12 km downstream plays a minor role in removing ^{137}Cs from the source term. The concentration of radionuclides attached to sediment (C_p) is then calculated from the known dissolved radionuclide concentration and the distribution coefficient (K_d) by:

$$C_{px,t} = K_d \times C_{x,t}. \quad (5\text{-}5)$$

$$C_{p\ 12\ km\ FMC} = 10{,}000 \text{ mL g}^{-1} \times 9.6 \text{ pCi L}^{-1} = 96 \text{ pCi g}^{-1}$$

The sediment transport rate, S_T, is found by knowing the flow rate and using constants for the particular sediment (Jirka et al. 1983):

$$S_T = aQ^b \quad (5\text{-}6)$$

The parameters, *a* and *b*, are constants that are estimated for each sediment size range. We assume that a = 0.0004 and b = 3, based on studies at the SRS (Ruby et al. 1981). Then the sediment transport rate becomes:

$$S_T = 0.0004\ (9 \text{ m}^3 \text{ s}^{-1})^3 \quad (5\text{-}7)$$
$$S_T = 0.29 \text{ g s}^{-1}$$

If we assume that the sediment carries all the particulate ^{137}Cs, then the sediment carries:

$$S_T \times C_{px} = 0.29 \text{ g s}^{-1} \times 96 \text{ pCi g}^{-1} = 28 \text{ pCi s}^{-1} \quad (5\text{-}8)$$

And the rate of dissolved ^{137}Cs carried downstream is:

$$C \times Q = 9.6 \text{ pCi L}^{-1} \times 9 \text{ m}^3 \text{ s}^{-1} \times 1000 \text{ L m}^{-3} = 86{,}400 \text{ pCi s}^{-1} \quad (5\text{-}9)$$

Finally, the total amount of ^{137}Cs being transported in FMC is the sum of dissolved and particulate ^{137}Cs so that

$$28 \text{ pCi s}^{-1} + 86{,}400 \text{ pCi s}^{-1} = 86{,}428 \text{ pCi } ^{137}\text{Cs s}^{-1} \quad (5\text{-}10)$$

With the release rate of 86,400 pCi s^{-1} for ^{137}Cs in Four Mile Creek water to the Savannah River, then over the period of a year, the total release to the river would be 2.7 Ci. The ^{137}Cs activity in sediment during that same year would be 880 mCi or 0.00088 Ci. This example does not incorporate the uncertainty distributions, which our source term estimates do. This model was applied to the estimation of annual release estimates for ^{137}Cs and ^{90}Sr. As described in the following sections, other sources of uncertainty were also considered.

Periodic flooding of the swamp

The SRS Swamp flooded about 20% of the time (74 days per year on the average) during 1958 to 1967. Historically the river overflowed its channel and flooded the swamps bordering the Site when its elevation rose higher than 27 meters above mean sea level at the SRS Boat Dock. This level corresponds to flows equal to or greater than about 438 cubic meters per second (or, about15,500 cfs). We have assumed that additional releases to the Savannah River occurred from the swamp when flooding occurred. We treat this uncertainty as a source of bias and increase releases for radionuclides that were retained in the swamp, like cesium and strontium, by 20% (with a range of 10%-30%) for most years. Because detailed records exist for rainfall amounts, this percentage varies. For years with very high rainfall amounts like 1964 and 1971, we assume a value of 40% (with a range of 25%- 60%. For years with low rainfall, we assume the swamp flooded only about 10% of the time (with a range of 5%-15%).

Sampling and Analytical Procedures

Sampling and analytical uncertainties are important to consider for all key radionuclides, but they are especially important for radionuclides, like tritium, that are not impacted heavily by flow through the SRS swamp. In this case, the sampling and analytical uncertainties are the major sources of uncertainty in the release estimates for tritium in surface waters. Nevertheless, the uncertainty associated with the release estimates must be considered for any radionuclides in surface water from the analytical errors in measurement of flow, and in sampling and determination of the radionuclide concentrations in the water. Appendix A describes the analytical and sampling procedures in detail.

Generally, there were differences of 10% or less in the volume going into the Site streams from the reactor basins. It appeared that the effluent volume to the Site streams was monitored reasonably well by both the Site and the USGS (Ashley 1959, 1964, 1967). Estimates of error for the routine concentration measurements varied with the radionuclide, the sample preparation and with the counting procedure. Nevertheless, SRS provided extensive documentation of its health physics sampling and analytical procedures over the years, both in standard procedures manuals

(Du Pont 1953, 1959, 1989) and also in frequent memoranda and interim reports (Albenesius and Meyer 1962, Ashley 1967a).

Table 5-9 summarizes the various factors that were considered in calculating revised release estimates of releases of tritium, ^{137}Cs, and ^{90}Sr in liquid effluents to Site streams that eventually entered the Savannah River. Several time periods are considered because of changes in the sampling procedures and analytical methods for the radionuclides. In addition, the retention within the swamp varied among the radionuclides and those factors were considered, as described above.

Table 5-9 Factors Considered in the Uncertainty Estimates for Surface Water Source Terms for Key Radionuclides

Radionuclide	Time period	Measured effluent release	Measured in stream at Road A	Estimated measurement uncertainty	Effect of swamp Kd values[a]	Effect of swamp Flooding
Tritium	1954-1957	yes	no	50%	none	none
	1958-1959	yes	yes	40%	none	none
	1960-1973	yes	yes	25%	none	none
	1974-1992	yes	yes	15%	none	none
^{137}Cs	1954-1958	no	no	60%	10,000	1.1 to 1.4[b]
	1959-1973	yes	yes	50%	10,000	1.1 to 1.4[b]
	1974-1992	yes	yes	25%	10,000	1.1 to 1.4[b]
^{90}Sr	1954-1960	some	no	60%	100	1.1 to 1.4[b]
	1961-1973	yes	yes	50%	100	1.1 to 1.4[b]
	1974-1992	yes	yes	25%	100	1.1 to 1.4[b]

[a] The median estimate is listed; the range of K_d values is: cesium (50–80,000) and strontium (8–4000).

[b] Based on rainfall amounts for each year; in general the swamp flooding about 20% of the time each year when the average rainfall was about 47 inches. 1964 had very heavy rainfall (73 in) and extensive flooding occurred, while 1954 was quite dry (28 in) with little swamp flooding.

TABULATION OF RELEASES

Releases of Beta-Gamma Emitters

Chapter 3 of this report describes our methods of selecting and ranking the key chemicals and radionuclides for in-depth source term development. Section 3.2 describes the results of our screening and ranking of radionuclides using the NCRP screening methodology (NCRP 1996). The results of that process showed that the for releases to surface water, the radionuclides, ^{137}Cs, tritium, ^{90}Sr, ^{60}Co, ^{32}P, and ^{131}I accounted for about 95% of the screening value to people living near the SRS. All of these radionuclides decay by beta emission. Furthermore, ^{137}Cs and tritium dominate the dose contributions for most major pathways. For that reason, we have focused most effort in the following sections on reconstructing releases of the beta emitters, tritium, ^{137}Cs, and ^{90}Sr. Release estimates are also described for ^{60}Co, ^{32}P, ^{131}I and uranium, which decays by alpha emission.

Tritium Release Estimates

The key processes potentially leading to tritium releases at the SRS are the:

- Reactor operations at the reactor facilities
- Recovery of transuranic elements in the separations facilities
- Recovery of tritium in the tritium facilities
- Laboratory research area
- Heavy water rework facility.

Tritium as a site product for SRS was created in the reactors by neutron capture in lithium-6 in the lithium/aluminum targets and control rods. This capture produced tritium in a concentrated form, which was then extracted and packaged for use for the weapons program. Most releases of this tritium product occurred as releases to the air during the extraction and packaging operations in the Tritium Facilities located in H-Area and constituted the majority of atmospheric releases from the SRS, ranging from 200,000 Ci to 520,000 Ci per year (see Chapter 4.1)

Tritium was also produced in SRS reactor moderator by activation of deuterium. The concentration of tritium in the heavy water was a function of the neutron flux in the reactor and the length of the irradiation, with the tritium concentration in the moderator building up slowly over the years. Some of this tritium was lost to air and to liquid effluents by evaporation of moderator leaks and by the carry over of tritium oxide on fuel and target elements during reactor discharge. Releases of tritium to air from each reactor were as high as 60,000 Ci per year depending on the tritium concentration in the moderator, roughly 10% to 25% of tritium releases to air from the Separations Area.. Direct releases of tritium to air from routine 400-D operations were about up to 2500 Ci per year (compared to about 200,000 Ci to 520,000 Ci of tritium from the Separations Area).

Direct releases of tritium to plant streams and the Savannah River from all SRS reactor areas ranged from about 5000 Ci in the 1980s up to 100,000 Ci in the 1960s. Tritium releases directly from the 400-D facilities to Beaver Creek Dam from the 400-D operation have ranged from about 2500 Ci in the 1980s to about 6000 Ci in the 1960s. Our approach to determining these overall release estimates for tritium is the topic of this section.

Tritium was released to onsite streams from numerous activities, but the largest quantities were primarily from reactor operations and associated activities (Figure 5-12). Tritiated water was released in liquid effluents directly to onsite streams and to the seepage basins, and from outcropping into streams from burial ground releases and shallow groundwater aquifers. Continuous proportional samples were taken of water leaving the separations areas to the seepage basins and to Four Mile Creek. A fraction of the composite sample was analyzed weekly with Beckman Model liquid scintillation counters having a tritium sensitivity of 300 pCi L^{-1} (± 10%) and calibrated with National Bureau of Standards samples (Jacober et al. 1973). Cooling water discharged to seepage basins and to the streams from the tritium facilities was also monitored but results showed very low concentration of tritium compared to other facilities onsite.

Releases of tritium from the reactor areas to onsite streams were monitored daily since the mid 1950s, and reported in Site documents such as aperture card printouts of weekly volume and concentration measurements of tritium. Table 5-10 summarizes the onsite tritium monitoring of releases to onsite streams and seepage basins through the 1970s. The Site made progress on

increasing the sensitivity of the tritium analysis, especially for large volume samples. Appendix A of this report contains a thorough description of these studies.

The basic tritium analytical procedure used the "tri-carb" liquid scintillation counter, which was placed in operation by June 1958 (Du Pont 1958). (See Appendix A). By September 1962, the analytical procedure used for tritium analysis was fairly well established at the SRS. For water samples, an initial detection limit of 5000 pCi L^{-1} with a counting time of 30 minutes was established. By the 1980s, liquid scintillation counting was still used for tritium determination in water but the recoveries were near 100% and the sensitivity or limits of detection were from 300 to 1000 pCi L^{-1} (Du Pont 1989).

Using these methods, tritium concentrations in water samples were determined from approximately 20 locations in onsite streams. There was extensive monitoring of tritium from facilities and at numerous locations in the Site streams. Tritium concentrations in the Site streams and streams flow rates were measured on a weekly basis beginning in 1959. These values were combined to provide estimates of tritium activity (in Ci) that were reported weekly or monthly.

Table 5-10. Tritium Monitoring of Liquid Effluent Releases at the SRS through the Mid-1970s[a]

Area	Emission point	Type and frequency of monitoring
Reactor areas	Disassembly basin	Once per day
	Thermal shield	Grab sample
	Heat exchanger	Once per week or more often
	Moderator spills	As they occurred
	Distillation cooling water	Daily
Separations areas		
To seepage basins	234-H cooling water	Grab sample weekly
	232-G lab drain	Grab sample weekly
	232-H floor drain	Grab sample when releases occurring
To streams	232-H cooling water	Grab sample weekly
	232-H floor drains	Grab sample weekly
	238-H cooling water	Grab sample weekly
	238-H floor drains	Grab sample weekly
	234-H cooling water	Grab sample weekly
	236-H floor drains	Grab sample weekly
Savannah River Laboratory	776-A low level system hold tank	Monthly composite sample analyzed
Heavy Water Plant	772-D	Continuous
	420-D waste water	Each drain sampled and analyzed
	421-2D	Hold tank grab sample

[a] Adapted from Towler (1980).

Correlation of Radionuclide Releases with Reactor Power Levels. The radionuclides released to air and water from the SRS resulted primarily from operations of the five reactors and separations areas. For times when radionuclides were not routinely measured in the effluent, it is reasonable to suppose that releases of some radionuclides could be correlated with power levels

or production operations at these facilities. For example, because radionuclides released to surface water originated primarily from the reactors, it seemed reasonable to correlate releases of radionuclides that result from reactor operation (such as fission and activation products) with the duration and power levels of each reactor. This correlation between power levels and measurement values could form the basis of a method to calculate releases of particular radionuclides that were not measured during certain time periods, especially in the 1950s. [The power levels for the five operating reactors at the SRS were compiled for all years of operation. Refer to Chapter 2, *SRS Facilities: History of Processes and Operat*ions, for details.]

Figure 5-14 shows the variation with time of the reactor power levels and the total tritium activity measured at Road A in the Site streams. The reactor power levels decline from about 250 to 300 gigawatt days (GWd) in the early 1960s to about 100 GWd in by 1970. Tritium losses to the Site streams averaged about 500 to 7000 Ci mo^{-1}; however, they varied widely with releases exceeding 20,000 Ci in February 1963 and in May, June and August 1964. This comparison shows that the total tritium activity measured at Road A in the Site streams does not correlate positively with reactor power levels. The Site noted that releases of radioactivity to the reactor area seepage basins and the Site streams were not easily correlated to operating time periods because many releases of liquid effluents occurred from the storage basins when the reactors were not operating (Evans et al. 1992).

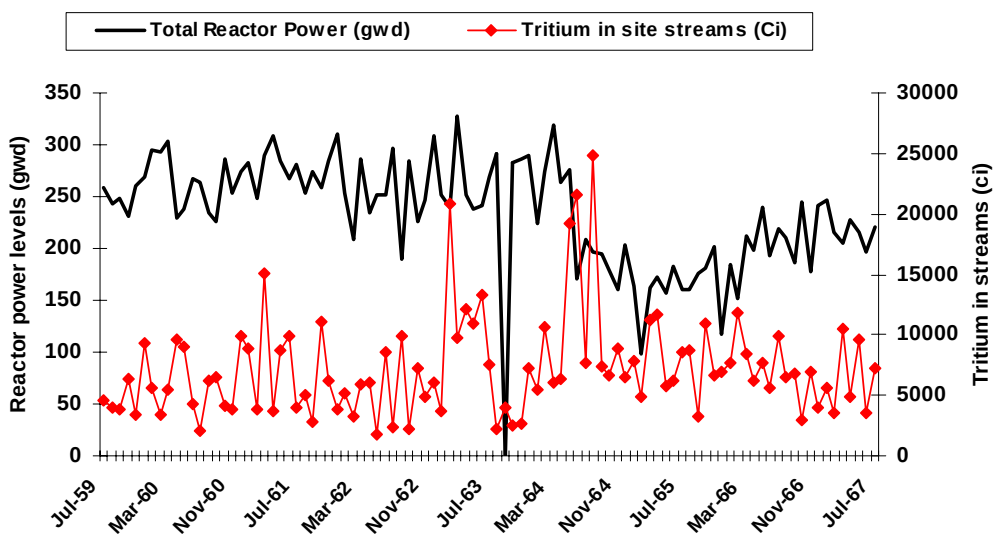

Figure 5-14. Variation with time of the reactor power levels and the total tritium activity measured at Road A in the Site streams. (See Chapter 2 for data source).

Tritium Release Data. We calculated the total tritium discharged each week to the Site streams multiplying the weekly tritium concentration (picocuries per liter) and the total flow measured that week. The datasets are available in the electronic Water Release Excel file. The tritium release data are presented in the following order:

1. data from the various streams will be compared to provide perspective on which Site streams had the highest levels of tritium.
2. the monthly time trends of tritium in Site streams reflect production trends and unplanned events or spills that occurred onsite.

3. we combined the monthly totals of activity measured at Road A for all Site streams with uncertainties to provide our annual release estimates.

Figure 5-15 shows the variation in weekly tritium concentrations measured in Four Mile Creek at the Road A sampling location. The figure shows tritium concentrations measured from July 1959 through December 1963 ranging from about 10,000 to 100,000 pCi L^{-1}, with peaks up to 1,000,000 pCi L^{-1} (Ashley 1959). For comparison, the broken lines indicate the general range of average background tritium levels measured in surface water throughout the U. S. during this period. Tritium levels were higher during that period throughout the U.S. because of fallout from weapons testing (see Chapter 6). Figure 5-16 covers a shorter time period, from 1962 through 1964, and compares tritium concentrations in Steel Creek with those measured in Savannah River during the same time. Tritium concentrations ranged from about 20,000 to 100,000 pCi L^{-1}, while those in the Savannah River were lower, 3000 to 30,000 pCi L^{-1}. This figure shows another point of reference, the current U.S. Environmental Protection Agency drinking water standard for tritium of 20,000 pCi L^{-1} (or, 20 pCi mL^{-1}) (Zeigler et al. 1985). This is an example of the type of analytical data that the SRS collected and maintained. We collected and used these types of measurement data along with flow rate measurements in these streams for reconstructing historic releases of tritium from the SRS.

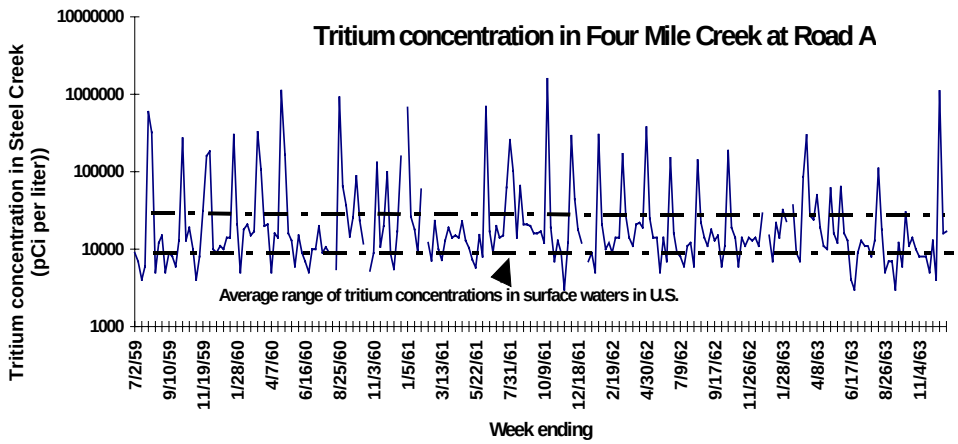

Figure 5-15. Weekly tritium concentrations measured in Four Mile Creek (FMC) at Road A (sampler location #6) from July 1959 through December 1963. This location was the final monitoring point on the FMC before it flowed into the Savannah River. The dashed lines represent the range of average tritium concentrations measured in surface waters around the U. S. during this time (Ashley 1959, Murphy and Carlton 1991).

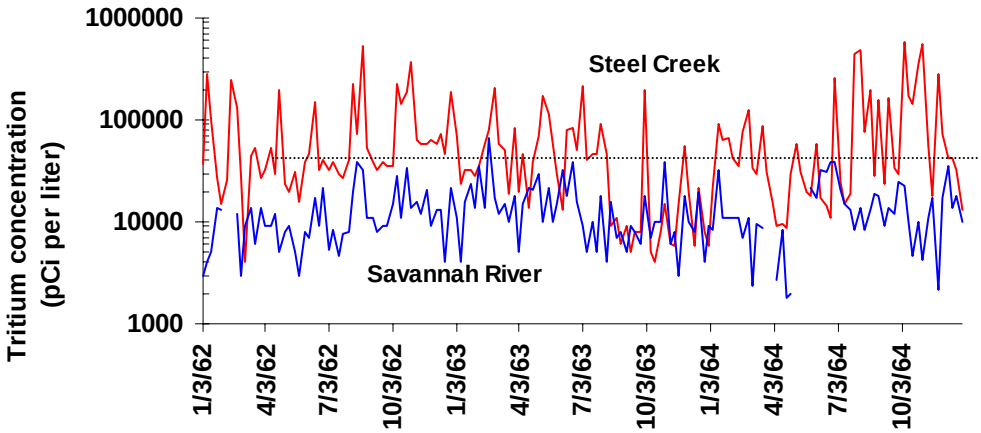

Figure 5-16. Weekly tritium concentrations measured in Steel Creek at Road A and in the Savannah River downstream of the SRS at U.S. Highway 301 from January 1962 through December 1964. The dashed line represents the current U.S. Environmental Protection Agency drinking water standard, and is added for reference purposes (Zeigler et al. 1985).

When the tritium concentrations are combined with the water volume data, then total activity can be compared among the Site streams. In Figure 5-17, the weekly tritium activities, plotted for the five major Site streams, show that the highest tritium activities were generally present in Four Mile Creek, Pen Branch, and Steel Creek. Historically, the Upper Three Runs and Lower Three Runs Creeks had lower levels of activity. These values are based on measurements taken at Road A, the final sampling location before the streams flowed into the Savannah River. Total tritium levels to streams ranged from about 5,000 to 12,000 Ci per month during this time. Par Pond received liquid effluents from the R and P Reactor areas beginning in 1958 and thus prevented their direct release to the surface streams.

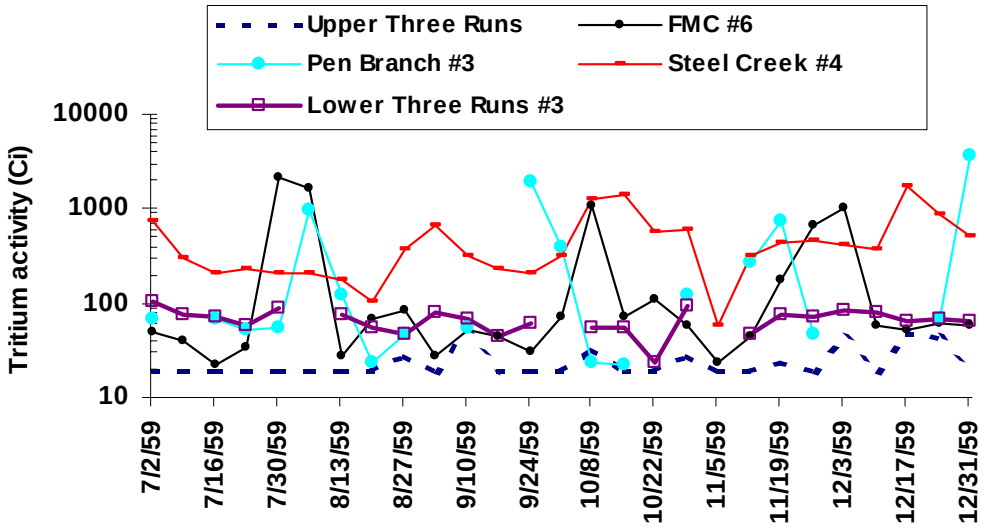

Figure 5-17. Tritium activity in the five Site streams at the SRS measured weekly beginning in the late 1950s. The pattern of higher tritium levels in Four Mile Creek, Pen Branch, and Steel Creek was seen throughout most of the operational history of the Site. In general, radioactivity levels in the Upper Three Runs and Lower Three Runs Creeks were lower. The figure establishes that Steel Creek, which received effluents from the reactor areas, and Four Mile Creek, which received effluents form the separations area, C Reactor area, and some from the D-Area, historically had the highest tritium activities.

Figure 5-18 shows this pattern over a longer period of time and in a different way. The pie chart shows that over 35% of the total tritium activity measured in transport in Site streams was found in Steel Creek, which received effluents from the L and P Reactors. Pen Branch and Four Mile Creek each carried roughly 25% to 30% of the activity. The Upper and Lower Three Runs carried much lower levels. Upper Three Runs historically transported lower levels of tritium because it received discharges only from the north F-Area (Du Pont 1966, Du Pont 1967, Du Pont 1968, Marter 1970).

The SRS began accounting for the amount of tritium released when it began operations in 1953. Actual measurements of the amount of tritium in transport in SRS streams and the Savannah River began in the latter half of 1959. There are 2 broad types of tritium releases from the SRS: (1) direct releases from the SRS facilities themselves and (2) migration of tritium mainly from the seepage basins in the F-area and H-area, the burial ground, and the K-Area containment basin. In the early years almost 100% of the tritium releases to streams was from direct releases. Figure 5-19 shows that releases due to tritium migration from the F-Area and H-Area seepage basins became more important in terms of accounting for releases to the Site streams and eventually to the Savannah River. Since the mid 1970s migration and outcropping to streams have accounted for most of the SRS tritium releases to surface water (Zeigler et al. 1985, Murphy et al 1991, Murphy and Carlton 1991). The Site has reported fairly good agreement in the inventory of tritium measured at the point of release, in the plant streams before entry into the river and in the Savannah River.

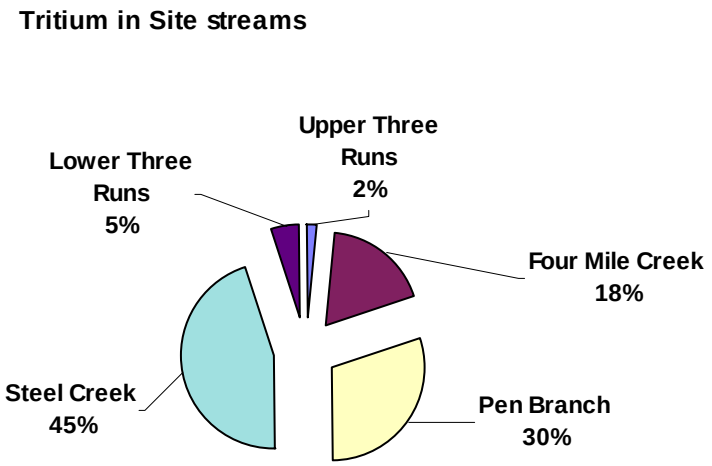

Figure 5-18. The relative amount of total tritium activity in the major Site streams, based on weekly measured values from 1959-1967 in the streams at the last onsite location before the streams emptied into the Savannah River. During this period, Steel Creek, Pen Branch, and Four Mile Creek carried similar levels of tritium in the water, while Upper Three Runs and Lower Three Runs (LTR) water had much lower tritium activity. One reason for lower levels in Lower three Runs was that liquid releases from the P and R Reactors were discharged to Par Pond instead of directly to LTR after 1958 Area (Du Pont 1966, Du Pont 1967, Du Pont 1968, Marter 1970).

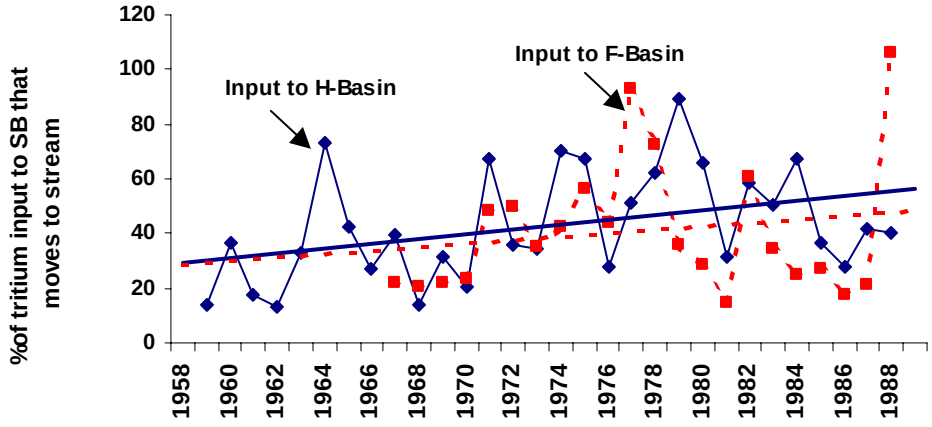

Figure 5-19. Gradual increase in tritium activity seeping from the F-Area and H-Area seepage basins to the Site streams as a percentage of the original input. Migration from the seepage basins and outcropping to Site streams has been the major source of tritium releases in more recent years after the reactor operations stopped.

We reconstructed releases from the SRS using the measured weekly values of concentration and flow rate. We considered several sources of uncertainty, with the analytical errors of sampling and analytical errors being the most important for tritium releases, as previously described in the Uncertainty section of this chapter. We used CrystalBall™ to account for those uncertainties in our calculations (Decisioneering 1995). All Excel spreadsheets with these calculations accompany this report. Figure 5-20 displays the results of our source term reconstruction for tritium, with associated uncertainty estimates. Annual median release estimates were 35,000 Ci or less before 1959 and after 1979. Over 80,000 Ci of tritium was released in 1961, 1963, 1964, 1965, 1966, 1967 and 1968. The highest median release of 121,000 Ci was in 1964 and the 5th and 95th percentiles of the distribution are 84,000 and 177,000 Ci. The median estimate of the total for all years is about 1,800,000 Ci with the 5th and 95th percentiles of the distribution of 1,300,000 and 2,500,000 Ci tritium. There is overall general agreement between our reconstructed tritium release estimates to the Site streams based on the original measurement data and supported by weekly and monthly reports from the 1960s and 1970s and the annual total reported by the SRS.

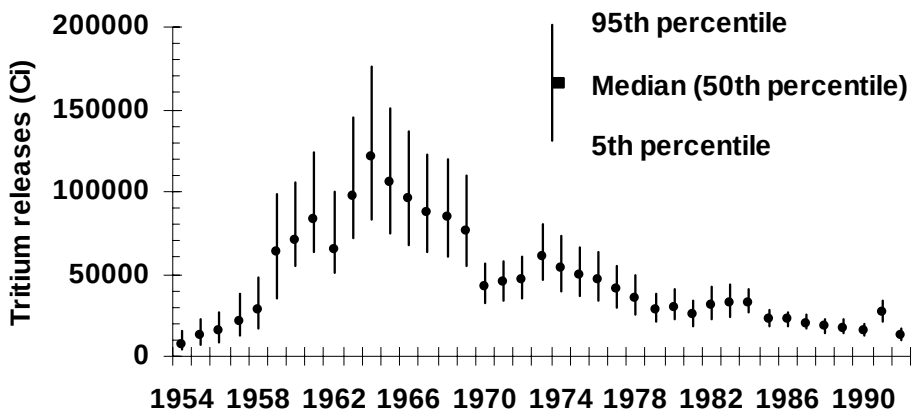

Figure 5-20. Estimates of tritium releases to surface water from the SRS with uncertainty estimates. Each year is represented by a vertical line that represents the 95th (top) and 5th (bottom) percentiles of the distribution of releases. The median or 50th percentile is shown as the filled shape in the center.

The time-dependent release pattern of tritium to surface water reflects the operations and events that were occurring onsite as shown in Figure 5-21. Tritium releases to surface water peaked in 1964 when all reactors were operating. Quite a large number of fuel failures and other unplanned events like spills occurred in the late 1950s through 1964, when the R Reactor was permanently shut down. In addition, the Site processed short-cooled fuel on several occasions during this time, which led to higher levels of radioactivity releases to the streams when the disassembly basins were purged (Murphy et al. 1991). This same pattern is seen with the releases of ^{137}Cs and ^{90}Sr. (see subsequent sections for details). In addition, rainfall levels affected releases of radionuclides in liquid effluents (Figure 5-22). Some of the largest rainfall amounts

occurred in 1964, which led to increased runoff and SRS swamp flooding. Whereas monthly average rainfall for the area is between 3 and 6 in, over twice that level fell in January (8 in), July (10 in) and August (12 in) 1964. Another factor that must be considered in putting Site releases into perspective is weapons fallout and the effect it may have had on contributing to radioactivity levels in the area. (See Chapter 6 of this report).

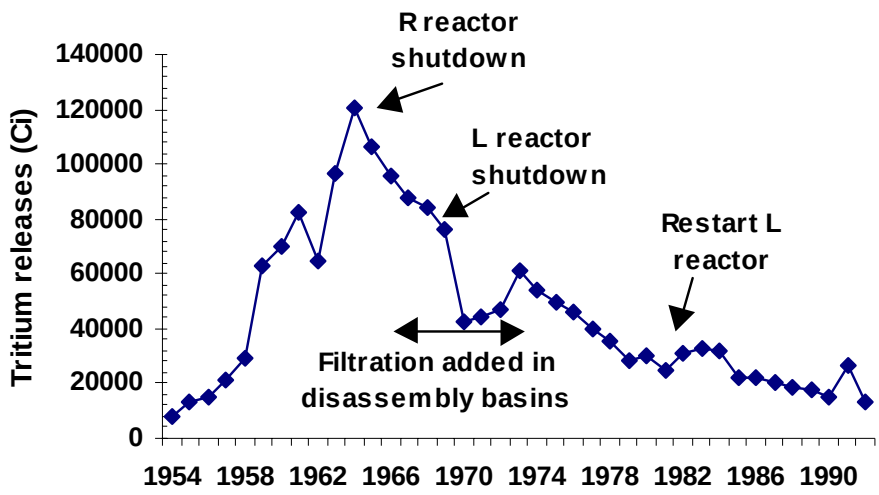

Figure 5-21. Comparison of annual tritium release estimates with some key operational events that affected releases of radioactivity to liquid effluents at the SRS (Du Pont 1958, Du Pont 1962, Du Pont 1966.

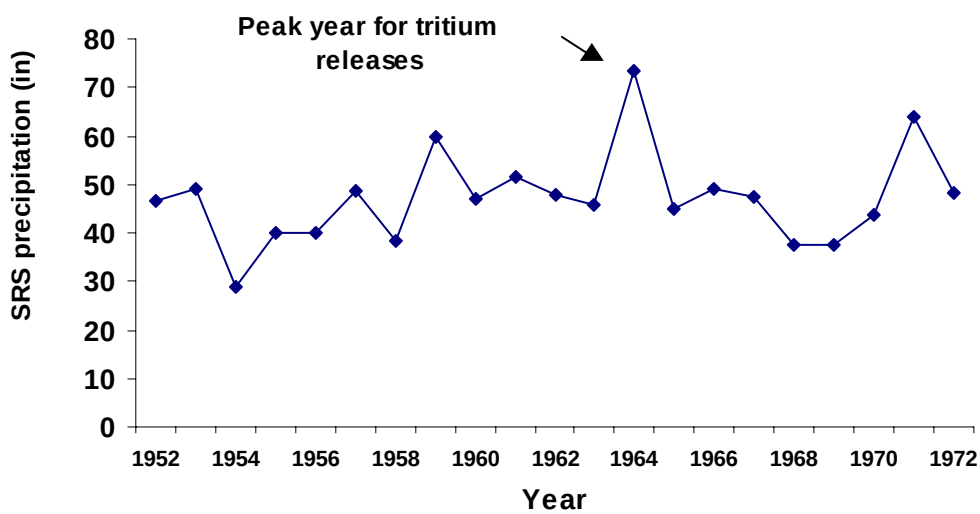

Figure 5-22 Annual rainfall levels in the SRS area from 1952 through 1972. The annual totals ranged from about 29 in. in 1954 to almost 75 in. in 1964, the year with the highest releases of tritium to Site streams.

Cesium Release Estimates

Most of the radiocesium at the SRS was formed as a byproduct of the nuclear fuel and targets during operation of the five production reactors. Many isotopes of cesium were produced in SRS reactors as activation products (e.g., ^{131}Cs, ^{132}Cs, and ^{134}Cs) and fission products (e.g., ^{136}Cs, ^{137}Cs, ^{138}Cs). Cesium-134 and ^{137}Cs are the most important for the purposes of offsite radiation exposure because of their longer half-lives of 2.1 years and 30 years, respectively. Of these, ^{137}Cs has had the greatest impact on the SRS environment. In the past, ^{137}Cs concentrations were usually measured in the:

- liquid effluent outfalls just downstream of the discharge point but before the discharged liquids drained into the Site stream
- streams at a number of locations, and
- Savannah River.

For source term reconstruction, we focused on measurements made in the Site streams and when available, in the effluent outfall.

Over the years, there were various steps taken in the reactor areas to minimize releases of radioactivity. In 1963, basin water was recirculated through the heat exchangers to be cooled and deionized to remove radionuclides, which reduced the discharge volumes considerably. Before 1963, basin water was continuously purged to Site streams. Later permanent sandfilters were installed to main water clarity. (see the reactor areas section of this chapter for details). Most of the radiocesium released in the early years originated in failed fuel elements. The largest ^{137}Cs releases appear to have occurred in 1957 in association with several fuel failures in the R Reactor. The Site monitored aqueous releases of cesium in several ways (Carlton et al. 1992a):

- Cooling water was monitored for beta-gamma activity beginning in 1955, and later, specific radiochemical analyses for radiocesium were done.
- The purge or wastewater from the disassembly basins was routinely analyzed for ^{137}Cs beginning in the mid 1950s
- Gamma spectrometry was introduced in the early 1960s; this technique along with the use of the ion exchange column enhanced the ability to detect low-levels of cesium in stream water (see Table 5-5).

The bases for our ^{137}Cs release estimates are the reported Site measurements made in the Site streams and in the effluent discharges from the facilities to the streams. As with the tritium measurements, weekly concentrations and flow rates were carefully tabulated for the source term reconstruction (see Tritium Release section in this chapter). The Site made weekly measurements of cesium activity at several locations in the Site streams beginning in the late 1950s. The reported concentration measurements and flow data from the aperture card printouts and from weekly records that Ashley (1959) compiled were the basis of our release estimates for ^{137}Cs. As shown in Figure 5-23 over 50% of the total ^{137}Cs activity measured in transport in the SRS streams was measured in Steel Creek, which received discharges from the L, P and R Reactors. Overall, each of the other streams contributed about 15% of the total quantity of ^{137}Cs.

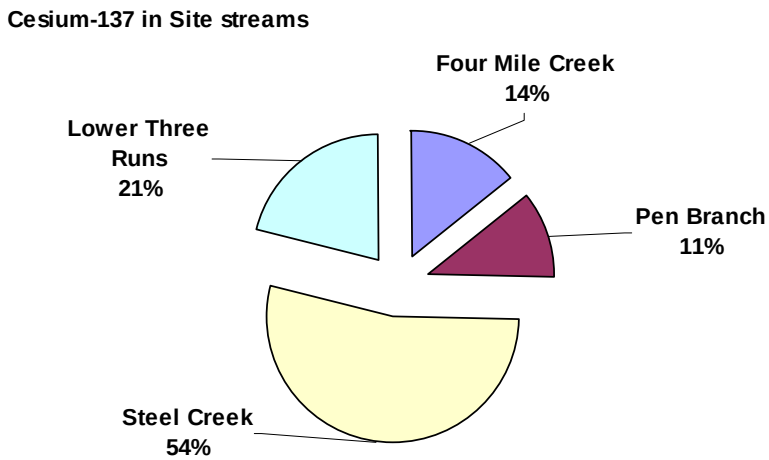

Figure 5-23. The relative amount of total cesium activity in the major Site streams measured weekly from 1959-1967 in onsite streams at Road A, the last onsite location before the streams emptied into the Savannah River. Over half of the ^{137}Cs activity released during this period was present in Steel Creek.

Radiocesium-specific measurements were made in the Site streams beginning in 1960. Before that time, only nonvolatile beta activity was measured in the preoperational survey and from the beginning of plant operations. To estimate ^{137}Cs releases for times when ^{137}Cs specific measurements were not made, we calculated a ratio of ^{137}Cs to nonvolatile beta activity when both measurements were made at the same time and same locations. We used this ratio, along with the nonvolatile beta activity measurements, to estimate levels of ^{137}Cs activity in the Site streams at Road A when ^{137}Cs -specific measurements were not made. Table 5-11 presents the data used to calculate the ratios, and the resulting average ratio and standard deviation for each stream that was used in the uncertainty calculations for cesium releases from the Site. For the annual release estimates for ^{137}Cs, there is more uncertainty associated with our release estimates prior to 1958 because the estimates are based on our calculated ratios and measured nonvolatile beta activity rather than on direct measurements of ^{137}Cs.

Figure 5-24 tracks the change in ^{137}Cs concentrations in the Site streams over time. From the time of the pre startup background survey (Reinig et al. 1953) through 1958, ^{137}Cs levels were higher in Lower Three Runs than in other Site streams that received discharges from the reactor areas. Prior to 1958 and the opening of Par Pond, water in the reactor disassembly basins was purged directly to the Site streams. This logarithmic scale shows a similar pattern of cesium concentrations in the Site streams from about 1958 when Par Pond was operational through the end of 1964. Then in late 1963 levels of ^{137}Cs in Steel Creek increased dramatically from about 25 pCi L^{-1} to a peak of almost 450 pCi L^{-1} in 1968 due to major spills and releases in the area. All reactor cooling water discharges to Steel Creek ceased when L Area reactor was shut down in February 1968.

Table 5-11. Ratio of ^{137}Cs to Nonvolatile Beta Activity in Site Streams from July 1958 through December 1962[a]

	Four Mile Creek (pCi L^{-1})			Pen Branch (pCi L^{-1})			Steel Creek (pCi L^{-1})			Lower Three Runs (pCi L^{-1})		
Date	NVB[b]	^{137}Cs	^{137}Cs/ NVB	NVB	^{137}Cs	^{137}Cs/ NVB	NVB	^{137}Cs	^{137}Cs/ NVB	NVB	^{137}Cs	^{137}Cs/ NVB
Jul-Dec 1958	110	11	0.10	470	7	0.01	120	12	0.10	130	38	0.29
Jan-Jun 1959	4300	14	0.00	640	6	0.01	300	16	0.05	74	19	0.26
Jul-Dec 1959	350	6	0.02	250	6	0.02	120	11	0.09	49	16	0.33
Jan-Jun 1960	400	7	0.02	360	8	0.02	530	8	0.02	20	8	0.40
Jul-Dec 1960	260	7	0.03	260	*7*	0.03	200	*9*	0.05	40	12	0.30
Jan-Jun 1961	260	14	0.05	190	6	0.03	220	14	0.06	24	9	0.38
Jul-Dec 1961	230	*9*	0.04	120	*13*	0.11	160	*10*	0.06	27	7	0.26
Jan-Jun 1962	120	12	0.10	540	10	0.02	230	26	0.11	50	11	0.22
Jul-Dec 1962	78	38	0.49	110	12	0.11	270	31	0.11	30.00	10.0	0.33
Average			0.11			0.04			0.07			0.31
Standard deviation			0.15			0.04			0.03			0.06

[a] The measurement results were reported monthly and semiannually in the Health Physics Regional Monitoring Semiannual Reports for those time periods; the ratio was used to estimate ^{137}Cs releases for early operational periods at SRS when only nonvolatile beta activity was measured.
[b] NVB = nonvolatile beta activity.

After that time, only L-Area and P-Area disassembly basin purges, undiluted by cooling water, were discharged to the creek. This action resulted in higher concentrations of radioactive materials (primarily ^{134,137}Cs from a few failed experimental fuel elements) in the stream water, even though the quantity of radioactive materials released to the creek began decreasing after 1968 (Marter 1970).

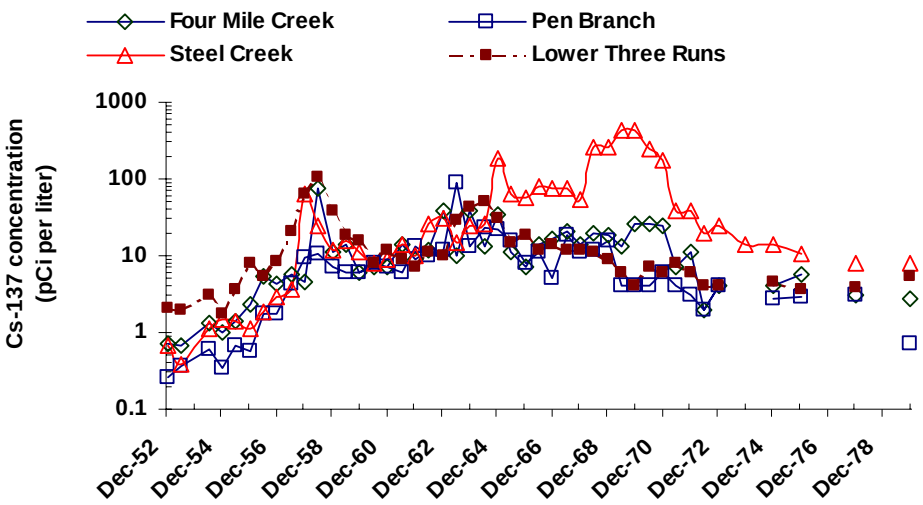

Figure 5-24. Semiannual average concentrations of ^{137}Cs in Four Mile Creek, Pen Branch, Steel Creek and Lower Three Runs measured at Road A. The figure shows a similar pattern of cesium concentrations in the Site streams from 1958 through 1964. From 1964 through 1975, the concentrations were much higher in Steel Creek than in the other Site streams.

These concentration measurements are then combined with weekly flow data to calculate the weekly reported releases of ^{137}Cs at Road A. Figure 5-25 shows the weekly releases of ^{137}Cs in the four main Site streams from April 1960, when measurements of ^{137}Cs began, through June 1967. Several peak releases are marked with the date and the stream where the high levels were measured. In June 1964, almost 50 Ci of ^{137}Cs were measured in Lower Three Runs Creek when a major spill occurred. These data are the basis of our source term calculations for ^{137}Cs releases.

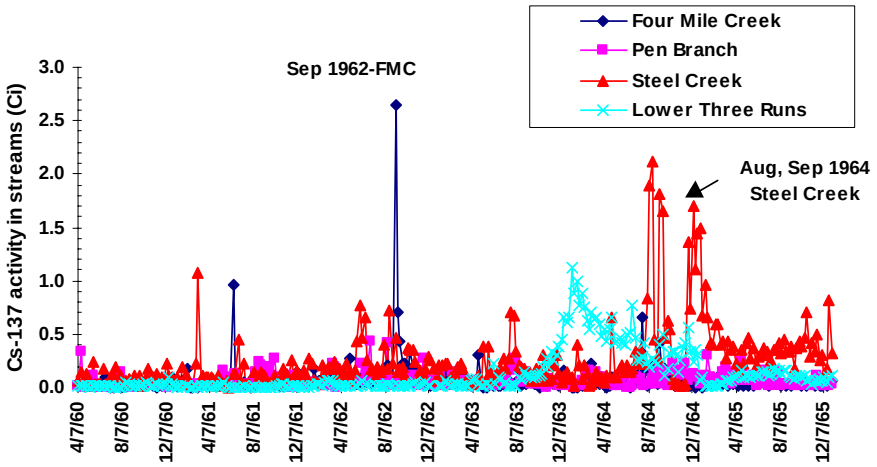

Figure 5-25. Comparison of ^{137}Cs activity in Site streams from 1960–1965. Several peak releases, marked with the date, correspond to unplanned but documented releases.

Studies of ^{137}Cs in the SRS streams and the swamp area have provided information to estimate uncertainties for our revised ^{137}Cs source terms. For example, the Site measured ^{137}Cs in Four Mile Creek (FMC) at Road 4 (just below where H-Area effluents entered), Road C (just below where F-Area effluents entered), and Road A. We used these measurements to calculate a fraction of Cs remaining at each location downstream from the facility discharge point to estimate a range of ^{137}Cs activity that may have reached the Savannah River (Du Pont 1967, 1968a). Roughly 70% (with a range of about 25% to 90%) of the ^{137}Cs measured at either Road 4 or Road C reached Road A (roughly 5 mi downstream in FMC). In this case, Four Mile Creek meanders at least another 7 mi., some through the SRS swamp before reaching the Savannah River. Some of that distance is similar to the conditions from the point of releases to the Road A location on Four Mile Creek and we can assume that under similar conditions that 70% of the activity measured at Road A might reach the river (or about 50% of the original activity measured at Road C).

Site studies support this general range of cesium retention in Site streams. A 1974 study and discussion of ^{137}Cs in effluent streams (Du Pont 1973) concludes that an average of 75% of annual Site releases of ^{137}Cs in liquid effluents are measured in transport in Site streams at Road A and 19% downstream in the river at Highway 301. These percentages were fairly consistent with a similar study in 1963 and 1964 where 80% of the ^{137}Cs released was measured in stream water and 30% in river water at Highway 301 (Du Pont 1964a, 1964b). We have used these estimates of retention in the Site stream beds in our estimates of cesium releases with associated uncertainty distributions.

The Site made a comprehensive radiological survey of the Lower Three Runs Creek corridor from May to August 1971 (Du Pont 1971). Researchers surveyed 11 stations beginning immediately below Par Pond dam and extending to just above the mouth of the main channel of the creek. The survey determined the flow of the Lower Three Runs Creek near its mouth, the distribution of radioactivity levels in sediments, soils, and terrestrial vegetation at various distances from the stream, and the concentrations of specific radionuclides in a few resident aquatic organisms in Lower Three Runs Creek. Measurable concentrations of ^{137}Cs were detected in streambed sediments and core samples. Usually the upper segments had higher concentrations of cesium. Maximum concentration of ^{137}Cs in sediment cores were less than maximum concentrations in soil cores collected at various distances from the stream in the flood plain bordering Lower Three Runs. This was attributed to the flooding conditions that occurred before R area reactor shutdown in 1964. Background levels of ^{137}Cs in soil cores were seen only in samples taken several hundred feet from the stream.

Figure 5-26 displays the results of our source term reconstruction for ^{137}Cs with associated uncertainty estimates. Annual median release estimates were approximately 0.1 Ci or less before 1955 and after 1976. Over 10 Ci of ^{137}Cs were released annually from 1964–1967, with the highest median release estimate in 1964 of about 20 Ci with the 5th and 95th percentiles of the distribution of 50 and 120 Ci. The median estimate of the total ^{137}Cs for all years is about 250 Ci with the 5th and 95th percentiles of the distribution of 100 and 600 Ci ^{137}Cs. There is overall general agreement between our reconstructed ^{137}Cs release estimates to the Site streams based on the original measurement data and supported by weekly and monthly reports from the 1960s and 1970s and the annual total reported by SRS. The small peak in the late 1980s corresponds to the restart of the L Reactor.

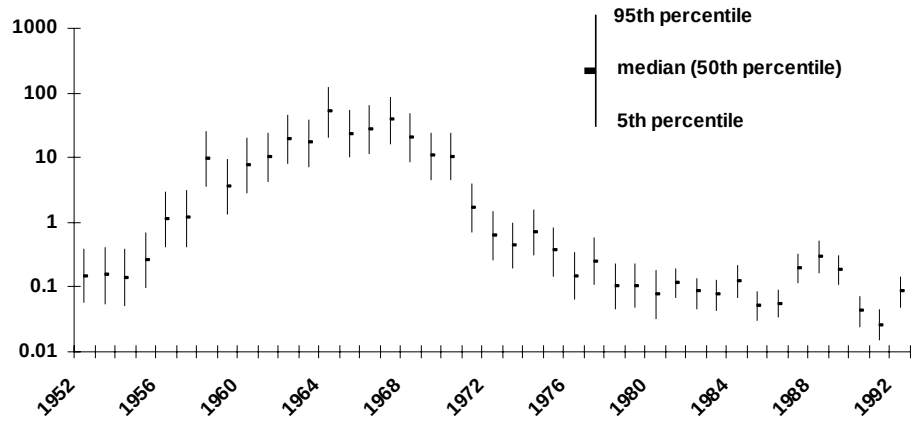

Figure 5-26. Estimates of ^{137}Cs releases to surface water from the SRS with uncertainty estimates, shown on a logrithmic scale. Each year is represented by a vertical line that represents the 95th (top) and 5th (bottom) percentiles of the distribution of releases with the median or 50th percentile shown as the filled shape in the center.

The time-dependent release pattern of ^{137}Cs to surface water reflects the operations and events that occurred onsite as shown in Figure 5-27. Releases of ^{137}Cs to surface water peaked in 1964 when all reactors were operating. Quite a large number of fuel failures and other unplanned events like spills occurred in the late 1950s through 1964, when the R Reactor was permanently shut down (Du Pont 1958, Du Pont 1962, Du Pont 1966). In addition, the Site processed short-cooled fuel on several occasions during this time, which led to higher levels of radioactivity releases to the streams when the disassembly basins were purged (Carlton et al. 1992a).

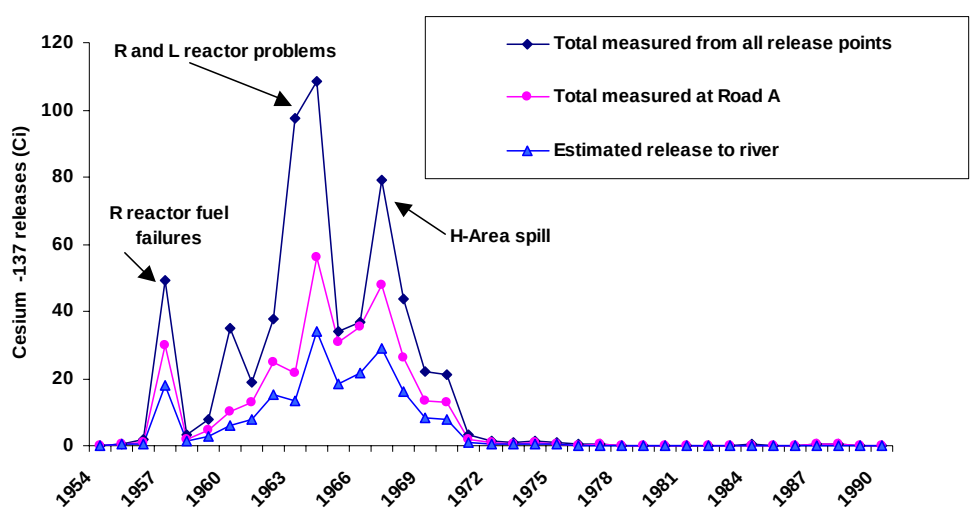

Figure 5-27. Comparison of ^{137}Cs activity from 1954 through 1990 released from the facilities, measured in onsite streams and estimated to have been released to the Savannah River.

Health Physics Regional Monitoring reports for 1957 and 1958 describe a number of fuel failures at the L, P and R Reactors that contributed to increased levels of nonvolatile beta activity in the Site streams. Further isotopic analyses were done in some instances and provided a rough estimate of levels of radiocesium and radiostrontium in the Site streams before routine monitoring for these specific radionuclides were done beginning in mid 1958. For example, the increased discharge of nonvolatile beta in disassembly basin water from L-Area and P-Area peaked in August and September 1957 due to failed fuel elements in those reactors. In addition, Site personnel had discontinued cooling water flow from L-Area in preparation for a process improvement shutdown, and concentrations of nonvolatile beta activity remained fairly high until mid-November 1957 when cooling water flow was resumed (Mealing et al. 1958).

Strontium Release Estimates

Natural mechanism for radiostrontium production is fission of naturally occurring uranium and thorium. Such fission occurs either by spontaneous fission decay or by neutrons in nature inducing fission (Carlton et al. 1992b). Two human-made sources have contributed to the radiostrontium in the global environment. These events are:

- Above-ground nuclear weapons test introduced about 22,000,000 Ci ^{90}Sr to the atmosphere, mostly through the mid-1960s
- Nuclear reactor operation (Satellite Cosmos 954 accident in 1978 released about 84 Ci ^{90}Sr at re-entry into earth's atmosphere and Chernobyl nuclear reactor accident in 1986 released 34,000 Ci ^{90}Sr.

Most radiostrontium at the SRS originated as fission products in the nuclear fuel and targets during the operation of the five production reactors. There are over 12 isotopes of radiostrontium produced but five of these have half-lives less than a second and, except for ^{89}Sr and ^{90}Sr, the others have half-lives of less than 10 hours. During normal reactor operations, radiostrontium is contained within the cladding of fuel and target elements during irradiation and cooling. Cooling is the interval between the end of irradiation and the beginning of chemical separations. The irradiated materials were stored underwater in large basins of water called disassembly and cooling basins. Beginning in the 1970s, the Site established a standard cooling for most irradiated materials of at least 200 days. This length of time limited the emissions of radioiodine significantly. About 200 days reduced the inventory of ^{89}Sr by a factor of about 16 but had little effect on the ^{90}Sr inventory.

Figures 5-28 and 5-29 show the general release pattern for radiostrontium to the Site streams, a pattern that is similar to that seen for tritium and ^{137}Cs releases. The pie chart indicates that over 60% of the total ^{89,90}Sr activity measured in transport in Site streams was found in Steel Creek, which received effluents from the L and P Reactors. Pen Branch carried roughly 15% of the activity, while Four Mile Creek and Lower Three Runs carried approximately 10% of the activity. Upper Three Runs transported very small amounts of ^{89,90}Sr, if any, because it received discharges only from the northside of the F-Area. The reactors were clearly the largest source of ^{90}Sr to the streams (Figure-29).

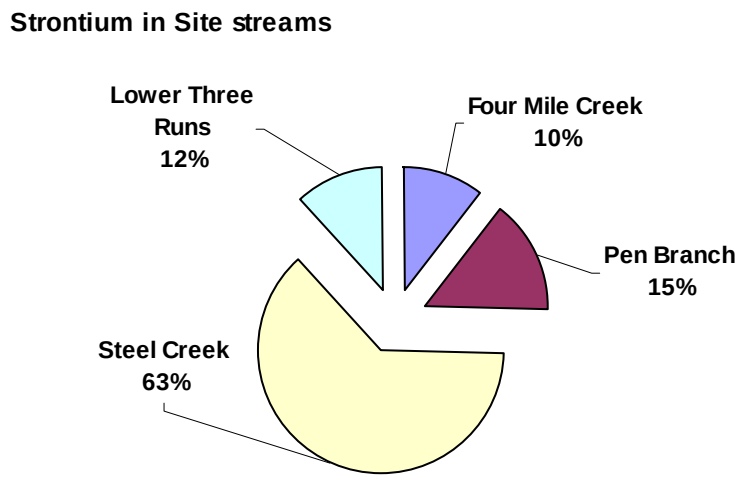

Figure 5-28. The relative amount of total strontium activity in the major Site streams measured weekly from 1960-1967 in onsite streams at Road A, the last onsite location before the streams emptied into the Savannah River. Over half of the ^{89,90}Sr activity released during this period was present in Steel Creek.

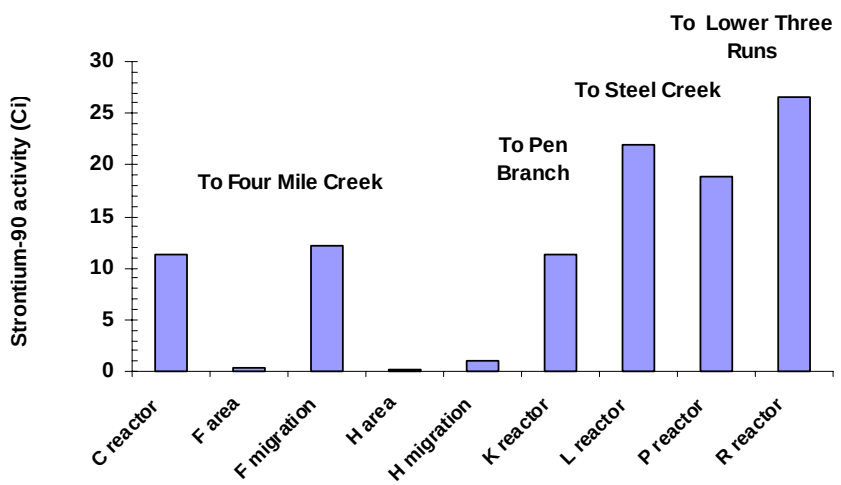

Figure 5-29. Releases of ^{90}Sr from various sources at the SRS over all years of operations. The reactors were the primary source of releases of ^{90}Sr (and other fission and activation products) at the SRS. Releases to Steel Creek were the highest because the reactors in the L-Area, P-Area, and R-Area discharged to that creek at different times over the years. However, liquid effluent releases from the P Reactor also went to the Lower Three Runs system.

Radiostrontium was measured in the reactor basin purge water beginning in the mid 1950s and-in the Site streams beginning in 1960. Before that time, only nonvolatile beta activity was measured in the preoperational survey and from the beginning of plant operations. To estimate ^{90}Sr releases for times when ^{90}Sr-specific measurements were not made, we calculated a ratio of ^{90}Sr to nonvolatile beta activity when both measurements were made at the same time and same locations. We used this ratio, along with the nonvolatile beta activity measurements to estimate levels of ^{90}Sr activity in the streams at Road A when ^{90}Sr-specific measurements were not made. This procedure is the same as we followed in estimating ^{137}Cs releases when specific measurements were not available (see Cesium release section in this chapter).

Our revised release estimates for ^{90}Sr were based on reported concentrations and flow measurements made in the streams, combined with uncertainties that have been described previously. Figure 5-30 shows the reported six-month totals of ^{90}Sr release to the Site streams. Figure 5-31 displays the results of our source term reconstruction for ^{90}Sr with associated uncertainty estimates. Annual median release estimates were approximately 1 Ci or less before 1957 and after 1976. We estimate that over 15 Ci of ^{90}Sr was released to the river in 1960 and over 5 Ci in 1962–1967 annually from 1964–1968. The median estimate of the total ^{90}Sr for all years is about 100 Ci with the 5th and 95th percentiles of the distribution of 45 and 250 Ci ^{90}Sr.

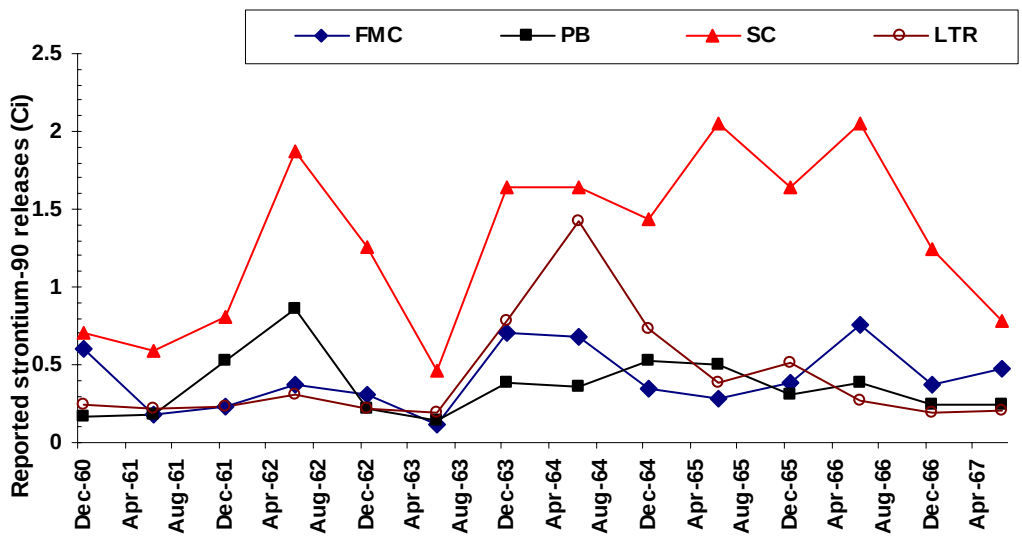

Figure 5-30. Semi-annual activity levels of ^{90}Sr in the streams based on the reported concentration measurements and flow rate in the Site streams at Road A.

The rate of migration from the seepage basins in the F-Area and H-Area have gradually declined since about 1970 when ^{90}Sr releases were estimated and measured, but the releases to Site streams from groundwater outcrops has been the biggest source of ^{90}Sr during that time.

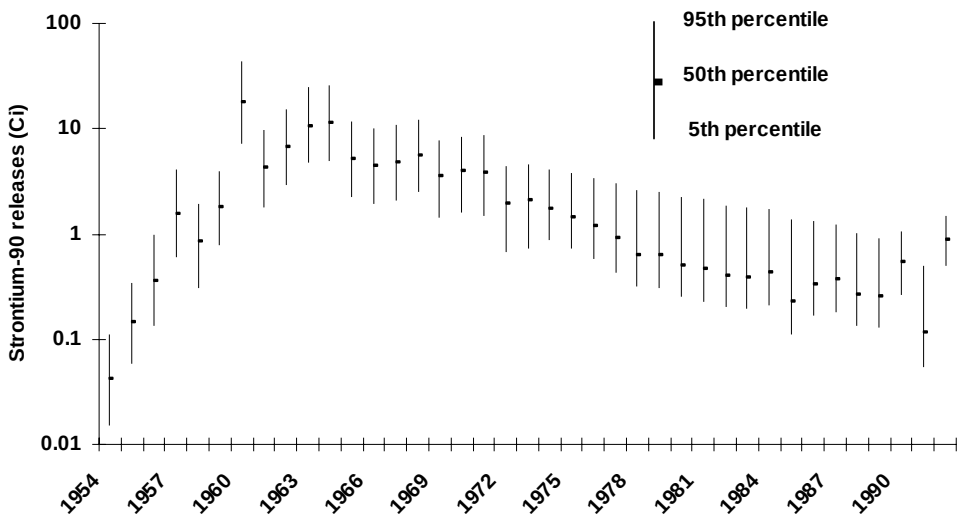

Figure 5-31. Estimates of ^{90}Sr releases to surface water from the SRS with uncertainty estimates, shown on a logrithmic scale. Each year is represented by a vertical line that represents the 95th (top) and 5th (bottom) percentiles of the distribution of releases with the median or 50th percentile shown as the filled shape in the center.

Releases of Iodine-131

As with the other fission and activation products important in SRS releases, the major sources of ^{131}I at the SRS were the chemical separations areas and the reactor areas. Figure 5-32 presents the releases of ^{131}I in liquid effluents to the streams and seepage basins from the SRS.

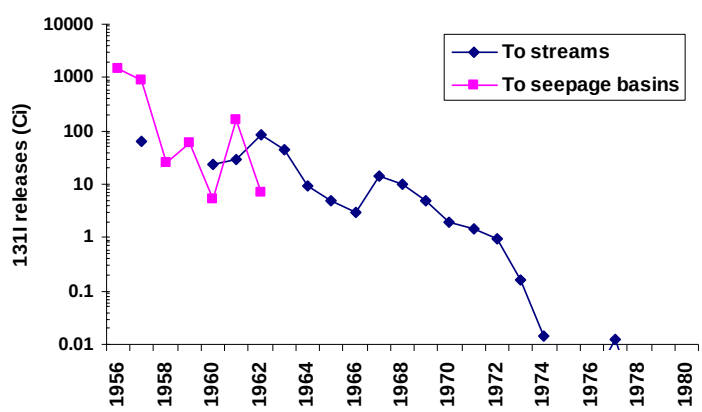

Figure 5-32. Reported releases of ^{131}I to Site streams and to seepage basins from the SRS. The large increase in releases to the seepage basins in 1961 resulted from the dissolution of short-cooled fuel elements in the separations area.

In the early years, a chloroform extraction technique was used to separate ^{131}I from stream and river water samples. This procedure had a sensitivity of about 6 pCi L^{-1}. In January 1963, a new technique was introduced for the analyses of ^{131}I in water that used an ion-exchange cation and anion resins to concentrate the radionuclides. The sensitivity of this method for ^{131}I in 40 L of sample was 1 pCi L^{-1} (Boni 1963).

Releases of Activation Products

The SRS released activation products in liquid effluents from facilities and operations that were similar to those responsible for releases of fission products to surface water. Chapter 4.3 in this report provides some background on the releases of activation products to air and surface waters from the SRS. Three activation products, ^{60}Co, ^{35}S and ^{32}P, may be of some importance for offsite residents if certain exposure pathways are considered (see Chapter 3 of this report). Our evaluation of reported releases of these radionuclides indicates that the historic record does include memos, and reports documenting interest in and monitoring of these materials. We have compiled measurement data for these radionuclides that can be used for later exposure assessment if such an assessment if warranted. The following sections provide summaries of information of these radionuclides.

Cobalt is chemically similar to iron and like iron, can exist in Co(II) and Co(III) oxidation states. However, Co(III) is only dominant at conditions beyond the oxidation limits of water at 25° C and 1 atmosphere, and thus in water Co(II) is dominant form. In the acidic conditions typical of SRS waters, cobalt is relatively mobile. Sorption to solids in the water can slow down cobalt migration. Factors such as pH, mineral content and grain size can strongly influence sorption. This is reflected in the range of K_d values (ratio of amount of constituent sorbed to solid phase over the concentration in water at equilibrium) of 0.2 to 3800 ml per gram (see Table 5-8).

The largest quantities of ^{60}Co were released from the SRS to the seepage basins in the F-Area and H-Area. Seepage from the basins released some activation products to the Site streams, particularly to Four Mile Creek. Figure 5-33 shows that more ^{60}Co was released to the Site streams than to the seepage basins in the early years. In the mid 1970s this trend generally changed when releases to the streams were more tightly controlled. This pattern is similar for the release of other activation products, too.

A second activation product released to surface water is ^{35}S. Prior to 1961, ^{35}S was not included in the reactor area release summaries. In the last quarter of 1961, specific analysis for ^{35}S was done and was detected in disassembly basin water from the reactor areas. The reason for doing ^{35}S analysis was because there was an imbalance between the nonvolatile beta activity in reactor effluents and the total estimated disassembly basins releases based on stream data during October 1961. The stream activity exceeded releases from the seepage basins.

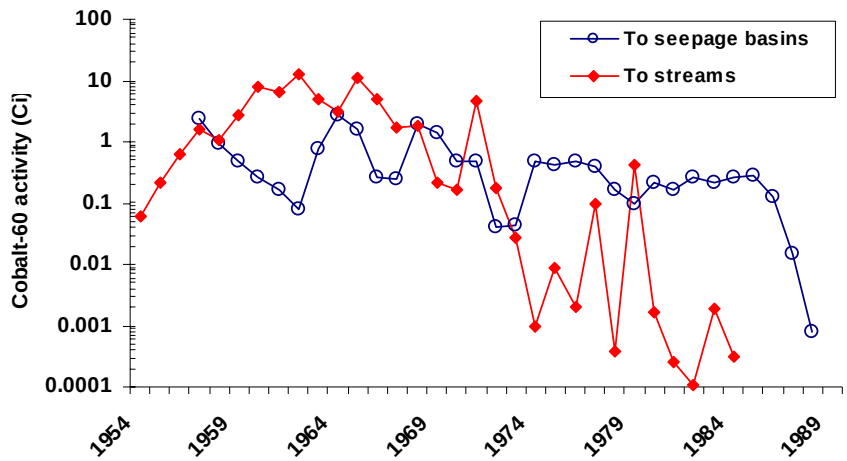

Figure 5-33. Reported releases of ^{60}Co to Site streams and seepage basins from the SRS. The releases of ^{60}Co to the streams were higher than releases to the seepage basins in the early years. The pattern is reversed in later years.

By January 1962, Site personnel were investigating the source of the ^{35}S. One thought was that the amount of ^{35}S found in the C-Area process water (0.015 uCi per ml) could have originated from chlorine and/or sulfur impurities introduced into the system in concentrations less than 1 ppm. The ^{35}Cl (n,p) ^{35}S reaction appeared to be the most likely source of ^{35}S, requiring levels of chloride impurities as low as 100 ppb (Du Pont 1961). In April 1962, it was demonstrated that the ^{35}S releases coincided with reactor outages and originated from fuel decontamination test tank that contained oxide layers from fuel assemblies. This was also a source for ^{32}P discharges.

The chemistry section at the SRS continued to standardize and optimize the chemistry methods for ^{35}S. Because the concentrations were generally very close to the lower detection limit (Du Pont 1961), the Site analysts continued to improve the method. Figure 5-34 compares the reported total releases of ^{60}Co, ^{32}P, and ^{65}Zn to streams from facilities at the SRS. This figure demonstrates that, just as with tritium and fisson product releases, the reactors were the main source of activation products released to Site streams. Measured releases of radionuclides in the various streams, combined with the known reactor operational times, indicate the while some reactors worked relatively well with minor complications over the years, others, like the R Reactor, were quite error and problem-prone. For example, the R Reactor operated only through 1964 but contributed the largest releases of some radionuclides, like ^{65}Zn to the Site streams.

In summary, the releases of key beta-gamma emitters (tritium, ^{137}Cs, ^{90}Sr) were greatest from the reactor areas. Although most releases from these areas to streams came from the disassembly basins and not directly from operations of the reactors, the overall pattern of releases over time are related to reactor operations. Figure 5-35 clearly demonstrates that the reactor power levels were highest in the early to mid-1960s because all five reactors were in full operation. Fuel failures and other problems in the R and the L Reactor areas accounted for some of the higher releases of radionuclides to water during the early 1960s. Finally, the quantities of activation

products released to air and water were quite small compared to the quantities of fission products, like ^{137}Cs, and ^{90}Sr that were released.

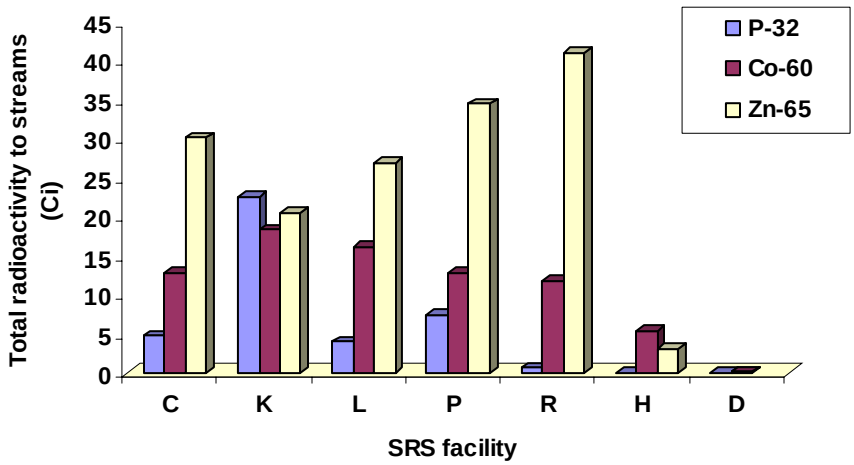

Figure 5-34. Comparison of total releases for all years from the SRS of ^{32}P, ^{60}Co, and ^{65}Zn. C, K, L, P, and R refer to the five operating reactors at the SRS. In this figure, H refers to the H-Area and D refers to the Heavy Water or D-Area.

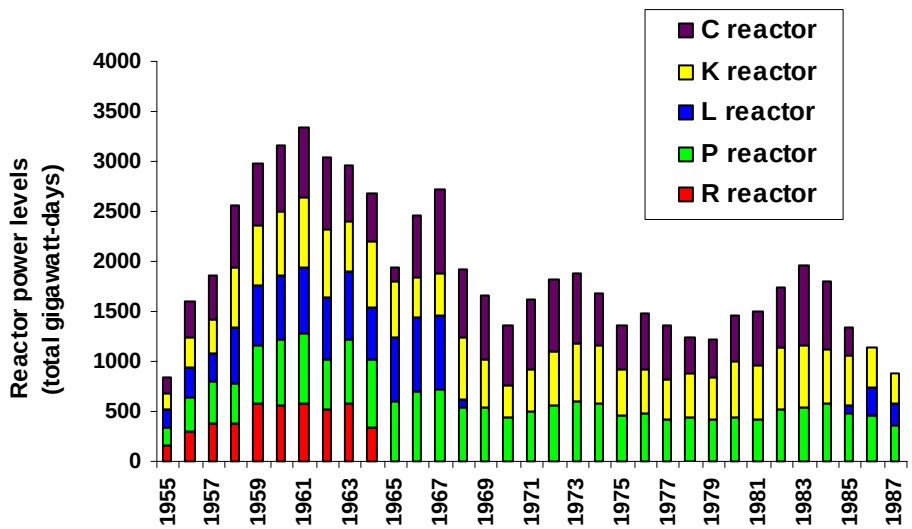

Figure 5-35. Reactor power levels for the operation reactors from 1952 through 1988. The operation periods for the reactors were: C Reactor 1954-1987, K Reactor 1954-1988, L Reactor 1954-1968 and 1985-1988, P Reactor 1954-1988 and the R Reactor 1954-1964. A restart of the K Reactor began in 1991. From 1954 through 1964, all reactors were operating.

Releases of Alpha Emitters: Uranium

Our radionuclide screening did not indicate that uranium would be a major contributor to offsite radiation doses; however, there were fairly large quantities of uranium released to Site streams, especially from the reactor materials area or M-Area. Furthermore, future exposure pathway analysis may indicate that although uranium may not be important radiologically, it may be important for its potential chemical toxicity (see Chapter 18 in this report). For these reasons, we summarize some of the release information that we have compiled for uranium.

In the 1950s, the SRS used an extraction method to separate uranium and plutonium from environmental samples like vegetation, soil, and water. This newer TBP extraction was based on a method commonly used at that time using nitric acid and tri-n-butyl phosphate in n-tetradecane diluent, and replaced the ether extraction method, which was more hazardous in the laboratory. From spiked sample studies, uranium and plutonium were nearly equally recovered. The average recoveries were about 76% for vegetation, 76% for soil and 82 ± 15% for water (Geiger circa1958). The procedure was used as part of the health physics regional monitoring program at the SRS. In the 1960s, uranium and plutonium were extracted by liquid ion exchange using tri-isooctulamine (TIOA) and then alpha counted (Johnson 1968a). One liter water samples were prepared for both extraction procedures with a final 10-15 ml volume transferred to a counting planchet for drying and alpha scintillation counting. The TIOA extraction method continued to be used for determining the concentration of uranium and plutonium in water to recent times (Du Pont 1989).

Uranium was produced and discharged from several areas at the SRS. The largest releases of uranium onsite occurred from the M-Area to Tim's Branch and to the seepage basins after they were put into service in 1973. Liquid overflow from several points in this process was discharged to the sewer system and directly to the surface streams, especially in the early years. The direct release of drain water from the uranium target production facility in M-Area was stopped in 1973 when the process sewer discharges were diverted to the newly opened M-Area seepage basin (Evans et al. 1992). In 1979, the primary uranium release streams were diverted to the settling basins and, by May 1982, all releases of untreated process wastewater were diverted to the settling basin.

Within the M-Area, the 300-Area was one of the first operations to begin at the Site. It was also one of the first sources of Site contamination to be of concern (Albenesius 1954). From startup in October 1952 through February 1953, an estimated 100 lb of uranium was discharged in the 300-Area effluent system to Tim's Branch. After that time, releases of uranium from the M-Area were routinely monitored and reported (Alexander and Horton 1956). Figure 5-36 shows that during the first 8 years, the 6-month uranium release totals varied from about 16 lb to over 4000 lb in late 1963, when a new etching process was introduced in September 1963. The 1960s began an upward trend in uranium releases to Tim's Branch that peaked in 1969 when over 25,000 lb of uranium was released. During this time, there was concern about the amount of uranium being released (Johnson 1968b).

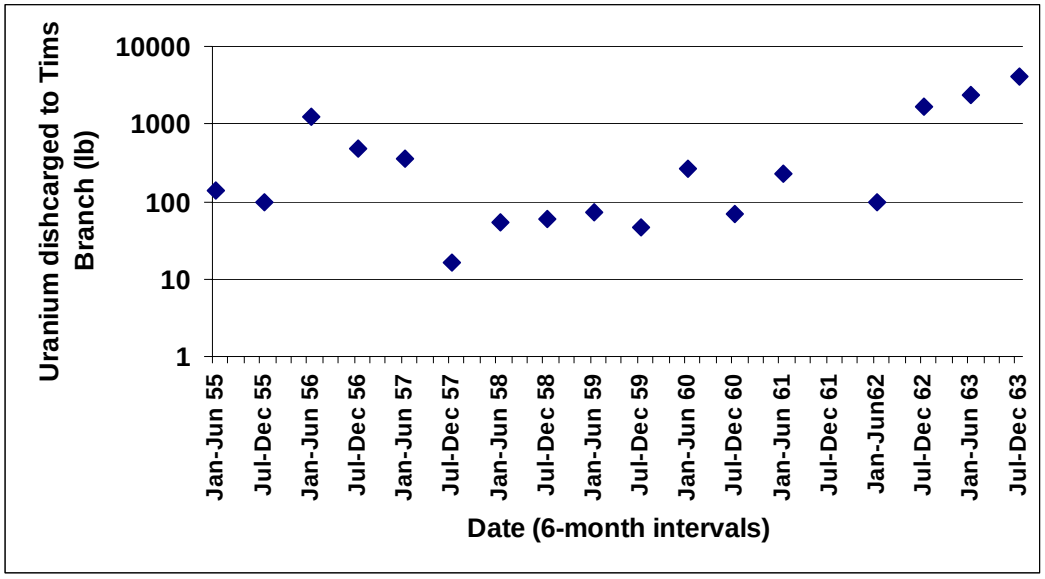

Figure 5-36. Measured releases of uranium to Tim's Branch from the M-Area from January 1955 through December 1963, reported in the health physics semi-annual progress reports.

Figure 5-37 illustrates that the largest releases of uranium to Tim's Branch occurred from 1964 to 1970. The high releases in the mid to late 1960s were due to the loss of uranium etching solution and the overflow of normal autoclave waste from the holding tank.

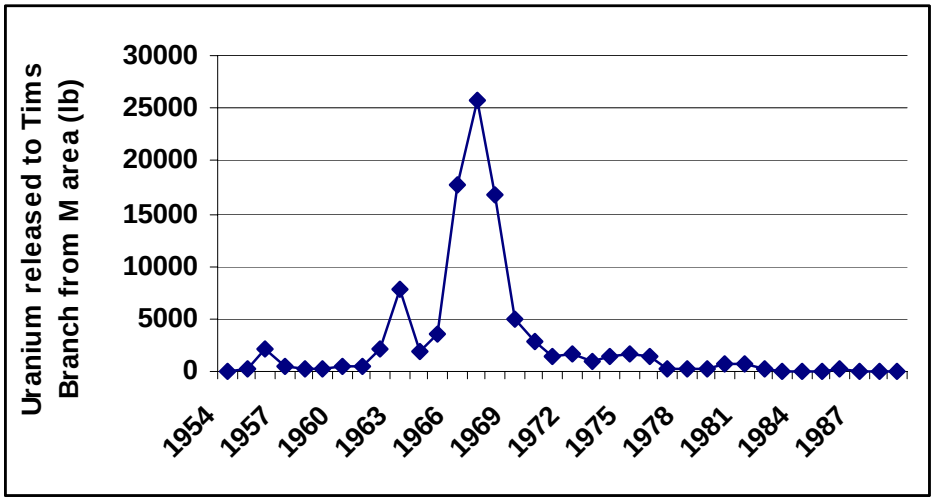

Figure 5-37. Reported annual uranium releases to Tim's Branch from M-Area. One curie uranium is equivalent to 6190 lb. uranium; therefore, 25000 lb uranium = ~4 Ci uranium. The total quantity of uranium release over this period was approximately 153,000 lb. (~25 Ci).

There were small releases of uranium from the separations area. Cooling water for portions of the F-Area and H-Area separations process line was pumped from deep wells and discharged to Four Mile Creek (FMC) after use. The water contained measurable amounts of radioactivity because of cooling coil leaks. Uranium was a predominant chemical constituent of this mixture (Evans et al. 1992). Cooling water discharged to Four Mile Creek was not analyzed specifically for uranium, and the activity was reported as "unidentified alpha." There were high releases of unidentified alpha reported in 1955 (0.045 C) and 1956 (0.037 Ci) to Four Mile Creek that were associated with evaporator coil leaks. Alpha spectrometry measurements on a few samples in 1955 attributed most activity to ^{238}U. If we assume all activity was due to ^{238}U then these activity levels correspond to about 300 lb. in 1955 and 240 lb in 1956. These conversions are based on a specific activity for ^{238}U of 1.24 Bq g^{-1}, or 6580 Ci lb^{-1}. (If we assume the uranium was natural uranium, then the activity levels correspond to about 140 lb. in 1955 and 120 lb. in 1956. The specific activity for "natural" uranium [99.27% ^{238}U, 72% ^{235}U and 0.0055% ^{234}U] is 692 nanocuries g^{-1}, or 3190 lb Ci^{-1}). High releases in 1984 (0.025 Ci unidentified alpha) were probably depleted uranium in runoff from the uranium recovery facilities (A-Line) to the storm sewer system. This activity corresponds to about 80 lb. of natural uranium.

For the reactor areas, the Site began measuring gross alpha in liquid effluents from the reactor areas in 1972, and there were no specific analyses for uranium. Figure 5-38 shows the pattern of alpha activity measured in wastewater from the reactors to the Site steams and to the seepage basins after 1972. These values are not corrected for the natural uranium content of the incoming river water, which is the source of the purge water for the reactor area

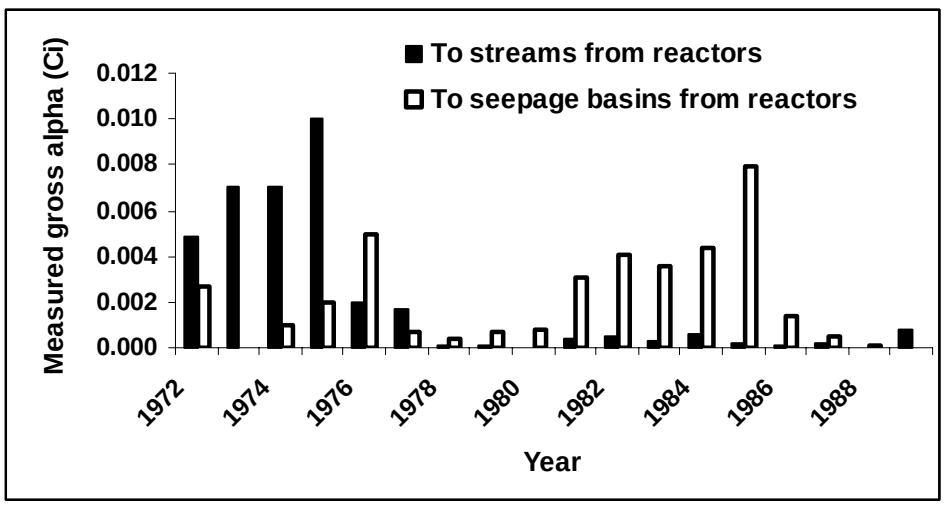

Figure 5-38. Measured releases of gross alpha from reactor areas to Site streams and the seepage basins from 1972–1989. These values are not corrected for the natural uranium content of the incoming river water, which was the source of the purge water. No measurements were made prior to 1972 and there were no specific analyses for uranium. The figure demonstrates a trend seen for other radionuclides discharged in liquid effluents, that is, that in later years, more effluents were discharged to the seepage basins rather than directly to the Site streams.

Looking at the concentration of uranium in stream water should provide insight into the quantity of uranium that may have reached the Savannah River. If we assume that all alpha activity is due to natural uranium, then this leads to release estimates of about 0.005 to 0.01 Ci (~15-30 lb of uranium) per year from 1972-1975. Much lower levels of alpha activity in wastewater from the reactor areas were reported in other years when measurements were made. These levels are for the wastewater entering Site streams. The uranium activity that reached offsite locations in the Savannah River would be much less. In the early years, only gross alpha was measured in Tim's Branch but this provides a good indication of the movement of uranium and other alpha emitters in Tim's Branch. Figure 5-39 shows that alpha activity increased dramatically in Tim's Branch after receiving the 700-Area effluent. Then a significant drop off in activity occurred as the water moved downstream. Near the confluence of Tim's Branch with Upper Three Runs and then at a point in Upper Three Runs 2 mi before Upper Three Runs discharges into the Savannah River, the alpha activity was near the levels measured in Tim's Branch upstream of the M-Area discharge points. While Savannah River water was not monitored specifically for uranium, alpha activity was measured routinely since plant startup. There were no differences in alpha activity levels upstream and downstream of the plant, even in the late 1960s, when the largest releases of uranium occurred from the M-Area.

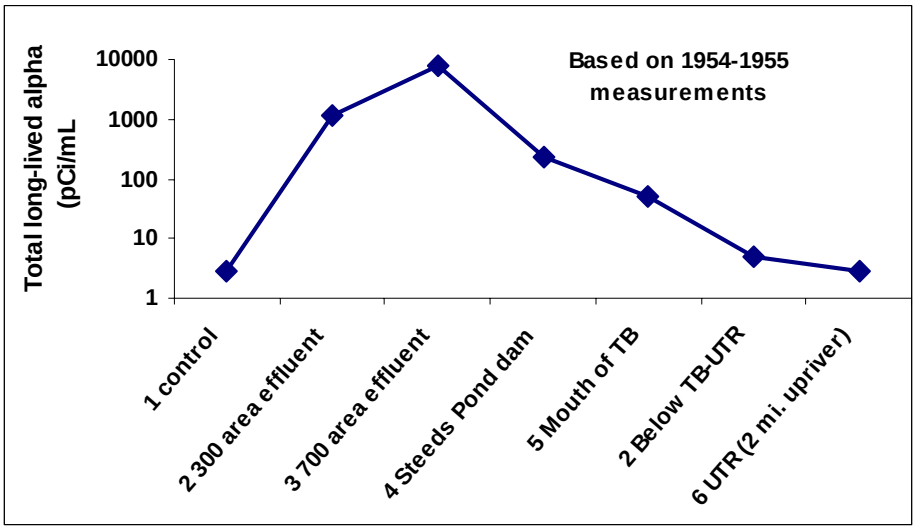

Figure 5-39. Alpha activity measured at various locations in Tim's Branch in 1954 and 1955. Alpha activity increased dramatically in Tim's Branch after receiving the 700 area effluent, then showed a significant drop off as the water moved downstream.

Studies have shown that uranium has deposited in the sediments of the streambed, thus accounting for the dramatic decrease in water with distance downstream. As early as June 1954, a study was underway to determine the distribution of uranium, released from the 300 and 700 areas, which had been removed from the water of Tim's Branch (Horton 1955a). The study included the determination of the distribution of uranium in the streambed, the uranium absorption by vegetation growing in and surrounding the stream, and the uranium content of the surrounding groundwater. There were 11 sampling stations at various locations. Horton 1955a contains complete soil sample data

Analysis of the collected data revealed no significant difference between the uranium content of the streambed at any of the sampling depths from 1 in. up to and including 6 to 9 in. There is a significant decrease in the uranium content of the samples collected below a depth of 9 in. in the streambed or at the water table at a distance of 6 ft from the edge of the stream. The maximum concentration of uranium in the streambed was at a location about 5 yd below the 300-Area discharge. The uranium content of the groundwater varied with sample location. At one point, uranium was detected at a distance of 45 ft from the edge of the stream. The uranium content of vegetation growing in the stream water was significantly higher than the uranium content of the vegetation growing near the stream. No uranium in vegetation was detected as the sampling locations became more removed from the effluent discharges.

SUMMARY

The Site had a fairly broad effluent monitoring program for releases of key radionuclides from the main facilities onsite to the streams and seepage basins. Measurements of alpha, beta, and some tritium were made from the beginning of operations and a preoperational survey helped to establish background levels of alpha and beta activity in some of the Site streams and in the Savannah River. We located many early records of measured concentrations and effluent flow to the streams for use in reconstructing past releases. The documentation, in the form of weekly and monthly reports, was quite extensive and there has been a great quantity of information to review.

To develop the surface water source terms, the process of screening using the NCRP methodology has focused our efforts and resources on those radionuclides that potentially are the largest contributors to radiation dose to those living offsite during the operational years at the SRS. Tritium, ^{137}Cs and ^{90}Sr are the main radionuclides of concern from past releases to surface streams that eventually reached the Savannah River, and we estimated detailed source terms for those radionuclides. Other radionuclides that also may be important depending upon the exposure pathways include ^{131}I, the activation products, ^{60}Co, ^{32}P, and ^{65}Zn, and uranium releases from the M-Area.

Surface water releases of radionuclides were highest in the early to middle 1960s and decreased into the 1980s. Our median estimate of release of tritium for all years (1952 through 1992) is 1.8 million Ci, with the 5th and 95th percentiles of the distribution of 1.3 million and 2.5 million, respectively. The median estimate of the total ^{137}Cs for all years is about 250 Ci with the 5th and 95th percentiles of the distribution of 100 and 600 Ci ^{137}Cs. There is overall general agreement between our reconstructed ^{137}Cs release estimates to the Site streams based on the original measurement data and supported by weekly and monthly reports from the 1960s and 1970s and the annual total reported by SRS. The median estimate of the total ^{90}Sr for all years is about 100 Ci with the 5th and 95th percentiles of the distribution of 45 and 250 Ci ^{90}Sr.

As would be expected, there is more uncertainty associated with release estimates in the 1950s and 1960s than in later years because of improvements in monitoring capabilities and techniques and in methods to prevent the release of materials from the reactors and processing facilities. The reactors were the source of the majority of radionuclide releases to surface water, primarily because most surface water releases came from the disassembly basins in the reactors areas, while releases of liquid effluents from the separations areas were discharged into storage tanks and seepage basins from the start of operations and not directly to the streams.

Nature, in the form of precipitation and the SRS swamp, played an important role in surface water releases. The year of highest releases to streams for tritium and ^{137}Cs (1964) was also the year with exceptionally high rainfall that increased runoff and led to heavy flooding of the SRS swamp. While the swamp was flooded cyclically about 20% of the time, the swamp also affected the mobility of some radionuclides flowing in stream water to the Savannah River by physical and chemical processes.

Finally, it is difficult to understand the full meaning of these release estimates before completing subsequent studies, where radiation doses and risks associated with these releases can be evaluated.

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CHAPTER 6

OTHER SOURCES OF CONTAMINATION IN THE ENVIRONMENT

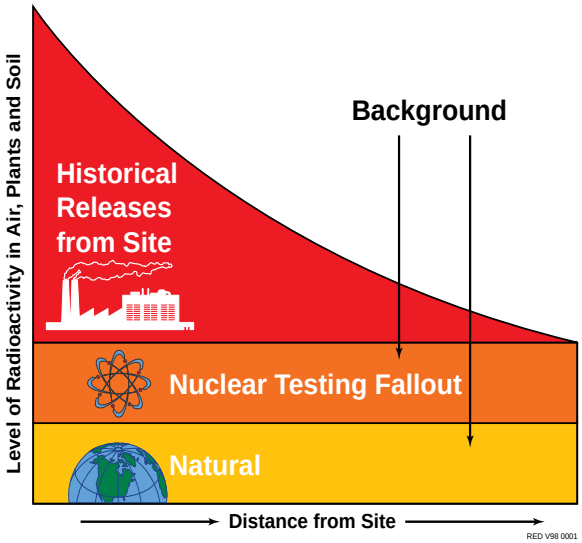
ABSTRACT

This chapter provides information about sources of radioactive contamination other than the Savannah River Site (SRS). This topic is important to dose reconstruction because other sources contribute to the total amounts of contamination measured by environmental monitoring programs. Definitive identification of the source of contamination is sometimes difficult. The most important non-SRS source of radionuclide contamination has been the aboveground testing of nuclear weapons in the 1950s and 1960s. Data from the Health and Safety Laboratory of the Atomic Energy Agency, the U.S. Geological Survey, and a Canadian fallout-monitoring program are provided to illustrate trends over time and space. In March 1955, a high deposition of radioactivity occurred in the vicinity of the SRS that appears to have resulted from rainout of debris from a nuclear test several days earlier in Nevada.

INTRODUCTION

The term contamination usually refers to unwanted radioactive or other material in the environment or other place where it may make surfaces or equipment unsuitable for some specific use. In this chapter we use the term contamination in a more general sense to denote the presence of certain radionuclides and chemicals in the environment. Some radioactive materials occur naturally in the environment, and others are produced by human activities. In Phase I of this study, other local man-made sources of contamination in the area of the SRS were identified (Stetar et al. 1995). However, the most significant contributor to radioactive contamination of the United States in the 20th century has been the detonation of nuclear weapons, which produced airborne radioactive debris called weapons fallout.

The term background often refers to amounts of materials that occur naturally in the environment, without input from human activities. Some radionuclides (such as tritium) have multiple sources: they occur naturally, are a component of weapons fall-out, and are also released from facilities such as the SRS. For the purposes of this chapter, "background" radionuclide concentrations in the environment around the SRS are defined as those levels occurring from *sources other than the SRS* (see schematic, right). Some background concentrations have changed over time and space, mainly because of trends in the deposition and decay of weapons fallout.



The first section of this chapter provides a general overview of weapons fallout, including the time trend of fallout deposition. The next section reviews a contamination incident that occurred in the SRS area in 1955, which has been determined to be most probably a hot-spot fallout deposition event. Natural, accidental, and other facility sources for radionuclides of interest are discussed next. Background levels of nonradiological chemicals of interest and associated monitoring data are presented in other chapters.

WEAPONS FALLOUT

Fallout is the radioactive debris that is subsequently deposited on the earth from a nuclear weapon that is detonated above ground. Weapons fallout has been distributed worldwide, sometimes into the upper layers of the atmosphere, gradually depositing to the earth and/or decaying to stable elements that are no longer radioactive. Some of the most important components of fallout have relatively long half-lives of 30 years or more.

Fallout was detected by historical environmental monitoring at the SRS. Environmental measurements conducted by SRS personnel are discussed in other chapters of this report, and the reader is referred to those sections to examine trends in the data. Separating radioactivity contributed by releases from the SRS from weapons fallout is difficult for some radionuclides.

It is not within the scope of this project to reconstruct the regional doses from weapons fallout in the region of the SRS. However, an understanding of the general trends in weapons fallout over time and space is necessary to interpret historical monitoring data.

Spatial and Temporal Trends

The first nuclear explosion occurred in 1945 in New Mexico. Fallout became recognized as a possible public health problem in the mid-1950s. In 1958, the U.S., United Kingdom (U.K.), and the Soviet Union, who were all conducting aboveground testing, declared a moratorium. However, in 1961, without advance warning, the Soviet Union unilaterally broke the moratorium agreement and exploded about 50 devices. The U.S. responded and major escalation of testing occurred. In early 1963, the U.S., U.K., and the Soviet Union signed a test ban agreement. France and China continued to conduct aboveground tests (particularly in 1968–1970) but not on as large a scale as the other countries had before 1963. In general, the highest levels of weapons testing fallout in air and deposition samples in the U.S. occurred during the years 1962 through 1964.

Spatial trend refers to how a material is distributed in the environment, for example, with distance away from the facility. Temporal trend refers to how the concentration of a material changes over time.

Nuclear fallout was injected both into the troposphere (lower atmosphere) and stratosphere (upper atmosphere). These two layers of the atmosphere circulate somewhat independently, with material injected into the stratosphere being distributed globally. The size of the explosion and the height of the detonation are the primary factors in whether fallout occurred close or far away from the bomb test location. The larger megaton bombs injected a large fraction of their fallout into the stratosphere. Some fallout debris stayed in the stratosphere for many months, coming down into the lower atmosphere during seasonal mixing of the atmospheric layers. Essentially all the debris

from smaller (kiloton) bombs was deposited within a few months following injection into the atmosphere because most fallout from these bombs was confined to the troposphere.

A long-term view of fallout deposition is provided by measurements made by the Health and Safety Laboratory (HASL) of New York City. This laboratory is now called the Environmental Measurements Laboratory. Operated by the U.S. Department of Energy (DOE) and its predecessors (the Energy Research and Development Administration and the Atomic Energy Commission), HASL was not independent of the DOE complex, but it was not involved in operating the SRS. HASL was known for its high-quality research and was instrumental in developing many techniques for radiochemical analyses. As opposed to a DOE production site's compliance-based monitoring program, the objective of the HASL environmental measurements program was to study the spatial and temporal distribution of natural and man-made radionuclides in surface air, deposition, and soil. Therefore, their historical results are very relevant to a dose reconstruction study.

In 1977, HASL provided a final tabulation of monthly monitoring of the fission product strontium-90 (^{90}Sr) deposition from a global network of stations started in 1954. During the early periods of nuclear weapons testing, ^{90}Sr was singled out as the most critical long-lived fission product produced by weapons testing, from the standpoint of dose to man. A gummed-film collector was operated by HASL from 1952 through 1959, and total beta radioactivity was measured. Because a more direct isotopic measurement was considered necessary, deposition collectors were added, first at HASL in New York City in 1954 and later at other locations. The collector was a high-walled stainless steel pot with an exposed area of 0.076 m^2. Collected precipitation and fallout material was transferred from the pot to laboratory glassware for evaporation and analysis for ^{90}Sr. A more practical method for other locations was developed that involved a funnel and ion-exchange column system with an exposed area of 0.072 m^2. With this collector, HASL was able to enlist the help of civilian and military weather installations and other government facilities throughout the world. This column system was used in Columbia, South Carolina, between April 1957 and June 1976. In all, 177 stations were established under the monthly radiostrontium fallout program. By 1976, 105 sites were operating; infrequent atmospheric testing and a subsequent decline in the stratospheric reservoir resulted in barely detectable levels of ^{90}Sr. In anticipation of a reduction in the number of stations and a longer collection time, the entire history of monthly strontium deposition data was presented in a summary report (HASL 1977).

Figure 6-1 shows the fallout depositions of ^{90}Sr in Columbia, South Carolina and New York City for 1954 through 1976. Table 6-1 provides the annual totals; individual monthly data are also available in HASL (1977). The monitoring record at New York City is quite complete; the only month of missing data was January 1954. At Columbia, missing data were more frequent (Table 6-1). Total deposition in Columbia for the years 1959, 1963, 1970–71, and 1973–1976 was estimated by assuming that deposition in months of missing data was the same as the average of the months that were monitored in the same year (Table 6-1). These data can be accessed directly in the Excel® workbook in the spreadsheet tab called "annual" by clicking on the following hyperlink: sr90dep.xls.

The years 1962 through 1964 were the highest for fallout deposition in the U.S. The total deposition at New York City was about 50% higher than that measured in Columbia. The fallout deposition relative to the highest year (1963) is given in Table 6-1 for the Columbia location. This relative trend is useful for interpreting time trends in environmental measurements collected by the SRS program.

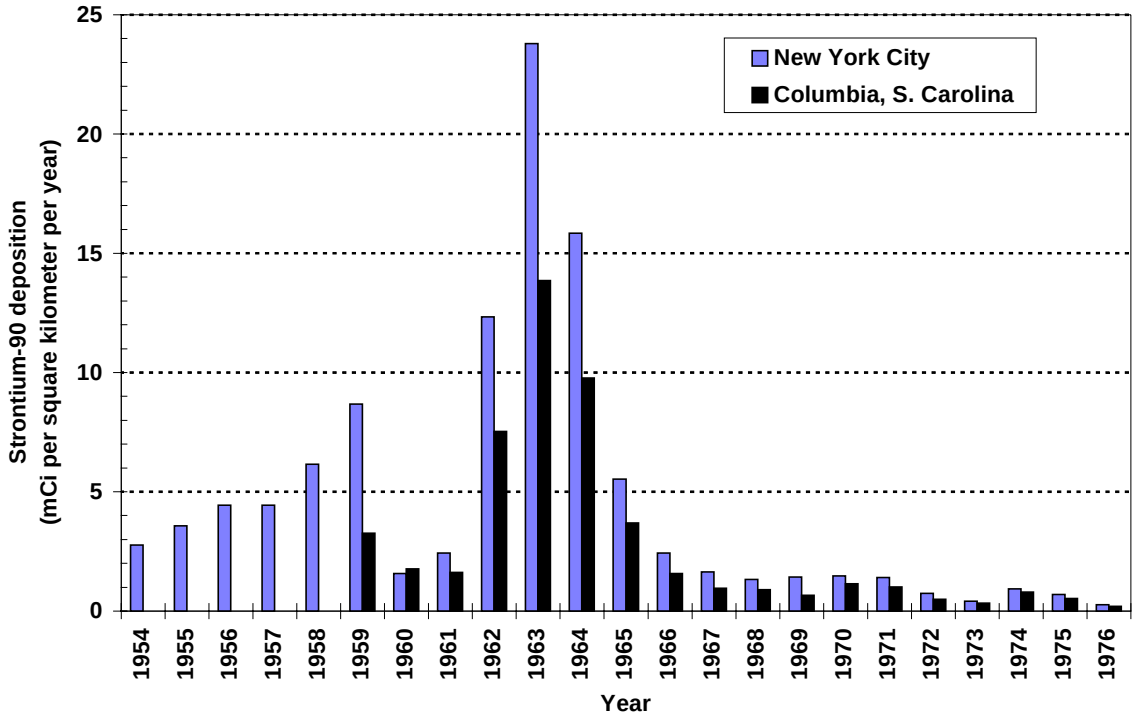


Figure 6-1. Deposition of the fallout radionuclide ^{90}Sr in New York City and Columbia, South Carolina (HASL 1977). Data in spreadsheet sr90dep.xls.

In the air monitoring chapter of this report (Chapter 8), the gross beta activity concentrations in particulate air samples within a 100-mi radius of the SRS are presented for a time interval that includes the peak fallout years (Figure 8-3). The highest monthly average concentrations of gross beta activity in air are observed in the spring of 1963, decrease about seven-fold in the fall of 1963, and increase again slightly in the spring of 1964. SRS monitoring data are compared with those from the Public Health Service monitoring of air in Columbia, South Carolina. The vast majority of beta activity in offsite air during these years is attributed to fallout from weapons testing.

Table 6-1. Deposition of ^{90}Sr in Columbia, South Carolina and New York City (HASL 1977)

Year	Reported deposition (mCi km^{-2})		Number of months of missing data for Columbia	Estimated deposition for Columbia (mCi km^{-2})	Ratio of annual deposition in Columbia to peak in 1963
	New York City	Columbia			
1954	2.76				0.12[a]
1955	3.57				0.15[a]
1956	4.43				0.19[a]
1957	4.44				0.19[a]
1958	6.16				0.26[a]
1959	8.68	2.17	4	3.26	0.23
1960	1.58	1.77	0	1.77	0.13
1961	2.43	1.62	0	1.62	0.12
1962	12.33	7.53	0	7.53	0.54
1963	23.79	12.71	1	13.87	1.00
1964	15.85	9.76	0	9.76	0.70
1965	5.53	3.7	0	3.70	0.27
1966	2.43	1.57	0	1.57	0.11
1967	1.64	0.96	0	0.96	0.07
1968	1.32	0.90	0	0.90	0.06
1969	1.43	0.67	0	0.67	0.05
1970	1.48	1.05	1	1.15	0.08
1971	1.41	0.93	1	1.01	0.07
1972	0.75	0.50	0	0.50	0.04
1973	0.42	0.30	1	0.33	0.02
1974	0.93	0.60	3	0.80	0.06
1975	0.69	0.49	1	0.53	0.04
1976	0.26	0.08	7	0.19	0.01

[a] Inferred from New York City measurements.

Tritium, a key radionuclide released from the SRS, has been increased well above its naturally occurring environmental concentrations because of nuclear weapons testing. A long-term perspective on this impact is provided by data collected by the Canadian Fallout Monitoring Program and published in Létourneau et al. (1994). These data can be accessed directly in the Excel® workbook in the spreadsheet tab called "data" by clicking on the following hyperlink: canada.xls.

Figure 6-2 illustrates the temporal trends in tritium concentrations in precipitation and in river water for 1955 through 1993 and shows the effect of weapons testing on environmental levels. As with the ^{90}Sr deposition data from Columbia, South Carolina, the peak year for tritium in precipitation was 1963, followed by 1964 (0.53 of 1963) and 1962 (0.33 of 1963). The tritium in river water shows a lag from the precipitation peak, with the highest levels measured in 1964–1965 in this Canadian valley at about 160 Bq L^{-1} (4300 pCi L^{-1}). Average concentrations in surface streams in the United States also reached about 4000 pCi L^{-1} in 1963 (NCRP 1979).

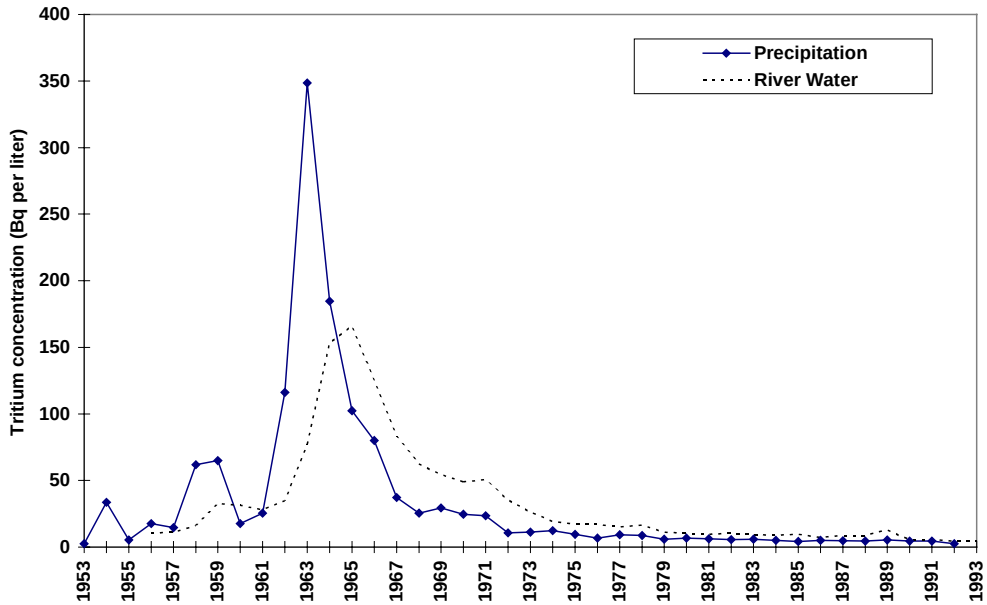


Figure 6-2. Annual average concentration of tritium in precipitation and river water from the Ottawa Valley, Canada (data from Létourneau et al. 1994, in spreadsheet canada.xls). The figure illustrates the effect of the injection of tritium into the atmosphere by the detonation of nuclear devices that increased concentrations in surface water by over 100 times the natural amounts in the early 1960s. Concentrations have gradually declined because of radioactive decay and mixing of atmospheric tritium into the oceans. Note the similar time trend as ^{90}Sr deposition in the U.S. shown in Figure 6-1.

Closer to the SRS, the U.S. Geological Survey monitored tritium in surface water of South Carolina streams, and others, since the early 1960s. The results and analytical procedures for this sampling program are tabulated in Wyerman et al. (1970). These data can be accessed directly in the Excel® workbook in the spreadsheet tab called "data" by clicking on the following hyperlink: streams.xls.

The data show the variations in stream tritium concentrations, caused principally by weapons fallout, as well as by seasonal, latitudinal, and continental effects. The four closest monitoring locations to the SRS are the Kissimmee River near Okeechobee, Florida; the Apalachicola River at Chattahoochee, Florida; the Savannah River near Clyo, Georgia; and the Neuse River near Vanceboro, North Carolina. The tritium content of surface waters varies greatly depending on the deposition of tritium as well as a variety of local hydrological factors. However, the impacts of weapons fallout as well as the SRS is clear (Figure 6-3).

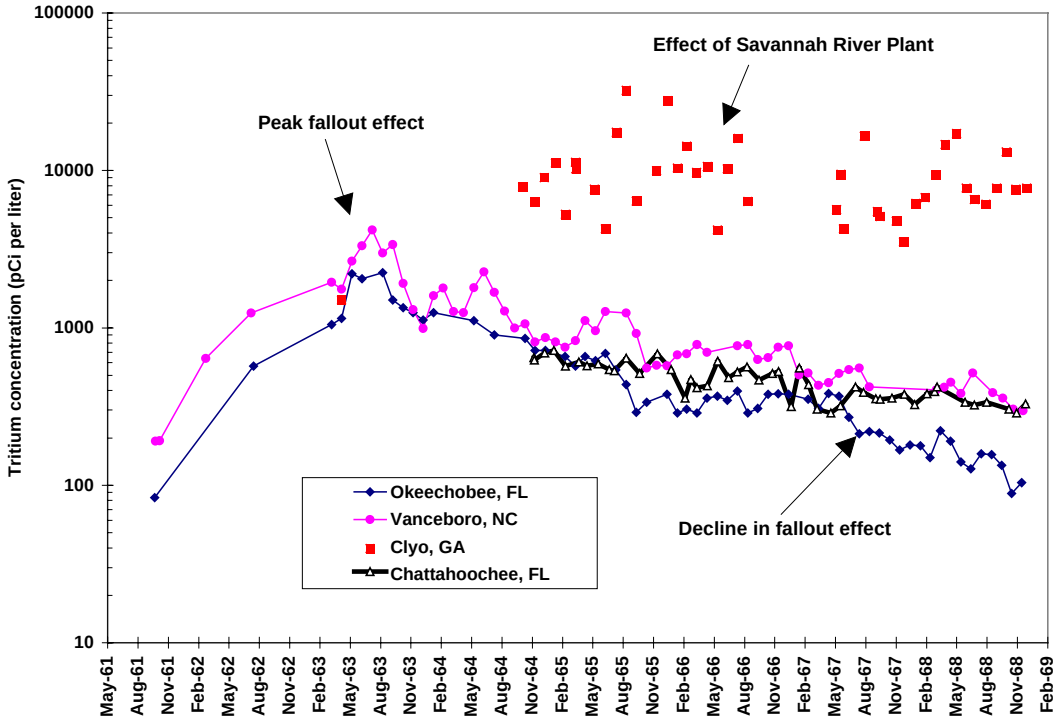


Figure 6-3. Tritium in four surface water streams in the southeastern U.S. Data collected by the U.S. Geological Survey (Wyerman et al. 1970), tabulated in spreadsheet streams.xls. The Clyo, Georgia, sampling location is downstream of the SRS.

The peak fallout years have been highlighted in the previous discussion. However, weapons fallout was detected into the following two decades, sometimes with better characterization of the material because of improved monitoring techniques. For example, the 1977 annual environmental report of the SRS (Du Pont 1978) notes an "extensive" special monitoring program undertaken in September following a Chinese atmospheric test. Fresh fallout was detected by a variety of devices 9 days after detonation. The 1978 annual report also noted that monitoring was conducted for additional Chinese tests.

In addition, the big-picture trends do not reflect the extensive spatial and temporal variability in fallout deposition, especially from the early lower-yield weapons tests that did not mix into atmosphere as extensively as later higher-yield tests. A contamination incident that illustrates the spotty distribution of weapons fallout, particularly in the 1950s and earlier, is reviewed in the next section. This incident has received much public scrutiny and interest. It also serves to underscore the importance of identifying other sources of radioactive contamination of the environment in any dose reconstruction study.

Contamination Incident at the Savannah River Plant, March 1955

Following a light rain between 2:00 and 3:00 p.m. on March 14, 1955, widespread ground contamination was detected at the Savannah River Plant (SRP, former name of Savannah River Site). The ground contamination was high enough that employees walking outside after the rain set off shoe monitors when they entered plant buildings (Sanders 1985). The Health Physics

Regional Survey Group completed an extensive survey the following day using portable radiation monitors.1 An oval area of radioactive surface contamination approximately 50 mi (80 km) long running generally from west to east, was deposited in the area of the SRP (Figure 6-4). Surface winds at the time of the deposition by light rain were from the north and northeast as reported by the weather station in Augusta (Marter 1985a).

The SRS environmental monitoring program detected high radioactivity in all media in the spring of 1955. The data were reported and analyzed in the semiannual report (Alexander and Horton 1956) and were discussed in the weekly environmental reports (Du Pont 1955b) as well as various internal memoranda (e.g., Mirshak 1983). Some of these SRS documents are discussed in more detail in a later subsection of this chapter. Additional original documentation for operational activities at this time was retrieved from archives of a senate investigation (Du Pont 1955a) and is reviewed later in this chapter.

Outside Scrutiny of 1955 Incident in the 1980s

In the 1980s, the 1955 contamination incident received additional scrutiny in a more public arena. This may have resulted partly because of the increasing declassification of historical documents. A brief chronology was outlined in an SRS internal memo titled "Mythical Reactor Accident at SRS on March 14, 1955" (Du Pont 1987):

- March 14, 1955 — Weapons test fallout arrives at SRS
- March 16, 1955 — Local media announcement of fallout
- 1956 — Arrival of fallout described in environmental monitoring semi-annual report for January–June 1955
- 1973 — Environmental monitoring reports declassified
- December 1984 — SRS learns about Alvarez and Franke paper
- January 1985 — Alvarez and Franke paper and Marter rebuttal presented at Health Physics Society meeting
- 1985 — Alvarez and Franke paper published in Ambio
- April 1986 — Chernobyl accident
- May 1986 — Formal rebuttal of Franke and Alvarez published in *Health Physics*
- May 1986 — Alvarez and EPA call for Federal investigation of "accident"
- May 1986 — Senator Thurmond requests an investigation by Armed Services Committee
- July 1986 — Senate probe finds no SRS accident.

[1] The type of radiation survey meter used for this special survey was a Thyac (Sanders 1985). These meters are sensitive to both penetrating beta and gamma radiation. Notes on radiation survey log sheets (Du Pont 1955) indicate that this instrument had a probe with a window that could be either open or closed, to shield out beta radiation. Routine ionization chambers used to detect penetrating radiation at various locations onsite and offsite were off-scale (14 mrep per week) when collected 1 week after the fallout. A survey made around 735-A with a gamma scintillometer 1 hour after the fallout showed a dose rate of 0.3 mrep per hour (Alexander and Horton 1956).

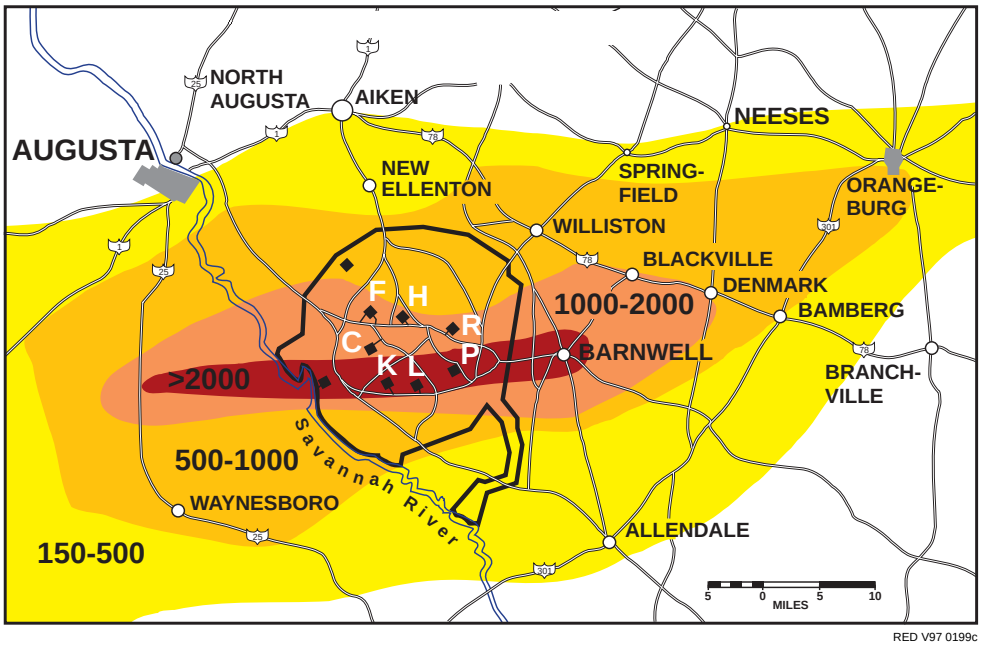


Figure 6-4. Radiation levels (counts per minute, cpm) measured with portable Thyac survey instruments on March 15, 1955 (redrawn from map in Alexander and Horton 1956). The darkest zone in the figure represents a region where measurements were more than 2000 counts per minute. Other zones are 1000–2000 cpm, 500–1000 cpm, and 150–500 cpm. Original documentation for this field survey was not located during our document search.

Alvarez and Franke Allegation and SRS Rebuttal. In a paper presented at a poster session on January 8, 1985, at the Health Physics Society midyear meeting and published in the proceedings, Drs. Bernd Franke and Robert Alvarez (1985a) claimed that the radioactive contamination in March 1955 around the SRS had resulted from an SRS reactor accident. In addition, the authors claimed that elevated offsite radiation levels for the 1954-1969 period were caused by atmospheric releases of radioactive materials from SRP (Franke and Alvarez 1985a). The first point will be discussed in this section.

With regard to the March 1955 incident, Drs. Franke and Alvarez (1985a) summarize the extent of contamination measured by SRS personnel that was documented in the SRS semiannual environmental report (Alexander and Horton 1956). However, Franke and Alvarez maintain, "As the center of the contamination was located around the reactors K, L, and P they could be considered as a more likely source of the contamination rather than rainfall deposition of a Nevada weapons test." The decay characteristics of the contamination, although consistent with fresh fission products, could also be explained by a release of 69% of ^{133}I (half-life 20.8 hours) and 31% ^{131}I (half-life 8.04 days). They claim that these nuclides "would be standard for a reactor incident. In all likelihood a major reactor accident happened at SRP on March 14, 1955."

An SRS critique of the first Franke and Alvarez paper (Marter 1984) was distributed at the same Health Physics Society midyear meeting, and a discussion session was held with conference attendees. According to McEnroe (1985), overwhelming support was demonstrated by the professional health physics community for the SRP position on the Franke and Alvarez paper. Numerous technical inaccuracies were noted.

A slightly expanded version of the Franke/Alvarez paper was published in the journal *Ambio* (Franke and Alvarez 1985b). There were some additions to the section that addressed the March 1955 contamination incident. A cumulative dose of 12 mrem was estimated, based on an exposure rate of 0.3 mR h^{-1} measured 1 hour after the rainfall (Alexander and Horton 1956) and (presumably) the observed rate of decay. In this article, they stated that the observed decay could be attributed to "halogens (iodine and bromium isotopes) from SRP reactor fuel at about 100 hours after unloading from the reactor." The authors point out that detailed records held by the SRS that might indicate if an unusual incident had occurred during this period were not publicly available.

Some review of the Teapot Hornet atmospheric nuclear test conducted at the Nevada Test Site is included in the *Ambio* article (Franke and Alvarez 1985b), probably in response to the arguments made by the SRS. Citing List (1956), which is reviewed later in this chapter, Franke and Alvarez (1985b) notes that the southern-most trajectory of radioactive debris from Teapot Hornet actually passed over North Carolina. Highest contamination measured from a gummed-film monitoring network was in Louisville, Kentucky, whereas levels in Atlanta, Georgia, and Charleston, South Carolina, were over three times less. Applying a ratio of empirical measurements at different locations, the authors calculated that more than 1.5% of the total fission products from the Teapot Hornet test would have to have been deposited in the vicinity of the SRS, which would be unlikely. Only 0.4% of the total fission products from the Upshot-Knothole weapons tests in 1953 had been deposited over the entire U.S. by the second day after the shots.

The SRP has always maintained that the gross contamination in March 1955 was not from the SRS, providing a formal rebuttal in the journal *Health Physics* (Marter 1986).

The main points made by the SRS personnel were

- Radiation monitoring data from the reactors (moderator monitors, process monitors, and continuous exhaust-stack air monitors) indicated no unusual activity during the month of March 1955. Investigations at other SRS facilities showed no unusual release of radioactivity.
- Recordings made by continuously operating environmental air monitors showed deposition of the activity in widely separated areas of the SRP site within a 30-minute period (Sanders 1985). This is not consistent with a site release from a single location. Surface winds of 10 mph were insufficient to disperse the activity that widely if a plant reactor were the source of the activity. The winds did not change direction 180 degrees, which would have been necessary to distribute the activity from the SRS in the observed pattern.
- The probable source was fallout from an atmospheric nuclear weapons test called Teapot Hornet that occurred in Nevada at 0820 hours, eastern standard time (EST), on March 12 (Carter and Moghissi 1977).
- The radioactive decay of activity was very consistent with fresh weapons test fallout at 2 days post-detonation. A mixture of 31% ^{131}I and 69% ^{133}I isotopes theorized by Franke and Alvarez is not supported by physical or chemical processes, and other short-lived radionuclides of iodine would be present if the source were an SRP reactor. Accidental iodine release from an SRS reactor, or from the chemical processing plant (as occurred

in 1961), would be expected to occur over a number of days. Radionuclides like ^{99}Mo and ^{239}Np, which were identified in precipitation from the March 1955 hotspot, would not be released from an SRS reactor except in a major fuel meltdown. In that case, long-lived radionuclides would have also been released, and these were not detected in the March 1955 fallout.

- Greater than 80% of the beta radioactivity measured in vegetation and rainwater following the fallout deposition was from nonvolatile beta-emitting radionuclides and less than 20% was radioiodine.

In response to a media request, Marter prepared an additional explanation (Marter 1985a) including a reference that documented numerous other fallout hot spots at other times (Sanders 1985) and an early attempt to identify the contributing radionuclides in the March 1955 fallout (Kinard 1955). Sanders shows that fallout hot spots were not uncommon, tabulating 31 incidents between 1951 and 1958 that illustrate localized surface contamination caused by atmospheric precipitation of nuclear fallout from weapons testing. These localized contamination incidents became less frequent after 1957 with the testing of larger yield bombs that carried radioactivity to higher elevations where it remained longer and became more evenly distributed.

Apparently the Marter (1986) rebuttal had already been submitted to the journal *Health Physics* when the *Ambio* article appeared. Marter (1985b) addresses some additional points in the *Ambio* article that had not been made in the previous Health Physics symposium paper. Marter (1985b) states that the high deposition on gummed film in the other states (Illinois and others) was primarily dry deposition from surface winds, which followed a different trajectory than winds at 18,000 ft.2 The deposition around SRP was caused by a short rainfall originating at high altitudes. Marter (1985b) does not address the additional magnitude issues raised by Franke and Alvarez (1985b), that is, that the contamination levels and initial dose rates measured near the SRS were quite high.

Senate Armed Services Committee Investigation. In July 1986, the Senate Armed Services Committee released the report of a staff investigation, conducted at the request of Senator Strom Thurmond, relating to the allegations of an unreported nuclear accident at the SRP on March 14, 1955 (Bott 1986). The investigation included discussions with U.S. Department of Energy officials, past and present employees of E.I du Pont de Nemours, Inc., and Bernd Franke and Robert Alvarez, as well as a thorough review of classified and unclassified documents. These documents, many recovered from archive facilities and not previously examined in this context, included health protection records (such as daily radiation survey logs, air sample logs, shift logs, and special work permits) and weekly operating reports. The report stated that there was no reason to conclude that an unreported nuclear accident at the SRP was responsible for the

[2] Marter is not entirely correct here. The highest depositions were associated with a different trajectory (see fallout maps later in this chapter). However, the high depositions in the midwestern states *were* associated with rainfall. In addition, recent reevaluations have shown the historical gummed-film monitors did intercept and retain both wet and dry deposition, although the collection efficiency was less with heavier rainfall (Beck et al. 1990). The estimated collection efficiency for dry deposition is believed to have been 20%. For a combination of dry and wet deposition, the collection efficiency of gummed-film ranged from 7 to 30% for high (>25 mm) and low (<0.75 mm) precipitation amounts in a 24-hour period, respectively.

radiation levels detected on March 14, 1955. This conclusion resulted from the following major findings:

1. The trajectories of the fallout from the Teapot Hornet test at the Nevada Test Site on March 12, 1955 were examined; one trajectory could have placed fallout over the vicinity of the SRP on March 14. Although the trajectory data for fallout from the test indicated passage several hundred miles north of the SRP, the Committee felt this did not discount the probability that fallout from the test was deposited at the SRP. The trajectories were not based on high altitude sampling of air, and the width of dispersion was not indicated.3
2. The direction and velocity of surface winds on the date in question were inconsistent with SRP as the source.
3. Monitoring at the SRP operating facilities did not indicate any release. There was no indication in any classified or unclassified documents of any unusual activity at the SRP on or about the date in question. There was an aggressive effort on the part of health physics personnel to determine the source of the contamination, although there was a presumption that it was related to a recent weapons test.
4. Increased radioactivity was not recorded on continuous air monitors until the rain began. Radioactivity from a SRP source would have been detected in the air before being deposited on the ground by rainfall.
5. The radioactivity decay pattern was consistent with fallout of approximately 2 days after detonation.
6. Other hot spots of this type have been measured in other parts of the country during the period of aboveground nuclear testing (e.g., see Sanders 1985, and gummed-film data reviewed later in this chapter).

The Senate Armed Services Committee noted that considerable resources had been expended in answering questions and in retrieving the documents and that further expenditure of government resources on this issue was unwarranted. They noted that the circumstantial evidence pointed to the possibility of an SRS release and that direct documentary evidence to the contrary had not been made available to the public. Consequently, they recommended that the unclassified record reviewed during this investigation be available for public review.

Since the investigation, those records have been maintained on a set of 14 microfilm cartridges, labeled "US Senate Investigation," in the public reading room at the University of South Carolina in Aiken. An index to the contents of the microfilm is available in paper form (Anonymous 1986). We reviewed the entire set of microfilms during two visits in November 1997 and February 1998, made in conjunction with other travel for this project. The films contain many hundreds of pages of SRS logbooks and onsite health physics monitoring records from the time period before, during, and after the March 1955 contamination incident. Most of the pages

[3] The meteorological trajectories are subject to error, particularly over regions of sparse data or in areas of variable or complex flow patterns. In general, over the U.S. for trajectories of the order of 1000 miles, the errors average 10–20% of the length of the trajectory (List 1956). The SRS is roughly 2000 mi from the Nevada Test Site. Thus, an error of 200 to 400 mi in the location of the trajectory might be expected at this distance from the detonation.

were routine operational monitoring records listed below for the following SRS areas: K-Area, L-Area, P-Area, R-Area, F-Area, H-Area, D-Area, and M-Area.

- Air sample log
- Monthly report
- Shift log
- Special work permit
- Special permit log time sheets
- Weekly report

These records were useful in documenting the arrival and extent of contamination that was deposited around and tracked into buildings on the SRS in March 1955. Each page of a logbook is numbered sequentially, so it can be confirmed that a record series is complete. For example, on microfilm roll number 1, the Radiation Survey Log Sheet pages numbered 3601 through 3800 contain surveys conducted between 3/12/55 and 3/23/55 in K-Area. Some of the key pages on these microfilms had been found previously during RAC's document search for this project. In addition, we made copies of 60 key pages from the microfilms that were particularly revealing as to the facts and the staff's interpretations of the March 1955 contamination event. These pages were assembled and given a SRS Phase II Database number (Du Pont 1955a). More detailed observations on the content of the microfilms are included in a later section of this chapter titled, "SRS Monitoring Relating to 1955 Incident."

Supporting Documentation for Evaluation of Source of Contamination

In our review of the 1955 contamination incident, the key references that documented the facts of the nuclear testing were obtained and reviewed. Operation Teapot, in the spring of 1955, was a series of 14 atomic detonations at the Nevada Test Site (List 1956). The fifth burst in the series, conducted on March 12, was named Hornet. Teapot Hornet was a 300-ft tower burst occurring at 13:20 Greenwich civil time (GCT), or 8:20 a.m. eastern standard time. The height of the top of the cloud was 37,000 ft above mean sea level. Only 4 of the 14 detonations in the Teapot series produced higher clouds of radioactive debris than the Hornet test on March 12, 1955. Trajectories for the atmospheric transport of the resulting debris were computed and compared to the fallout observed by gummed film at 87 locations in the continental U.S., 12 stations in Canada, and 6 stations elsewhere in North America (List 1956).

The immense heat from a nuclear detonation in the atmosphere produces a bubble of intensely hot gases. This buoyant bubble carries not only the debris resulting from the nuclear fission and the disintegration of the bomb casing and auxiliary equipment, but also great amounts of soil and dust, much of which becomes radioactive, that are drawn into the cloud. The term fallout refers to the deposition, on or near the surface of the earth, of radioactive particles resulting from the detonation of a nuclear device. It includes deposition from direct gravitational fallout of large particles, vertical currents and eddies in the atmosphere, and particles scavenged from the atmosphere and deposited by falling precipitation (which is referred to as rainout).

The movement of the atomic cloud is determined by the wind field, which can produce rapid dispersion or carry concentrated patches of debris for long distances in the upper atmosphere. The

deposition of particles from the atomic cloud is dependent on their size and density; these factors were not well known when the initial trajectories of atomic bomb debris were produced.

Meteorological Trajectories. The trajectories of the atomic cloud from weapons tests were computed from meteorological data from more than 150 stations in the U.S. and Canada. These stations reported the direction and speed of the winds every 6 hours. About one-half of these stations were equipped with electronic devices enabling soundings to be made at 6- or 12-hour intervals to very high altitudes even in the presence of clouds and also to measure the pressure, temperature, and relative humidity. The trajectories were prepared from maps from the National Weather Analysis Center (supplemented by additional site-specific wind reports).

For the March 12, 1955 burst, the trajectories indicate that the debris was carried eastward and fanned out to cover most of the central and eastern U.S. (Figure 6-5). A large precipitation area in the central states on March 14 resulted in deposition of debris from the Gulf states northward to the Great Lakes, and radioactivity continued to be associated with the precipitation area as it moved eastward on the following day.

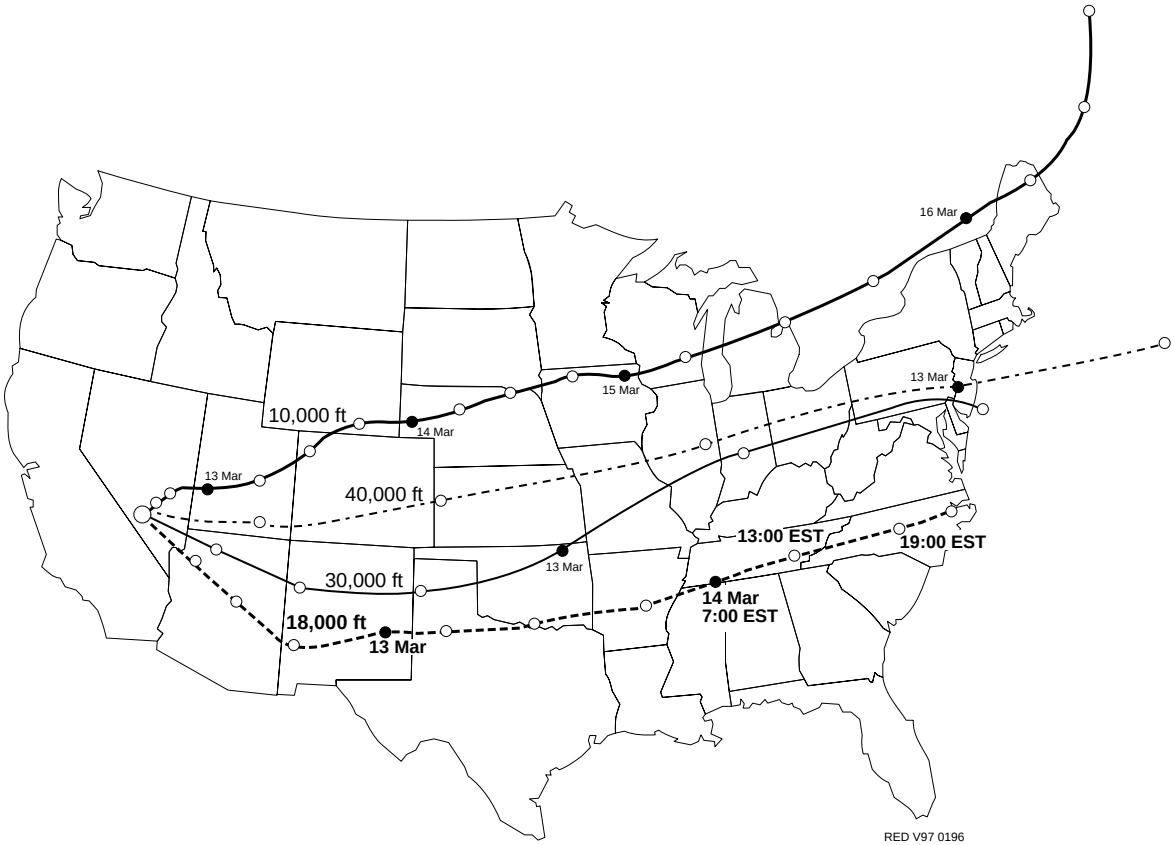


Figure 6-5. Trajectories of radioactive debris produced by the March 12, 1955, nuclear test Teapot Hornet (redrawn from figure in List 1956). The four trajectories represent the directions that atmospheric debris from the test traveled at different heights in the atmosphere. The center of the 18,000-ft trajectory passed over North Carolina between 1:00 and 7:00 p.m. EST on March 14 (shown as 13:00 and 19:00, in military time, on the figure). An error of several hundred miles is associated with the trajectory position at this distance from the blast. Rain occurred at the SRS between 2:00 and 3:00 p.m. EST.

Gummed Film Network. A simple and inexpensive monitoring technique was used in the 1950s to measure fallout of radioactivity to ground surfaces. A 1-ft square of gummed cellulose-acetate film was placed on a horizontal stand. Duplicate films were exposed at most stations on stands about 6 ft apart. The film was exposed for a 24-hour period and then mailed with a field data collection card to the Atomic Energy Commission's Health and Safety Laboratory (HASL) in New York City. At the HASL, the film was ashed and counted for beta activity. For the Teapot series, the average time between collection and analysis was about 3 weeks (List 1956). Even in the 1950s, the limitations and uncertainties of the gummed-film monitoring techniques were recognized. For example, List (1956) mentions

- Unknown efficiency of film during rain and snow
- Uncertainty in how film represents deposition on natural surfaces
- Possibility of concentration and re-deposition in dusty regions.

For the Teapot series, 18 of the northern stations were equipped with heated stands to melt the snow that might fall on the film. The closest stations to the SRS were located at Atlanta, Georgia, and Charleston, South Carolina.

Monitoring of weapons test fallout with gummed film led to the conclusion that precipitation was a major factor in depositing debris that was more than a day or two old. About 10 times more activity was found on film exposed during precipitation as on dry days. In general, the heavier the rainfall, the more material collected (List 1956). Although wet deposition was a scavenging process for fallout particles, recent reevaluations have shown that the collection efficiency of gummed-film monitors was less with heavier rainfall (Beck et al. 1990). The estimated collection efficiency for dry deposition is believed to have been 20%. For a combination of dry and wet deposition, the collection efficiency of gummed film ranged from 7 to 30% for high (>25 mm) and low (<0.75 mm) precipitation amounts in a 24-hour period, respectively.

Fallout Maps. List (1956) presents maps showing daily gummed-film fallout data at each location for 24-hour periods beginning at 12:30 GCT (7:30 EST); the map for March 14, 1955 is reproduced here as Figure 6-6. The units are millicuries (mCi) deposited per 100 mi^2. Areas with more than a trace of precipitation are shaded on the map. All radioactivity measurements were corrected to the expected radioactivity remaining on January 1, 1956, according to Equation (6-1):

$$A_t = A_0 \, t^{-1.2} \qquad (6\text{-}1)$$

where

A_t = the activity on January 1, 1956

A_0 = the activity extrapolated to 1 day after the most recent blast

t = the time in days between blast and January 1, 1956.

On March 14, 1955, the highest depositions of beta activity were recorded in the midwestern states of Illinois, Indiana, Michigan, and Wisconsin. These deposition data and the resulting map (Figure 6-6) do not include or consider the survey monitoring conducted by the SRS. The closest HASL gummed-film monitoring stations to the SRS were in Atlanta, Georgia, and Charleston, South Carolina.

Table 6-2 lists the daily fallout measured in Atlanta and Charleston in March of 1955. The data are from maps in List (1956). Values from only one station in Atlanta were presented; however two replicate results are given for Charleston during this period. There were seven separate bursts (No. 3 through 9 of the total series) in March of 1955, occurring on the 1st, 7th, 12th, 22nd, 23rd, and 29th (two bursts) of the month. All extrapolations of radioactivity remaining by January 1, 1956 were made by assuming that the fallout was from the most recent burst (List 1956).

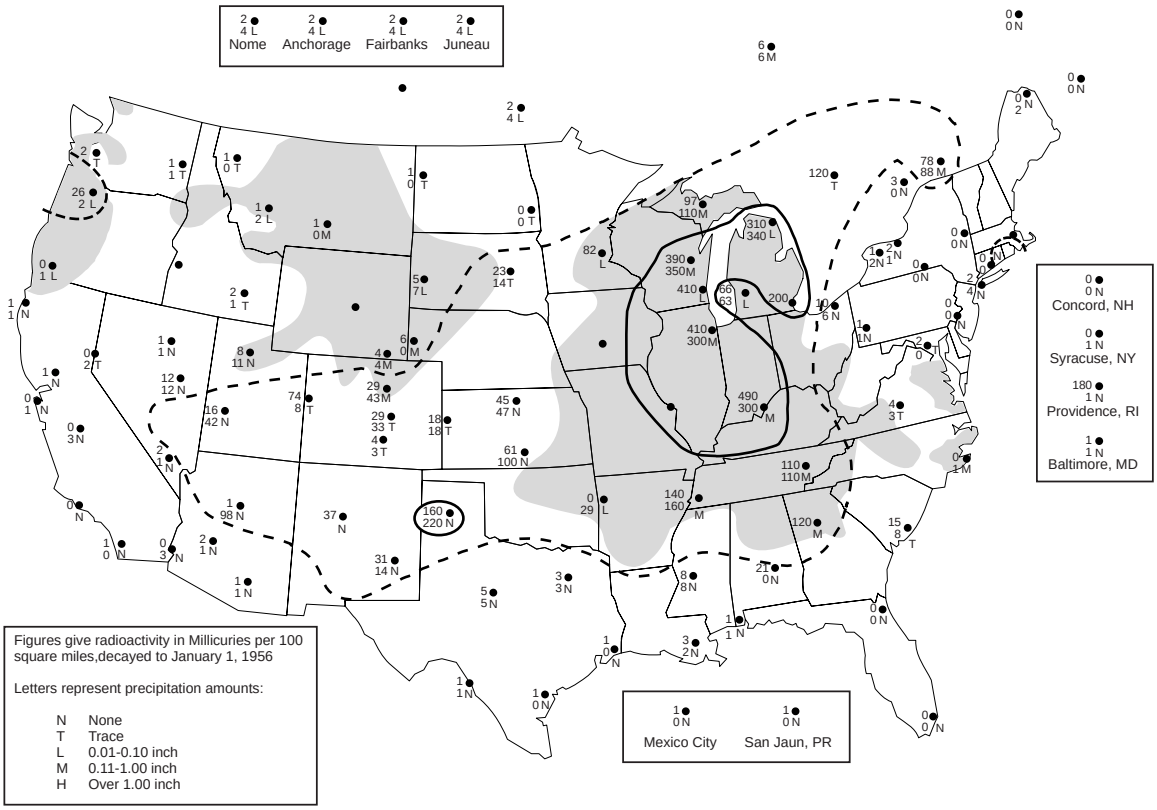


Figure 6-6. Radioactive fallout in the U.S. on March 14, 1955 (redrawn from figure in List 1956). The solid line outlines areas with more than 200 mCi/100 mi^2 on the most active film from each station in the area. The dashed line similarly outlines areas of more than 20 mCi/100 mi^2.

Table 6-2. Daily Radioactive Fallout (Beta Activity) in Atlanta, Georgia and Charleston, South Carolina in March 1955 (data from List 1956)

	Atlanta		Charleston		
Date in March[a]	Deposition (mCi/100 mi^2)[b]	Precipitation[c]	Replicate 1 (mCi/100 mi^2)[b]	Replicate 2 (mCi/100 mi^2)[b]	Precipitation
1	3	low	na[d]	na	na
2	3	none	2	1	none
3	1	trace	0	0	none
4	0	trace	0	0	none
5	na	na	0	0	none
6	na	na	4	3	trace
7	na	na	0	0	none
8	2	none	3	9	low
9	4	moderate	1	0	none
10	4	trace	0	1	none
11	na	na	2	1	none
12	1	moderate	0	0	none
13	12	low	0	0	none
14	120	moderate	15	8	trace
15	2	trace	na	na	na
16	0	moderate	na	na	na
17	1	none	0	0	none
18	na	na	3	4	low
19	2	moderate	5	3	high
20	na	na	88	0	none
21	9	low	6	5	trace
22	25	low	9	4	low
23	0	moderate	0	0	none
24	15	none	15	31	none
25	55	low	na	na	na
26	3	none	na	na	na
27	1	none	na	na	na
28	3	none	57	65	low
29	9	none	2	3	none
30	3	none	1	10	none
31	2	none	1	1	none

[a] Twenty-four hour period beginning 7:30 EST.
[b] Millicuries beta activity per 100 mi^2, corrected to activity on January 1, 1956.
[c] Low = 0.01 to 0.10 in.; moderate = 0.11 to 1.00 in.; high = over 1.00 in.
[d] na = not available.

Figure 6-7 plots the daily deposition data tabulated in Table 6-2. The highest deposition rate at these two stations occurred on March 14 in Atlanta. This was the same day of the observed

contamination at the SRS. The timing of the arrival of fallout in the area is also consistent with the predicted arrival based on trajectories from wind field data. Fallout at the 18,000-ft level was projected to occur at the longitude of the SRS between 1:00 p.m. and 7:00 p.m. EST on March 14. The light rain, which is believed to have carried the contamination to the ground, occurred between 2:00 and 3:00 p.m. on March 14 at the SRS.

High beta activity was seen in deposition at Atlanta, Georgia, on the same day in March 1955 that contamination was observed near the SRS.

The highest measured depositions of beta radioactivity from this test were recorded at locations in Kentucky, Illinois, Wisconsin, and Michigan. Two monitoring stations closer to the latitude of the SRS (at Amarillo, Texas, and Memphis, Tennessee) showed elevated depositions probably associated with the test. With the exception of the Texas station, all of the areas of highest deposition in the U.S. on March 14, 1955 were associated with low or moderate rainfall (Figure 6-6).

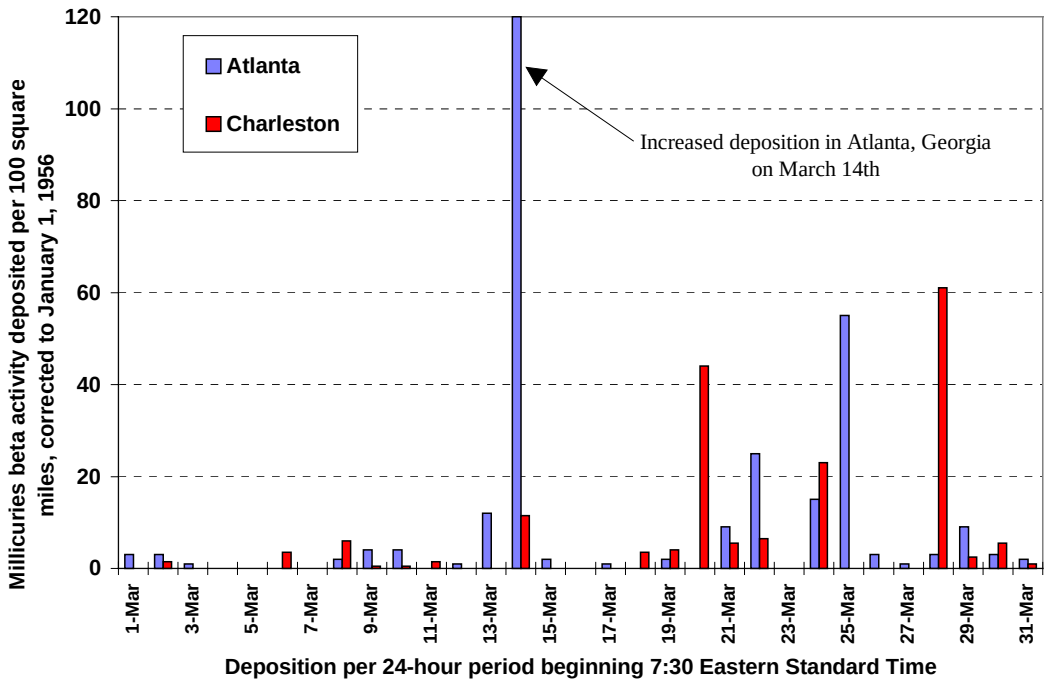


Figure 6-7. Daily deposition of beta activity in Charleston, South Carolina, and Atlanta, Georgia, in March 1955. Data taken from maps in List (1956) and presented in Table 6-2. The Charleston value plotted is the average of two replicates shown in Table 6-2.

Isotopic Composition of Fallout from Operation Teapot. The deposition measurements that were made on the gummed-film collections in the 1950s were all gross beta activity. The radiation survey conducted by SRS personnel measured total penetrating radiation, which would have included both gamma and beta-emitting radionuclides.

There are many different individual radionuclides that make up fresh weapons fallout; the composition varies with the type of nuclear test and with the time after detonation. Relatively recently, individual weapons tests have been analyzed, and an estimate of the various radionuclides produced has been published. The relevant report for the Teapot test series is Hicks (1981). For 30 times after the blast (10 times from 1 to 21 hours; 10 times from 1 to 300 days, and 10 times from 1 to 50 years), the following data are presented:

- The external gamma radiation exposure rate normalized to 1 mR per hour at 12 hours after the event, 1 m above the surface of the ground
- The deposition of each radionuclide in TCi m^{-2}
- The total deposition for all radionuclides in TCi m^{-2}.

Hicks (1981) presents the data for 128 radionuclides for the time interval of 1 to 300 days. By 1 year post-detonation, many of these radionuclides have decayed. At 1 year and beyond, the data are presented for 37 remaining radionuclides. Table 6-3 includes the data for the 33 radionuclides that contributed most to the external gamma exposure rate from widespread debris from the Teapot Hornet test. This list includes all radionuclides contributing at least 2% of the total exposure rate at 2 days or 5 days post-detonation. In addition, some longer-lived radionuclides were added to show those radionuclides making up 95% of the exposure rate after 1 year. The familiar long-lived radionuclides from weapons fallout, such as $^{137}Cs/^{137m}Ba$, $^{106}Ru/^{106}Rh$, and $^{144}Ce/^{144}Pr$, are relatively insignificant contributors to the total exposure rate at 2 days post-detonation. The data illustrate how the decay of short-lived radionuclides results in a rapid decrease in the total exposure rate from the debris in the first few days after the blast. The exposure rate versus time relationship for this blast is illustrated in Figure 6-8. The gamma exposure rate a week after the deposition around the SRS would have been less than one-half that observed on the third day after the blast when the survey was conducted.

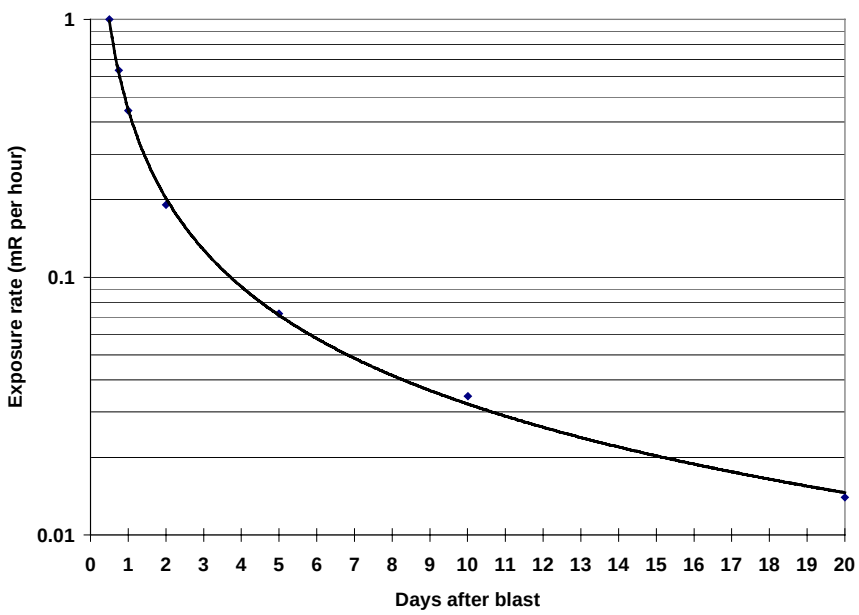


Figure 6-8. Exposure rate versus time after blast for the Teapot Hornet weapons test in Nevada (data from Hicks 1981). The curve shows the time trend for a hypothetical location at which the exposure rate was 1.0 mR/h at 12 hours after detonation.

Table 6-3. Isotopic Composition of Debris from the Teapot Hornet Test (Hicks 1981)

Nuclide	Time post-detonation					
	2 days		5 days		1 year	
	μCi m^{-2}	%	μCi m^{-2}	%	μCi m^{-2}	%
^{64}Cu	2.14	7.5	0.0435	0.4		
^{89}Sr	0.101	0.4	0.0971	1.0	0.000801	2.2
^{90}Sr	0.000692	0.0	0.000692	0.0	0.000674	1.9
^{90}Y	0.000281	0.0	0.000504	0.0	0.000674	1.9
^{91}Y	0.105	0.4	0.105	1.1	0.0015	4.1
^{95}Zr	0.0723	0.3	0.07	0.7	0.0015	4.1
^{95}Nb	0.0028	0.0	0.0067	0.1	0.00323	8.9
^{97}Zr	0.926	3.2	0.049	0.5		
^{97m}Nb	0.889	3.1	0.0473	0.5		
^{97}Nb	0.929	3.3	0.0493	0.5		
^{99}Mo	1.13	4.0	0.537	5.5		
^{99m}Tc	1.07	3.8	0.514	5.3		
^{103}Ru	0.193	0.7	0.183	1.9	0.000334	0.9
^{103m}Rh	0.193	0.7	0.183	1.9	0.000335	0.9
^{105}Rh	1.55	5.4	0.386	4.0		
^{106}Ru	0.0116	0.0	0.0115	0.1	0.00584	16.1
^{106}Rh	0.0116	0.0	0.0115	0.1	0.00584	16.1
^{131}I	0.701	2.5	0.563	5.8		
^{132}Te	1.62	5.7	0.854	8.7		
^{132}I	1.67	5.9	0.878	9.0		
^{133}I	2.64	9.3	0.245	2.5		
^{133}Xe	1.42	5.0	1.27	13.0		
^{135}Xe	1.91	6.7	0.0109	0.1		
^{137}Cs	0.000958	0.0	0.000958	0.0	0.000936	2.6
^{137m}Ba	0.000894	0.0	0.000894	0.0	0.000878	2.4
^{140}Ba	0.566	2.0	0.479	4.9		
^{140}La	0.333	1.2	0.461	4.7		
^{143}Ce	1.11	3.9	0.245	2.5		
^{143}Pr	0.183	0.6	0.236	2.4		
^{144}Ce	0.0126	0.0	0.0125	0.1	0.0052	14.4
^{144}Pr	0.0126	0.0	0.0125	0.1	0.0052	14.4
^{147}Pm	0.000219	0.0	0.0005	0.0	0.00145	4.0
^{239}Np	2.7	9.5	1.12	11.5		
Total of nuclides in this table	21.5	84.9	7.56	88.9	0.0343	95.0
Total of all nuclides in Hicks	28.5		9.77		0.0362	

Total Fallout from Teapot Series. Figure 6-9, from List (1956), shows the total fallout deposition from the Teapot series between February 18 through May 20, 1955, corrected to activity remaining on January 1, 1956. Most of the debris was carried by prevailing westerly winds. In general, fallout deposition decreased with distance from the Nevada Test Site. Total deposition in South Carolina, around 20 mCi/mi^2, was 5 to 10 times less than that deposited over large areas of Oklahoma, Colorado, and Utah. The Atomic Energy Agency estimated that less than 5% of the total beta activity released during the Teapot series was deposited in the U.S., exclusive of close-in fallout, from February 18 through May 20. This finding was consistent with that from two previous Nevada test series. Consequently, the residual activity remained in the atmosphere, either to decay over time or to be deposited to the ground with precipitation or with seasonal mixing of the layers of the atmosphere.

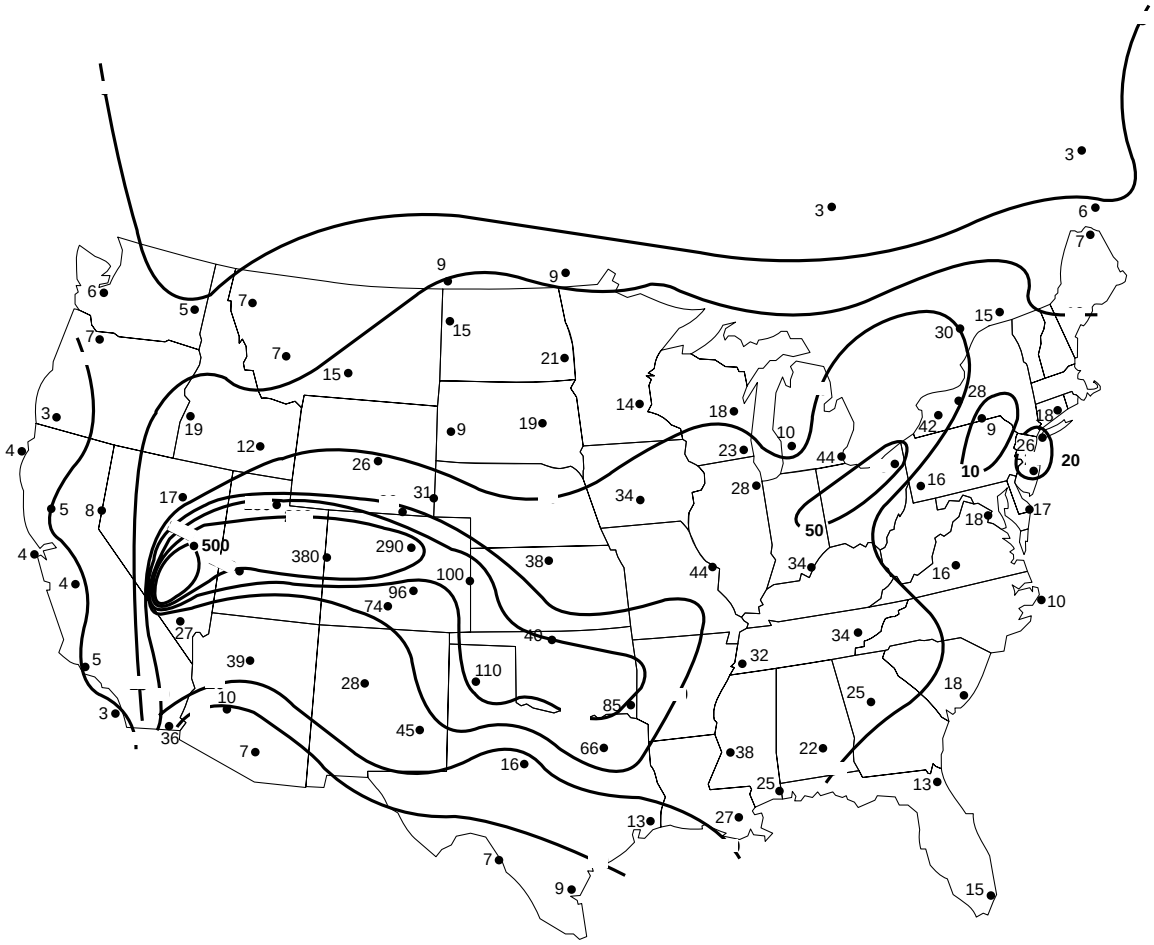


Figure 6-9. Total deposition of beta activity from the Teapot series, February 18–May 20, 1955. Each data point represents the measured total millicuries deposited per square mile, corrected to the amount of radioactivity that would have remained on January 1, 1956. The solid lines approximate equal deposition rates ranging from 5 to 500 mCi/mi^2. This figure was redrawn from one in List (1956).

The total deposition in March 1955 was roughly 0.3 mCi/mi^2 in Atlanta and 0.2 mCi/mi^2 in Charleston, or about 1/10 of the regional deposition from the entire test series (20 mCi/mi^2 or 2000 mCi/100 mi^2). Therefore, relatively significant depositions must have occurred at other times. Table 6-4 lists the deposition rates for these two locations for all days having a deposition of at least 10 at either location. Dates having a deposition of 100 or greater at either location are highlighted in bold. There are 9 such days over this 3-month period. Only one very high deposition day occurred in March; the remainder were in April and May.

The variation in daily deposition rates is apparent. In addition, regional hot spots of activity deposition from Teapot tests are evident from the maps in List (1956) for some days. For example, on May 7, 1955, the activity deposition at Jacksonville, Florida (south of the SRS) was around 200 mCi/100 mi^2 (with replicates of 170 and 250 mCi/100 mi^2). At St. Louis, Missouri, and Louisville, Kentucky (north of SRS), depositions were around 1350 mCi/100 mi^2 (with replicates of 1300 and 1400 mCi/100 mi^2). Yet, at Atlanta and Charleston on that day, depositions were 33 and 17 mCi/100 mi^2, respectively. It is not known whether significant ground contamination occurred at the SRS as a result of depositions of Teapot series fallout on days other than March 14, 1955. It does seem likely that this would have been the case.

SRS Monitoring Relating to 1955 Incident. The HASL fallout maps discussed in the previous section did not incorporate any data produced by the SRS environmental monitoring program. The isoactivity map reproduced earlier in this chapter (Figure 6-4) was published by the SRS in the semiannual environmental report (Alexander and Horton 1956) along with other site monitoring data. Alexander and Horton stated that the deposition of 1×10^{-7} Ci/ft^2 of beta activity in this area from bomb fallout between late February and the end of June obscured radioactivity releases from plant operations. All types of samples collected contained unusually high quantities of beta activity. Daily variations in the amount of fallout were determined by collecting particulate fallout on flypaper, collecting fallout in a water-filled tray, and from constant air monitors.

Alexander and Horton estimated that 68–77% of bomb fallout deposited on the plant site in the first half of 1955 was deposited on March 14. However, charts of the monitoring data illustrate many elevated concentrations at other times, including during April and May when contamination was also observed at the non-SRS gummed-film monitoring stations (Table 6-4).

More original documentation for these environmental monitoring results was not found during our document search. However, the weekly environmental monitoring reports for the first half of 1955 were located and reviewed (Du Pont 1955b). These reports have the title “Weekly Report – Control” followed by the date. They are typically three pages in length for each week and contain the number of samples collected by Regional Survey for routine monitoring and special problems; as well as notes on the week’s activities from the Health Physics Control Laboratory, the Development Group, and Bioassay. In general, what was accomplished is described, but few data are presented. The weekly report ending February 4 indicated that statistical analysis of the environmental data had begun for a bomb fallout report. The report ending February 18 stated that the regional survey group had made preparations for collecting data on fallout from the current Nevada tests. Similar general statements were made throughout the first quarter of 1955. For the week of March 14 through March 18, 1955, the regional survey group “surveyed surrounding territory to determine extent of fallout in rain on March 14.” Reference is also made to rain samples collected from five onsite locations that contained “unusually large quantities of short half-life radioactive fission products.”

Table 6-4. Daily Deposition Amounts for Days with at Least 10 Millicuries per 100 Square Miles at Atlanta, Georgia, or Charleston, South Carolina, for the Period February 18 through May 20, 1955[a]

Date	Deposition (mCi/100 mi^2)[b]	
	Atlanta	Charleston
March 14	**120**	12
March 20	na	44
March 22	25	6
March 24	15	23
March 28	3	61
April 1	**210**	0
April 2	65	18
April 3	24	4
April 4	17	2
April 5	62	**170**
April 6	21	58
April 7	4	44
April 9	10	1
April 10	36	10
April 11	26	15
April 13	17	5
April 14	1	26
April 19	8	28
April 20	2	10
April 21	9	11
April 22	18	0
April 24	12	6
May 1	10	6
May 7	33	17
May 8	**170**	**230**
May 9	22	72
May 10	88	16
May 11	9	**290**
May 12	**300**	43
May 13	**150**	**102**
May 14	na	17
May 15	na	16
May 16	80	6
May 18	**400**	0
May 20	**130**	98

[a] Source: List (1956).

[b] Dates having a deposition rate of 100 or greater at either location are presented in bold type.

In these weekly reports, the notes of the Bioassay group include actions taken for all significant internal exposures of personnel to radioactive materials. If an accident had occurred during the week of March 14, 1955, some description of personnel overexposures would be expected to show up in this section of the weekly report. Instead, the following observations are made for that week: "After the above average fallout was reported Tuesday morning, shoe covers were required for all people entering the laboratory. The Area Survey group made smears of the area. Activity was found on the floor of the offices and outside halls, but none in the laboratory. A fission product analysis was also made on a composite urine sample collected Tuesday. No activity could be found due to excretion or contamination of equipment." Shoe covers were discontinued the following Monday. During May 9–13, two surveys to determine ground contamination due to bomb fallout showed negligible contamination as determined with a Thyac instrument.

Kinard (1955) attempted to determine the radioisotopes present by gamma spectroscopic analysis of an evaporated rain sample supplied on March 17, 1955. Because the sample was evaporated, little volatile activity would be expected to be remaining in this sample. A spectrum was obtained daily on this sample between March 17 and 28. The resolution of gamma energies possible with the technology available (a 1-1/2 × 1-in. sodium iodide crystal and photomultiplier) was not good by later standards. However, the energies, half-lives, and possible radionuclides are listed. There were two broad half-life groups: one about 70 hours and the other about 110 hours. Most probable nuclides identified were ^{99}Mo; ^{95m}Nb (daughter ^{95}Zr); ^{97}Zr (daughters ^{97m}Nb, ^{97}Nb); ^{132}Te (daughter ^{132}I); and ^{133}Xe.

Lapsley (1955) notes three abnormal changes that were observed in the pattern of local airborne alpha activity. The changes, which could not be explained by natural radon and thoron decay products, occurred after three recent nuclear weapons tests in Nevada. The dates of the tests were March 12, March 29, and May 15, 1955. Abnormally high alpha activity was observed at the SRS on the fourth night, the third night, and the fourth night, respectively, after these tests. The abnormal alpha activity had a longer half-life, with one component having an 8.5-hour half-life contributing about 3×10^{-5} dpm/cm^3 initially. Another decay curve indicated a component with about 2.4-hour half-life. A tentative evaluation of ^{234}Pu was made for the 8.5-hour half-life material. It is a possible product of the tests, being formed by alpha reactions with ^{233}U, ^{235}U, and ^{239}Pu. The half-life of the short-lived daughter of radon and thoron in the air of Building 773-A had been determined to be about 40 minutes (filterable alpha activity). Activity concentrations of the natural activity ranged from 10^{-5} dpm/cm^3 to about 10^{-3} dpm/cm^3.

The detailed operational records from the SRS, which were obtained and reviewed during the Senate Armed Services Committee investigation, are available for review on microfilm at the Aiken public reading room (Anonymous 1986). We examined the entire set at that location and made copies of about 60 key pages (Du Pont 1955a). Some of these records on the microfilms had been obtained by *RAC* previously as part of the overall dose reconstruction document review and retrieval process, for example, a radiation survey logbook from P-Area (Anonymous 1955a). The microfilms contain many hundreds of pages of SRS logbooks and onsite health physics monitoring records from the time period before, during, and after the March 1955 contamination incident. Most of the pages were routine operational monitoring records, of the types in the list below, for the following SRS areas: K-Area, L-Area, P-Area, R-Area, F-Area, H-Area, D-Area, and M-Area.

- Air sample log
- Monthly report
- Shift log
- Special work permit
- Special permit log time sheets
- Weekly report.

For the most part, these records deal with measurements and observations that were taken inside buildings during operations for routine safety purposes. A Thyac or Cutie Pie survey instrument was most often used for the radiation surveys. Typical background count rates with the Thyac instrument (open probe) were 150–200 counts per minute (cpm). The probe was held 1 in. from the surface in question (for example, a floor or table). Both penetrating radiation exposure rates and removable contamination levels (such as counts per minute of beta-gamma contamination on paper towel smears) were recorded on the log sheets. Following are notes from P-Area radiation surveys relevant to the March 1955 environmental contamination incident. All times are Eastern Standard Time. Unless noted otherwise, the P-Area surveys are contained within the document referenced as Anonymous (1955a). These notes are presented to give the reader an idea of the eyewitness observations made by a number of workers during the time of the ground contamination incident.

The description of survey number P11223 of the lunchroom, at 5:50 p.m. on March 14, was to make "routine and special check of tables, seats and entrance as result of fallout contamination." Survey number P11230 of the general area inside building 105-P at 6:30 p.m. on March 14 states, "contamination spread thru-out bldg. from fall-out." Survey number P11224 on March 14 detected 2.9×10^{-4} µCi/cc in rainwater as compared with 9.5×10^{-7} µCi/cc in water from the 186-1 basin and 2.4×10^{-7} µCi/cc in tap water. Survey number P11222 at 3:40 p.m. on March 15, 1955, was a survey of loading a scrap cask. High contamination (5000 counts per minute), which was reduced to 800 cpm after one washing with water, was "probably due to fall out as the casks were setting on the truck outside the transfer area during the rain and were put into the transfer area about 1535." Survey number P11230 at 6:30 p.m. on March 15 in Building 105-P remarks that there were "no unusual rad. levels found or shielding leaks. Contamination spread thru-out bldg. from fall-out." Survey number P11247 was of the "fall out on concrete in front of 105-P E side." Spots reading 4000 to 5000 cpm on the Thyac were smeared. Nine smears ranged from 412 cpm to 954 cpm beta-gamma. The fallout seemed to be equally distributed over this area of ground. A routine paper smear survey of the floor of the lunch room (survey P11273 on March 16) found that the "floor was found contaminated due to fall out." Survey P11321 of the Inst. Shop (Purif. Area) on March 18 showed "contamination found on floor attributed to fall-out. Personnel carried some into area on soles of their shoes." The routine smear survey of the 105-P lunchroom on March 19 (P11347) showed that "the floor is still contaminated from the fall out." By March 21 (P11396) the survey of the lunchroom showed <100 cpm beta-gamma, and <2 cpm alpha (alpha counts were never positive).

Similar radiation survey log sheets for F-Area, R-Area, K-Area, and L-Area were obtained from the Senate investigation microfilms (Du Pont 1955a). In L-Area, two lunchroom surveys (L5636, L5643) in building 105-L on March 12 showed no contamination; however at 2:10 a.m. on March 15, the lunchroom survey (L5681) showed beta-gamma "contamination on floor of

lunch room due to 'fallout.' Sign put up telling people to keep feet on floor." Levels were considerably less than those seen in P-Area. On March 16, no action was taken on floor contamination in the 704-L building cafeteria "due to conditions in area from radioactive fallout" (survey L5713). In R-Area, a sidewalk outside the cafeteria read 4000 cpm at 6:00 p.m. on March 14 (survey R15427). There was no contamination in lunchroom of 105-R at 6:45 p.m. on March 14 (R15363), but at 2:00 a.m. on March 15 there was contamination there (survey R15371). A survey of the shoes of two personnel at 8:20 a.m. on March 15 (R15385) showed Thyac readings up to 2000 cpm above background (200 cpm). The technician states, "The men had not been in an RDZ so it was decided the contamination was brought in from outside of the building." Over one-half of 42 smears of the floors of all levels of building 105-R on March 15 were contaminated (survey R15406). In F-Area, at 3:45 p.m. on March 14, the technician notes on survey F5799, "Survey made to verify reports of high activity fall out. Paper towel smears show contamination ranging from 1500 c/m4 to as high as 15000 c/m. There is doubt at the present time as to the source of the contamination. At the time of this survey it was raining." At 9:30 p.m. on March 14, at the railroad area west of the 221 stack, a Thyac reading was 2250 cpm at ground level on the ties and 500 cpm at 3 ft above ground level.

Thyac count rates at ground level (usually 1 in. from surface) were over four times higher than those made at a height of 3 ft above ground.

Over 97% of the radioactivity measured by the Thyac radiation monitors on March 15 was from beta-emitting radiation; the remainder was gamma radiation.

In K-Area, contamination was found over all the floor of the 704-K cafeteria at 9:00 p.m. on March 14 (survey 3652). The area was "not cleaned up since entire area (outdoors) was above the limits. The contamination was due from the radioactive fallout." At 5:30 a.m. on March 15 in K-Area, a survey was made of roads and walkways that showed up to 3000 cpm above background (K3657). Radiation survey K3661 was the only one found for this contamination event that showed the Thyac readings expressed both as beta-gamma and as gamma alone. The latter measurement would have been made with the instrument probe window closed. Fifteen locations outside 105-K building at 10:00 a.m. on March 15 had beta-gamma readings of 1500 to 4100 cpm whereas gamma-alone readings were 50 to 140 cpm. The average ratio of gamma:beta-gamma was 0.028 (range 0.016–0.067). This is an important finding that indicates that the wide area survey shown in Figure 6-4 represents mostly beta activity. These data can be accessed directly in the Excel® workbook in the spreadsheet tab called "survey K3661" by clicking on the following hyperlink: 100kwr.xls.

Dozens of radiation survey log sheets, produced by many different technicians at separate locations, illustrate clearly that contamination was widespread around the SRS on March 14 and 15, 1955, and originated *outside* the SRS buildings. The hundreds of survey log sheets reviewed show no indication of a large-scale accident at this time.

Another source of original data during the time period of interest for the March 1955 contamination incident is air sample log sheets. The data sheets in Anonymous (1954) record radioactivity in air collected within Building 105-P between 9/2/54 and 8/27/55. An initial 10-minute count of beta gamma activity is recorded as well as a 10-minute count taken 4 hours later, which allows short-lived natural radon and thoron products time to decay. The calculation

[4] c/m is counts per minute, also abbreviated as cpm.

showing the conversion from counts per minute to activity concentration for fission products and for plutonium and uranium is preprinted on the data sheets. There were no samples taken between March 10, 1955, at 7:15 a.m. (log sheet P2920) and March 15, 1955, at 3:45 a.m. (survey P-2921).

Similar Air Sample Log Sheets for K-Area (Anonymous 1955b) end with sheet number K1250 dated 4/11/55. A sample taken outside the window of the health physics office on March 15, 1955, (log sheet K1193) had a higher 4-hour count (160 cpm net of background) than observed at locations inside the building during that same general time period (approximately 20–40 cpm). We reviewed air sample log sheets from all areas from the microfilm record (Anonymous 1986) and copied key pages from P-Area, R-Area, K-Area, and F-Area (Du Pont 1955a). The location of the samples varies, there is not always a delayed (4-hour) count, and there are few comments. It does not appear that any insight can be gained from the Air Sample Log Sheets for the March contamination incident.

Shift logs give a verbal confirmation to notes from formal data sheets. The M-Area shift log notes for March 14, 1955 (Du Pont 1955a) state, "Building in normal operation. Large number of shoe contaminations due to radioactive rainfall. Made surveys at barricades and gate house." The F-Area shift log notes for March 14 contain the following:

> "Background has been high with RR cut reporting around 1000 cts/min. Background at 700 Area and 100 Areas has been 1000–1300 cts/min. It is presumed this is 'fall out' from atomic tests. A line vicinity showed 800 cts/min background Thyac, but 13,500 cts/min from paper towel smear of rails west of stack in clean area. (Illegible) near exclusion area fence west of A line read 7000 cts/min paper towel smear."5 "High background thought to be coming from stack or Nevada tests." "Today until 8:50 tomorrow morning let men go home on contamination of shoes up to 1000 cts/min, unless you can determine contamination is caused by the srp process, rather than Nevada." "Stack samples pulled at 3:30 p.m. to determine if activity of project was due to us: apparently not; sample read only 10–12,000 c/m (Thyac). This fallout is known to be project-wide & presumed to be a result of bomb tests."

On March 15, 1955, notes from the F-Area shift log relative to the contamination were as follows:

> "Shoe contamination due to yesterday's fallout remains common. Certain types of shoe soles absorb up to 3000 c/m. Procedure for release: at A-line, check for ∝, release if negative; at 211-F determine contamination level & base release on contamination history. In no case is plant-caused contamination to be released. Check of fallout indicates half-life in 30–36 hr range.6 Roadway reading maximum of 2000-2500 c/m, towel smearable contam. to 25000 on pipes, hand-rails, etc. Rubber gloves were required for work in outside areas where process contamination is posible [sic] as it is not determinable whether skin contamination is caused by fallout or process. No skin decontamination record is necessary for decontamination of personnel known to have been contaminated by non-process, i.e. fallout, material."

[5] Break in quotes indicates an area where unrelated text appears in the logbook.

[6] Shorter half-life than would be expected based on exposure rate decay curve from Teapot Hornet (Figure 6-8). No data were located to support this statement, so it is not known what isotopes were being measured. Some components of the fallout have half-lives in this range. (Table 6-3).

Weekly reports of reactor area activities were reviewed for the time period of interest. A brief mention of the contamination is made in Kauffman (1955a):

> "The fall out which occurred on the afternoon of 3/14/55 caused difficulty with contamination surveys such as the hand and foot checks. Lectures were given to a number of personnel on the nature of the fall-out. Decay and absorption studies are being made on samples of fall-out material. A special report will be issued on the fall-out incident in 100-P area."

The 100-P weekly report for March 21–March 27 (Kauffman 1955b) states, "On the morning of March 25, 1955 fall-out activity was observed for the second time during the month. Thyac readings ranging from 800 c/m to 1000 c/m, above background, were found by spot-checks outside of the 105-P building." No mention of contamination is made in the weekly reports from 100-L for March 1955. However, at 100-C, the weekly report ending March 20, 1955 (Catlin 1955) states:

> "Numerous cases of foot contamination from radioactive fallout were detected by the Hand and Foot Counters. One hand contamination case was also reported. Smear surveys outside the 105-C Building gave contamination levels as high as 5000 c/m. This material was tracked into the 105-C Building to give contamination levels as high as 500 c/m. Half-life of the fallout was found to be approximately 30 hours. Notices were posted at all Hand and Foot Counters to acquaint building personnel with the problem."

The next week, contamination was observed again on March 25, 1955. Smear survey spot checks outside the 105-C building revealed contamination levels up to 2000 cpm.

The 100-K weekly report for March 14 through 20 (Du Pont 1955a) contained fallout data for six areas outside the building. The original data were on Radiation Survey Log Sheet K3661. On March 15, Thyac readings were 3100–4000 cpm; five days later on March 20 they ranged from 650–1100 cpm at the same locations. On average, the March 20 readings were 24% of the March 15 readings, for both Thyac measurements and for removable contamination measured by smears (data in spreadsheet 100kwr.xls). This is in reasonable agreement with the decay trend illustrated for the Teapot Hornet test in Figure 6-8 of this chapter; between 3 days and 8 days after the blast, a somewhat smaller reduction of 30–35% in the exposure rate is expected. However, we do not know the precise time of day that the exposure rate measurements were taken at SRS. If taken early in the day on March 15 and late in the day on March 20, a decrease of around 25% is expected, based on Figure 6-8. In addition, some weathering of the contamination either surficially or into the ground would tend to further reduce the exposure rate beyond the theoretical amount based on decay alone. Therefore, we have concluded that the exposure rate reduction observed in K-Area between March 15 and 20 is consistent with the decay characteristics of the Teapot Hornet fallout event.

A monthly report of Miscellaneous Areas for March 1955 (contained in Anonymous 1986) states that "occurrence of fallout on 3-14-55 necessitated the use of shoe covers in the 735-A Bioassay labs and counting rooms." A summary of important items for 100-P area in March 1955 (Du Pont 1955a) contained the following comments:

> "The 'fallout' from the Nevada Tests proved to be one of the biggest problems during the month. For a number of days the Hand and Foot counters would indicate 'Decontamination Required' for all personnel in the building. Special checks had to be given to all personnel working in RDZ locations with a Thyac in the H.P. Office. A number a [sic] special indoctrination sessions on the fall-out were given to different groups in the area."

In general, we have found that internal documents reporting on SRS operations frequently note contamination incidents, personnel overexposures, unplanned releases, and other unusual incidents. There does not appear to be any attempt to hide this type of information. An example during the time period of interest is Radiation Survey Log Sheet F1917 from March 15, 1955 (Du Pont 1955a), in which an incident involving product from the FB line resulted in significant contamination within that working area as well as radiation exposure to personnel. The previous paragraphs of this section have reviewed original, primary records that confirm widespread radioactive contamination around the SRS buildings on March 14, 1955 and the days that follow. However, in the records of special work permits, weekly reports, and monthly reports contained in the Senate investigation microfilms (Anonymous 1986) as well as weekly reports of the control group (Du Pont 1955b), there was nothing to indicate that a major problem in SRS facilities caused this contamination.

Stephens and Ross (1984) summarizes sites of contamination on the SRP. It includes a list of many sites contaminated with radioactivity from SRS activities. There were 25 locations in F-Area between November 1954 and November 1978; 9 areas in H-Area between July 1956 and 1983; 28 areas in G-Area, M-Area, and A-Area between December 1953 and 1973; and 3 areas in the reactor areas between 1957 and 1983. This demonstrates that the personnel were cataloging areas believed to be contaminated by their operations. The sites listed for 1955 were small in extent relative to the March 1955 contamination area. No mention is made of any SRS event leading to significant environmental contamination in March 1955.

National Cancer Institute Report on Exposure of the American People to ^{131}I from Nuclear Weapons Tests. The National Cancer Institute (NCI) used the gummed-film monitoring data from the 1950s in a recent retrospective analysis of estimated exposures and thyroid doses received by the American people from ^{131}I in fallout following the Nevada atmospheric nuclear bomb tests (NCI 1997). A correction was made to estimate the fraction of iodine from each of the tests because the monitoring data were reported as total beta activity. The projected trajectories from the Teapot Hornet test were the same as shown in List (1956), reproduced in this chapter as Figure 6-5.

Our Conclusion About the 1955 Contamination Incident

We conclude that it is highly probable that the ground contamination measured at the SRS on March 14–15 came from deposition of debris from the Teapot Hornet test conducted on March 12 in Nevada. This conclusion is based on a number of factors reviewed in the previous sections, perhaps most importantly (1) the timing of the deposition relative to the predicted arrival of Nevada fallout, (2) the measurement of elevated radioactivity in Atlanta on the same date, and (3) the decay characteristics of the material. The main argument supporting an SRS accidental release is that localized contamination centered on the reactor areas at the SRS. However, there were no other nearby areas being monitored for radioactivity, so the complete extent of the contamination

will never be fully known. This incident underscores the difficulty in interpreting environmental monitoring data from the 1950s, when isotopic analyses were much less frequent than gross activity analyses, and when hot spots from fallout were more frequent than in the 1960s and later.

NATURAL, ACCIDENTAL, AND OTHER FACILITY SOURCES OF ENVIRONMENTAL RADIOACTIVITY

Although fallout from weapons testing has been the most important man-made contributor to radioactive contamination of the global environment, there are other sources. In addition, some radionuclides occur naturally in the environment. To focus our discussion, it is necessary to identify the radionuclides of interest for this dose reconstruction. In Phase I of this study, a screening calculation was conducted to identify the radionuclides that probably contributed the largest proportion of offsite doses from past SRS releases to air and water (Meyer et al. 1995, Table 25). There were 14 radionuclides in the most important group: ^{241}Am, ^{41}Ar, ^{60}Co, ^{137}Cs, ^{3}H, ^{131}I, ^{129}I, ^{32}P, ^{90}Sr, ^{35}S, ^{238}Pu, ^{239,240}Pu, ^{65}Zn, and ^{95}Zr/Nb. In Chapter 3 of this Phase II report, some of the radionuclides identified in Phase I are highlighted as prime candidates for future detailed source term work. Those eight nuclides, which should be evaluated further for their offsite release potential, are ^{3}H, ^{137}Cs, ^{131}I, ^{90}Sr, ^{60}Co, ^{32}P, ^{41}Ar, and ^{129}I.

Only two of these most important radionuclides, tritium, (^{3}H), and iodine-129 (^{129}I), are produced naturally in the environment.

Natural Sources

Natural tritium is produced by the interaction of cosmic rays with nitrogen in the upper atmosphere. The natural production rate has been calculated to be 4 million curies per year, leading to an inventory of 70 million curies (NCRP 1979). Since the mid-1950s, the global inventory of tritium from aboveground nuclear weapons tests has overwhelmed the natural inventory. This inventory from weapons tests was (within a factor of 2) 3100 million curies in 1963 (NCRP 1979).

The average concentration of tritium in environmental waters from natural tritium production is 3.2 to 16 pCi L^{-1} (NCRP 1979). Average concentrations up to 4000 pCi L^{-1} were measured in U.S. surface waters in the peak fallout years (see previous section).

Radioactive isotopes of iodine are produced naturally by the interaction of cosmic rays with the earth's atmosphere and by rare spontaneous fission of uranium and thorium in the earth's crust. Essentially the only radioactive isotope of iodine to survive the transit time between production in the atmosphere and deposition on the earth's surface is ^{129}I (with a half-life of 16 million years). The global inventory of ^{129}I accumulated over the lifetime of the earth is estimated to be 10 Ci in the terrestrial environment and 30 Ci in the oceans and atmosphere (NCRP 1983). Fallout from nuclear weapons testing has added about 10 Ci of ^{129}I into the atmosphere (NCRP 1983).

There are no other key radionuclides in this dose reconstruction that occur naturally except in rare instances. For example, ^{239}Pu and fission products such as ^{137}Cs can occur in minute quantities in some naturally occurring ores containing uranium or thorium. Based on the Phase I screening analysis (Meyer et al. 1995), another 11 minor radionuclides were selected that probably contributed between 0.1 and 1% to offsite dose from the SRS releases: ^{140}Ba,La,

^{141,144}Ce, ^{14}C, ^{51}Cr, ^{85}Kr, ^{239}Np, ^{106}Ru, ^{99}Tc, ^{232}Th, uranium, and ^{91}Y. Of these, ^{14}C, thorium, and uranium isotopes occur naturally in significant quantities. Minor amounts of ^{85}Kr are produced naturally.

Environmental measurements that do not distinguish individual radionuclides (gross beta, nonvolatile beta, external gamma, and gross alpha) always include some background level of naturally occurring radioactivity such as ^{40}K, ^{226}Ra, radon, and other radionuclides in the uranium and thorium series. The background radioactivity concentrations depend on the sample collected and how it is prepared and analyzed. When appropriate, background levels of gross radioactivity are discussed in other sections of this report that address the individual environmental media.

Accidental Sources

A significant source of ^{238}Pu in the global environment was the burnup of a satellite (SNAP-9A) in April 1964 (Hardy et al. 1972). This malfunction resulted in an estimated 17 kilocuries (kCi) of ^{238}Pu being released to the upper atmosphere. Although most of this material ended up falling out in the Southern Hemisphere, there were detectable amounts in the Northern Hemisphere that affected the ratio of ^{238}Pu to ^{239,240}Pu in fallout after that time.

Plutonium-238 from the April 1964 burnup of the SNAP-9A satellite was first detected by the HASL in ground-level air in Ispra, Italy in early 1966. In New York City, the elevated levels were first detected in April of that year. After 1970, HASL stopped reporting ^{238}Pu at many stations, because concentrations were barely detectable and soil sampling indicated that 95% of the SNAP-9A ^{238}Pu had been deposited by that time. Because of the low levels of ^{238}Pu in the air after October 1971, the errors associated with the measurements are large (Figure 6-10).

The Chernobyl Unit 4 nuclear reactor accident in April 1986 in the Ukraine released radioactivity to the atmosphere that was detected at locations throughout the U.S. Table 6-5 lists the amounts believed to have been released, for those longer-lived radionuclides having significant activity 10 years later. In addition to these, Bradley (1997) gives estimated release quantities for the following shorter-lived radionuclides: ^{133}Xe (45 million curies), ^{131}I (7.3 million curies), ^{85m}Kr, ^{140}Ba, ^{95}Zr, ^{103}Ru, ^{99}Mo, ^{141}Ce, ^{89}Sr, ^{132}Te, ^{239}Np, and ^{242}Cm. Based on ^{131}I/^{129}I isotope ratios measured by Paul et al. (1987), the accidental release of ^{129}I is estimated to be 0.2 Ci.

Other nuclear reactor accidents such as Three-Mile Island (in Pennsylvania, 1979); SL-1 (in Idaho, 1961); and Windscale (in the U.K., 1957) released smaller amounts of radioactivity that were essentially limited to their regions.

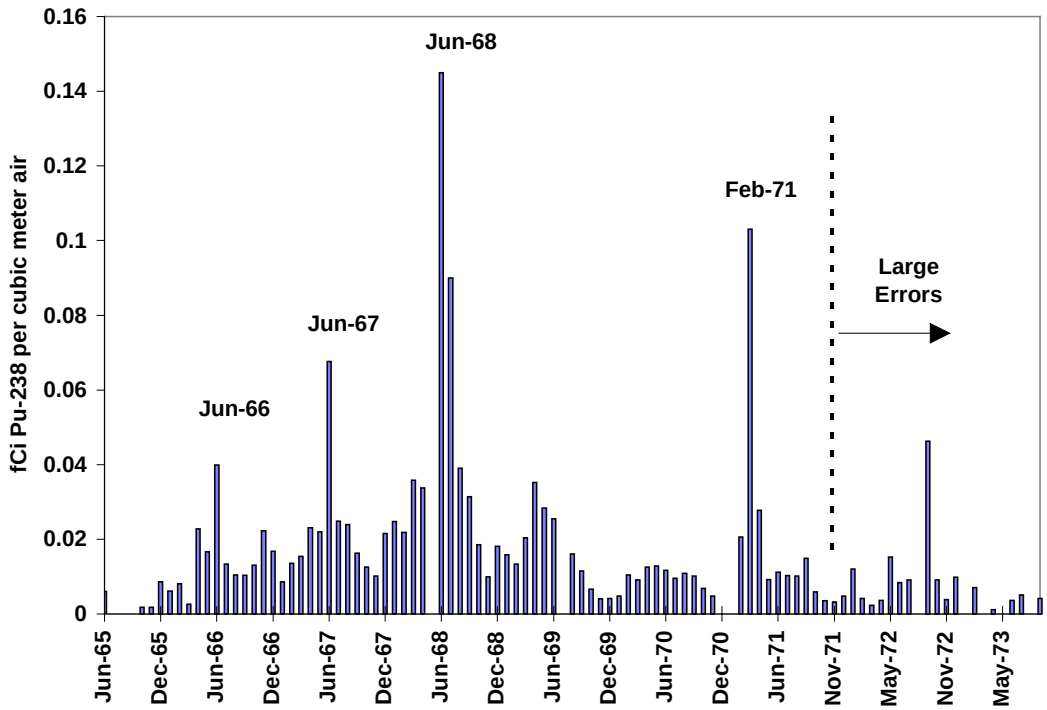


Figure 6-10. Plutonium–238 in air in New York City, measured by the Health and Safety Laboratory surface air-monitoring program (HASL 1975). Concentrations in the late 1960s were affected by the accidental burnup of a satellite that contained ^{238}Pu. These data can be accessed directly in the Excel® workbook by clicking on the following hyperlink: puair.xls.

Table 6-5. Estimated Release of Radionuclides from the Chernobyl Accident (Bradley 1997)[a]

Nuclide	Released Activity by May 6, 1986 (Ci)
^{137}Cs	1,000,000
^{85}Kr	950,000
^{90}Sr	220,000
^{241}Pu	140,000
^{134}Cs	500,000
^{106}Ru	1,600,000
^{240}Pu	1,000
^{238}Pu	800
^{239}Pu	700
^{144}Ce	2,400,000
^{242}Pu	2

[a] Estimated error ± 50%.

Other Facility Sources

Other facility sources of man-made radioactivity include commercial nuclear reactors, medical and industrial facilities, and other government sites in the weapons complex. Their contributions to environmental radioactivity levels usually can not be distinguished from ambient background conditions at distances very far from the facilities. They need not be considered as contributors to concentrations measured near the SRS.

RELATIVE CONTRIBUTIONS OF VARIOUS SOURCES OF RADIOACTIVITY

Some radionuclides identified in the screening analysis (see Chapter 3), if detected in the SRS regional environment (within about 10 km of the source), would be expected to have come almost exclusively from SRS releases. These include ^{85}Kr, ^{41}Ar, ^{32}P, ^{35}S, ^{60}Co, ^{129}I, ^{99}Tc, and ^{51}Cr. In addition, ^{238}Pu is mainly released to the environment from facilities that produce or process nuclear materials (such as the SRS), although lesser amounts have been contributed by weapons fallout. The accidental burnup of the SNAP satellite is an important contributor to atmospheric ^{238}Pu in the late 1960s.

Natural sources are unimportant contributors to environmental concentrations for the key radionuclides identified for this dose reconstruction. Similarly, other government and commercial facilities would not contribute significantly to environmental radioactivity levels near the SRS.

Radionuclides in weapons fallout such as ^{241}Am, ^{137}Cs, ^{3}H, ^{131}I, ^{90}Sr, ^{239,240}Pu, ^{65}Zn, and ^{95}Zr/Nb were also released in significant quantities from the SRS. A careful examination of source term amounts and trends as well as environmental concentrations and trends is necessary before an informed judgment can be made about the relative contributions for radionuclides produced from multiple sources.

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CHAPTER 7

ENVIRONMENTAL MONITORING FOR RADIONUCLIDES

ABSTRACT

This chapter summarizes the history of the environmental monitoring program at the Savannah River site (SRS). The SRS program provides the largest source of environmental monitoring data for radionuclides originating from the SRS. This chapter describes the sources and types of documents reporting environmental monitoring data, and provides an overview of the methods used to compile and cross-check the data. Chapters 8, 9, 10, 11, 12.1, 12.2, 13, and 14 describe the environmental monitoring data for radionuclides identified as potentially useful for the dose reconstruction in detail by media. The environmental monitoring data for chemicals are discussed separately in Chapters 19 and 20. Quality assurance and quality control issues related to environmental monitoring for radionuclides are also discussed in this chapter.

Chapters 8, 9, 10, 11, 12.1, 12.2, 13, and 14 focus on the radionuclides that were determined to be most important by screening calculations (Chapter 3), and those radionuclides that were monitored on a regular basis, especially during the early years and that can be used to confirm the magnitude of releases and/or the fate and transport in the environment. These chapters evaluate the available environmental monitoring data by media from the beginning of SRS operations through approximately 1991. This does not mean that environmental monitoring data do not exist after 1991, but the focus of this dose reconstruction project is on historical exposures so the earlier data are generally most useful.

This chapter stresses the importance of environmental monitoring data collected by groups and organizations independent of the Site, and summarizes the useful datasets that have been identified and compiled. Specific details concerning these independent data are provided in Chapters 8, 9, 10, 11, 12.1, 12.2, 13, and 14.

INTRODUCTION

Environmental monitoring data are a valuable resource for dose reconstruction because they provide direct information about the concentrations of contaminants in various environmental media (e.g., air, water, vegetation, and biota) at particular points in space and time. Depending on the nature and quality of these data, they may be used in several important areas in the dose reconstruction. For example, the data may be used to independently check source term estimates or environmental transport model calculations, or to derive specific parameters used in the environmental transport models.

The current phase of the SRS Dose Reconstruction Project focuses on reconstructing the releases of radionuclides and chemicals and, therefore, does not yet use environmental transport models. Each chapter discusses the media-specific potential usefulness as well as limitations of each compiled data set. These discussions include the impact of such things as original data availability, measurement uncertainties, spatial and temporal resolution, the ability to establish appropriate background concentrations, and other media-specific factors. However, making definitive statements about precisely how the data will be used is beyond the scope of this phase of the project.

REGIONAL MONITORING PROGRAM

The SRS has carried out environmental monitoring activities since before operations at the Site commenced. A preoperational background survey designed to establish background levels of naturally occurring radionuclides before plant startup was carried out from June 1951 to January 1953 (Reinig et al. 1953). Indigenous terrestrial and aquatic animals, vegetation, and food crops were collected and analyzed for total alpha and beta content. Once operations commenced in 1953, this program was adopted for routine monitoring (Harvey 1964). The following excerpt from Harvey (1964) describes the development of the program through the early 1960s.

> Emphasis was placed on the collection and radioanalysis of terrestrial plants and animals through 1955 due to the dominance of fallout radioactivity. Radioiodine (^{131}I), radiostrontium (^{89,90}Sr), and radiocesium (^{134,137}Cs) analyses were begun for certain samples. In 1956, 1957, and 1958, a series of fuel element failures released low concentrations of radioactive material to the Plant streams, so sampling emphasis was shifted to the aquatic environment. In 1956, gamma spectrometry was first used to identify gamma-emitting radionuclides in environmental samples. Data collected through 1959 indicated that the program could be improved by selective sampling. For example, routine collection of seven species of fish not normally eaten by man was discontinued because they contained essentially the same or lower concentrations of radionuclides than the three most common species of edible fish. The analyses of flesh and bone only were continued because the other tissues are not normally eaten. The use of gamma spectrometry to identify gamma-emitting radionuclides was a major factor in developing a selective sampling program for aquatic and terrestrial specimens. The value of spectrographic analyses was subsequently enhanced by the development of computer programs for routine determinations of radionuclide concentrations.
> In the current program, radionuclide uptake and retention patterns of biota are determined from specific analyses of selected indicator organisms, including algae, bluegill sunfish, ring-necked ducks, and cottontail rabbits.

The routine environmental monitoring program was designed to ensure that the amounts of radioactivity released to the environs of the Site did not exceed the maximum permissible concentrations (MPC) that had been established by the National Committee on Radiation Protection (Horton 1957). MPCs were established for various radioactive isotopes in water and air from which the Site calculated MPCs for foods (Horton 1957).

In reviewing the Regional Monitoring Program in 1960, Horton (1960) states that the program was established to accumulate information necessary for determining the effectiveness of Plant controls for radioactive wastes and for an authoritative record of environmental conditions in the event of litigation. The environmental monitoring staff also considered that the purpose of the program should be extended to include collecting scientific information concerning the fate of radionuclides in the environment and to determine better methods of waste management (Horton 1960; Marter and Boulogne 1961).

Horton (1960) also stated that the program needed to emphasize isotopic analysis of samples and, more scientifically, defined MPCs in foods before the best interpretation of the data could be

made. The automation of gamma spectrometry analyses in the early 1960s permitted routine analysis of effluent streams, river water, reactor area effluents, biological specimens, and many other samples not previously analyzed isotopically (Marter and Boulogne 1961). As a result, gross analyses of many samples were supplemented with quantitative data on specific isotopes.

In 1965, the number of samples obtained for the routine environmental monitoring program was reduced by 1500 to approximately 14,000 samples (Marter and Johnson 1966). The reduction in sampling load permitted more duplicate analyses of certain "critical" samples and made the program more cost effective (Marter and Johnson 1966). The memo also indicated that a further reduction of 5000 samples a year could be made without compromising the effectiveness of the environmental monitoring program.

Although the Health Physics Environmental Monitoring Program at SRS was initiated primarily to measure radioactivity, the schedule was expanded in 1973 to include nonradioactive measurements (Johnson and Zeigler 1972). Environmental monitoring for chemicals is discussed further in Chapters 19 and 20.

DATA SOURCES

The environmental monitoring data relevant to SRS are obtained from a number of different sources. The largest source is the various series of environmental monitoring reports that SRS produced throughout its operation. These are summarized in Table 7-1 and include weekly, monthly, quarterly, semiannual, and annual report series; however, none has been produced throughout the entire time period. Probably the earliest study is Reinig et al. (1953), which describes a preoperational background survey that established background levels of naturally occurring radionuclides before plant startup. That survey was carried out from June 1951 to January 1953, during which time approximately 6600 samples from the environs of the SRS were collected and analyzed. Water, mud, air, and biological specimens were sampled. The biological specimens included aquatic and terrestrial animals and a wide range of vegetation.

The first reports concerned with environmental monitoring activities were a series of monthly reports (Table 7-1). These were first produced in August 1951 (Reinig 1951a) by the Health Instrument Group, which was renamed the Health Physics Group by November 1951 (Reinig 1951b). The early reports document the progress of the Site survey. Environmental monitoring monthly reports continued to be produced through 1979. During this time, the name of the group producing them changed periodically. For example, from Health Physics to Control in July 1956 (Du Pont 1956) to Control and Methods in November 1959 (Du Pont 1959) and to Environmental and Allied Studies by March 1961 (Du Pont 1961b). Starting in July 1965, three separate monthly reports were produced by what had become the Radiological and Environmental Sciences Division: (1) Environmental Monitoring (Du Pont 1965a), (2) Earth Sciences (Du Pont 1965b), and (3) Radiological Sciences (Du Pont 1965c). We reviewed all of these reports for relevant environmental monitoring data.

Weekly environmental monitoring reports cover the time period from the end of May 1954 (Du Pont 1954) through October 1962 (Du Pont 1962). However, from around mid-August 1961, the reports covered 2-week periods (Du Pont 1961c), and reports from May through October 1962 appear to have been produced monthly. These reports were separate from the monthly reports described above (Table 7-1). Quarterly environmental monitoring reports were produced briefly for a 2-year period that covered the first quarter of 1960 (Du Pont 1960) through the last

quarter of 1961 (Du Pont 1961a). Semiannual reports were produced over a significantly longer time period than the quarterly reports (Table 7-1). The first semiannual report covers the first 6 months of 1953 (Albenesius 1954). No semiannual report was produced for the last 6 months of 1953, and the next report covered January to July 1954 (Horton 1954). The final environmental monitoring semiannual report covered the first 6 months of 1971 (Du Pont 1971).

An annual environmental monitoring report titled Health Physics Regional Monitoring Annual Report was first produced for the year 1963 (Du Pont 1964). The title was changed the following year (Ashley 1964) to Environmental Monitoring at the Savannah River Plant Annual Report and remained the same for the annual reports produced through 1984. Beginning in 1971, a second annual report series titled Environmental Monitoring in the Vicinity of the Savannah River Plant was produced (Du Pont 1972). This series was replaced in 1984 by a series titled Savannah River Plant Environmental Report that is produced to the current day. In 1988 the title was revised to Savannah River Site Environmental Report (Table 7-1).

Table 7-1. Summary of Savannah River Site Environmental Monitoring Reports

Report title	Period of publication	Frequency of publication	SRS Doc. Series #
Health Physics Regional Monitoring Annual Report	1963	Annual	DPSPU 64-11-12
Environmental Monitoring at the Savannah River Plant	1964–1984	Annual	DPST 65-302 through DPST 85-302
Environmental Monitoring in the Vicinity of the Savannah River Plant	1971–1983	Annual	DPSPU 72-30-1 through DPSPU 84-30-1
Savannah River Plant Environmental Report	1984–1987	Annual	DPSPU 85-30-1 through DPSPU 88-30-1
Savannah River Site Environmental Report	1988–present	Annual	WSRC-RP-89-59-1
Health Physics Site Survey Data	1953 (Jan-Jul)	Semiannual	DPSPU 54-11-12
Radioactivity in the Environs of the Savannah River Plant	1954 (Jan-Jul)	Semiannual	DP-92
Semi-Annual Progress Report	1954 (Jul–Dec)	Semiannual	DPSP 55-25-34
	1955 (Jan-Jun)		DPSP 56-25-13
Health Physics Regional Monitoring Semiannual Report	1955 (Jul–Dec)	Semiannual	DPSP 56-25-54
	1956 (Jan-Jun)		DPSP 56-25-4
	1956 (Jul–Dec)		DPSP 57-25-15
	1957 (Jan-Jun)		DPSP 57-25-43
	1957 (Jul–Dec)		DPSP 58-25-17
	1958 (Jan-Jun)		DPSP 58-25-38
	1958 (Jul–Dec)		DPSPU 59-11-23
	1959 (Jan-Jun)		DPSPU 59-11-30
	1959 (Jul–Dec)		DPSPU 60-11-9
	1960 (Jan-Jun)		DPSP 60-25-26

Report title	Period of publication	Frequency of publication	SRS Doc. Series #
	1960 (Jul–Dec)		DPSP 61-25-4
	1961 (Jan-Jun)		DPSP 62-25-2
	1961 (Jul–Dec)		DPSP 62-25-9
Health Physics Environmental Monitoring Semiannual Report	1962 (Jan-Jun)	Semiannual	DPSP 63-25-3
	1962 (Jul–Dec)	Semiannual	DPSP 63-25-10
Effect of the Savannah River Plant on Environmental Radioactivity Semiannual Report.	1962 (Jan-Jun)	Semiannual	DPSPU 62-30-24
	1962 (Jul–Dec)		DPSPU 63-30-12
	1963 (Jan-Jun)		DPSPU 63-30-1
	1963 (Jul-Dec)		DPSPU 64-30-1
	1964 (Jan-Jun)		DPSPU 64-30-2
	1964 (Jul-Dec)		DPSPU 65-30-1
Effect of the Savannah River Plant on Environmental Radioactivity Semiannual Report.	1965 (Jan-Jun)	Semiannual	DPSPU 65-30-2
	1965 (Jul–Dec)		DPSPU 66-30-1
	1966 (Jan-Jun)		DPSPU 66-30-2
	1966 (Jul–Dec)		DPSPU 67-30-1
	1967 (Jan-Jun)		DPSPU 67-30-2
	1967 (Jul–Dec)		DPSPU 68-30-1
	1968 (Jan-Jun)		DPSPU 68-30-2
	1968 (Jul–Dec)		DPSPU 69-30-1
	1969 (Jan-Jun)		DPSPU 69-30-2
	1969 (Jul–Dec)		DPSPU 70-30-1
	1970 (Jan-Jun)		DPSPU 70-30-2
	1970 (Jul–Dec)		DPSPU 71-30-1
	1971 (Jan-Jun)		DPSPU 71-30-16
The Effect of the Savannah River Plant on Environmental Radioactivity Quarterly Report	1960 (Jan-Mar)	Quarterly	no number
	1960 (Apr-Jun)		no number
	1960 (Jul-Sep)		no number
	1960 (Oct-Dec)		no number
	1961 (Jan-Mar)		no number
	1961 (Apr-Jun)		no number
Effect of the Savannah River Plant on Environmental Radioactivity Quarterly Report	1961 (Jul-Sep)	Quarterly	no number
	1961 (Oct-Dec)		DPSPU 62-30-11
Health Instrument Group Site Survey Monthly Progress Report	1951 (Aug – Oct)	Monthly	no number
Health Physics Group Monthly Report	1951 (Nov) – 1956 (Jun)	Monthly	no number
Control Monthly Report	1956 (Jul) – 1959 (Oct)	Monthly	no number
Control and Methods Monthly Report	1959 (Nov) – 1961 (Feb)	Monthly	no number

Report title	Period of publication	Frequency of publication	SRS Doc. Series #
Environmental Monitoring and Allied Studies Monthly Report	1961 (Mar) – 1965 (Jun)	Monthly	no number
Environmental Monitoring Monthly Report	1965 (Jul) – 1979 (Jan)	Monthly	no number
Works Technical Department Monthly Report	1979 (Feb – Dec)	Monthly	no number
Earth Sciences Monthly Report	1965 (Jul) – 1972 (Oct)	Monthly	no number
Radiological Sciences Monthly Report	1965 (Jul) – 1972 (Nov)	Monthly	no number
Radiological Chemistry Monthly Report	1971 (Dec) – 1972 (Aug)	Monthly	no number
Weekly Report – Control	1954 (May) – 1959 (Nov)	Weekly	no number
Weekly Report – Control and Methods	1959 (Nov) – 1961 (Feb)	Weekly	no number
Weekly Report – Environmental Monitoring and Allied Studies	1961 (Feb – Aug)	Weekly	no number
Weekly Report – Environmental Monitoring and Allied Studies	1961 (Aug) – 1962 (~May)	Biweekly	no number
Weekly Report – Environmental Monitoring and Allied Studies	1962 (~May) – 1962 (Oct)	Monthly	no number

A primary source of environmental monitoring data is handwritten compilations of effluent and environmental monitoring results on ledger sheets. At some point in time, each sheet was photographed by the Site in a format similar to microfiche and mounted on a computer card with a hole cut in the center. These cards are called aperture cards. The time period covered by the aperture cards varies depending on the media (Table 7-2) and the type of measurements. Not all of the data on the aperture cards are useful for dose reconstruction; therefore, we reviewed the aperture cards to determine if the data should be compiled in an electronic format. Data that were not relevant included measurements of feral coliform bacteria, river algae, area film badge thermoluminescent dosimeter measurements, and fly paper autoradiographs. Other data were determined not to be useful because individual sample locations could not be identified. Table 7-2 summarizes the environmental monitoring data for radionuclides available from the aperture cards for the media of interest.

The data on the aperture cards have been cross-checked with the data presented in the various weekly, monthly, quarterly, semiannual and annual environmental monitoring reports. In general, good agreement is observed between the two sources of data. Further details about the specific media are provided in Chapters 8, 9, 10, 11, 12.1, 12.2, 13, and 14.

Table 7-2. Summary of Useful Aperture Card Data for Radionuclides

Environmental media	Time period of data	Measurements[a]
Air	1957–1973	Gross alpha and beta, fission products
Rainwater	1959–1973	^{131}I, ^{90}Sr, ^{137}Cs, tritium
Surface water	1962–1973	Gross alpha and beta, tritium
Fish	1970–1981	^{137}Cs, ^{90}Sr
Vegetation	1957; 1959–1973	Gross alpha and beta, fission products, tritium
Milk	1959–1973	^{131}I, ^{90}Sr, ^{137}Cs, tritium
Agricultural products	1966–1973; 1982–1983	Gross alpha and beta, fission products
Wild game	1965–1966; 1970–1982; 1984–1985	Nonvolatile beta, ^{137}Cs, radioiodine

[a] Available measurements may vary by year.

Independent (non-SRS) Data Sources

In a public study such as this dose reconstruction, independent monitoring by groups can be important for developing trust in the Site's monitoring results. These monitoring data are valuable because they have been generated by organizations or groups independent of the day-to-day running of the Site and can be used to determine the quality and reliability of the SRS data. An understanding of background levels can be particularly important to the dose reconstruction process at U.S. Department of Energy sites, because often the time periods of highest releases from the sites overlap with the time of highest depositions of fallout from weapons testing. These independent monitoring programs are rarely as comprehensive, in terms of localized monitoring, as the Site's program, but they can provide a cross-check of results generated by the site at a similar location and time.

In Chapters 8, 9, 10, 11, 12.1, 12.2, 13, and 14, environmental monitoring datasets for radionuclides identified as potentially useful for the dose reconstruction are evaluated. Where possible, environmental monitoring data useful for the dose reconstruction that have not been collected by the SRS are also evaluated. In general, the independent data are most useful for those years when atmospheric fallout from nuclear weapons testing was high and when they provide information about background concentrations of radionuclides at locations away from the Site. A summary of the independent data that have been compiled is provided in Table 7-3.

One source of useful independent monitoring data is the U.S. Environmental Protection Agency (EPA), started by the U.S. Public Health Service (PHS) almost 40 years ago. The U.S. EPA has monitored rainwater and has reported tritium concentrations since 1974 for various locations, including Columbia and Barnwell, South Carolina. See Chapter 8 for additional details.

The PHS also had a milk monitoring program that started in 1957. Milk was sampled from locations throughout the United States. The three monitoring locations closest to the SRS were Atlanta, Georgia; Charleston, South Carolina; and Charlotte, North Carolina. The PHS milk monitoring data provide an independent source of information about the concentrations of fallout radionuclides found in milk in the southeastern United States and allow comparison with the

concentrations measured in milk samples from around the SRS. See Chapter 10 for more detailed information.

The South Carolina Department of Health and Environmental Control (SCDHEC) and the Georgia Department of Natural Resources (GDNR) generated additional information regarding radionuclide concentrations in fish from the Savannah River. These organizations began routine fish collection programs in the early 1970s and 1980s and provide ^{137}Cs, ^{90}Sr, and tritium concentrations for fish collected from various locations along the river. These data provide independent sources of information that can be used verify concentrations reported by the SRS. See Chapter 14 for a more detailed discussion of data provided by these organizations.

Table 7-3. Summary of Compiled Independent Monitoring Data[a]

Environmental Media	Data source	Measurements	Time period of data[b]
Air	U.S. PHS	beta	1959–1964
Rainwater	U.S. EPA	tritium	1982–1990
Milk	U.S. PHS/ U.S. EPA	^{137}Cs, ^{140}Ba, ^{131}I, ^{89}Sr, ^{90}Sr	1960-1993
Offsite surface water:			
Savannah River	U.S. PHS	gross alpha, beta	1956–1960
Savannah River	U.S. PHS	^{90}Sr	1956–1957, 1961—1964
Savannah River	USGS[c]	tritium	1962–1964
Kissimmee River (FL)	USGS	tritium	1962–1964
Neuse River (NC)	USGS	tritium	1962–1964
Stream (Waynesboro, GA)	USGS	tritium	1962–1964
Fish	GDNR	tritium, ^{137}Cs, ^{90}Sr	1982–1997
	SCDHEC	^{137}Cs	1975–1995

[a] See Chapters 8, 9, 10, 11, 12.1, 12.2, 13, and 14 for specific details.
[b] Available measurements may vary by year.
[c] USGS = United States Geological Survey.

Quality Assurance and Quality Control

The early sampling and analytical techniques used by SRS were limited by the quality of the sampling equipment and detectors available at the time. Furthermore, limitations are associated with some of the SRS environmental datasets, such as inadequate control locations, improper calibrations, and nonrepresentative sampling techniques. These limitations, along with the overall quality of the monitoring program at any given time, must be considered as datasets are identified for use in a specific dose assessment.

The semiannual and annual environmental reports before 1975 do not mention quality assurance or quality control. Discussions with personnel involved in the environmental monitoring program indicate that there were no interlaboratory comparisons in the 1950s and 1960s. However, beginning with the 1958 reports, a table of sensitivities and standard deviations of the laboratory analyses is provided in each semiannual or annual report. The standard deviations are based on spike recovery values.

The 1975 annual report (Ashley and Ziegler 1976) includes a "Monitoring Results Treatment and Quality Control" section, which provides a description of the quality control program including participation in the EPA Interlaboratory Comparison Program. No cross-check data are reported.

In 1976, in addition to participation in the EPA Interlaboratory Comparison Program, SRS participated in an Energy Research and Development Administration (ERDA)-wide quality assurance program conducted by the Health and Safety Laboratory of New York City. The results of the cross-check programs are not provided. The 1976 annual report (Ashley and Ziegler 1978) discusses quality control for nonradiological analyses and provides a comparison of SRS and Georgia Department of Natural Resources data for certain water quality parameters.

Results of SRS participation in the interlaboratory cross-check program are provided in annual reports starting in 1977. An appendix detailing the SRS quality control program can be found in the annual reports starting in 1980.

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CHAPTER 8

RADIONUCLIDES IN AIR AND RAINWATER

ABSTRACT

This chapter discusses radionuclide concentrations measured in air and rainwater collected on or in the vicinity of the Savannah River Site (SRS). We evaluated these data with regard to their potential usefulness for dose reconstruction. We also attempted to validate the reported concentrations by making comparisons among as many data sources as possible. These data included original handwritten compilations for many years. We have found these original data to consistently correspond to the information reported in monthly, semiannual, and annual summary reports.

We compiled and examined air and rainwater data to determine their usefulness for source term verification, model validation, and direct exposure assessment. In general, the air data are most valuable for source term verification and model validation. However, the potential usefulness of the data may be limited in many cases by spatial resolution. The rainwater data may be useful for identifying long-term trends; however, they are limited in their use for source term verification and model validation. Appendix K further discusses potential uses for environmental monitoring data.

INTRODUCTION

Air and rainwater have been sampled at various locations on or in the vicinity of the SRS since measurements of natural background levels of radioactivity began in 1951. Air represents a major pathway for transport of SRS releases and exposure of offsite individuals, and so can be extremely important to a historic dose reconstruction. Radionuclide-specific data from air filters located in the effluent stream from stacks may be used to validate airborne source terms and dispersion models. In the absence of specific effluent data, air monitoring results could be used to develop airborne source terms, particularly in cases of unplanned releases. If the data are of high quality and above detection limits, they may also be used directly to quantify human exposure via the airborne pathways.

This chapter summarizes reported information regarding radionuclide concentrations in air and rainwater on or in the vicinity of the SRS. We examined several sets of routine semiannual and annual environmental monitoring reports prepared by the SRS contractor, which span the years 1953 through 1991. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series. We reviewed additional documents for information pertaining to air and rainwater monitoring at the SRS. These include monthly monitoring reports from 1951 through 1956, from 1960 through 1971, and from 1976 through 1990; weekly monitoring reports from 1954 through 1964; and handwritten compilations of original environmental monitoring results from 1959 through 1973 that were photographed in a format similar to microfiche.

The handwritten compilations of original effluent and environmental monitoring results, recorded on ledger sheets, are the primary sources of original data. The data represent results reported for each time period of measurement (e.g., weekly or biweekly). The sheets were photographed in a format similar to microfiche and mounted on computer cards with a hole cut in

the center. These cards, called aperture cards, were found for August 1959 through December 1973. The aperture card data were compiled in Excel spreadsheets and were cross-checked with the data presented in the semiannual and annual Health Physics monitoring reports. Generally, average concentrations reported for a specific time period (usually 6 months or a year) in a report were compared with averages computed using the aperture card data on the Excel spreadsheets. In general, good agreement is observed between the aperture cards and reports. For the time period before August 1959, data are available from the weekly and semiannual monitoring reports. These data are incomplete and are typically reported as semiannual averages. From 1974 onward, data are available on computer printouts, called monthly reports, in frequencies that correspond to the sampling periods.

A synopsis of the types of data available, their sources, and the name of the Excel workbooks containing the aperture card data are shown in Table 8-1. Data from semiannual, annual, and monthly reports were not electronically compiled for two main reasons. First, they are easily obtained from the reports. On the other hand, aperture card data are not easily handled (one must either read the cards with a special reader or handle oversized photocopies) and tend to be illegible. Second, data from semiannual and annual reports represent averages for each 6-month period or year and do not provide the degree of resolution needed for a dose reconstruction that individual sample results will provide.

The results were evaluated in light of their potential usefulness to the dose reconstruction. Usefulness was considered in terms of the following:

- The contaminant is a key contaminant (i.e., screened in Task 3 of Phase I of the dose reconstruction to potentially contribute significantly to dose [Meyer et al. 1995])
- The contaminant was monitored during the period of interest (i.e., 1951 through 1990)
- Contaminant concentrations were above detection limits
- The results demonstrate expected trends (spatially and temporally)
  - Concentrations were generally higher onsite than offsite
  - Concentrations were generally higher during periods of elevated releases
- The medium is directly impacted by releases.

The potential usefulness of the data was considered to ensure that efforts were focused on those contaminants that would most likely be addressed in the next phase of the dose reconstruction.

The following sections present a description and evaluation of the air monitoring program and a description and evaluation of the rainwater monitoring program. Following the discussions of the air and rainwater data, data usefulness and limitations are addressed. Appendix A contains details related to the analytical procedures associated with collecting and measuring air samples.

Table 8-1. Description of Airborne Radionuclide Data

Radionuclide	Time period	Description of data	Source of data	Name of Excel workbook
Gross alpha	1955—1958	Semiannual average and maximum for all locations	Semiannual reports	Data not compiled
	1959—1973	Individual results[a]	Aperture cards	Alpha-Beta-all.xls[b]
	1974—1975	Annual average and maximum for each location	Annual reports	Data not compiled
	1976—1990	Individual results	Monthly reports	Data not compiled
Gross particulate beta	1954—1958	Semiannual average and maximum for all locations	Semiannual reports	Data not compiled
	1959—1973	Individual results	Aperture cards	Alpha-Beta-all.xls[b]
	1974—1975	Annual average and maximum for each location	Annual reports	Data not compiled
	1976—1990	Individual results	Monthly reports	Data not compiled
^{131}I	1954—1958	Semiannual average and maximum for all locations	Semiannual reports	Data not compiled
	1959—1973	Individual results	Aperture cards	Iodine-all.xls[b]
	1974—1975	Annual average and maximum for each location	Annual reports	Data not compiled
	1979—1990	Monthly composite results for onsite (three sites), perimeter, 25-mi, and 100-mi areas	Monthly reports	Data not compiled
$^{89, 90}Sr$	1967—1975	Semiannual averages of monthly composite results for F-Area and H-Area, plant perimeter, 25-mi radius, and 100-mi radius areas	Annual reports	Data not compiled
	1976—1990	Monthly composite results for F-Area, H-Area, and 3/700 Area, perimeter, 25-mi, and 100-mi areas	Monthly reports	Data not compiled
	1974—1990	Annual, minimum, maximum, and average values for each location	Annual reports	Data not compiled
^{3}H	1956—1958	Maximum result	Semiannual reports	Data not compiled
	1959—1973	Individual results	Aperture cards	Tritium-all.xls[b]
	1974—1975	Annual minimum, maximum, and average for each location	Annual reports	Data not compiled
	1976—1990	Individual results	Monthly reports	Data not compiled

Table 8-1. (Continued)

Radionuclide	Time period	Description of data	Source of data	Name of Excel workbook
Gamma nuclides (^{137}Cs, ^{7}Be, ^{54}Mn, $^{95}Zr/Nb$, ^{106}Ru, ^{144}Ce	1964–1973	Monthly composite results for F-Area, H-Area, and 3/700 Area, perimeter, 25-mi, and 100-mi areas	Aperture cards	Gamma-all.xls[b]
	1974–1990	Annual minimum, maximum, and average for each location	Annual reports	Data not compiled
	1979–1990	Monthly composite results for F-Area, H-Area, and 3/700 Area, perimeter, 25-mi, and 100-mi areas	Monthly reports	Data not compiled
^{238}Pu, ^{239}Pu	1977–1990	Monthly composite results for F-Area, H-Area, and 3/700 Area, perimeter, 25-mi, and 100-mi areas	Monthly reports	Data not compiled
^{3}H in rainwater	1954-1963, 1974-1990	Semiannual or annual maximum, and average for each location	Semiannual and annual reports	Data not compiled
^{131}I in rainwater	1954-1963	Semiannual or annual maximum, and average for each location	Semiannual and annual reports	Data not compiled
Gamma radio-nuclides, Sr , and Pu in rainwater	1974-1990	Semiannual or annual maximum, and average for each location	Semiannual and annual reports	Data not compiled
Gross alpha in rainwater	1956-1963	Semiannual or annual maximum, and average for each location	Semiannual and annual reports	Data not compiled
Nonvolatile beta in rainwater	1954-1963	Semiannual or annual maximum, and average for each location	Semiannual and annual reports	Data not compiled
Specific radionuclides in rainwater	1977-1990	Individual results	Monthly reports	Data not compiled
Specific radionuclides in rainwater	1959—1973	Individual results	Aperture cards	Tritium-all.xls, Iodine-all.xls, Gamma-all.xls[b]
Particle deposition	1960	Individual results	Aperture cards	Particles-all.xls

[a] Individual results are for each location and for each period of collection (typically weekly).

[b] A worksheet is available for each year. For example, the gross beta activity data for 1959 may be found in the workbook entitled Alpha-Beta-all.XLS, in the worksheet entitled “1959 – beta”.

RADIONUCLIDE CONCENTRATIONS IN AIR

Tables 8-2, 8-3, and 8-4 (located in Addendum 8A at the end of this chapter) present summaries of the types of air monitoring data obtained by the SRS contractor at onsite, 25-mi, and 100-mi sampling locations. Each horizontal line in the tables represents a change in the routine monitoring program, such as, the addition of new sampling sites. A brief history of the air monitoring program is presented below.

During the preoperational monitoring program (Reinig et al. 1953), in June 1951–January 1953, air was sampled continuously at two onsite locations using CWS #6 filter paper and a dual pump unit. One pump was running while the other pump was off, at 6-h intervals, and the filter of the inactive pump was analyzed for gross beta. A Geiger-Mueller (GM) counter constantly recorded the activity on air filters. Radon and thoron concentrations were estimated using buildup and decay curves. Radioautographs were made of filters weekly to estimate the quantity of particles suspended in the air sampled at 12 inner perimeter and three 25-mi radius locations. Also, during the preoperational period, fallout particles were trapped through settling on adhesive flypaper emplaced at the 12 inner perimeter locations and counted weekly using radioautographs. Deficiencies in the preoperational survey are reviewed in *The 1951 Preoperational Environmental Survey for the Savannah River Plant - In Retrospect* (Du Pont 1962a).

Routine air monitoring began on January 1, 1953. The routine monitoring program was essentially a continuation of the preoperational monitoring program, conducted at selected onsite and 25-mi locations. However, additional sampling sites were gradually added to the onsite monitoring network to collect more gross beta and suspended particle data (Albenesius 1954).

Beginning in 1955, air filters were counted weekly for gross alpha, using alpha scintillation counters. Continuous air samplers with a flow rate of 2.0 cfm and 2-in. CWS #6 filters were used for this purpose. Also, in the same year, silver nitrate impregnated HV70 filters were used to collect radioiodine (Alexander and Horton 1956).

Starting in 1956, tritium in air was collected at onsite and 25-mi radius stations using silica gel. The design basis of the regional air monitoring program at that time can be found in Horton (1957).

Major changes in the air monitoring program were implemented in 1960 and 1961. Air filters were no longer analyzed for suspended particles in 1960 and the particulate fallout study was discontinued in 1961. The regional monitoring program was expanded to include background sites. Sampling at 100-mi radius locations (Columbia, South Carolina; Greenville, South Carolina; Macon, Georgia; and Savannah, Georgia, was initiated in 1961. This background monitoring program began with sampling and analysis for gross alpha and gross beta and was expanded in 1962 to include ^{131}I and gamma-emitting radionuclides. Gamma spectroscopy was also used to analyze air filters collected at onsite and 25-mi radius locations beginning in 1962. These and other changes, especially in procedures, may be found in Marter and Boulogne (1962).

By 1973, air filters were routinely analyzed for ^{89,90}Sr. Routine analysis of filters for ^{239,240}Pu followed in 1974. Iodine-131 was eliminated from the routine monitoring program from 1974 through 1979, but no documented rationale could be found for this deletion. However, it is possible that the analyses were discontinued because this radionuclide was rarely detected in air samples. The data from aperture cards show that ^{131}I was usually below the detection limits.

The 1980 air monitoring program consisted of 19 onsite, 8 offsite (25-mi radius), and 4 background (100-mi radius) monitoring locations (Du Pont 1981). The locations of the onsite and 25-mi perimeter sampling sites, referenced above, are shown in Figure 8-1.

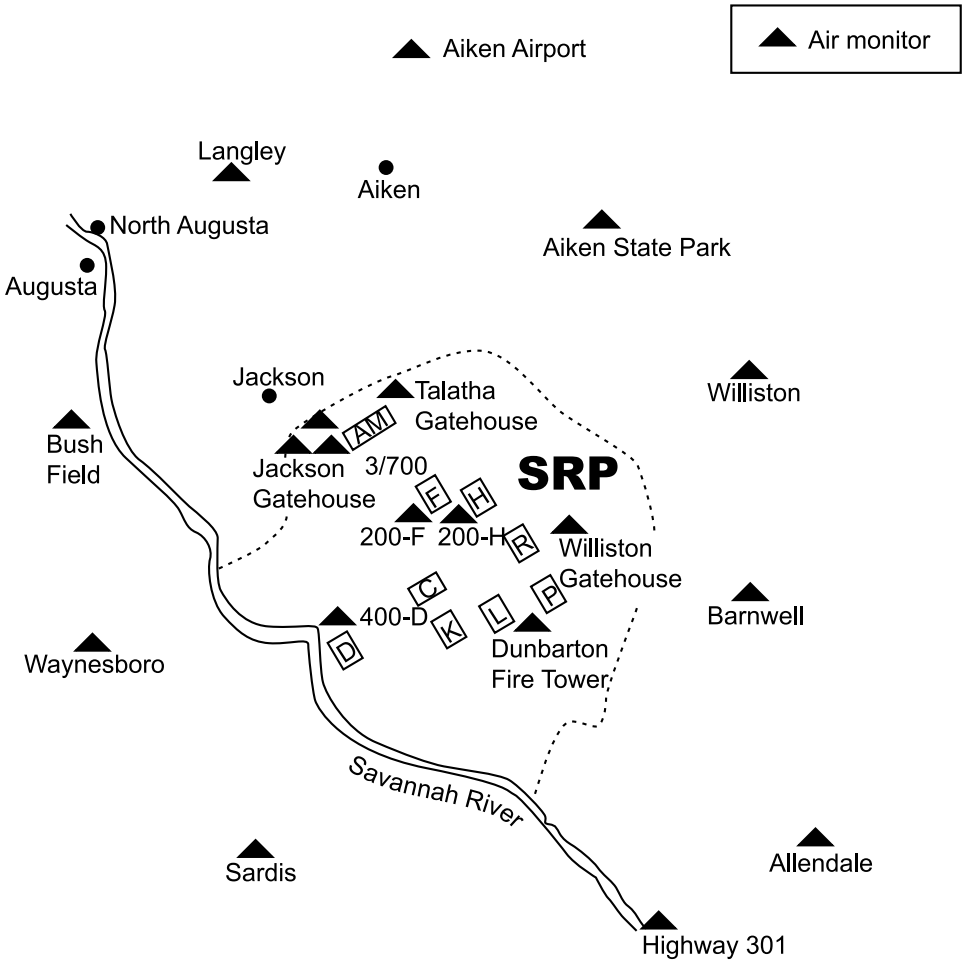

Figure 8-1. Onsite and 25-mi radius air monitoring locations.

The following sections present the results of the air monitoring program by type of radionuclide measured. The SRS environmental monitoring program results of air sampling for radionuclides discussed in this chapter have been compiled in Excel spreadsheets.

Measurements of Beta-Gamma Emitters in Air

The beta-gamma emitters that were routinely monitored in air by SRS were radioiodine, strontium, tritium, and ^{137}Cs. Collection periods varied as monitoring program personnel determined the need. In addition, gross particulate alpha and gross particulate beta activity was measured during preoperational monitoring and weekly thereafter. Two of these radionuclides (i.e., radioiodine and tritium) were identified in the initial screening process as the largest contributors to the screening dose from airborne releases (Meyer et al. 1995).

On the other hand, little knowledge can be obtained from measurements of gross alpha activity. Neither of the key radionuclides released to air is an alpha-emitter. While great effort could be made to use the gross alpha data, it is inefficient given the relative insignificance of potential doses because of releases of the key alpha emitters (plutonium and uranium). Gross alpha data were used as a screening technique to detect anomalies (i.e., sudden increases over background), not to identify specific radionuclides. For these reasons, gross particulate alpha measurements were not examined.

Air filters that were used to measure emissions from SRS facilities were not analyzed for specific isotopic alpha activity until the early 1970s because of the low release rates and the small potential for plutonium release. Instead, filters were submitted for laboratory analysis for specific alpha emitters only if gross beta-gamma activity was detected above background.

Gross Beta Measurements in Air

Gross particulate beta activity concentrations in air can be useful to demonstrate patterns, especially when the source of activity is known. This pattern was evident during the years when global fallout from nuclear weapons testing was elevated. Figure 8-2 demonstrates temporal trends for the years 1959 through 1964, peak fallout years. The peaks in activity compare well at all locations and with measurements made by the U.S. Public Health Service (PHS) at Columbia. This figure confirms that peak measurements were primarily due to global fallout and not to SRS operations.

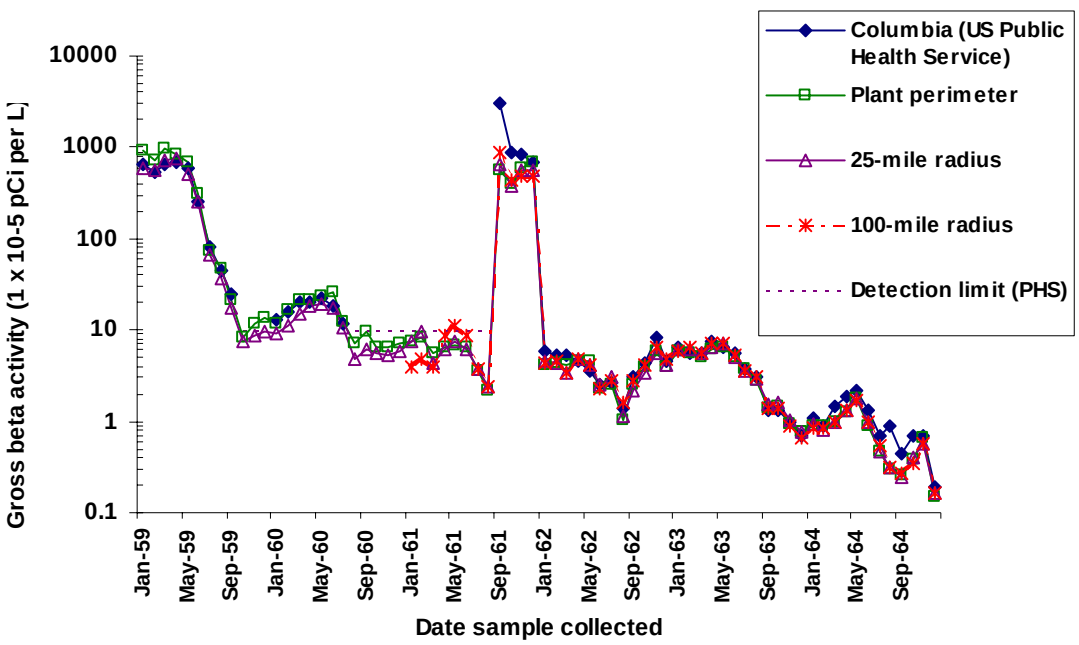

Figure 8-2. Average monthly concentrations of particulate beta radioactivity measured on and off the SRS by the SRS and by the U.S. Public Health Service from 1959 through 1964. The highest gross beta activity (2970 × 10-5 pCi L^{-1}) was measured in Columbia in September 1961. Link to tabulated figure data.

The close correspondence between PHS and SRS data particularly during the period from June 1962 through 1964 serves to verify SRS measurements.

Radioiodine Measurements in Air

Sampling for volatile and particulate forms of ^{131}I at onsite, plant perimeter, and 25-mi radius locations began in 1955. Sampling at locations 100 mi from the SRS was initiated in 1962 to provide data on background concentrations of radionuclides (Du Pont 1962b).

According to Kantelo et al. (1990), ^{131}I has been detected at locations close to the F and H separations areas in most years. At the plant perimeter and 25-mi locations, ^{131}I was detected from 1955 through 1963. Near the 100-mi radius, ^{131}I was detected only during 1962 and 1963.

Original data (i.e., from aperture cards) could not be found for years before 1959. This is unfortunate because the highest average concentrations of ^{131}I in air were reported during 1956 (see Table 8-5.) Figure 8-3 presents average monthly ^{131}I concentrations measured in air at onsite, perimeter, 25-mi, and 100-mi areas from 1959 through 1966. We calculated these averages by using original data. The highest monthly averages estimated for this period are generally far less than the semiannual averages reported for 1956 (Mealing 1957a, 1957b).

Table 8-5. Average Semiannual ^{131}I Concentrations in Air from 1955 through 1958

	Average radioiodine concentration in air (10^{-14} μCi mL^{-1})						
Location	Jul.–Dec. 1955	Jan.–Jun. 1956	Jul.–Dec. 1956	Jan.–Jun. 1957	Jul.–Dec. 1957	Jan.–Jun. 1958	Jul.–Dec. 1958
400-F[a]	7	700	580	150	94	6	22
200-H[a]	28	570	470	390	110	8	28
Dunbarton Fire Tower[b]	7	Data incomplete	47	54	49	8	26
Talatha Gatehouse[b]	3	67	38	40	54	8	24
400-D[b]	6	60	36	41	42	8	12
Williston Gatehouse[b]	10	64	34	45	53	8	25
700-A[b]	3	70	20	27	42	7	31
Waynesboro[c]	4	14	19	30	43	7	24
Aiken Airport[c]	2	19	16	27	20	8	22
Allendale[c]	3	13	12	21	28	8	19

[a] Onsite location.
[b] Plant perimeter location.
[c] 25-mi radius location.

Figure 8-3 also indicates that SRS releases impacted concentrations of ^{131}I measured in air at perimeter and 25-mi locations during the first halves of 1959 and 1961. Interestingly, ^{131}I concentrations appear to be equivalent during the last part of 1961, 1962, and the first part of 1963. This pattern confirms that shown in Figure 8-3 and may reflect increased global fallout. After 1963, only onsite concentrations were consistently measured above the detection limit.

The impact of SRS releases on perimeter locations can also be examined by looking at the ratios of the monthly ^{131}I concentrations at different areas (see Figure 8-4). For example, the ratio

of onsite to 25-mi radius ^{131}I concentrations are, with few exceptions, greater than 1. This indicates that F-Area and H-Area concentrations are elevated relative to 25-mi radius locations from 1959 through 1963, reflecting emissions from F-Area and H-Area stacks. The times when the ratio approaches or equals 1 indicates equivalent concentrations at both areas, possibly reflecting global fallout (e.g., late 1959, early 1961, late 1962, and early 1963). Figure 8-4 also shows ^{131}I concentrations at plant perimeter locations relative to concentrations measured at 25-mi radius locations. Radioiodine concentrations have been elevated at plant perimeter locations relative to 25-mile radius locations only a few times (most notably, during June and July 1959, the summer of 1961, and October 1961).

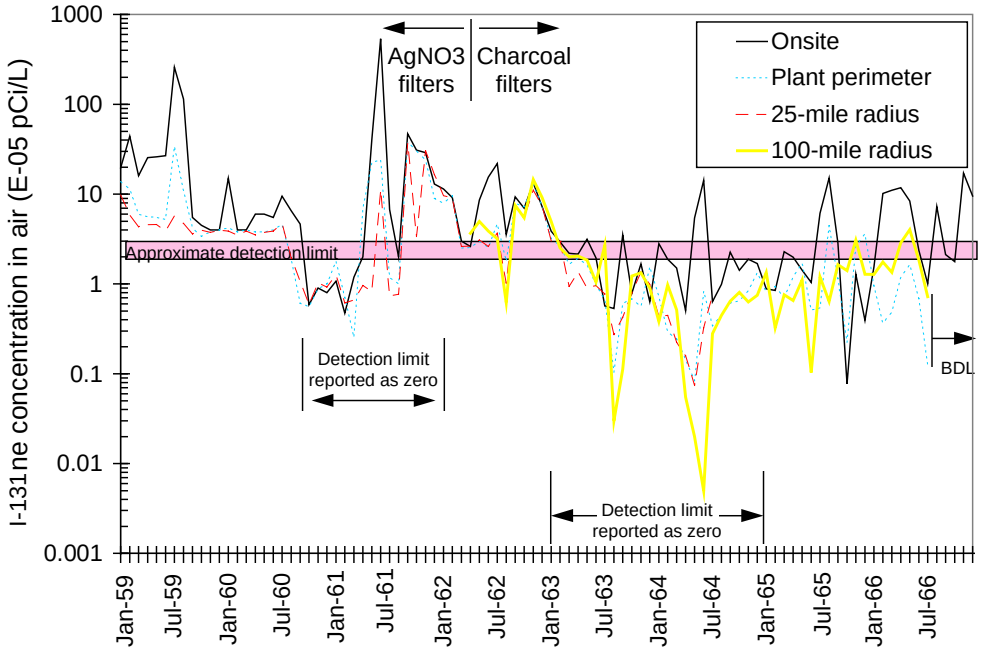

Figure 8-3. Average monthly concentrations of ^{131}I measured on and off the SRS by the SRS from 1959 through 1966 (BDL=all individual measurements below detection limits). The approximate detection limit during this time was about 2×10^{-5} pCi L^{-1}. Link to tabulated figure data.

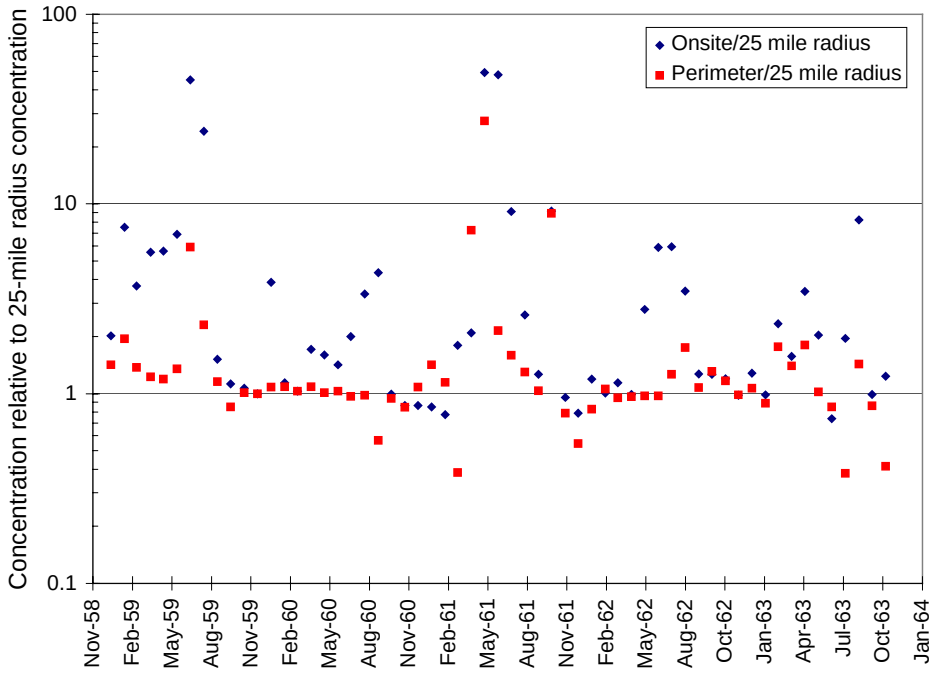

Figure 8-4. Iodine-131 concentrations measured at F-Area, H-Area, and 25-mi radius locations relative to concentrations measured at 25-mi radius and perimeter locations. Link to tabulated figure data.

Although original data were not available for all years (e.g., before 1956), other reports were found that assessed original data to ascertain temporal and spatial patterns. Kantelo et al. (1990) used a slope analysis technique to determine that the SRS was the principle source of radioiodine in the local atmospheric environment during 1956. Annual average ^{131}I concentrations were estimated for each onsite, perimeter, and 25-mi sampling location and plotted against distance (Figure 8-5). The distances used were measured from the center of the two Chemical Separations Areas because greater than 99% of the atmospheric radioiodine released from the SRS originated from this area (Kantelo et al. 1990). Logarithmic scales were used so that a straight line would result. The slope of this line was then used to indicate whether the Site emissions impacted the perimeter and offsite locations. A slope of one indicates no difference between concentrations measured at onsite, perimeter, and 25-mi stations. A large negative slope implies that SRS releases influenced ^{131}I concentrations measured at offsite areas. In Figure 8-5, it is obvious that releases from the separations area had a much greater impact on the local offsite environment in 1956 than in 1962. Slopes calculated in this manner by Kantelo et al. (1990) show the greatest impact from the SRS occurred during 1956 (see Table 8-6). Slopes approached zero during 1962 and 1963, reflecting global fallout concentrations.

Figure 8-6 illustrates the slopes derived, from aperture card data, for the years 1959 through 1963. Figure 8-7 is another way of illustrating the impact of SRS releases on the immediate environment. In this case, the chart axes are expressed using an arithmetic, rather than a geometric, scale.

Table 8-6. Slopes of Annual Average ^{131}I Concentrations in Air as a Function of Distance[a]

Year	Slope
1955	-0.46
1956	-1.05
1957	-0.52
1958	-0.27
1959	-0.62
1960	-0.11
1961	-0.32
1962	-0.06
1963	0.00

[a] Source: Kantelo et al. (1990).

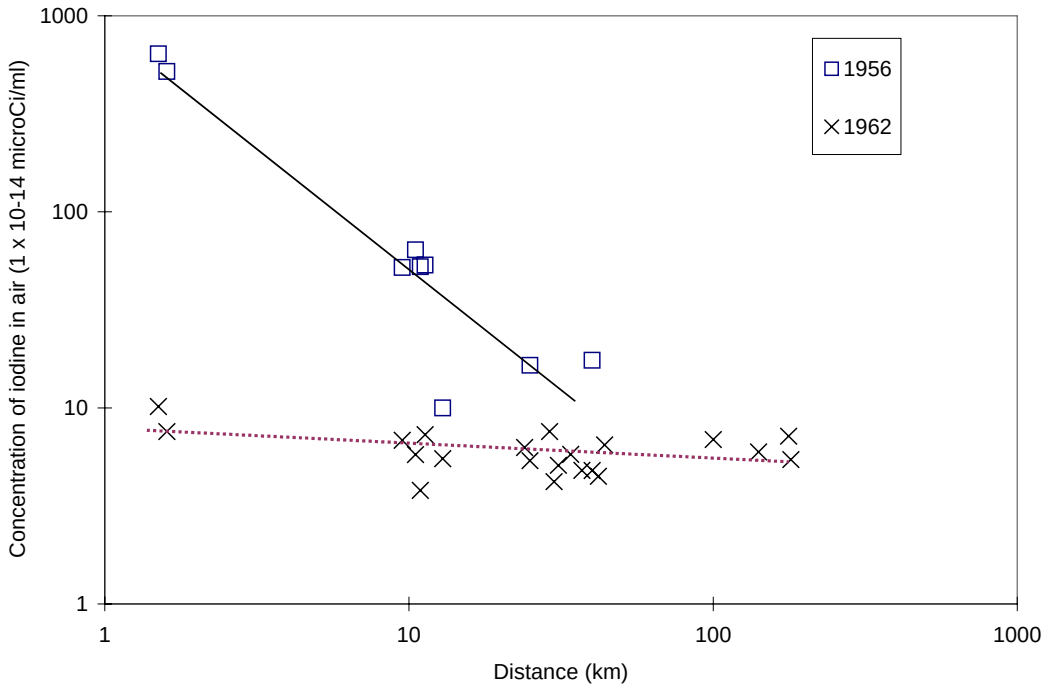

Figure 8-5. Annual average concentrations of ^{131}I in air with distance from the center of the Chemical Separations Area at SRS for two representative years (source: Kantelo et al. 1990). Link to tabulated figure data.

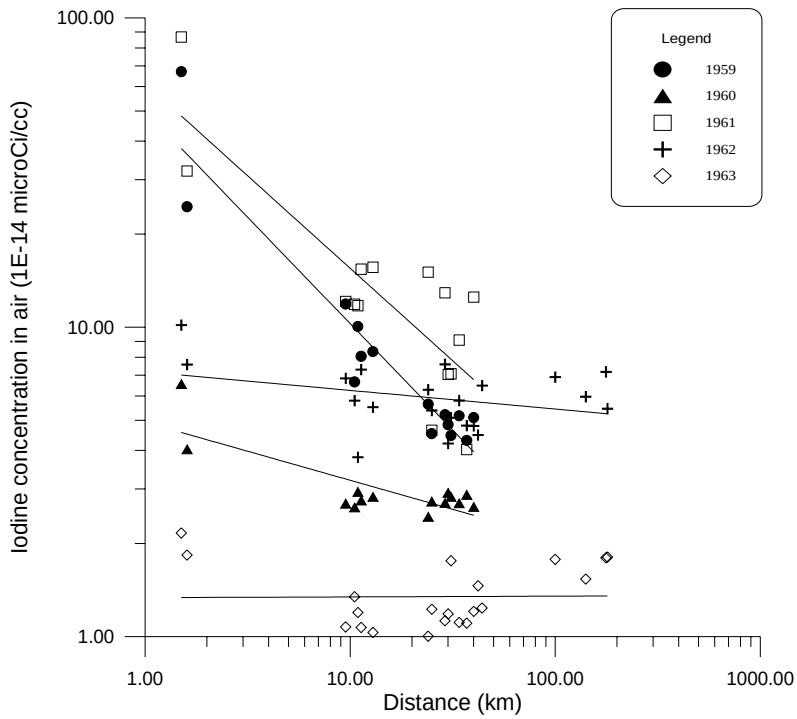

Figure 8-6. Annual average concentrations of ^{131}I in air with distance from the center of the Chemical Separations Area at SRS for the years 1959–1963 (data derived from aperture cards). The figure shows that he highest concentration was measured closest to the site in 1961 at about 90×10^{-14} μCi ml^{-1} (9×10^{-5} pCi L^{-1}). Link to tabulated figure data.

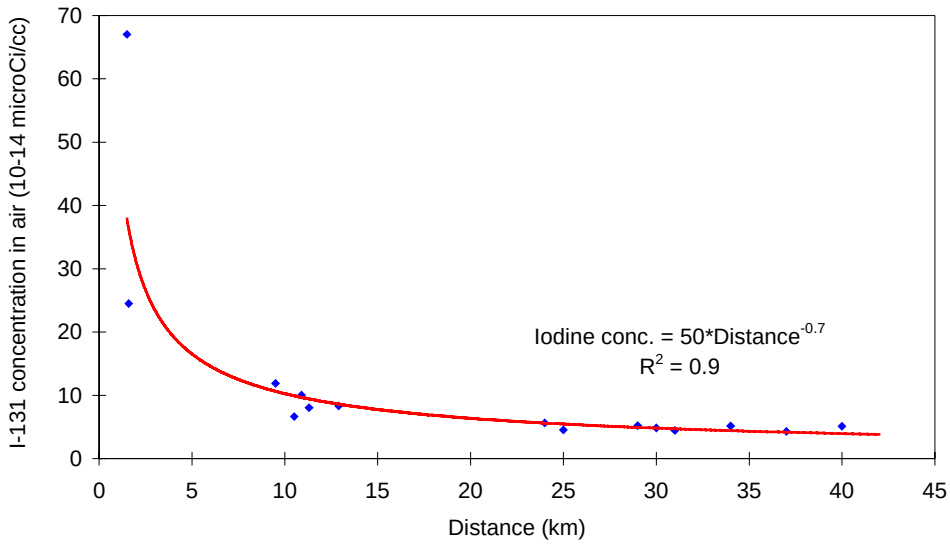

Figure 8-7. Annual average concentrations of ^{131}I in air with distance from the center of the Chemical Separations Area at SRS during 1959 (data derived from aperture cards). Link to tabulated figure data.

There are no apparent spatial dispersion patterns associated with ^{131}I releases to the atmospheric environment as illustrated in Figures 8-8 through 8-10. Figure 8-8, which represents 1959, shows elevated concentrations near the source of release (1.5 km), somewhat elevated concentrations at the plant perimeter (9.5–13 km), and background concentrations at the 25-mi (25–44 km) radius locations. However, no one direction appears to predominate in terms of concentration. This reflects the variable wind patterns at the SRS. Figures 8-9 and 8-10, which represent the fallout years of 1962 and 1963, show fairly equivalent concentrations as a function of distance and direction; however, the onsite concentrations appear to be slightly higher.

Ideally, the original data (collected weekly and biweekly) can be used to verify or identify routine and unplanned releases. For example, according to Kantelo et al. (1990), the only acute release incident because of the inadvertent reprocessing of very short-cooled materials occurred during May and June 1961. A plot of weekly concentrations of ^{131}I in air measured at onsite and perimeter locations clearly shows elevated concentrations during this period (see Figure 8-11). Figure 8-11 also indicates elevated releases from October through December, reflecting increases in production rates.

Figure 8-12 compares release data with data monitored at the two onsite locations for the year 1964. Elevated concentrations at 200-F and 200-H during May and June tracked increased releases during this period. However, higher releases of ^{131}I during August and September are not closely correlated with concentrations measured at the two onsite locations during the same period. The small number (two) of onsite stations decreases the probability that an elevated release event will be detected in the environment.

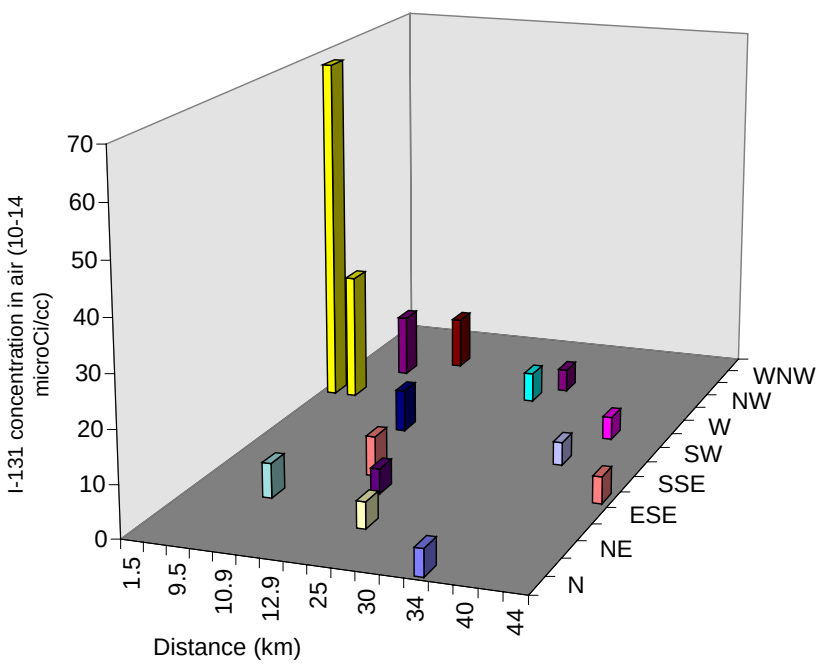

Figure 8-8. Annual average concentrations of ^{131}I in air with distance and direction from the center of the Chemical Separations Area at SRS during 1959 (note that X value axis is not to scale) (data derived from aperture cards). Link to tabulated figure data.

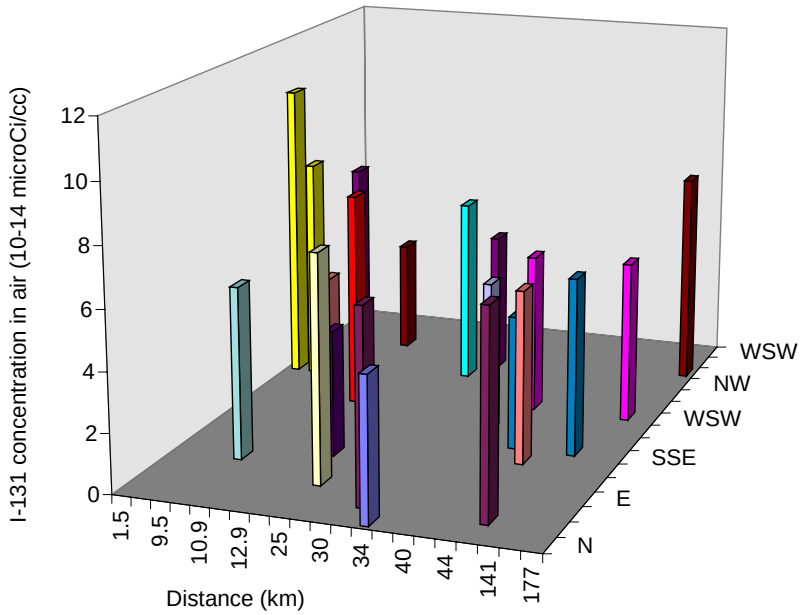

Figure 8-9. Annual average concentrations of ^{131}I in air with distance and direction from the center of the Chemical Separations Area at SRS during 1962 (note that X value axis is not to scale) (data derived from aperture cards). Link to tabulated figure data.

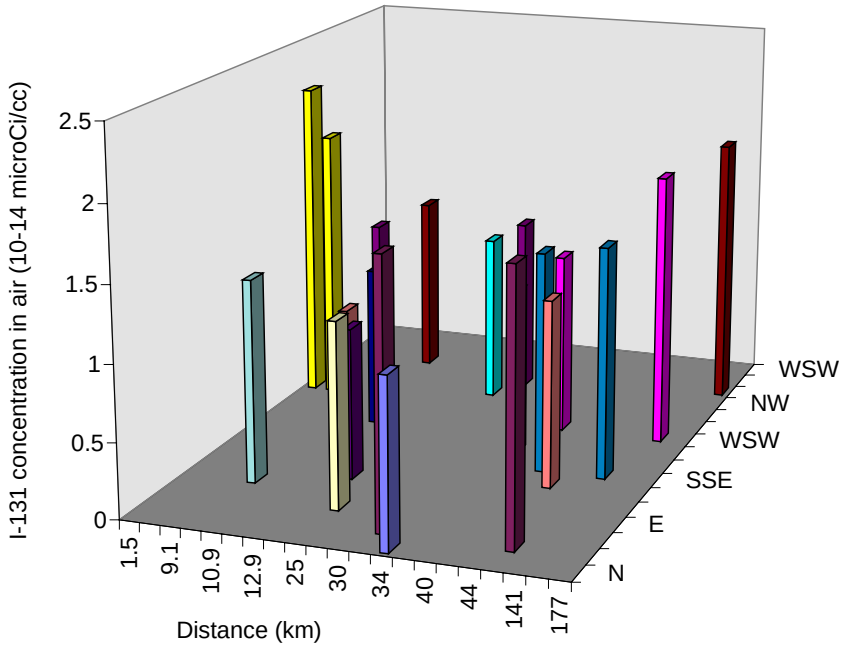

Figure 8-10. Annual average concentrations of ^{131}I in air with distance and direction from the center of the Chemical Separations Area at SRS during 1963 (note that X value axis is not to scale). Data derived from aperture cards. Link to tabulated figure data.

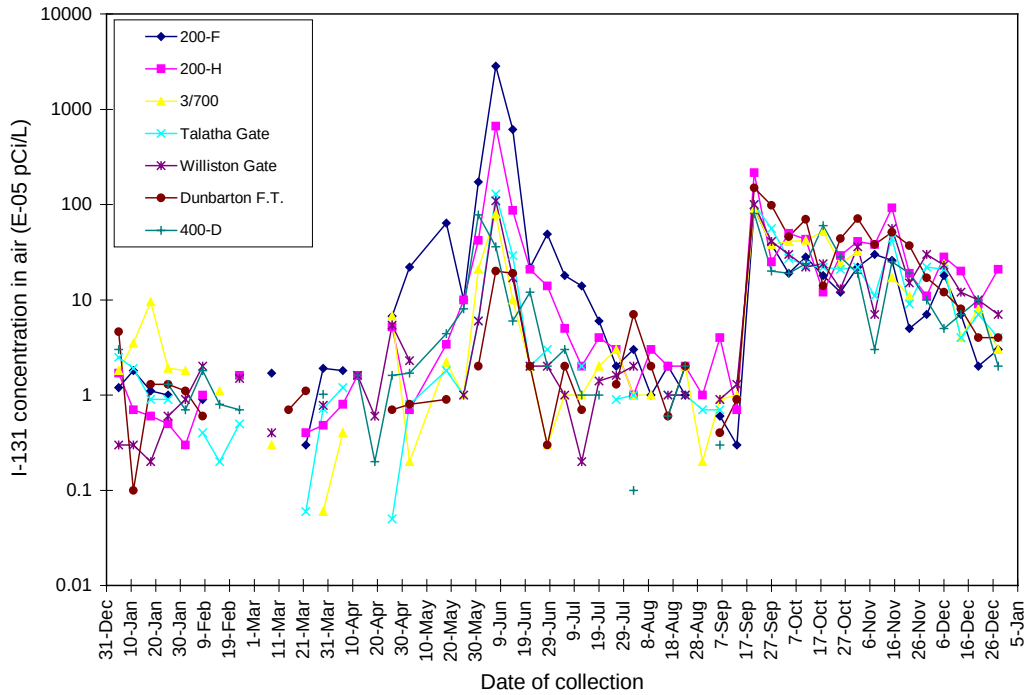

Figure 8-11. Concentrations of ^{131}I measured in air at onsite and perimeter locations during 1961 showing the impacts of incidents during May and June and increased atmospheric releases from the SRS during October through December (data derived from aperture cards). Link to tabulated figure data.

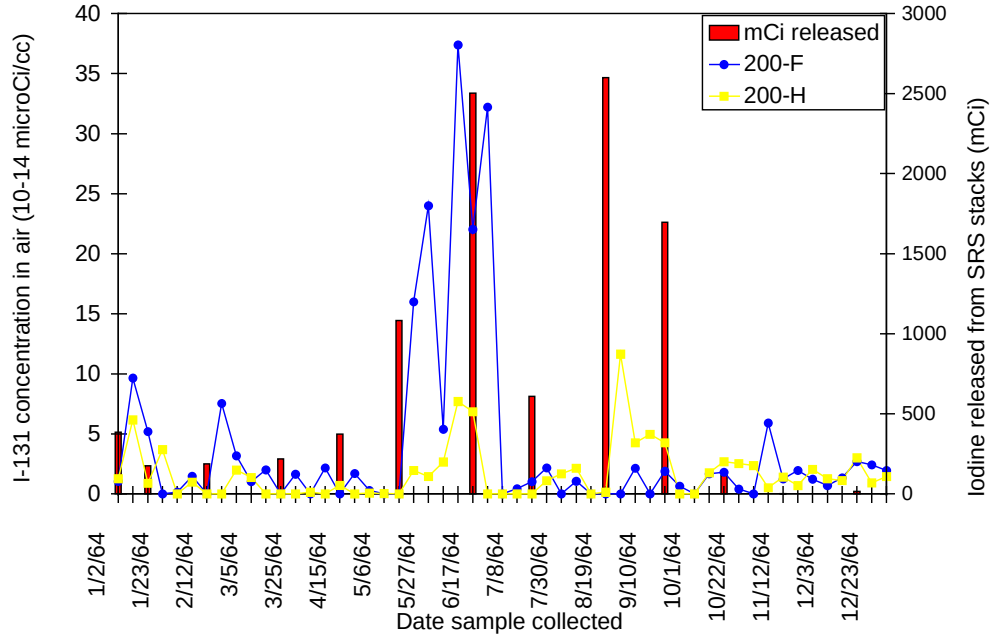

Figure 8-12. Comparison of monthly ^{131}I releases with ^{131}I concentrations measured in air effluents and two onsite locations during 1964 (data derived from aperture cards). Link to tabulated figure data.

Radiostrontium Measurements in Air

Analysis of air filters for ^{90}Sr at onsite, plant perimeter locations, 25-mi radius, and 100-mi radius began in 1967. Carlton et al. (1992) evaluated the spatial pattern with distance for two time periods: during (1967–1975) and after (1976–1986) atmospheric nuclear testing. They found no consistent spatial pattern with distance from the SRS. However, they observed that the average ratio of $^{90}Sr/^{137}Cs$ inside the SRS boundary was similar to that found in bomb fallout during the earlier period and more like those from SRS releases after 1976. They also noted that measurements made after 1976 were very low and had correspondingly high measurement errors.

Unfortunately, individual sample measurements were not available and, thus, an independent, in-depth data evaluation could not be made.

Tritium Measurements in Air

Measurements of the tritium content of atmospheric water vapor have been made since 1960 at and around the SRS. Tritium is the only radionuclide that has been consistently measured above detection levels by the air monitoring network. The major source of tritium in the environment during the period of SRS operations is from nuclear testing, which began in 1945 and peaked in the early 1960s (Murphy et al. 1991). Most of the tritium released from the SRS has been from the tritium facilities (F-Area and H-Area). Tritium released from these areas is in two forms: elemental tritium (HT) and the oxide form (HTO). The oxide form (tritiated water) is approximately 25,000 times more radiotoxic than elemental form because it is easily absorbed and incorporated into living tissue (Elwood 1971). Tritium released from the reactor areas is almost entirely HTO. The SRS air monitors contain silica gel that absorbs water and, therefore, sample the HTO form of tritium. Studies of the HT-to-HTO ratios in atmospheric releases from the separations areas were conducted from 1976 through 1979. These studies indicated that the overall percentages for the 1976–1979 period were 44% HT and 56% HTO (Murphy et al. 1991). However, significant variations in release rates and HT-to-HTO ratios were observed because of differences in the nature of the work and in the operating periods of the various facilities.

There appears to be little correlation between reported atmospheric releases with environmental measurements of tritium in air (see Figures 8-13 and 8-14.) The primary factors that can result in environmental measurements that do not track release measurements include the form of tritium released and variable meteorological conditions over the period of release. The environmental samplers only collected tritiated water, not tritiated hydrogen gas. Meteorological factors, such as temperature, wind speed, and wind direction, determine the direction, dispersion, and height of a contaminant plume. The environmental samplers, thus, may not have always been in the plume path or they sampled concentrations that were not necessarily representative of mean concentrations in the plum. However, air data consistently show a decrease in tritium concentration relative to distance from the Site (Figures 8-15 and 8-16).

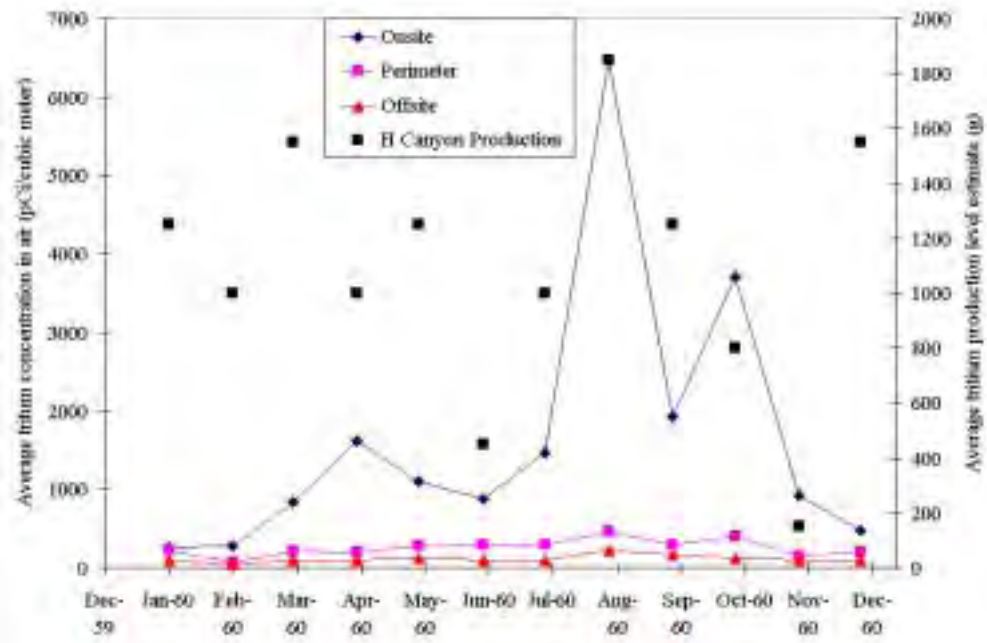

Figure 8-13. Average monthly tritium concentrations in moisture in air measured at onsite (F-Area and H-Area), SRS perimeter, and offsite (25-mi radius) locations during 1960. Production rates projected for the H-Area facility are also shown (see Chapter 2 of this report) (data derived from aperture cards). Link to tabulated figure data.

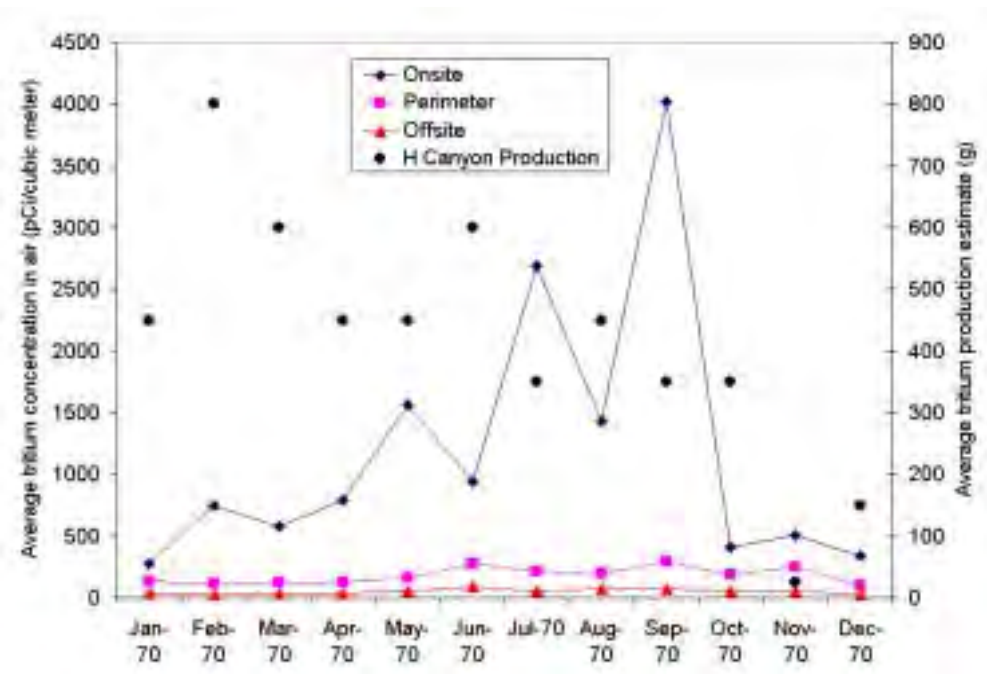

Figure 8-14. Average monthly tritium concentrations measured in moisture in air at onsite (F-Area and H-Area), SRS perimeter, and (25-mi radius) offsite locations during 1970. Production rates projected for the H-Area facility are also shown (see Chapter 2 of this report) (data derived from aperture cards). Link to tabulated figure data.

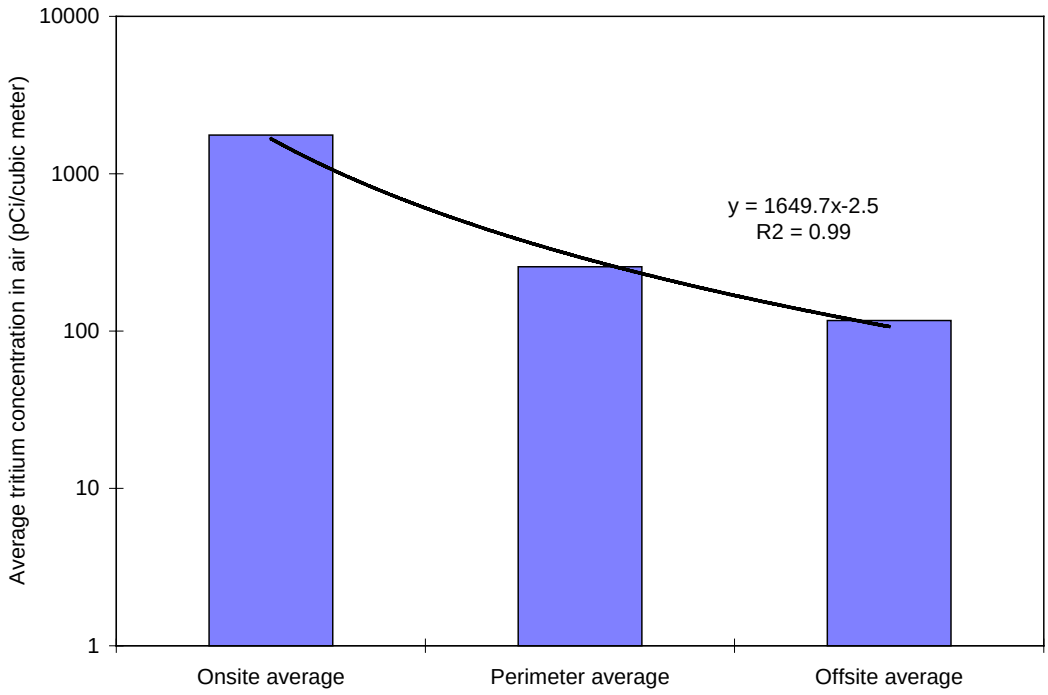

Figure 8-15. Relationship of average annual tritium concentrations measured in moisture in air during 1960 with distance from F-Area and H-Area (onsite locations) (data derived from aperture cards). Link to tabulated figure data.

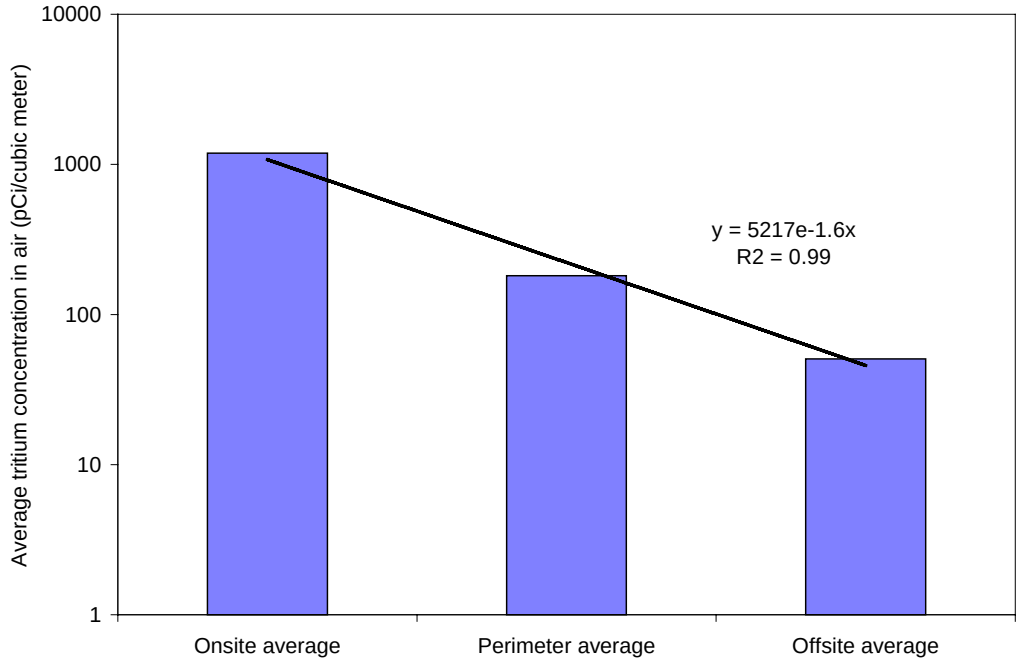

Figure 8-16. Relationship of average annual tritium concentrations measured in moisture in air during 1970 with distance from F-Area and H-Area (onsite locations) (data derived from aperture cards). Link to tabulated figure data.

Figures 8-17 and 8-18 show individual results for F-Area and H-Area monitors during 1960 and 1970. It is clear that the H-Area monitor consistently measures higher concentrations of tritium in the air than the F-Area monitor. This may reflect higher releases from the production facility in that area. Unfortunately, no weekly or biweekly release data are available for comparison. However, Murphy et al. (1991) shows a positive correlation between the amount of tritium released and concentrations of tritiated water in air at 20 km.

Cesium Measurements in Air

Measurements of gamma-emitting radionuclides began in July 1964 with monthly composites made of samples collected at F-Area, H-Area, perimeter, and 25-mi locations. Only one series of measurements was made in 1964. Beginning in 1965, monthly composites were analyzed routinely. Cesium-137 is the only gamma-emitting radionuclide that has been consistently measured above detection limits. The exception is during the early years, when other radionuclides (e.g., ^{54}Mn, ^{103}Ru, ^{106}Ru, and ^{144}Ce) from nuclear testing fallout were measured. It is expected that the concentrations of ^{137}Cs measured in the earliest years are associated with fallout from atmospheric weapons testing. Figure 8-19 shows measurements obtained from July 1964 through 1965. The impact of global fallout is obvious in the pattern of similar concentrations measured at each distance from the SRS.

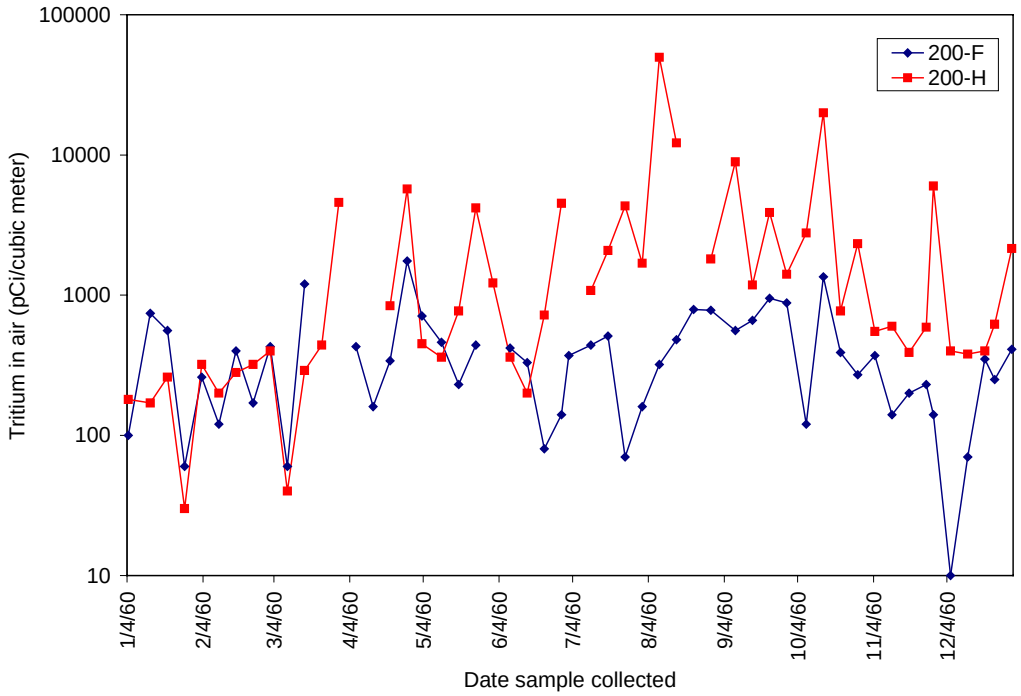

Figure 8-17. Weekly tritiated water measurements made in air at F-Area and H-Area during 1960 (data derived from aperture cards). Link to tabulated figure data.

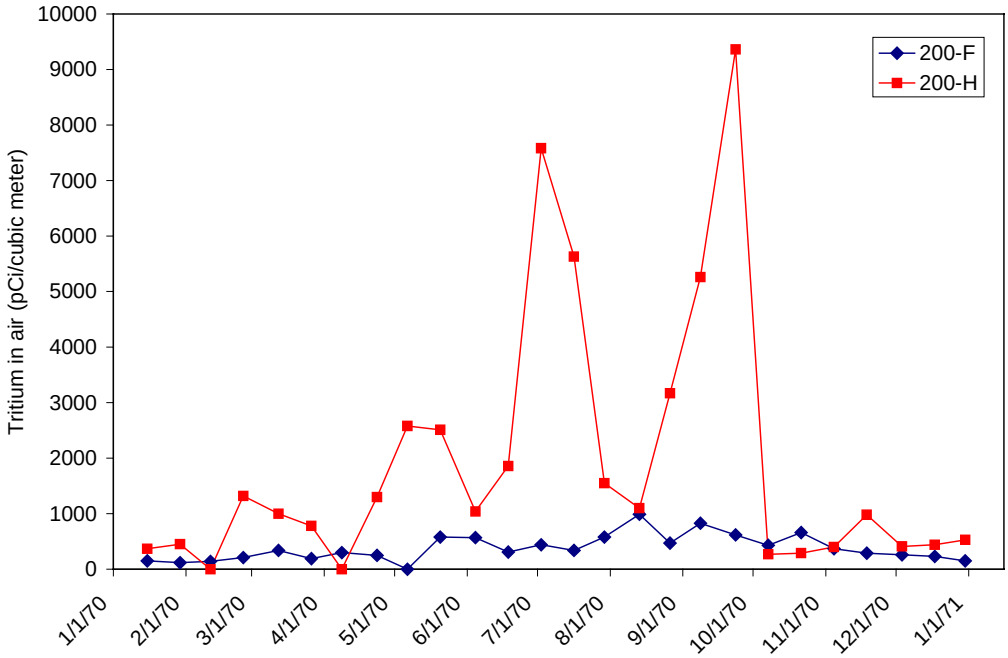

Figure 8-18. Biweekly tritiated water measurements made in air at F-Area and H-Area during 1970 (data derived from aperture cards). Link to tabulated figure data.

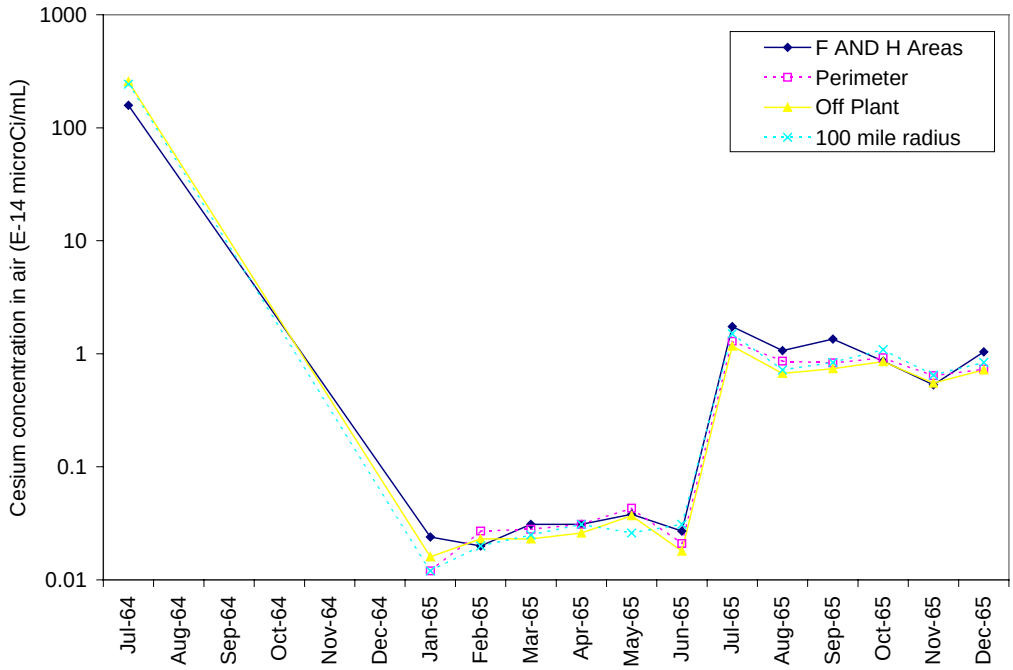

Figure 8-19. Monthly averages of ^{137}Cs measured at onsite, perimeter, 25-mi radius, and 100-mi radius locations from July 1964 through 1965 (data derived from aperture cards). Link to tabulated figure data.

During later years, Site releases appear to impact measurements made close to the SRS. Figure 8-20 shows higher concentrations of ^{137}Cs at F-Area and H-Area locations than at perimeter, 25-mi radius, and 100-mi radius locations during 1973.

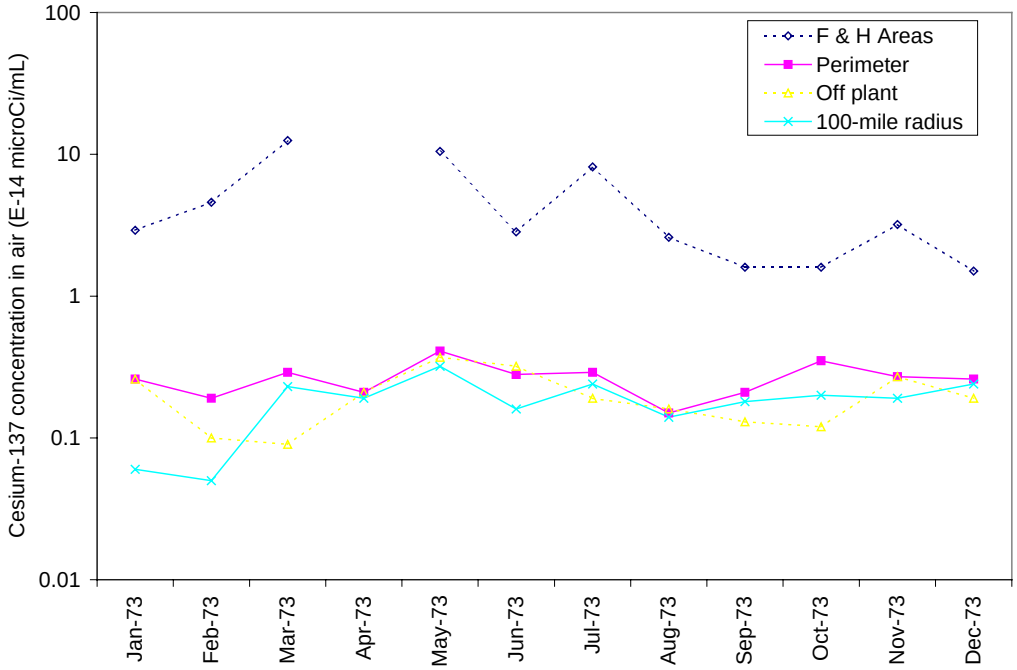

Figure 8-20. Monthly averages of ^{137}Cs measured at onsite, perimeter, 25-mi radius, and 100-mi radius locations during 1973 (data derived from aperture cards). Link to tabulated figure data.

Background Measurements of Radionuclides in Air

The SRS was not the only potential source of radionuclides in the environment (see Chapter 6 of this report). Fallout from atmospheric weapons testing was a source of both radionuclides in the environment and, therefore, in milk samples. Fallout concentrations of these radionuclides were greatest in the early 1960s. The concentrations in fallout decreased significantly from 1963 onward following the bomb-testing moratorium. The PHS (currently the U.S. Environmental Protection Agency [EPA]) initiated an air monitoring program for the southeast United States in 1955, and it provides the only independent source of air monitoring data. The monitoring location that is closest to the SRS is Columbia, South Carolina. Unfortunately, only particulate beta activity was measured (Figure 8-2). However, it is possible to compare these data with SRS particulate beta activity results for validation of these data during fallout years. At present, no other sources of independent monitoring data are available for comparison with SRS data.

RADIONUCLIDE CONCENTRATIONS IN RAINWATER

This section summarizes reported information regarding tritium concentrations in rainwater collected from locations on and in the vicinity of the SRS. Deposition of tritium in rainfall can be the result of two processes: rainout and washout. Rainout incorporates tritium in precipitation as it forms in clouds. Washout occurs when precipitation falling from the cloud passes through air containing tritium. Washout of tritium from air that has received tritium from facility stacks or evaporation from seepage basins is the process that is most important in the vicinity of SRS.

Rainwater data were found in aperture cards that were dated 1962 through 1965, 1968, and 1972 through 1973. The aperture cards represent the primary sources of original data. The data available from aperture cards were transcribed and recorded in Microsoft Excel workbooks entitled "Tritium-all.xls." Five sets of environmental monitoring reports spanning the years 1954 through 1991 were also examined. Only three sets of monitoring reports had information regarding tritium concentrations in rainwater: (1) *Health Physics Regional Monitoring, 1954–1963*, (2) *Environmental Monitoring at the Savannah River Plant, 1964–1991, Environmental Monitoring in the Vicinity of the Savannah River Plant, 1971–1983*, and (3) the *Savannah River Plant Environmental Report, 1984–1989*. The other two sets of monitoring reports (*Effect of the Savannah River Plant on Environmental Radioactivity, January 1962–June 1971* and *Environmental Monitoring at the Savannah River Plant, 1964–1976*) did not contain any information regarding tritium concentrations in rainwater.

Sample Collection, Preparation, and Analysis

Since 1962, rainwater has been collected at air monitoring stations located at onsite, plant perimeter, and 25-mi radius locations. During the period from 1961 through 1972, there were three onsite, five plant perimeter, and ten 25-mi radius locations. The number of onsite locations was increased to 6 in 1974 and to 10 in 1983. The number of plant perimeter locations was increased to 10 in 1973, to 12 in 1974, and to 13 in 1976. The number of 25-mi radius locations increased to 12 in 1974. This sampling regimen was consistent through 1985. In 1986, the number of routine plant perimeter and 25-mi radius locations was reduced to one station at Darkhorse. In 1978, rainwater collection began at four 100-mi radius locations, including Columbia and Greenville, South Carolina, and Macon and Savannah, Georgia.

Rainwater was collected in 2 × 2-ft fallout collection pans. The water was passed through an ion exchange column and collected in plastic jugs for weekly analyses through 1980. Analysis frequencies were not reported from 1981 through 1991, but sample numbers are indicative of bimonthly analysis for plant perimeter and 25-mi radius locations and quarterly analysis for 100-mi radius locations. Following collection, tritium concentrations were determined by liquid scintillation counting. Beginning in 1962, rainwater samples were also analyzed for iodine; beginning in 1965, they were analyzed for other radionuclides. The data for these radionuclides do not appear to be useful for dose reconstruction purposes and, therefore, are not discussed in this chapter.

Tritium Concentrations in Rainwater

Figures 8-21 and 8-22 show a decrease in tritium concentration in rainwater deposition as function of distance from the Site. This pattern is similar to that for tritium in air (see Figure 8-14) and indicates that the tritium measured in rainwater resulted from the washout of tritiated water vapor. Figure 8-23 shows a generally close correlation between the concentration of tritiated water vapor in air and tritium in rainwater. This helps to confirm that the source of tritium in rainwater is from the washout of tritium in water vapor.

There were no apparent directional trends for the plant perimeter, 25-mi radius, and 100-mi radius locations. This is consistent with the tritium concentrations measured in vegetation.

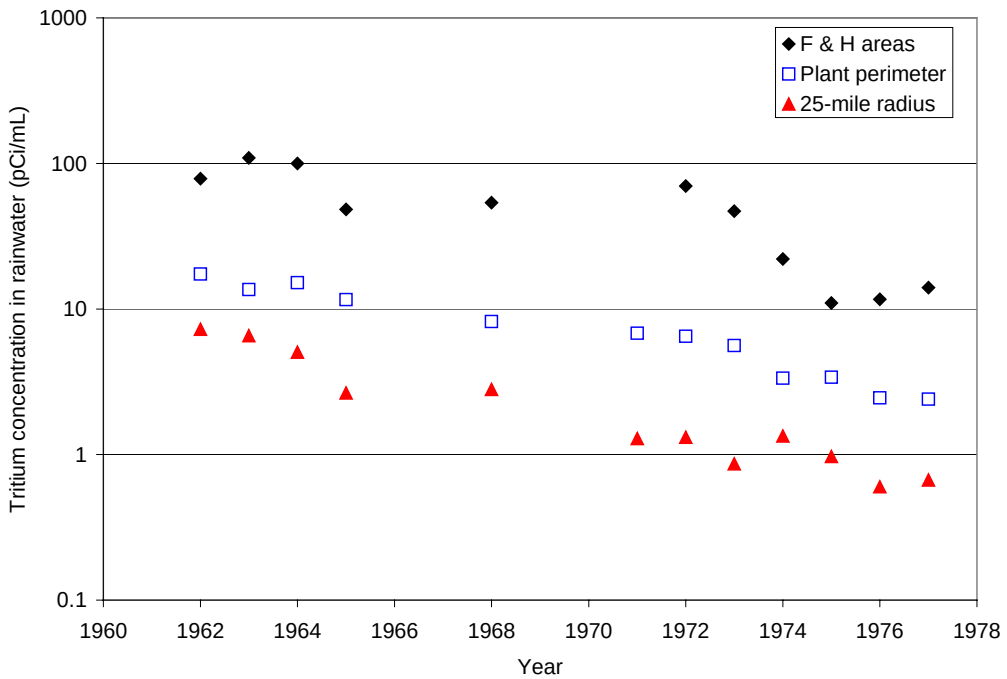

Figure 8-21. Annual averages of tritium measured in rainwater collected at onsite (F-Area and H-Area), site perimeter, and 25-mi radius locations from 1962 through 1977. Data were not available for 1966, 1967, 1969, and 1970 (data derived from aperture cards). Link to tabulated figure data.

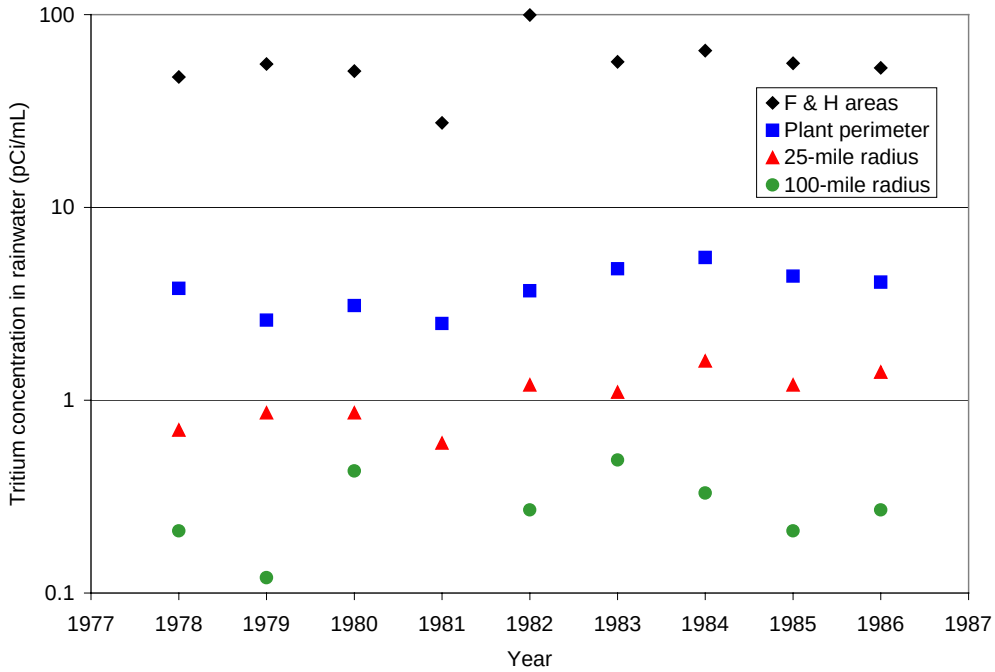

Figure 8-22. Annual averages of tritium measured in rainwater collected at onsite (F-Area and H-Area), Site perimeter, 25-mi radius and 100-mi radius locations from 1978 through 1986 (data derived from aperture cards). Link to tabulated figure data.

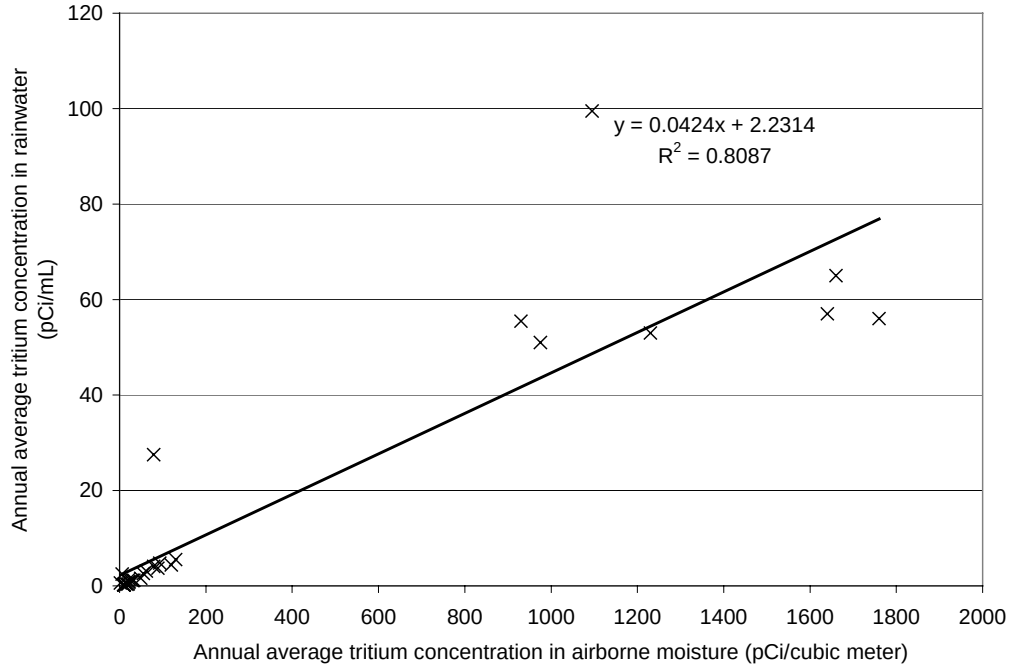

Figure 8-23. The annual average concentration of tritium in rainwater compared to the concentration in water in air at each monitoring location (1979–1986) (data derived from aperture cards). Link to tabulated figure data.

In general, reported tritium concentrations in rainwater are well correlated with reported tritium concentrations in vegetation. Table 8-7 shows average reported concentrations for rainwater and vegetation from 1984 through 1986. Vegetation and rainwater concentrations are within a factor of 2 for all locations.

Table 8-7. Average Tritium Concentrations in Vegetation and Rainwater from 1984 through 1986[a]

Location	Rainwater (pCi mL^{-1})	Vegetation (pCi mL^{-1})
F-Area and H-Area	58	44
Plant perimeter	4.7	9.1
25-mi radius	1.4	2.3
100-mi radius	0.34	0.52

[a] Source: Annual monitoring reports.

Background Measurements of Radionuclides in Rainwater

The EPA has measured tritium in rainwater collected at various locations throughout the United States since 1974. Before 1974, only gross beta concentrations were measured. Since 1975, this information has been reported in a series of reports published by EPA entitled *Environmental Radiation Data*. Concentrations were reported for Columbia and Barnwell, South Carolina, for a number of years. These data were useful for comparing and verifying the concentrations reported by the SRS for the same locations. Table 8-8 shows reported concentrations by the EPA and SRS for Columbia, South Carolina, from 1982 through 1990 and for Barnwell, South Carolina, and the Barnwell gate location from 1979 through 1986.

Table 8-8. Comparison of SRS and EPA-reported Tritium Concentrations[a]

	Columbia		Barnwell[b]	
Year	SRS	EPA	SRS	EPA
1979			2.6	0.65
1980			3.9	2.23
1981			3.1	1.16
1982	0.48	0.61	4	1.13
1983	1.4	0.40	5.5	1.31
1984	0.68	0.60	6.6	1.74
1985	0.51	0.56	4.1	1.14
1986	0.70	0.29	3.4	1.63
1987	0.19	0.29		
1988	0.12	0.27		
1989	0.06	0.29		
1990	0.11	0.23		

[a] Data from annual reports and from EPA Environmental Radiation Data.
[b] SRS reported concentrations for the Barnwell gate and EPA reported concentrations for Barnwell, South Carolina.

The concentrations reported by the EPA and SRS correlate well for the samples collected at Columbia and are near the limits of detection (0.2 pCi/g for the EPA). The SRS reported concentrations at the Barnwell gate are higher than those reported by the EPA for Barnwell, South Carolina. This would be expected since Barnwell, South Carolina, is approximately 10 km east of the Barnwell gate at the plant perimeter, and concentrations decrease rapidly with distance from the Site.

ELECTRONICALLY COMPILED AIR AND RAINWATER DATA

The various data summarized in this chapter are electronically compiled in several Microsoft Excel® workbooks. One workbook (Ch8-Figure_data.xls) contains the figures depicted in this chapter as well as the tabulated data that were used to produce the figures. In this workbook, there is a separate chart for each figure and each worksheet contains the tabulated data for one or a group of figures. Iodine.xls, Beta.xls, and Tritium.xls contain worksheets that summarize data tabulated from aperture cards that were used to generate the figures found in Ch8-Figure_data.xls, and they are referenced within this workbook.

The remaining data consist of workbooks containing all the aperture card data we compiled that are related to monitoring of air and rainwater, described in Table 8-1. These data can be accessed by clicking on the following hyperlinks: Iodine-all.xls, Tritium-all.xls, Gamma-all.xls, Alpha-Beta-all.xls, and Particle_data-all.xls. The file name reflects the contaminant(s) measured, and a separate spreadsheet exists for each year. For example, the spreadsheet entitled "1965 – air" in the workbook entitled "Tritium-all.xls" contains tritium measurements made in during 1965. Most of the aperture card data were not used to generate figures in this chapter but are included here to provide the reader with a complete electronic set of original data. It should also be noted that much of the data on the aperture cards is unreadable or barely legible. *RAC* recopied many of these sheets very carefully on a separate trip to the SRS, but many of the cards are not capable of adequate enhancement. The data entered on the worksheets represent a best attempt at recreating the handwritten ledgers. If specific data could not be read, that was noted on the spreadsheet.

USEFULNESS OF AIR AND RAINWATER MONITORING DATA

Factors that determine how useful the air and rainwater monitoring data may be for reconstructing historical releases from the SRS include

- The availability of original monitoring datasets for detailed analyses
- Sufficient datasets to allow spatial and/or temporal trends in the data to be identified
- The ability to differentiate between Site releases of contaminants and other sources of the same contaminants in the environment.

The following sections discuss these factors in more detail.

Availability of Original Monitoring Measurements

Air Monitoring Data

One of the problems with the air monitoring data that are reported in the quarterly, semiannual, and annual reports is that the results are often averaged over large spatial areas or

over more than one sample period for a given location. For example, for the period from 1955 through 1956, the ^{131}I results are reported as a semiannual average and a maximum for each 6-month period. Any releases that occur during a shorter time period will be masked in the 6-month average. This averaging limits the data evaluation that can be performed to confirm elevated releases. In addition to limiting the ability to perform source term verification, it restricts the use of the data in detecting any unplanned releases. Because these averages provide poor resolution, it is unlikely that the semiannual data can be used for more than confirming long-term trends and averages estimated using original data that are available from 1959 on.

The aperture cards provide individual monitoring data from 1959 through 1973. Only summary data are available from annual reports for 1974 and 1975. For the time period after 1975, computer printouts of individual monitoring data exist. However, except for onsite locations, concentrations of ^{131}I were below detection limits after 1963. It appears that only the onsite ^{131}I data will be useful for the dose reconstruction after this year. Gross beta and tritium measurements are consistently above detection limits for all years. Although the number of air sampling stations fluctuated over the history of the SRS operations, a comprehensive dataset of gross beta, ^{131}I, tritium, and gamma-emitting radionuclide measurements in air has been compiled in the aperture cards and the computer-generated monthly reports.

Rainwater Monitoring Data

Results in the semiannual and annual monitoring reports are reported as averages and maxima for the reporting period (i.e., semiannual or annual) at each monitoring location. Releases that occur during a shorter time period cannot be discerned. This limits the data evaluation that can be performed to confirm elevated releases. In addition to limiting the ability to perform source term verification, it restricts the use of the data in detecting any unplanned releases. Because these averages provide poor resolution, it is unlikely that the data provided in the environmental monitoring reports can be used for more than identifying long-term trends and averages.

The aperture cards located during Phase I of the dose reconstruction provide individual monitoring data from 1962 through 1965, 1968, and 1972 through 1973. These data will perhaps be useful in determining more detailed spatial and temporal trends. Only summary data are available from annual reports for 1967, 1969 through 1971, and 1974 through 1986.

Availability of Spatial and Temporal Datasets

Air Monitoring Data

Figures 8-2 through 8-20 demonstrate that the datasets can provide spatial and temporal information for use in the dose reconstruction. It remains to be determined in a subsequent phase of the dose reconstruction if there are enough data to be useful for source term verification or development or for validation of dispersion models. In some cases, the frequency of measurements may not provide the resolution necessary for these uses. For example, ^{137}Cs data are measurements of monthly composite samples. Most of the data sets for gross beta activity, ^{131}I, and tritium represent weekly or biweekly measurements at individual locations, and as such, they may be particularly useful to a dose reconstruction.

Factors that determine the value of these data include the availability of original data for detailed analyses, sufficient datasets to allow trends in the data to be identified, and the ability to differentiate between Site releases and other sources of the same contaminants in the environment. Weekly and biweekly datasets are available for the key radionuclides (i.e., tritium and radioiodine) for most years of concern (1959–1990), providing good temporal resolution. Spatial resolution is more limited, however, as there were few samplers located onsite during the early years, when releases were higher. Results generally indicate a trend of decreasing radionuclide concentrations from the Site to offsite locations, except during periods of high global fallout. This implies that the air sampling data can be used to verify SRS releases.

Rainwater Monitoring Data

Figures 8-21 through 8-23 demonstrate that the data sets can provide some spatial and temporal information for use in the dose reconstruction. Tritium in rainwater data indicate a trend of decreasing radionuclide concentrations from the Site to offsite locations, indicating SRS is the source of tritium. Unfortunately, data are not available for all years, and there are few onsite samplers during the early years of monitoring. It remains to be determined in a subsequent phase of the dose reconstruction if there are enough data to be useful for source term verification or development or for validation of dispersion models.

Summary: Data Usefulness

The historic air and rainwater monitoring data that are available demonstrate spatial and temporal patterns that reflect what is known about SRS releases. The key contaminants detected in air and rainwater during the period of interest (1951–1990) are radioiodine and tritium. As expected, concentrations decrease with distance from the Site and in many cases track known periods of elevated releases. The datasets may, thus, be useful for verifying source term estimates. Further analysis in a subsequent phase of the dose reconstruction can help to determine if there are enough data to be useful for source term or validation of dispersion models. The data may be limited by insufficient temporal and spatial resolution because weekly or biweekly datasets are not available for all years, particularly during the earliest years of Site operations. The highest reported releases occurred during the operational period before 1959. The measurements also represent a limited number of sampling locations, particularly onsite during the early years of monitoring.

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ADDENDUM 8A

Table 8-2. Onsite air monitoring program summary (1951–1990)

Table 8-3. 25-mile air monitoring program summary (1951–1990)

Table 8-4. 100-mile air monitoring program summary (1961–1990)

Note: The information contained in the following tables was obtained primarily from SRS monitoring reports. See chapter 7, table 7-1 for a complete description of the various monitoring report series.

Table 8-2. Onsite Air Monitoring Program Summary (1951–1990) (per Chapter 7, Table 7-1)

			Analytical parameters						
From date	To date	Collection frequency	α,β[a]	^{131}I	^{3}H	^{90}Sr	γ[b]	Gamma-emitting, other nuclides	Specific sampling locations
9/1/51	9/30/52	Weekly	x					Radon and thoron	CMX and Dunbarton fire tower
1/1/54	6/30/55	Weekly	x					Radon and thoron	Talatha gatehouse, Williston gatehouse, Dunbarton fire tower, 200-F, 200-H, 400-D, and 300-700
7/1/55	6/30/57	Weekly	x	x					Talatha gatehouse, Williston gatehouse, Dunbarton fire tower, 200-F, 200-H, 400-D, and 300-700
1/1/56	12/31/56	Weekly			x				Four locations along the 200-F 1-mile perimeter
1/1/57	6/30/57	Weekly			x				Four locations on the F-Area 1-mile radius perimeter, and at F-Area and H-Area air monitor buildings
7/1/57	12/31/57	Weekly	x	x	x				Talatha gatehouse, Williston gatehouse, Dunbarton fire tower, 200-F,200-H, 400-D, and 3/700
1/1/60	6/30/60	Weekly	x	x	x		x	Gamma analysis of 200-F and 400-D samples	Talatha gatehouse, Williston gatehouse, Dunbarton Fire Tower, 400-D, 200-F, 200-H, 300/700, Green Pond Church, Military Recreation Site, and Jackson
7/1/60	6/30/62	Weekly	x	x	x		x	Gamma spectroscopy performed when counting time available.	Talatha gatehouse, Williston gatehouse, Dunbarton Fire Tower, 400-D, 200-F, 200-H, 300/700, Green Pond Church, Military Recreation Site, and Jackson
7/1/62	12/31/62	Weekly	x	x	x		x	Gamma spectroscopy performed when counting time available.	Talatha gatehouse, Williston gatehouse, Dunbarton Fire Tower, 400-D, 200-F, 200-H, 300/700, and Jackson
1/1/65	12/31/66	Weekly	x	x	x		x	Cs-137, Ce-144, Ru-106, Zr/Nb-95, Mn-54	Dunbarton Fire Tower, 400-D, 200-F, 200-H, 3/700, and Jackson
1/1/67	12/31/67	Biweekly	x	x	x	x	x	Cs-137, Ce-144, Ru-106, Zr/Nb-95, Mn-54	Dunbarton Fire Tower, 400-D, 200-F, 200-H, 3/700, and Jackson
1/1/68	12/31/69	Biweekly	x	x	x	x	x	Cs137; Ce141,144; Ru-103,106; Zr-95,Nb-95; Be-7	Dunbarton Fire Tower, 400-D, 200-F, 200-H, 3/700, and Jackson
1/1/70	12/31/70	Biweekly	x	x	x	x	x	Cs-137; Ce-141,144; Ru-103,106; Zr-95,Nb-95; Be-7.	Talatha gatehouse, Williston gatehouse, Dunbarton Fire Tower, 400-D, 200-F, 200-H, 3/700, and Jackson
1/1/71	12/31/71	Biweekly	x	x	x				Talatha gatehouse, Williston gatehouse, Dunbarton Fire Tower, 400-D, 200-F, 200-H, 3/700, and Jackson
1/1/71	12/31/71	Monthly				x	x	Cs-137; Ce-141,144; Ru-103,106; Zr-95,Nb-95; Be-7	Talatha gatehouse, Williston gatehouse, Dunbarton Fire Tower, 400-D, 200-F, 200-H, 3/700, and Jackson

Table 8-2. (continued)

			Analytical Parameters						
From date	To date	Collection frequency	α,β[a]	^{131}I	^{3}H	^{90}Sr	γ[b]	Gamma-emitting, other nuclides	Specific sampling locations
1/1/72	12/31/72	Biweekly	x	x	x				Talatha gatehouse, Williston gatehouse, Dunbarton Fire Tower, 400-D, 200-F, 200-H, 700-A, 735-A, Jackson, and Green Pond
1/1/72	12/31/72	Monthly				x	x	Cs-137; Ce-141,144; Ru-103,106; Zr-95,Nb-95; Be-7	Talatha gatehouse, Williston gatehouse, Dunbarton Fire Tower, 400-D, 200-F, 200-H, 700-A, 735-A, Jackson, and Green Pond
1/1/73	12/31/73	Weekly	x		x	x	x	Cs-137; Ce-141,144; Ru-103,106; Zr-95,Nb-95; Be-7.	Eight stations near the plant perimeter (one of which appears to actually be offsite)
1/1/74	12/31/74	Monthly					x	Pu-238; Pu-239.	Twelve stations near the plant perimeter
1/1/74	12/31/74	Weekly	x		x	x	x	Cs-137; Ce-141,144; Ru-103,106; Zr-95,Nb-95; Be-7.	Twelve stations near the plant perimeter
1/1/75	12/31/75	Biweekly			x				A-Area, F-Area, H-Area, Dunbarton (DFT), Williston Gate, Par Pond, Allendale Gate, A-14, Barnwell Gate, D- Area, Dark Horse, Talatha Gate, Green Pond, Highway 21/167, Jackson, Pattersons Mill, East Talatha, and Windsor Road
1/1/75	12/31/75	Monthly				x	x	Be-7; Zr-95,Nb-95; Ru-103,106; Cs-137; Ce-141,144; Pu-238; Pu-239.	A-Area, F- Area, H-Area, Dunbarton (DFT), Williston Gate, Par Pond, Allendale Gate, A-14, Barnwell Gate, D- Area, Dark Horse, Talatha Gate, Green Pond, Highway 21/167, Jackson, Pattersons Mill, East Talatha, and Windsor Road
1/1/75	12/31/75	Weekly	x						Eighteen stations, at A-Area, F-Area, H- Area, Dunbarton (DFT), Williston Gate, Par Pond, Allendale Gate, A-14, Barnwell Gate, D-Area, Dark Horse, Talatha Gate, Green Pond, Highway 21/167, Jackson, Pattersons Mill, East Talatha, and Windsor Road
1/1/76	12/31/76	Biweekly			x				A-Area, F-Area, H-Area, Dunbarton, Par Pond, Williston Gate, Allendale Gate, A-14, Barnwell Gate, D- Area, Dark Horse, East Talatha, Green Pond, Highway 21/167, Jackson, Pattersons Mill, Talatha Gate, West Jackson, and Windsor Road
1/1/76	12/31/76	Monthly				x	x	Be-7; Zr-95,Nb-95; Ru-103,106; Cs-137; Ce-141,144; Pu-238; Pu-239.	A-Area, F-Area, H-Area, Dunbarton, Par Pond, Williston Gate, Allendale Gate, A-14, Barnwell Gate, D- Area, Dark Horse, East Talatha, Green Pond, Highway 21/167, Jackson, Pattersons Mill, Talatha Gate, West Jackson, and Windsor Road

Table 8-3. 25-mile Air Monitoring Program Summary (1951–1990) (per Chapter 7, Table 7-1)

			Analytical parameters						
From date	To date	Collection frequency	α,β[a]	^{131}I	^{3}H	^{90}Sr	γ[b]	Gamma-emitting, other nuclides	Specific sampling locations
9/1/51	6/30/54	Weekly	x				x	Radon and thoron	Aiken Airport, Allendale, and Waynesboro
7/1/54	6/30/55	Weekly	x						Aiken Airport,Allendale, and Waynesboro
7/1/55	12/31/56	Weekly	x	x					Aiken Airport,Allendale, and Waynesboro
1/1/57	12/31/57	Weekly	x	x	x				Aiken Airport, Allendale, and Waynesboro (all). Langley, Bush Field, Williston, Barnwell, and Sardis (radiodine only)
1/1/58	12/31/60	Weekly	x	x	x				Aiken Airport, Allendale, Waynesboro, Langley, Bush Field, Williston, Barnwell, and Sardis
1/1/61	12/31/61	Weekly	x	x	x				Aiken Airport, Allendale, Waynesboro, Langley, Williston, Barnwell, Sardis, Bush Field, Aiken State Park, and Highway 301
1/1/62	12/31/62	Weekly	x	x	x		x	Gamma spectroscopy performed when counting time available	Aiken Airport, Allendale, Waynesboro, Langley, Williston, Barnwell, Sardis, Bush Field, Aiken State Park, and Highway 301
1/1/63	12/31/64	Weekly	x	x	x		x	Cs-137, Ce-144, Ru-103, Ru-106, Zr/Nb-95, Mn-54, Ba/La-140	Aiken Airport, Allendale, Waynesboro, Langley, Williston, Barnwell, Sardis, Bush Field, Aiken State Park, and Highway 301
1/1/65	12/31/65	Weekly	x	x	x		x	Cs-137, Ce-144, Ru-106, Zr/Nb-95, Mn-54	Aiken Airport, Allendale, Waynesboro, Langley, Williston, Barnwell, Sardis, Bush Field, Aiken State Park, and Highway 301
1/1/66	12/31/66	Biweekly	x	x	x		x	Cs-137, Ce-144, Ru-106, Zr/Nb-95, Mn-54	Aiken Airport, Allendale, Waynesboro, Langley, Williston, Barnwell, Sardis, Bush Field, Aiken State Park, and Highway 301
1/1/67	12/31/71	Biweekly	x	x	x	x	x	Cs-137, Ce-144, Ru-106,-103, Zr/Nb-95	Aiken Airport, Allendale, Waynesboro, Langley, Williston, Barnwell, Sardis, Bush Field, Aiken State Park, and Highway 301
1/1/72	12/31/72	Biweekly	x		x	x	x	Cs-137, Ce-144, Ru-106,-103, Zr/Nb-95	Aiken Airport, Allendale, Waynesboro, Langley, Williston, Barnwell, Sardis, Bush Field, Aiken State Park, and Highway 301

Table 8-3. (continued)

			Analytical parameters						
From date	To date	Collection frequency	α,β[a]	^{131}I	^{3}H	^{90}Sr	γ[b]	Gamma-emitting, other nuclides	Specific sampling locations
1/1/73	12/31/74	Weekly	x		x	x	x	Cs-137; Ce-141,144; Ru-103,106; Zr-95,Nb-95; Be-7.	Aiken Airport, Allendale, Waynesboro, Langley, Williston, Barnwell, Sardis, Bush Field, Aiken State Park, and Highway 301
1/1/74	12/31/74	Monthly					x	Pu-238; Pu-239.	Augusta, Langley, Aiken Airport, Aiken State Park, Springfield, Lees, Olar, Allendale, Perkins, Waynesboro, South Richmond, and Highway 301
1/1/75	12/31/80	Biweekly			x				Augusta, Langley, Aiken Airport, Aiken State Park, Springfield, Lees, Olar, Allendale, Perkins, Waynesboro, South Richmond, and Highway 301
1/1/75	12/31/83	Monthly				x	x	Be-7;Zr-95,Nb-95; Ru-103,106; Cs-137; Ce-141,144; Pu-238; Pu-239.	Augusta, Langley, Aiken Airport, Aiken State Park, Springfield, Lees, Olar, Allendale, Perkins, Waynesboro, South Richmond, and Highway 301
1/1/75	12/31/81	Weekly	x						Augusta, Langley, Aiken Airport, Aiken State Park, Springfield, Lees, Olar, Allendale, Perkins, Waynesboro, South Richmond, and Highway 301
1/1/79	12/31/81	Monthly		x					25-mi perimeter: Augusta, Langley, Aiken Airport, Aiken State Park, Springfield, Lees, Olar, Allendale, Perkins, Waynesboro, South Richmond, and Highway 301
1/1/80	12/31/83	Quarterly			x				Four distant locations, about 100 mi from the plant, in Columbia and Greenville, South Carolina, and Macon and Savannah, Georgia
1/1/81	12/31/83	Biweekly	x						25-mi perimeter: Augusta, Langley, Aiken Airport, Aiken State Park, Springfield, Lees, Olar, Allendale, Perkins, Waynesboro, South Richmond, and Highway 301
1/1/84	12/31/90								

[a] Gross beta activity.
[b] Gamma-emitting and other radionuclides.

Table 8-4. 100-mile Air Monitoring Program Summary (1961–1990) (per Chapter 7, Table 7-1)

			Analytical parameters						
From date	To date	Collection frequency	α,β[a]	^{131}I	^{3}H	^{90}Sr	γ[b]	Gamma-emitting, other nuclides	Specific sampling locations
1/1/61	12/30/61	Weekly	x						Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/62	12/31/62	Weekly	x	x			x	Gamma spectroscopy performed when counting time available.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/63	12/31/65	Weekly	x	x			x	Cs-137, Ce-144, Ru-103, Ru-106, Zr/Nb-95, Mn-54, Ba/La-140	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/66	12/31/66	Biweekly	x	x			x	Cs-137, Ce-144, Ru-106, Zr/Nb-95, Mn-54	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/67	12/31/67	Biweekly	x	x		x	x	Cs-137, Ce-144, Ru-106,-103, Zr/Nb-95	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/70	12/31/70	Biweekly	x		x	x	x	Cs137; Ce141,144; Ru-103,106; Zr-95,Nb-95; Be-7.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/71	12/31/71	Biweekly	x	x	x	x	x	Cs-137; Ce-141,144; Ru-103,106; Zr-95,Nb-95; Be-7.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/72	12/31/72	Biweekly	x		x	x	x	Cs-137; Ce-141,144; Ru-103,106; Zr-95,Nb-95; Be-7.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/73	12/31/74	Weekly	x		x	x	x	Cs-137; Ce-141,144; Ru-103,106; Zr-95,Nb-95; Be-7.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/74	12/31/74	Monthly					x	Pu-238; Pu-239.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/75	12/31/79	Biweekly			x				Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/75	12/31/78	Monthly				x	x	Be-7;Zr-95,Nb-95; Ru-103,106; Cs-137; Ce-141,144; Pu-238; Pu-239.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/75	12/31/83	Weekly	x						Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/79	12/31/79	Biweekly			x				Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/79	12/31/79	Monthly		x		x	x	Be-7;Zr-95,Nb-95; Ru-106; Cs-137; Ce-144; Pu-238; Pu-239.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA

Table 8-4. (continued)

			Analytical Parameters						
From date	To date	Collection frequency	α,β[a]	^{131}I	^{3}H	^{90}Sr	γ[b]	Gamma-emitting, other nuclides	Specific sampling locations
1/1/80	12/31/82	Monthly		x		x	x	Be-7;Zr-95,Nb-95; Ru-106; Cs-137; Ce-144.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/81	12/31/83	Quarterly			x				Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA
1/1/83	12/31/90	Monthly					x	Be-7; Sr-89,90; Zr-95,Nb-95; Ru-106; Cs-137; Ce-144.	Columbia, SC; Greenville, SC; Macon, GA; and Savannah, GA

[a] Gross alpha activity and gross beta activity.

[b] Gamma-emitting and other radionuclides.

CHAPTER 9

RADIONUCLIDES IN VEGETATION AND AGRICULTURAL PRODUCTS

ABSTRACT

This chapter discusses radionuclide concentrations measured in vegetation and agricultural products collected on or in the vicinity of the Savannah River Site (SRS). We evaluated these data with regard to their potential usefulness for dose reconstruction. We also attempted to validate the reported concentrations by making comparisons among as many data sources as possible. These data included original, hand-written compilations for many years. We have found these original data to consistently correspond to the information reported in monthly, semiannual, and annual summary reports.

We compiled and examined vegetation data and other environmental data to determine their usefulness for source term verification, model validation, and direct exposure assessment. In general, the vegetation data are most valuable for source term verification and model validation. However, the potential usefulness of the data is limited in many cases by spatial or temporal resolution. Appendix K further discusses potential uses for environmental monitoring data.

INTRODUCTION

Since 1953, vegetation has been sampled at various locations on or in the vicinity of the Savannah River Site (SRS). Bermuda grass has been the preferred species for sampling because of its use as food for dairy cattle and its year-around availability. In May 1961, the SRS began collecting local agricultural products, including collards, plums, peaches, oats, wheat, soybeans, rye, corn, and meat (chicken and beef), at several vicinity locations. Radionuclide concentrations in vegetation and agricultural products are important to evaluate because they serve as sensitive indicators of aerial deposition resulting from plant operations and atmospheric weapons testing fallout. Examining concentrations measured in vegetation and agricultural products at various distances from the SRS can help to determine the source of atmospherically deposited radionuclides and may also be useful for source term and model validation.

This chapter summarizes reported information regarding radionuclide concentrations in vegetation and agricultural products on or in the vicinity of the SRS. We examined several sets of routine semiannual and annual environmental monitoring reports, prepared by the SRS contractor and spanning the years 1953 through 1992. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series.

We reviewed several additional documents for information pertaining to vegetation monitoring at the SRS. These include monthly monitoring reports from 1962 through 1965, weekly monitoring reports from 1959 through 1962, and aperture cards (handwritten compilations of original environmental monitoring results that were photographed in a format similar to microfiche) from 1957 and 1959 through 1973.

Annual environmental monitoring reports provide only average annual results for all collection locations for each agricultural product and radionuclide. However, we reviewed aperture cards for 1966–1973 and 1982–1983 and these documents do provide concentrations

measured in agricultural products at individual collection locations for each collection period. In general, although sometimes illegible, the information reported in the aperture cards is consistent with information provided in the monitoring reports.

Data that are available for vegetation and agricultural products include gross alpha activity; nonvolatile beta activity; and concentrations of ^{7}Be, ^{144}Ce, ^{137}Cs, ^{40}K, ^{54}Mn, ^{106}Ru, ^{95}Zr/Nb, ^{140}Ba/La, ^{131}I, and ^{3}H. Appendix A details analytical and counting procedures for vegetation and agricultural products. We focussed the majority of our efforts on the evaluating vegetation data, rather than food or agricultural product data, because vegetation has been collected since the early days of plant operations. Atmospheric releases from SRS were generally the highest and the potential for offsite deposition was the greatest during these early years. Agricultural products, on the other hand, were not collected until May 1961. Most of the significant atmospheric releases at SRS occurred before this time. Additionally, there were fewer agricultural product collection locations, and concentrations were reported in the annual environmental monitoring reports as average annual concentrations for all locations. This precludes any spatial or temporal analysis of these data and limits their usefulness. The vegetation data are somewhat more adequate for spatial or temporal analyses and span a greater number of years. It can be assumed, however, that spatial deposition patterns inferred from vegetation monitoring would be similar to those based on agricultural product monitoring. We provide the summary results for the F-Area and H-Area, plant perimeter, 25-mi radius, and 100-mi radius locations in this chapter.

RADIONUCLIDE CONCENTRATIONS IN VEGETATION

Since 1953, vegetation has been sampled at various locations on or in the vicinity of the SRS. Before 1971, the environmental monitoring reports do not specify the type of vegetation that was sampled, and it appears likely that several different species were included in the sampling protocol. The monitoring reports give results for vegetation in general, but it is assumed that the majority of results since 1971 apply to Bermuda grass (Du Pont 1972a). Bermuda grass was indicated as the preferred species for sampling because of its importance as a pasture grass for dairy cattle and its year-around availability. Vegetation samples have been routinely collected from F-Area and H-Area, plant perimeter, 25-mi radius, and 100-mi radius locations.

Beginning in July 1955, radionuclide concentrations were reported for vegetation samples collected at 24 F-Area and at 24 H-Area locations. The number of locations was reduced to 10 at each area from 1957 through June 1964. Between July 1964 and 1971, vegetation was sampled at a total of four locations; two locations were approximately 1 mi distant from each of the areas. Two locations were positioned approximately 1 mi northwest and 1 mi west of the H-Area, and the other two were positioned approximately 1 mi southeast of the F-Area. In 1972, one location southeast of the F-Area was repositioned to approximately 1 mi west of the F-Area, and the location west of the H-Area was repositioned to approximately 1 mi southeast of the H-Area. From 1972 through 1991, the same four F-Area and H-Area locations were sampled. Samples were collected and composited for weekly analysis through 1972. Since 1972, the sampling frequency has been monthly. The monitoring reports provided semiannual averages through 1972 and annual averages from 1973 through 1991. Aperture cards provided weekly and monthly composite values for 1957 and 1959 through 1973.

There were 15 inner perimeter locations from 1953 through June 1955 and 14 from July 1955 through 1958, after which inner perimeter samples were no longer collected. There were 21

plant perimeter locations from 1953 through June 1954, 17 from July 1954 through June 1955, 14 from July 1955 through June 1964, 7 from July 1964 through 1981, 13 from 1982 through 1986, and 14 from 1987 through 1991. There were ten 25-mi radius locations from 1953 through June 1954, 11 from July 1954 through June 1955, 14 from July 1955 through June 1964, 7 from July 1964 through 1981, 12 from 1982 through 1985, 6 in 1986; and 4 from 1987 through 1991.

Samples from the plant perimeter and 25-mi radius locations were collected weekly and composited for analysis from 1954 through June 1965. Samples were collected monthly and composited for analysis from July 1965 through 1983. Samples were collected quarterly and composited for analysis from 1986 through 1991. The sampling frequency is unclear in 1984 and 1985. However, based on the reported number of samples, the sampling frequency has been quarterly since 1983. The monitoring reports provided semiannual averages through 1972 and annual averages from 1973 through 1976 and from 1983 through 1991. Aperture cards provided monthly and weekly composite values from 1957 and 1959 through 1973.

The monitoring reports began providing annual averages at four 100-mi radius locations in 1974, including Macon and Savannah in Georgia and Columbia and Greenville in South Carolina. The collection frequency was monthly through 1978 and quarterly from 1979 through 1991. Concentrations were reported rather sporadically in the aperture cards beginning in 1962. The reporting frequency varied from weekly to semiannually.

Figure 9-1 shows locations that were sampled at F-Area and H-Area, the plant perimeter, and the 25-mi radius distances from 1982 through 1985. The number of locations has varied somewhat historically, as discussed above, but Figure 9-1 provides a general representation of the various locations discussed in this chapter. The general reduction in sample numbers over time was an attempt to create a cost effective program without compromising its utility, and it permitted more duplicate analyses of certain “critical” samples (Marter and Johnson 1966). Table 9-1 provides a general summary of the routine sampling and analyses that have been completed for vegetation since the inception of the monitoring program.

Tritium

Because tritium concentrations were reported for individual vegetation samples collected at the plant perimeter, 25-mi radius, and 100-mi radius locations, spatial trend analysis is possible. Table 9-2 compiles mean reported concentrations since 1976 at all locations, along with the approximate direction from the center of the SRS. There were no apparent trends for any direction although concentrations decreased as a function of distance in all directions. This was expected given the lack of a dominant prevailing wind direction at the SRS. However, only annual average concentrations were reported for each location, which precluded examining temporal trends throughout the year and, consequently, during episodic releases.

According to the reports reviewed, tritium has been the only radionuclide of SRS origin to be detected offsite or beyond the plant perimeter in vegetation. Figure 9-2 shows the annual average tritium concentrations for the years 1974 through 1991. Concentrations measured at the F-Area and H-Area are clearly greater than those measured at the plant perimeter, 25-mi, and 100-mi locations. The plant perimeter concentrations are consistently higher than 25-mi radius concentrations, which are slightly higher than 100-mi radius concentrations. Concentrations for all locations decreased significantly from 1974 through 1977 and have fluctuated since that time.

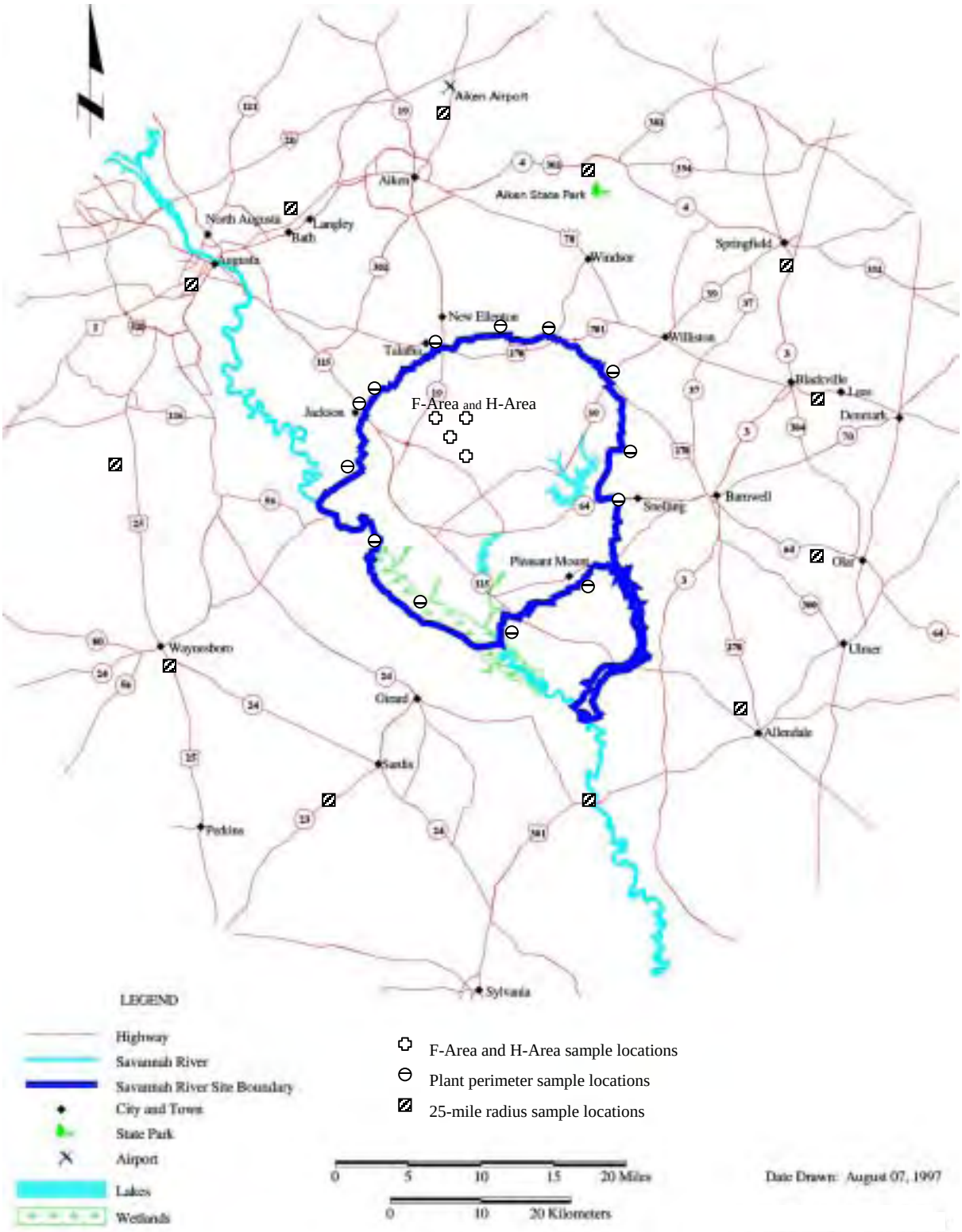

Figure 9-1. Map showing vegetation sampling locations during 1985.

Table 9-1. Summary of Routine SRS Radiological Monitoring for Vegetation[a]

	Number of routine locations					Radiological analyses				
Year	F- and H-Area	Inner perimeter	Plant perimeter	25-mile radius	100-mile radius	Gross alpha	Gross nonvolatile beta	Radioiodine	Tritium	Gamma spectroscopy
1953		15	21	10		x	x			
June 1954		15	21	10		x	x			
July 1954		15	17	11		x	x			
June 1955		15	17	11		x	x	x		
July 1955	48	14	14	14		x	x	x		
1956	48	14	14	14		x	x	x		
1957	20	14	14	14		x	x	x		
1958	20	14	14	14		x	x	x		
1959	20	b	14	14		x	x	x		
1960	20		14	14		x	x	x		
June 1961	20		14	14		x	x	x		
July 1961	20		14	14		x	x	x		x
1962	20		14	14	4	x	x	x		x
1963	20		14	14	4	x	x	x		x
June 1964	20		14	14	4	x	x	x		x
July 1964	4		7	7	4	x	x	x		x
1965 - 1973	4		7	7	4	x	x	x		x
1974	4		7	7	4	x	x	x	x	x
1975	4		7	7	4	x	x		x	x
1976	4		7	7	4	x	x		x	x
1977	4		7	7	4	x	x		x	x
1978	4		7	7	4	x	x	x	x	x
1979	4		7	7	4	x	x	x	x	x
1980	4		7	7	4	x	x	x	x	x
1981	4		7	7	4	x	x	x	x	x
1982	4		13	12	4	x	x	x	x	x
1983	4		13	12	4	x	x	x	x	x
1984	4		13	12	4	x	x		x	x
1985	4		13	12	4	x	x	x	x	x
1986	4		13	6	4	x	x		x	x
1987	4		14	4	4	x	x	x	x	x
1988	4		14	4	4	x	x	x	x	x
1989	4		14	4	4	x	x	x	x	x
1990	4		14	4	4	x	x		x	x
1991	4		14	4	4	x	x		x	x

[a] This table is intended to provide the reader with general time periods only for collection and analysis of samples at routine locations and is not an exact representation of all collected data
[b] sampling was discontinued

Table 9-2. Median, Maximum, and Minimum Annual Average Tritium Concentrations Measured in Vegetation Samples Collected at Individual Plant Perimeter, 25-Mi Radius, and 100-Mi Radius Locations Since 1976[a]

Location (years reported)	Direction[b]	Concentration (pCi mL^{-1})		
		Median	Maximum	Minimum
Plant perimeter				
East Talatha (1982–1991)	N	4.7	12	1
Windsor Road (1982–1991)	NNE	3.1	8	1.7
Darkhorse (1976—1991)	NE	4.4	15	0.8
Highway 21/167 (1982–1991)	ENE	3.4	36	0.8
Barnwell Gate (1982–1991)	E	1.9	13	0.6
Pattersons Mill (1976–1991)	ESE	3.0	18	0.4
Allendale Gate (1976–1991)	SSE	2.0	6.9	0.3
A/14 (1976–1991)	SSW	6.6	13	1.4
D area (1976–1991)	WSW	6.8	30	2.1
West Jackson (1982–1991)	WNW	3.1	27	1
Jackson (1982—1991)	NW	3.8	24	0.8
Greenpond (1976—1991)	NW	4.2	19	0.8
Talatha Gate (1976–1991)	NNW	4.2	15	0.6
Plant perimeter mean		*3.9*	*10.6*	*1.3*
25-mi radius				
Aiken Airport (1976–1985)	N	3.5	5	1.2
Aiken State Park (1976–1985)	NNE	1.8	4.9	0.6
Springfield (1976–1991)	NE	1.5	4.5	0.2
Lees (1976–1985)	ENE	2.3	3.1	1.5
Olar (1976–1985)	E	1.9	4.2	0.01
Allendale (1976–1991)	SE	1.1	4	0.2
Highway 301 (1976–1987)	SSE	1.6	2.9	0.7
Perkins (1976–1987)	SSW	1.4	4.6	0.7
Waynesboro (1976–1991)	WSW	1.6	4.8	0.3
South Richmond (1976–1986)	W	1.8	3.7	0.4
Augusta (1976–1986)	NW	1.7	4.1	0.8
Langley (1976–1991)	NNW	1.5	3.3	0.1
25-mi radius mean		*1.6*	*3.2*	*0.4*
100-mi radius				
Columbia	NNE	0.4	2.4	0.09
Greenville	NNW	0.4	2.9	0.03
Macon	WSW	0.4	8	0.04
Savannah	SSE	0.4	2.3	0.01
100-mi radius mean		*0.4*	*2.8*	*0.09*

[a] Data are from Du Pont (1977, 1978, 1979, 1980, 1981, 1982, 1983, 1984), Zeigler et al. (1985), Zeigler et al. (1986b, 1987b), Mikol et al. (1988b), Davis et al. (1989b), Cummins et al. (1990, 1991), and Arnett et al. (1992).

[b] Approximate direction relative to the center of the SRS.

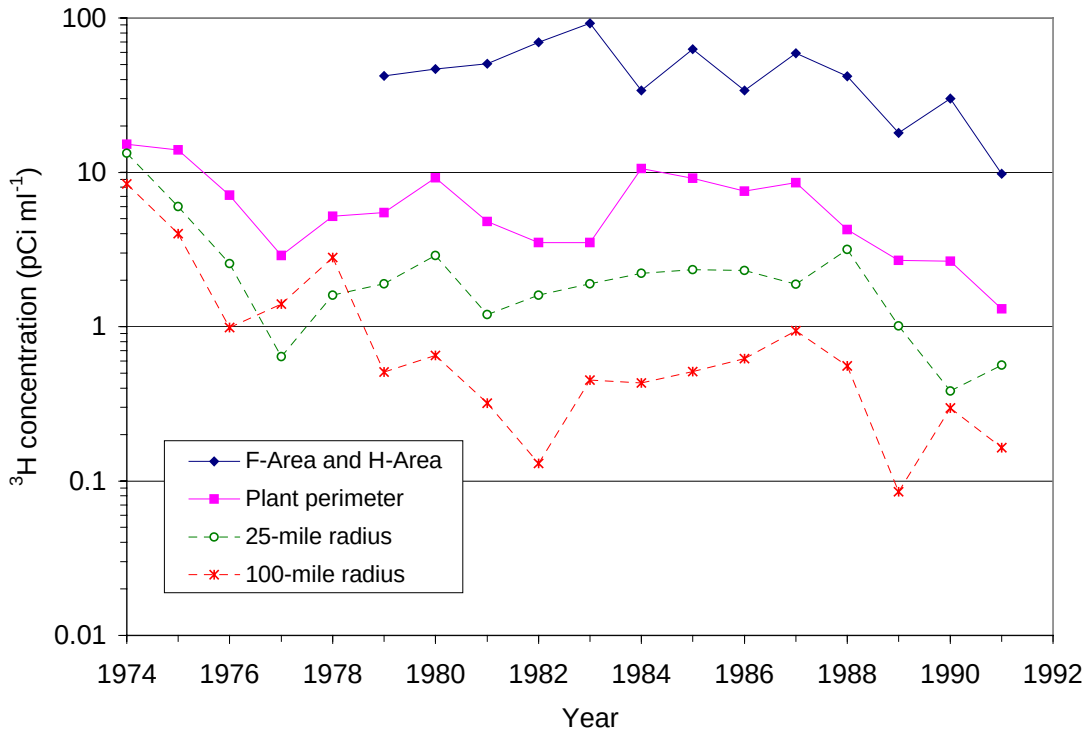

Figure 9-2. Tritium concentrations measured in water extracted from vegetation at the F-Area and H-Area, plant perimeter, 25-mi radius, and 100-mi radius locations. Link to tabulated figure data.

Because tritium concentrations were reported for 1-mi distant locations surrounding the F-Area and H-Area and many tritium releases originated from F-Area and H-Area stacks, it is instructive to look at concentrations as a function of distance from the F-Area and H-Areas (Figure 9-3). While the distance estimates are not precise, they do provide a good approximation. Average concentrations for 1979 through 1991 were calculated for approximately 1-, 10-, 25-, and 100-mi distances. Concentrations decreased as a power function of distance for the four locations, and regression analysis of the log-transformed data yielded a very good fit (R^2 = 0.997). This is consistent with information reported on special studies in 1973 and 1974 for tritium in vegetation at distances up to 8 mi in four directions around H-Stack (see special studies section).

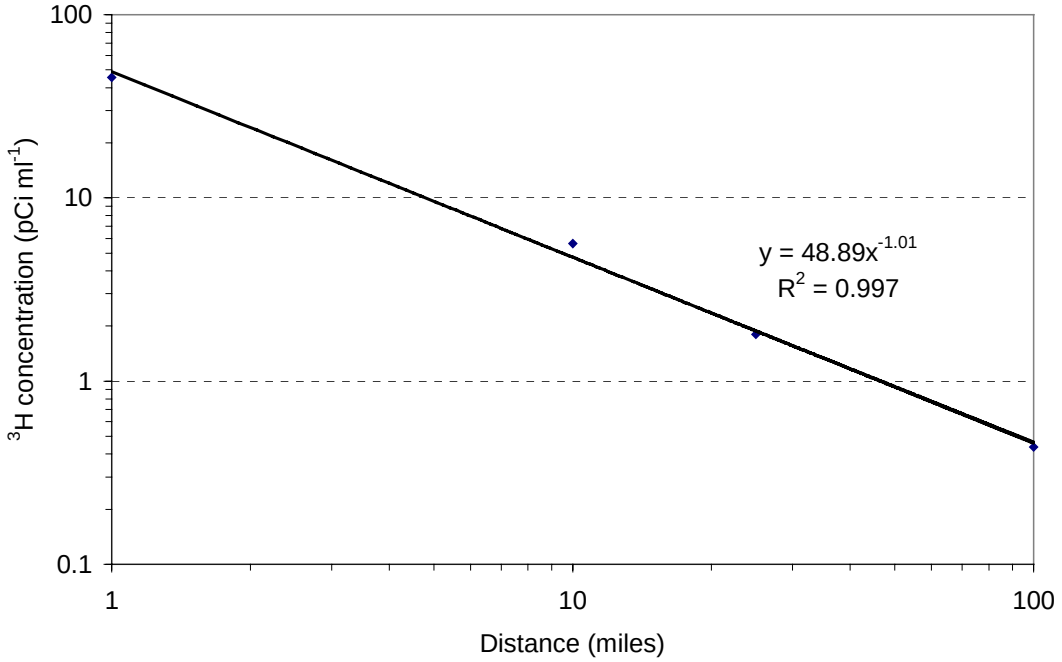

Figure 9-3. Average tritium concentration as a function of distance from the center of the SRS for 1979 through 1991. Link to tabulated figure data.

Figures 9A-1–9A-4 (Addendum 9A) show tritium concentrations for the individual plant perimeter, 25-mi radius, and 100-mi radius locations as a function of time since 1976. There are no apparent temporal or spatial trends for any of the locations at the 25-mi and 100-mi radius locations. The high concentration reported for the west-southwest direction at the 100-mi radius location in 1978 was due to a reported concentration of 30 pCi mL^{-1} in one sample collected at Macon, Georgia. At the plant perimeter, concentrations appear somewhat elevated in a westerly direction from 1983 through 1986. The wind was blowing in a northeast direction for most of the reported releases during this time period (see Table 9-3). An abnormally high concentration (36 pCi mL^{-1}) was reported at the Highway 21/167 perimeter location (east-northeast direction) in 1987. The wind was blowing in a northeast direction during a June 31, 1987 reported release (see Table 9-3). Comparisons based on annual average concentrations are of limited usefulness, however, because it is not possible to examine spatial or temporal trends during episodic releases.

Radioiodine, Gross Alpha, Nonvolatile Beta, and Gamma-Emitting Radionuclides

The environmental monitoring reports maintained that the remaining radionuclides for which concentrations have been reported are either naturally occurring or the result of atmospheric fallout from weapons testing. The following four sections examine these claims for radioiodine, gross alpha, nonvolatile beta, and gamma-emitting radionuclides. Naturally occurring radionuclide concentrations, including ^{7}Be and ^{40}K, are also examined.

Before the Test Ban Treaty of 1963 was signed by the U.S., United Kingdom, and Soviet Union, extensive atmospheric weapons testing was conducted. In addition, the Chinese conducted

atmospheric weapons tests on May 13, 1965; May 9 and October 27, 1966; September 26 and November 17, 1976; September 17, 1977; March 14 and December 14, 1978; and October 16, 1980 (Du Pont 1965b, 1966b, 1967a; Ashley and Zeigler 1978a, 1978b, 1981, and 1983). Carter and Moghissi (1977) provides a comprehensive discussion of all nuclear detonations occurring between July 1945 and June 1975.

Several radionuclides associated with atmospheric testing as well as reactor and dissolution facility operations have been periodically detected in vegetation (e.g., ^{103,106}Ru, ^{95}Zr/Nb, ^{140}Ba/La, and ^{131}I). However, with the exception of the longer-lived radionuclides (including ^{141,144}Ce and ^{134,137}Cs) and nonvolatile beta activity, concentrations have in most cases been near the lower limit of detection (LLD)[1]. In general, reported concentrations for all fallout radionuclides have been indistinguishable at the plant perimeter, 25-mi radius, and 100-mi radius locations. Based on the data that have been reviewed, there is little evidence to suggest that SRS operations have resulted in elevated perimeter or offsite concentrations of any fission products, with the exception of radioiodine. There is some indication that concentrations have historically been higher at F-Area and H-Area locations, which is likely the result of F-Area and H-Area releases. With the exception of radioiodine, there is also evidence that suggests this pattern may partially be the result of variations in areal deposition rates. These topics are discussed in the following sections.

Radioiodine Concentrations

Vegetation was analyzed for ^{131}I from 1955 through 1991. Before 1961, radioiodine concentrations were determined using undried samples of vegetation, so concentrations reported in this chapter during that time period are in units of picocuries per gram wet weight. Beginning in 1961, vegetation samples were dried before analysis, so concentrations from 1961 through 1991 are in units of picocuries per gram dry weight. This should be noted when comparing concentrations measured during these two time periods.

The monitoring reports state that tritium is the only radionuclide of SRS origin to be detected in offsite vegetation. To examine the credibility of this claim, it is helpful to compare concentrations of other radionuclides measured in vegetation collected at the F-Area and H-Area (many of the radioiodine releases at the SRS originated from the F-Area and H-Area stacks), plant perimeter, and 25-mi radius locations (Figure 9-4). Concentrations measured before July 1961 were reported on a wet weight basis, and concentrations measured since July 1961 were reported on a dry weight basis. For comparison, activity reported on a wet weight basis is approximately 3 times lower than the same amount of activity reported on a dry weight basis. For the purpose of viewing the data in Figure 9-4, we have increased the wet weight data before July 1961 by a factor of 3 to allow for a more direct comparison to the dry weight data after July 1961. F-Area and H-Area concentrations were not reported before July 1955, so inner perimeter concentrations were substituted for analysis of iodine, gross alpha, and nonvolatile beta comparisons during that time period. The lower limit of detection (LLD) is also shown.

[1] The lower limit of detection (LLD) values are provided in various figures throughout this chapter whenever they were reported in the environmental monitoring reports. In many cases, however, the reports state "...the sensitivity of analysis varied due to differing sample size..." and no LLD values were provided.

Radioiodine concentrations measured in vegetation have generally been highest at the F-Area and H-Area locations, particularly during the 1950s. Elevated concentrations at all locations during 1961, 1962, 1965, 1966, and 1978 may be related to atmospheric weapons tests conducted by the Soviet Union, U.S., and China during those years. However, limited data were available for F-Area and H-Area locations from September 1960 through December 1961, and concentrations have been infrequently reported since 1974. A concentration of 245 pCi g^{-1} was reported for F-Area and H-Area locations in June 1961, compared to concentrations of 5.6 and 5.2 pCi g^{-1} at the plant perimeter and 25 mi-radius locations, respectively.

Figures 9-5 and 9-6 show ^{131}I concentrations at the F-Area and H-Area and plant perimeter locations, respectively, compared to concentrations measured at the 25-mi radius locations. This comparison is useful to establish whether concentrations at the F-Area and H-Area and plant perimeter locations are elevated compared to concentrations at the 25-mi radius locations. Ratio values consistently greater than 1 suggest higher concentrations than at the 25-mi radius locations, and ratio values averaging close to 1 suggest similar concentrations to the 25-mi radius locations. The values are plotted on a logarithmic scale in Figure 9-5, but it is clear that F-Area and H-Area concentrations are elevated relative to 25-mi radius locations, particularly through 1960. This reflects emissions from F-Area and H-Area stacks. Conversely, concentrations do not appear to be elevated at plant perimeter locations relative to 25-mi radius locations based on these data.

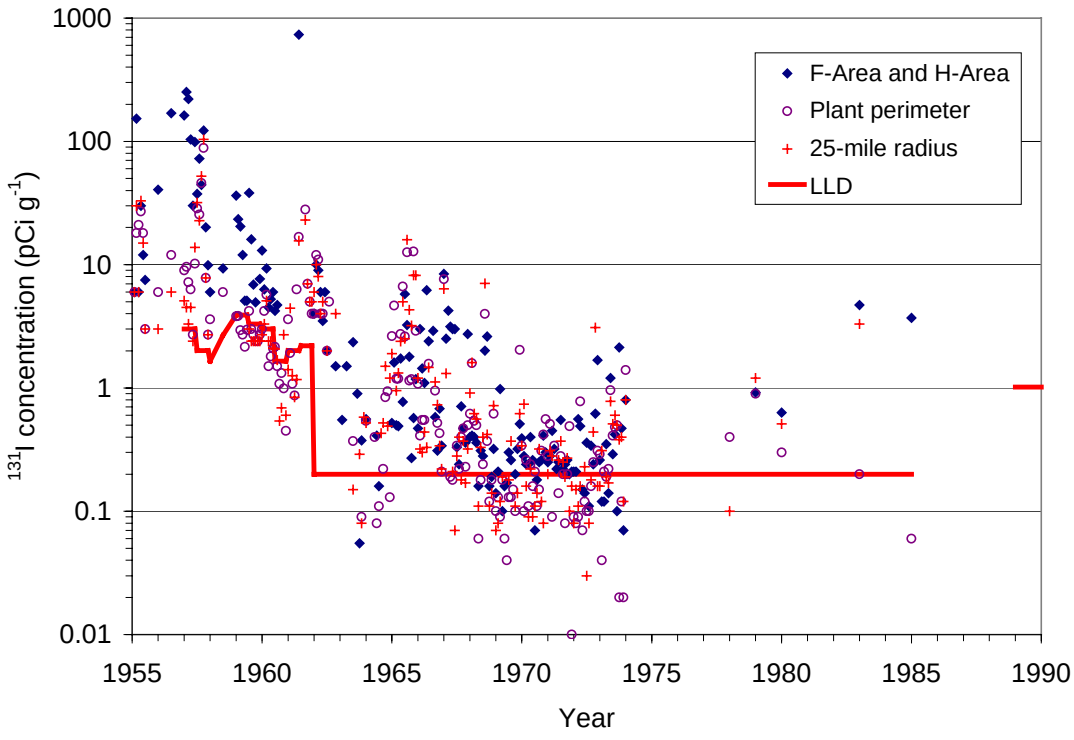

Figure 9-4. Iodine-131 concentrations measured in vegetation (dry weight) from the F-Area and H-Area, plant perimeter, and 25-mi radius locations since 1955 shown with the reported LLD. For the period before July 1961, we have increased the reported wet weight concentrations by a factor of 3 to allow for a more direct comparison to the dry weight data (see text for details). Link to tabulated figure data.

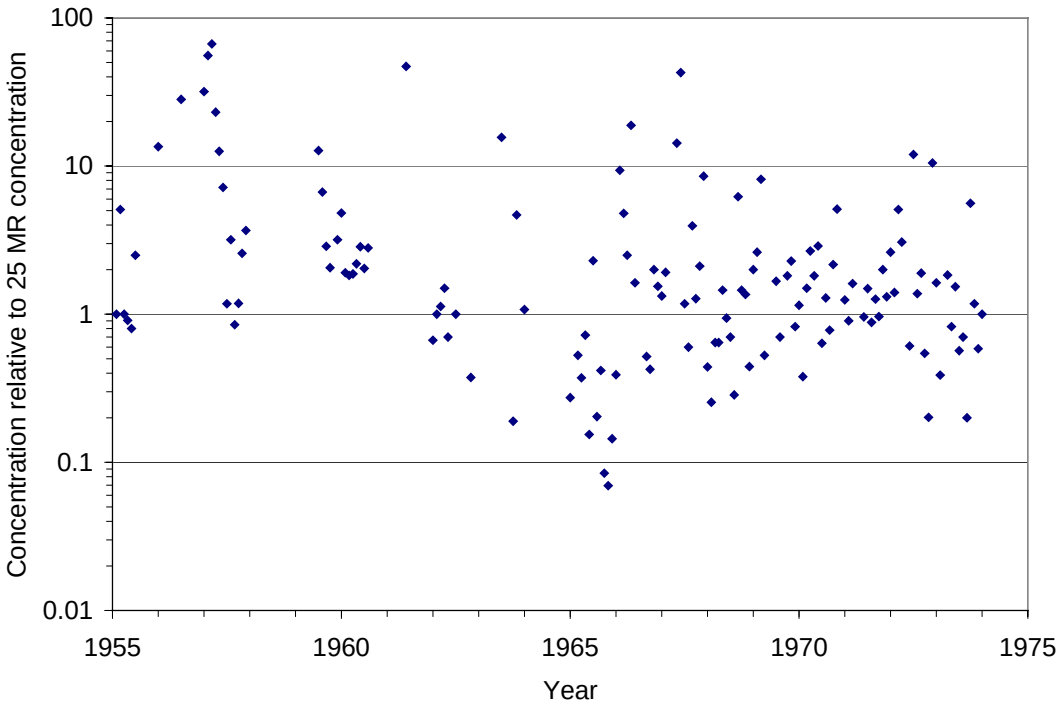

Figure 9-5. Ratio of ^{131}I concentrations measured in vegetation at the F-Area and H-Area to those measured at 25-mi radius locations. Link to tabulated figure data.

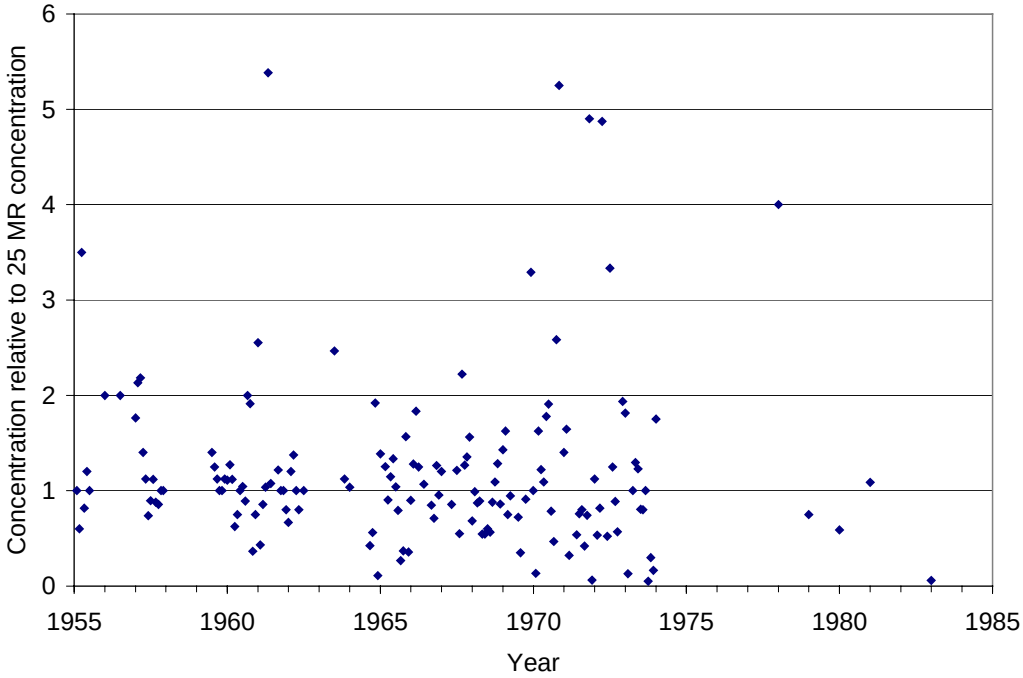

Figure 9-6. Ratio of ^{131}I concentrations measured in vegetation at the plant perimeter to those measured at 25-mi radius locations. Link to tabulated figure data.

Since 1970, no concentrations greater than 1 pCi g^{-1} have been reported for either of these locations. The higher ratio values since that time in Figure 9-6 are likely the result of comparing measured concentrations near or below the detection limit of 0.2 pCi g^{-1}. A higher ratio value (5.4) in May 1961 resulted from comparing reported concentrations of 2.1 and 0.39 pCi g^{-1} at the plant perimeter and 25-mi radius locations, respectively. The concentration for F-Area and H-Area locations was not reported. A higher ratio value (3.5) in April 1955 resulted from comparing reported concentrations of 7 and 2 pCi g^{-1} at the plant perimeter and 25-mi radius locations, respectively. For comparison, the inner perimeter concentration was 2 pCi g^{-1}.

Weekly average ^{131}I concentrations are available for 1957 and 1959 through 1961 in aperture card printouts for those years (Figures 9-7, 9-8, 9-9, and 9-10) (Du Pont 1957, 1959a, 1960c, and 1961b). Concentrations appear elevated at the plant perimeter locations compared to 25-mi radius locations through the first half of 1957. This suggests that SRS operations in 1957 may have impacted ^{131}I concentrations in vegetation to at least the plant perimeter. During the second half of 1957 and from January 1959 through June 1961, plant perimeter and 25-mi radius concentrations appear similar and/or near the LLD. This indicates that the majority of ^{131}I peaks detected in vegetation during these time periods resulted from atmospheric deposition. Similar peaks in concentration at the plant perimeter and 25-mi radius locations occurred in June 1961, following a substantial increase in the daily release rate of ^{131}I from the Building 291-F stack. It is difficult to determine if this increase resulted from weapons testing or plant releases, but similar concentrations at both distances suggest deposition related to weapons testing. Unfortunately, weekly or monthly data for 1958 or for years before 1957 are not available. Significant plant releases of ^{131}I occurred in 1956, but reported releases in 1958 were similar to reported releases in 1960, and elevated concentrations at the plant perimeter are not evident in 1960. Average monthly and semiannual concentrations in 1955 and 1956 suggest little SRS impact, but the temporal resolution is severely limited considering the relatively short half-life of ^{131}I (about 8 days).

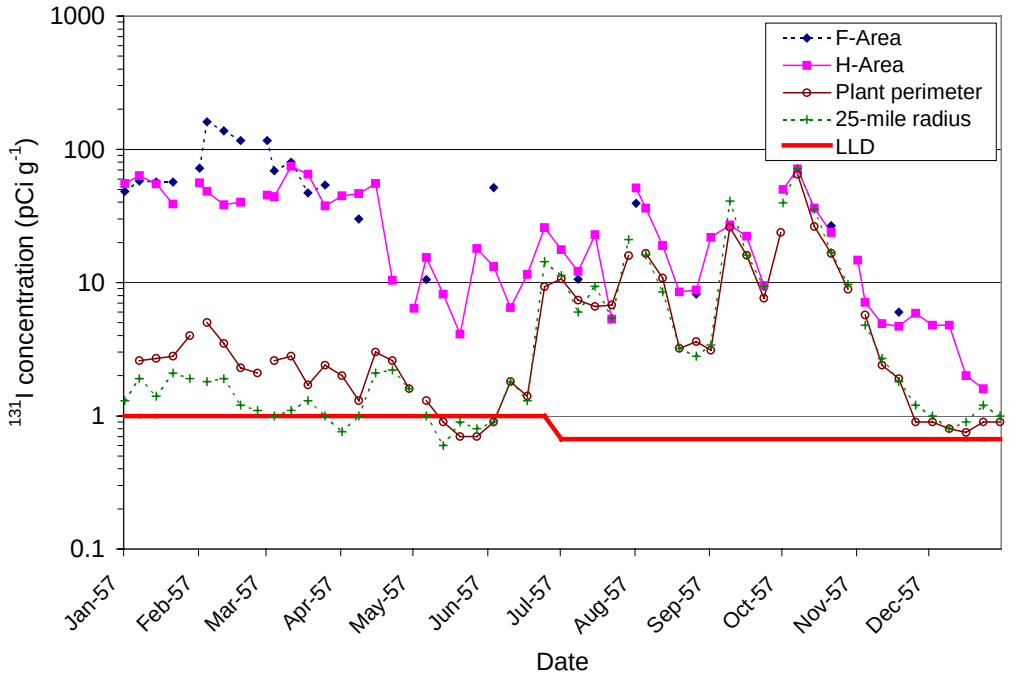

Figure 9-7. Weekly ^{131}I concentrations measured in vegetation (wet weight) at the F-Area and H-Area, plant perimeter, and 25-mi radius locations during 1957. Link to tabulated figure data.

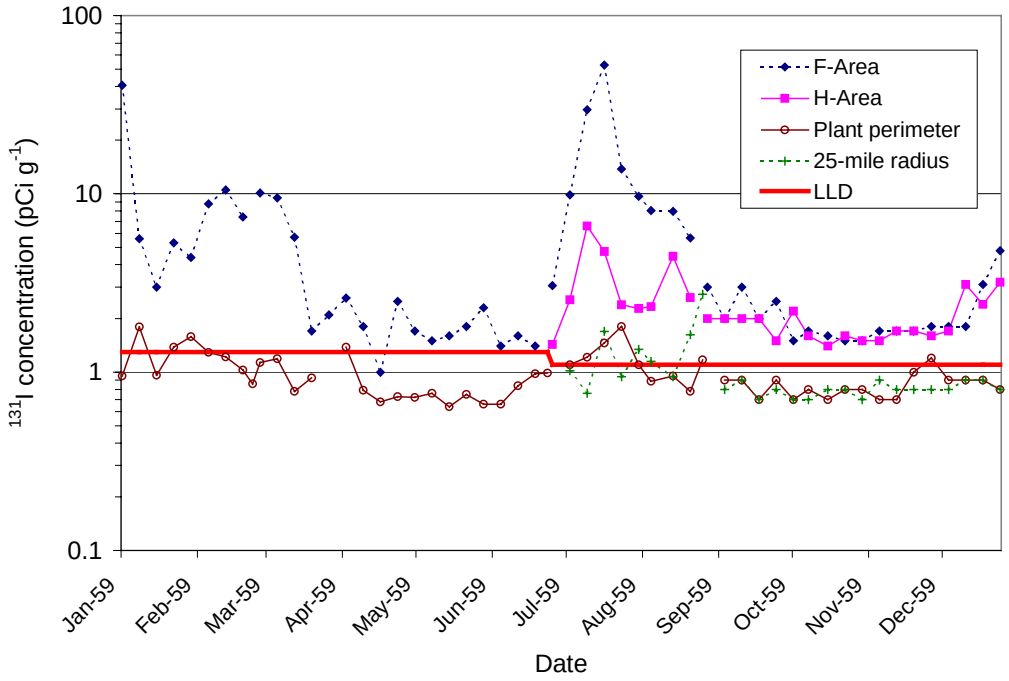

Figure 9-8. Weekly ^{131}I concentrations measured in vegetation (wet weight) at the F-Area and H-Area, plant perimeter, and 25-mi radius locations during 1959. Link to tabulated figure data.

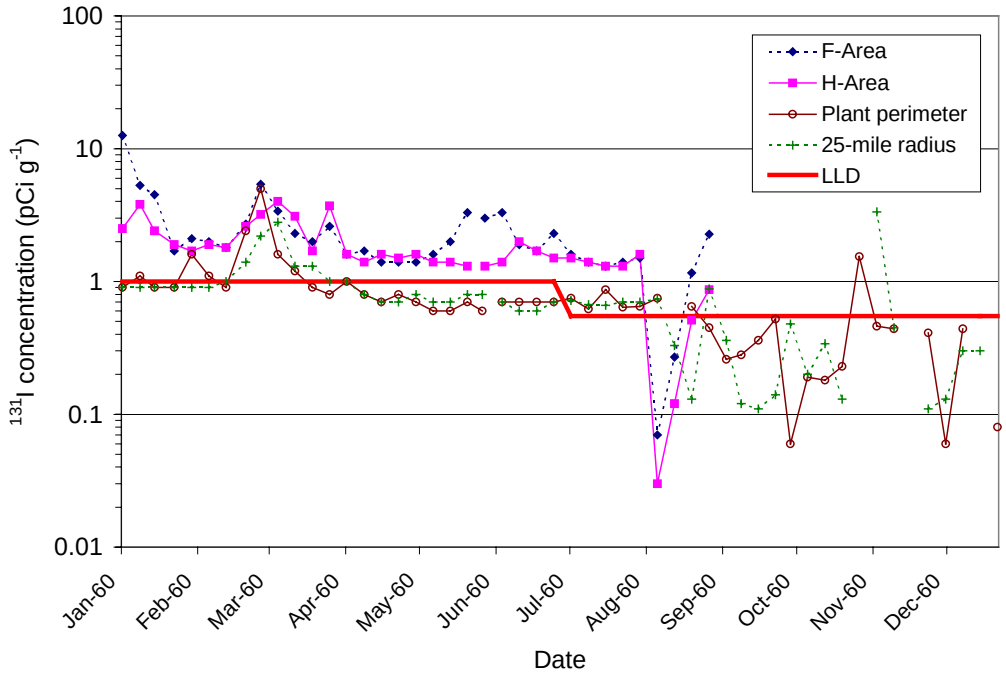

Figure 9-9. Weekly ^{131}I concentrations measured in vegetation (wet weight) at the F-Area and H-Area, plant perimeter, and 25-mi radius locations during 1960. Link to tabulated figure data.

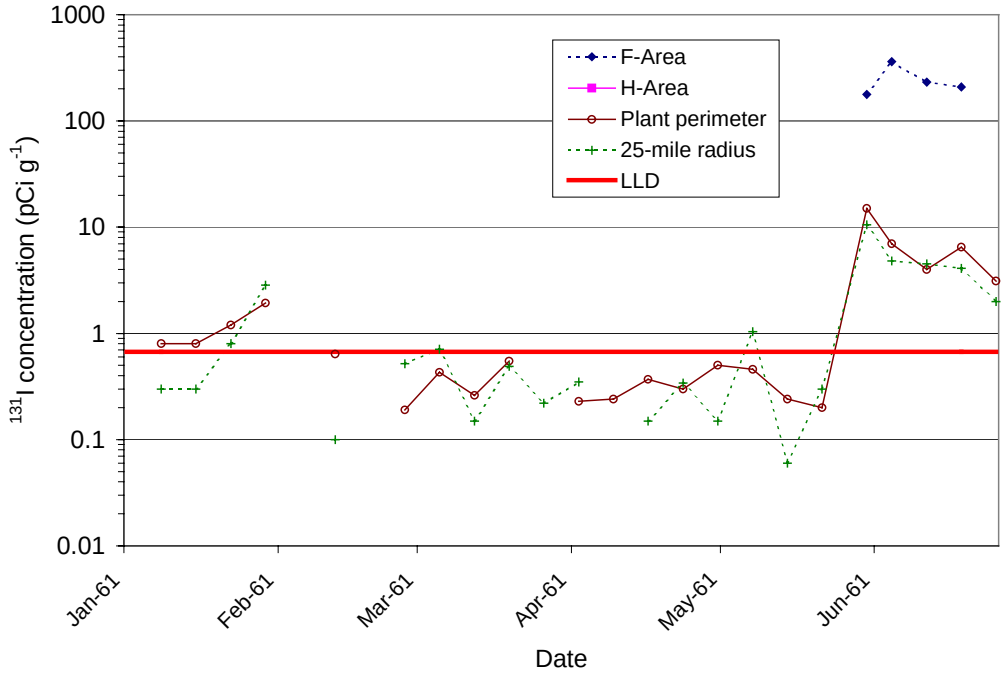

Figure 9-10. Weekly ^{131}I concentrations measured in vegetation (wet weight) at the F-Area and H-Area, plant perimeter, and 25-mi radius locations during the first half 1961. Link to tabulated figure data.

Comparing vegetation and air ^{131}I concentrations during 1959 and January through June of 1961 reveals generally close correlation between the two media, which would be expected (Figures 9-11 and 9-12). Vegetation concentrations do not appear to show the same increase seen for air concentrations during April, May, and June 1959. An explanation for this is not apparent, but trend differences between the two media could result from variations in wind rose, precipitation, and collection dates.

Based on the concentrations reported in the documents we reviewed, ^{131}I concentrations have clearly been elevated at the F-Area and H-Area locations compared to both the plant perimeter and 25-mi radius locations. However, it is difficult to distinguish between reported concentrations at the plant perimeter and 25-mi radius locations since the second half of 1957, and it is likely that the majority of ^{131}I detected at these locations was the result of atmospheric deposition. Plant operations appear to have impacted plant perimeter concentrations during at least the first half of 1957. Based on reported releases for 1955 and 1956, which were greater than for 1957, it is likely that ^{131}I concentrations in vegetation at the plant perimeter locations were also impacted by SRS operations during these years.

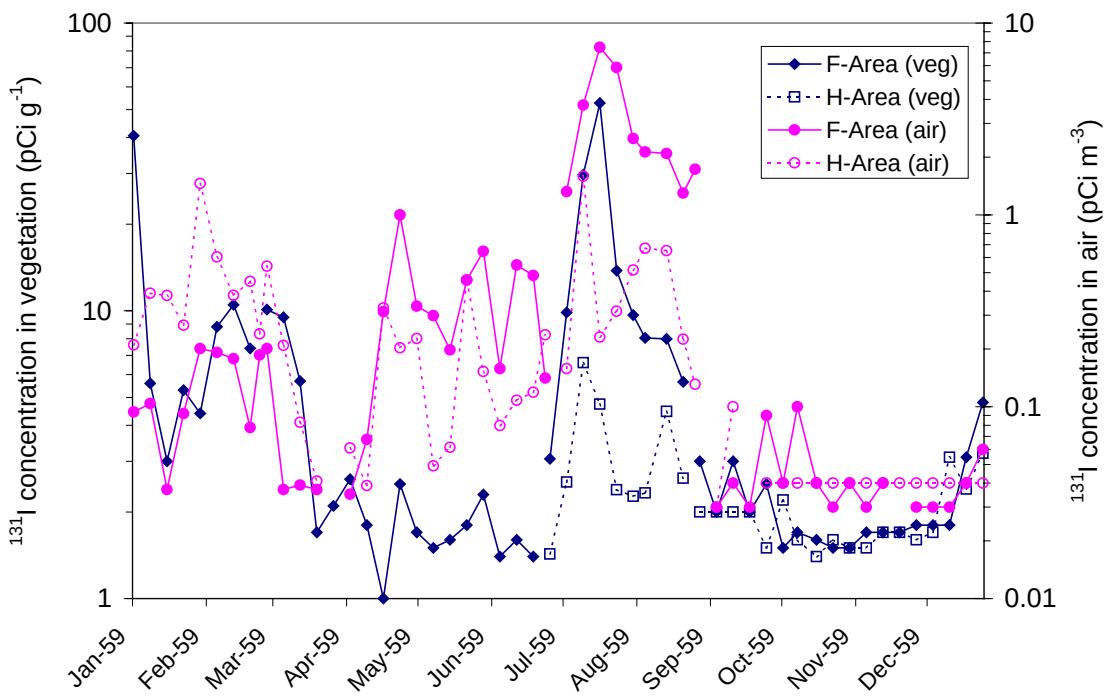

Figure 9-11. Comparison of ^{131}I concentrations measured in vegetation (wet weight) and air samples collected from the F-Area and H-Area locations during 1959. Link to tabulated figure data.

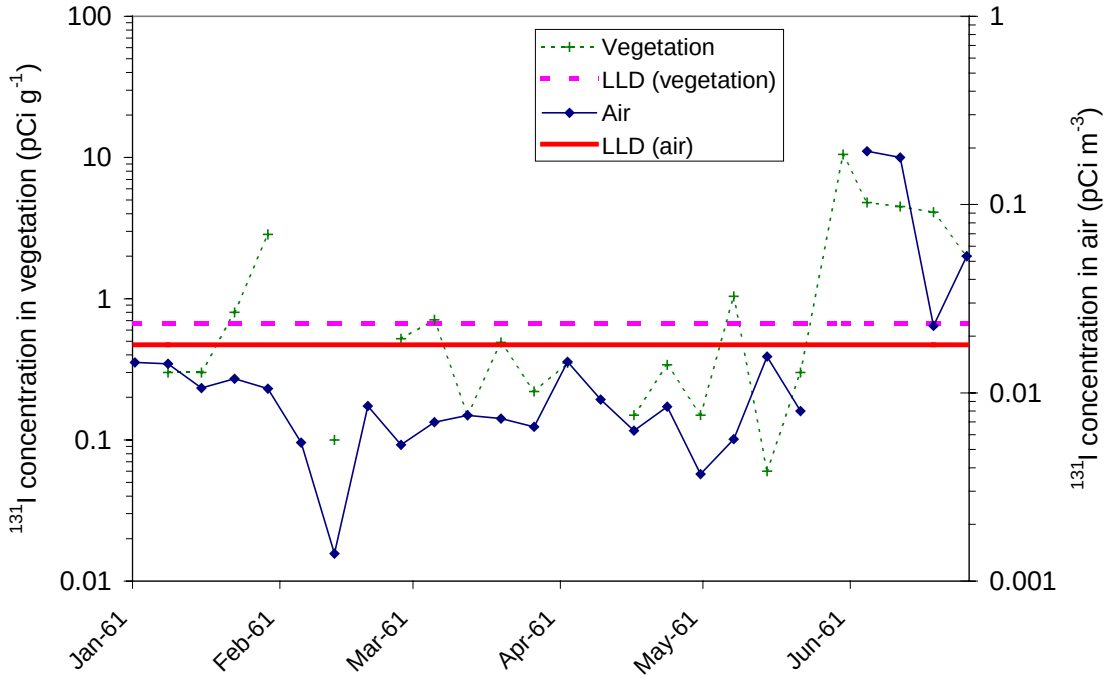

Figure 9-12. Comparison of ^{131}I concentrations measured in vegetation (wet weight) and air samples collected from the 25-mi radius locations during the first half of 1961. Link to tabulated figure data.

Gross Alpha Concentrations

Gross alpha concentrations were reported for vegetation from 1954 through 1991. It is again helpful to examine concentrations that have been reported for the F-Area and H-Area and plant perimeter locations to assess potential SRS impact (Figure 9-13). To present the data as clearly as possible, 25-mi radius concentrations are not included, but they have been generally indistinguishable from plant perimeter concentrations. Concentrations appear consistently higher at the F-Area and H-Area locations.

Comparing gross alpha concentrations at the F-Area and H-Area and plant perimeter locations to concentrations at 25-mi radius locations (Figures 9-14 and 9-15, respectively) confirmed consistently higher concentrations at F-Area and H-Area locations but generally indistinguishable concentrations at the plant perimeter and 25-mi radius locations.

A higher ratio value (5.5) in October 1961 (Figure 9-15) resulted from comparing reported concentrations of 0.22 and 0.04 pCi g^{-1} at the plant perimeter and 25-mi radius locations, respectively. For comparison, the concentration reported for F-Area and H-Area locations was 0.1 pCi g^{-1}. A higher ratio value (4.1) in 1991 (Figure 9-15) resulted from comparing reported concentrations of 0.29 and 0.07 pCi g^{-1} at the plant perimeter and 25-mi radius locations, respectively. For comparison, the concentration reported for the 100-mi radius locations was 0.22 pCi g^{-1}.

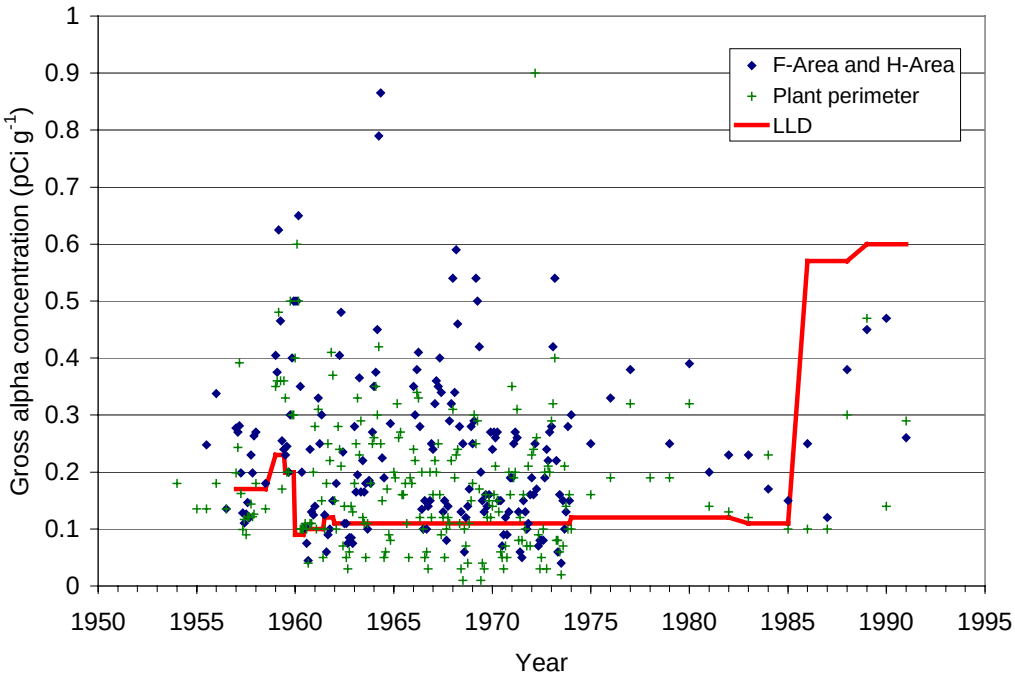

Figure 9-13. Gross alpha concentrations measured in vegetation (dry weight) at the F-Area and H-Area and plant perimeter locations shown with the reported LLD. Link to tabulated figure data.

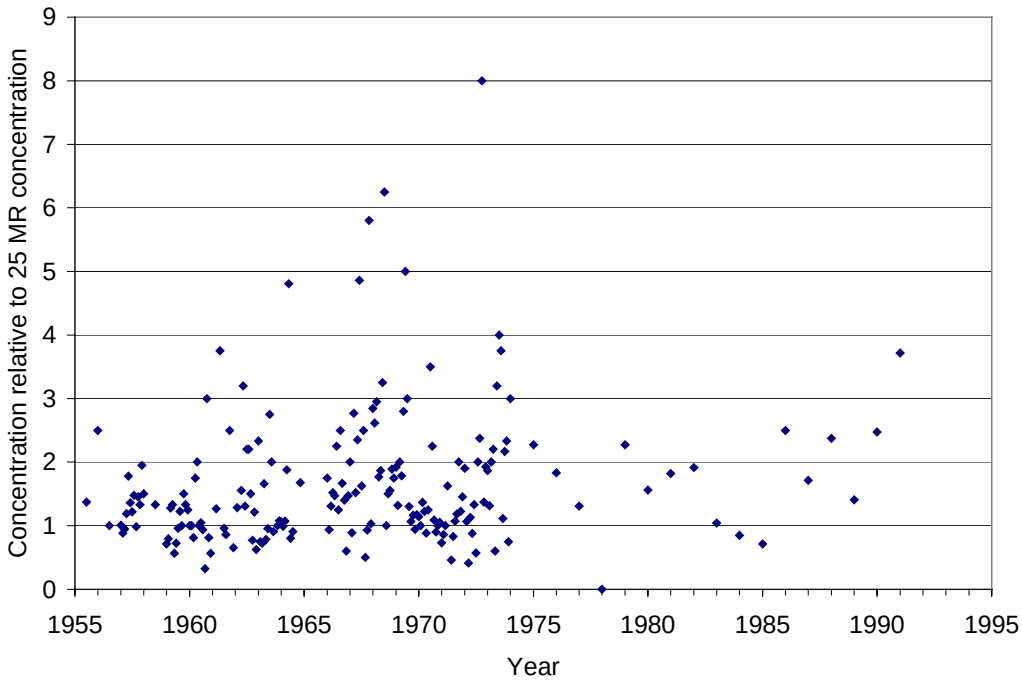

Figure 9-14. Ratio of gross alpha concentrations measured in vegetation (dry weight) at the F-Area and H-Area to those measured at 25-mi radius locations. Link to tabulated figure data.

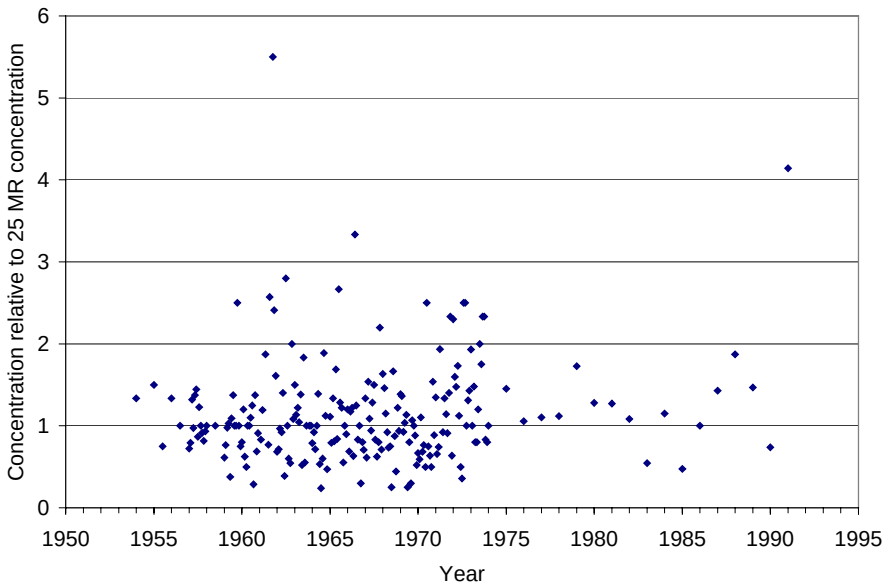

Figure 9-15. Ratio of gross alpha concentrations measured in vegetation (dry weight) at the plant perimeter to those measured at 25-mi radius locations. Link to tabulated figure data.

Based on the concentrations reported in the documents we reviewed, gross alpha concentrations appear to be elevated at the F-Area and H-Area locations compared to both the plant perimeter and 25-mi radius locations. However, it is difficult to distinguish between reported concentrations at the plant perimeter and 25-mi radius locations, and it is likely that the majority of gross alpha activity detected at these locations has been the result of atmospheric deposition.

Nonvolatile Beta Concentrations

Nonvolatile beta concentrations were reported for vegetation from 1954 through 1991. The primary radionuclides that could potentially contribute to nonvolatile beta activity at the SRS include ^{137}Cs, ^{65}Zn, ^{90}Sr, ^{32}P, and naturally occurring ^{40}K. It is again helpful to examine concentrations that have been reported for the F-Area and H-Area and plant perimeter locations to assess potential SRS impact (Figure 9-16). To present the data as clearly as possible, 25-mi radius concentrations are not included, but they have been generally indistinguishable from plant perimeter concentrations. Concentrations appear consistently higher at the F-Area and H-Area locations.

A significantly higher concentration (2600 pCi g^{-1}) was reported for the F-Area and H-Area locations (actually inner perimeter locations, see the iodine section) in March 1955. The high concentration was attributed to fallout from Nevada Test Site testing on March 12 (Teapot Hornet), which reportedly arrived on March 14 (Alexander and Horton 1956). Stack air filters indicated no unusual discharge, and constant air monitors showed that the material arrived at widely separated areas on the plant site between 2:00 and 2:30 p.m. A wind speed of 10 mph recorded at 200-F would likely not have been sufficient for such widespread contamination. Half-life determinations made by several Health Physics Area Survey Groups showed the half-life to

be between 30 and 40 hours, which is consistent with fission products that have had a few days to decay during atmospheric transport from the Nevada Test Site. The half-life for fission products released from the 200-Area would be significantly shorter since the shorter-lived radionuclides would not have had time to decay. The material appeared to be fallout, and lower concentrations measured at the plant perimeter and 25-mi radius locations were attributed to later sampling dates. The fallout reportedly arrived on March 14, and inner perimeter, outer (plant) perimeter, and 25-mi radius locations were sampled on March 15, 23, and 30, respectively. The reported concentrations for these locations were 2600, 220, and 700 pCi g^{-1}, respectively. It is not clear why the plant perimeter concentrations were lower than the 25-mi radius concentration. However, variations in precipitation patterns could be a factor. See Chapter 6 for a more detailed discussion regarding this event.

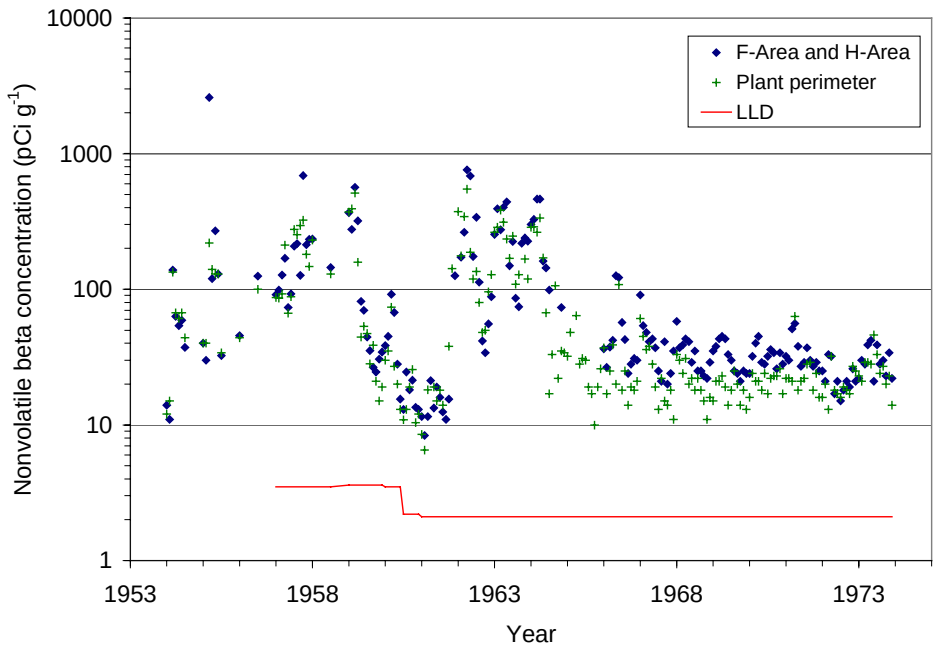

Figure 9-16. Nonvolatile beta concentrations measured in vegetation (dry weight) at the F-Area and H-Area and plant perimeter locations shown with the reported LLD. Link to tabulated figure data.

The nonvolatile beta concentrations at the F-Area and H-Area and plant perimeter locations compared to concentrations at the 25-mi radius locations (Figures 9-17 and 9-18) confirm consistently higher concentrations at the F-Area and H-Area locations but generally indistinguishable concentrations at the plant perimeter and 25-mi radius locations. A higher ratio value (3.4) in April 1957 (Figure 9-18) resulted from comparing reported concentrations of 212 and 62 pCi g^{-1} at the plant perimeter and 25-mi radius locations, respectively. For comparison, the concentration reported for the F-Area and H-Area locations was 169 pCi g^{-1}. A higher ratio value (3.3) in 1991 (Figure 9-18) resulted from comparing reported concentrations of 106 and 32 pCi g^{-1} at the plant perimeter and 25-mi radius locations, respectively. The concentration at the F-Area and H-Area locations was not reported.

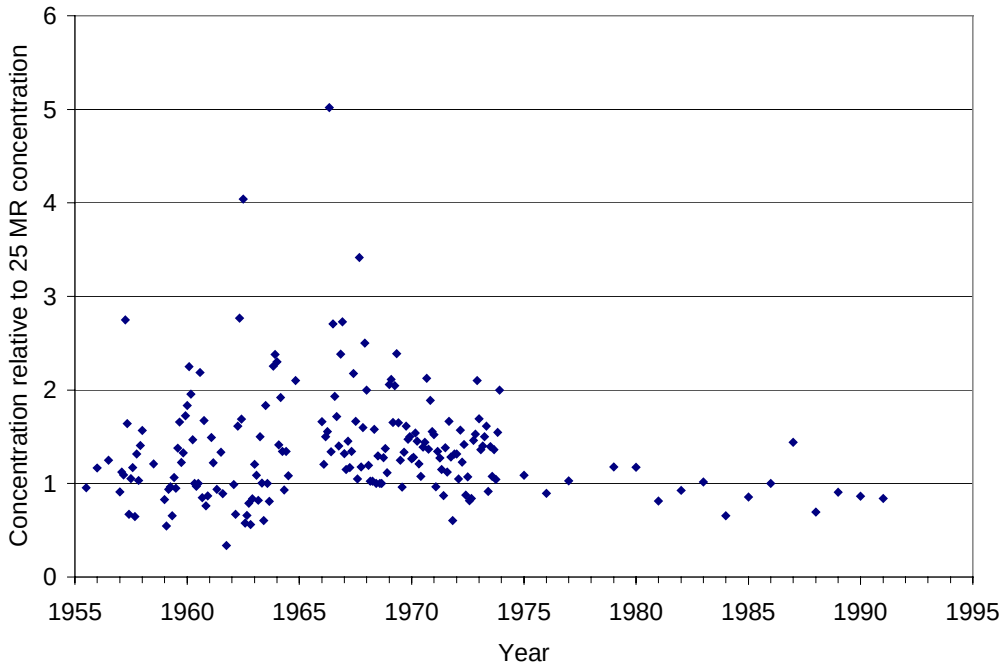

Figure 9-17. Ratio of nonvolatile beta concentrations measured in vegetation (dry weight) at the F-Area and H-Area to those measured at 25-mi radius locations. Link to tabulated figure data.

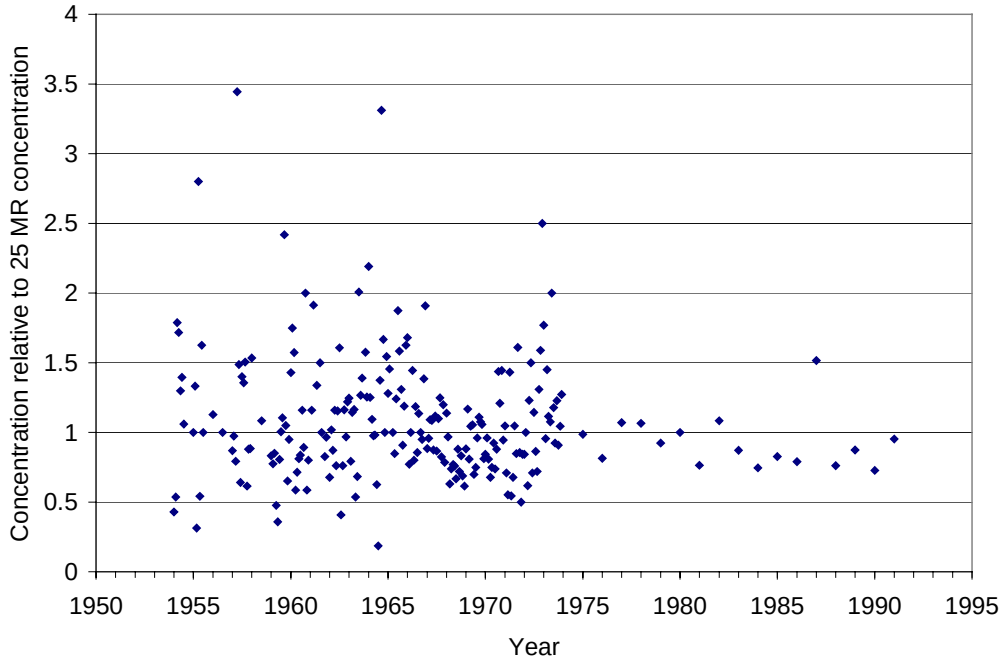

Figure 9-18. Ratio of nonvolatile beta concentrations measured in vegetation (dry weight) at the plant perimeter to those measured at the 25-mi radius locations. Link to tabulated figure data.

Based on the concentrations reported in the documents we reviewed, nonvolatile beta concentrations appear to be elevated at the F-Area and H-Area locations compared to both the plant perimeter and 25-mi radius locations. However, it is difficult to distinguish between reported concentrations at the plant perimeter and 25-mi radius locations, and it is likely that the majority of nonvolatile beta activity detected at these locations was the result of atmospheric deposition.

Gamma-Emitting Radionuclide Concentrations

Gamma-emitting radionuclides (^{144}Ce, ^{137}Cs, ^{106}Ru, and ^{95}Zr/Nb) have routinely been detected in vegetation and have been reported since July 1961. Concentrations of ^{137}Cs, ^{144}Ce, ^{106}Ru, and ^{95}Zr/Nb are shown in Figures 9-19, 9-20, 9-21, and 9-22, respectively. There do not appear to be any significant differences between concentrations of these radionuclides measured at the plant perimeter and 25-mi locations. However, as was noted for gross alpha and nonvolatile beta activity, F-Area and H-Area locations appear elevated compared to the 25-mi radius and plant perimeter locations. Where lower limits of detection (LLDs) were reported, they are included in the figures. However, from approximately 1973 through 1985, these values were not reported. Ashley and Zeigler (1984) states, "Where samples are analyzed by gamma spectrometry, the lower level of detection of a given radionuclide varies with the background of each individual channel grouping, with the geometry and volume of the sample analyzed, and with the number of radionuclides present in the sample."

Comparing plant perimeter and F-Area and H-Area concentrations to 25-mi radius concentrations again confirmed elevated concentrations at the F-Area and H-Area locations and generally indistinguishable concentrations at the plant perimeter and 25-mi radius locations. For brevity, the plots are not shown, but the same general trends discussed for alpha and nonvolatile beta activity were evident. F-Area and H-Area concentrations appear elevated for the four radionuclides, as illustrated in Figures 9-14 and 9-17, and plant perimeter concentrations appear generally indistinguishable from 25-mi radius concentrations for the four radionuclides, as illustrated in Figures 9-15 and 9-18.

Based on the concentrations reported in the documents we reviewed, gamma-emitting radionuclide (^{144}Ce, ^{137}Cs, ^{106}Ru, and ^{95}Zr/Nb) concentrations appear to be elevated at the F-Area and H-Area locations compared to both the plant perimeter and 25-mi radius locations. However, it is difficult to distinguish between reported concentrations at the plant perimeter and 25-mi radius locations, and it is likely that the majority of gamma-emitting radionuclides detected at these locations were the result of atmospheric deposition.

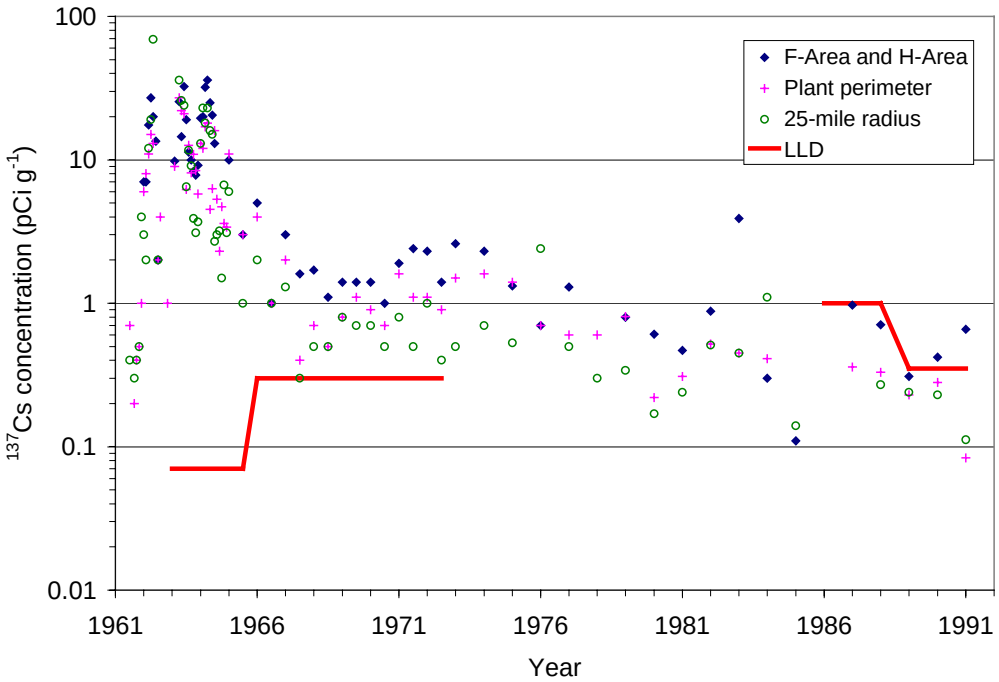

Figure 9-19. Cesium-137 concentrations measured in vegetation (dry weight) at the F-Area and H-Area, plant perimeter, and 25-mi radius locations. Link to tabulated figure data.

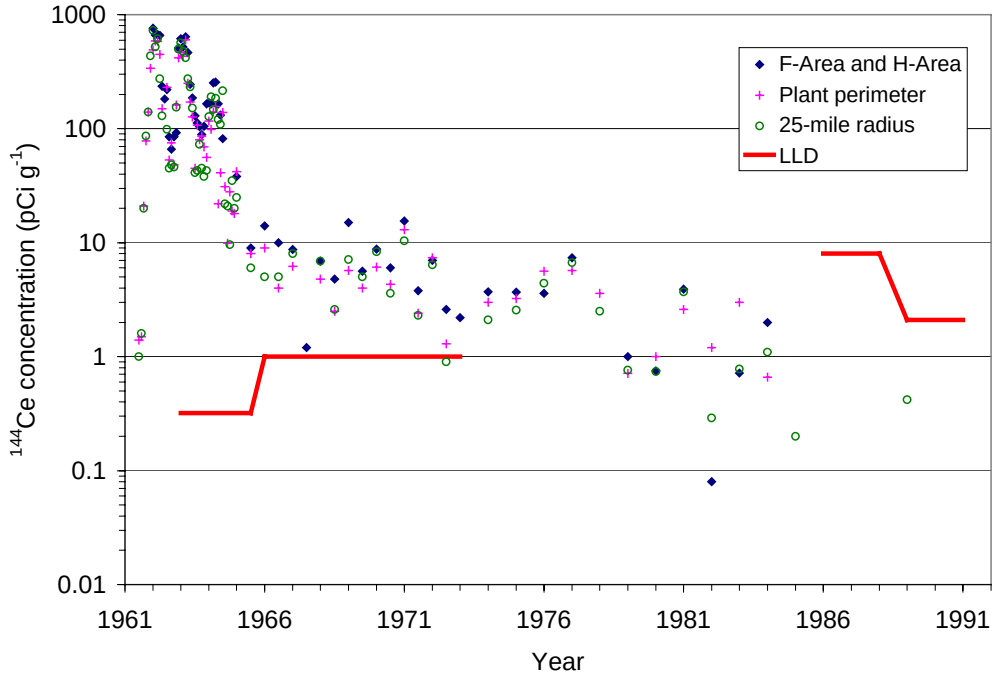

Figure 9-20. Cerium-144 concentrations measured in vegetation (dry weight) at the F-Area and H-Area, plant perimeter, and 25-mi radius locations. Link to tabulated figure data.

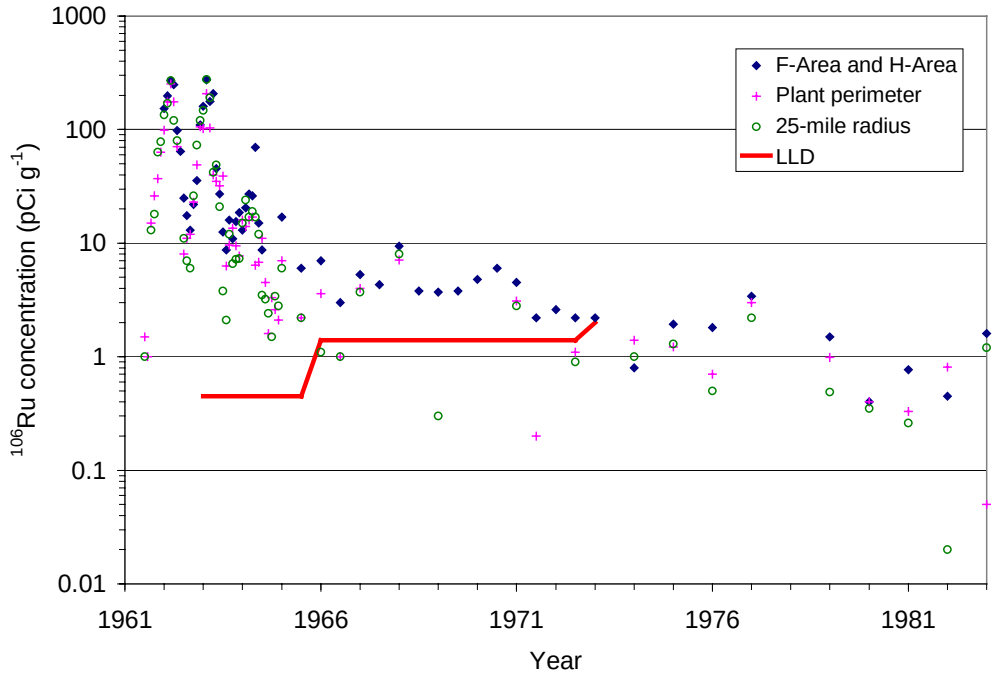

Figure 9-21. Ruthenium-106 concentrations measured in vegetation (dry weight) at the F-Area and H-Area, plant perimeter, and 25-mi radius locations. Link to tabulated figure data.

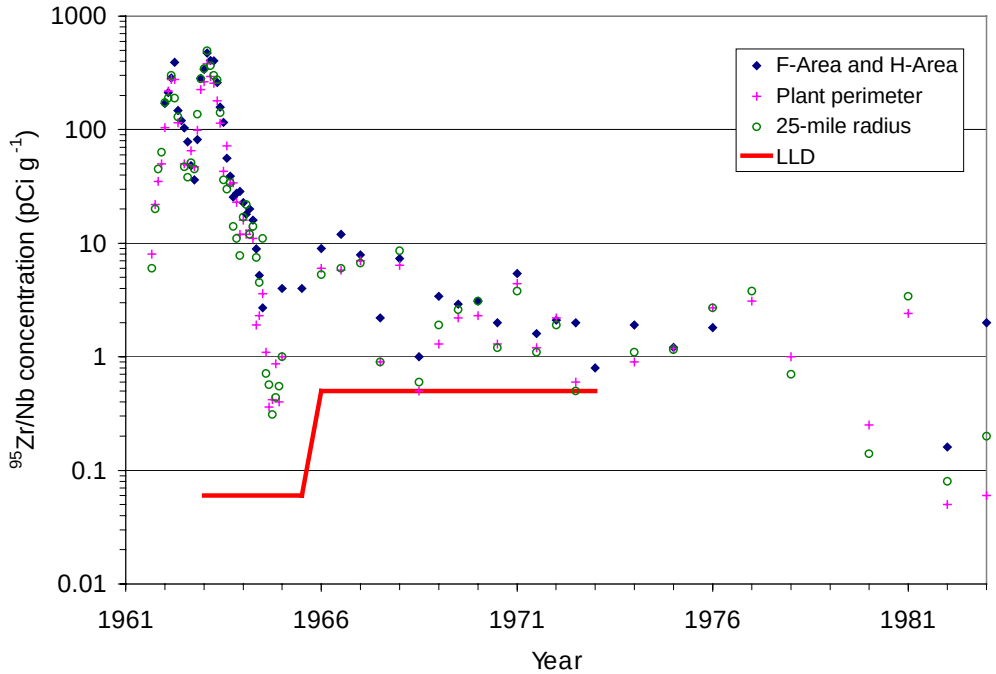

Figure 9-22. Zirconium/Niobium-95 concentrations measured in vegetation (dry weight) at the F-Area and H-Area, plant perimeter, and 25-mi radius locations. Link to tabulated figure data.

Naturally Occurring Radionuclide Concentrations

It is interesting to note that gross alpha activity, nonvolatile beta activity, and gamma-emitting radionuclide concentrations measured at the F-Area and H-Area locations have typically been elevated compared to the plant perimeter and 25-mi radius locations. Naturally occurring ^{7}Be concentrations have been measured at all locations since 1965. Because ^{7}Be is primarily a product of cosmic interactions in the atmosphere and is not extensively associated with weapons testing or SRS activities, it is instructive to examine concentrations at the various locations to determine whether the same trends are evident (Figure 9-23).

The same general trends are evident, with F-Area and H-Area concentrations consistently higher than plant perimeter and 25-mi radius concentrations. Examining F-Area and H-Area and plant perimeter concentrations compared to 25-mi radius concentrations (Figures 9-24 and 9-25) confirms higher concentrations at the F-Area and H-Area locations, and plant perimeter and 25-mi radius concentrations appear indistinguishable. Close inspection of the data also reveals higher concentrations during the first half of the year, which is consistent with expected patterns of atmospheric deposition.

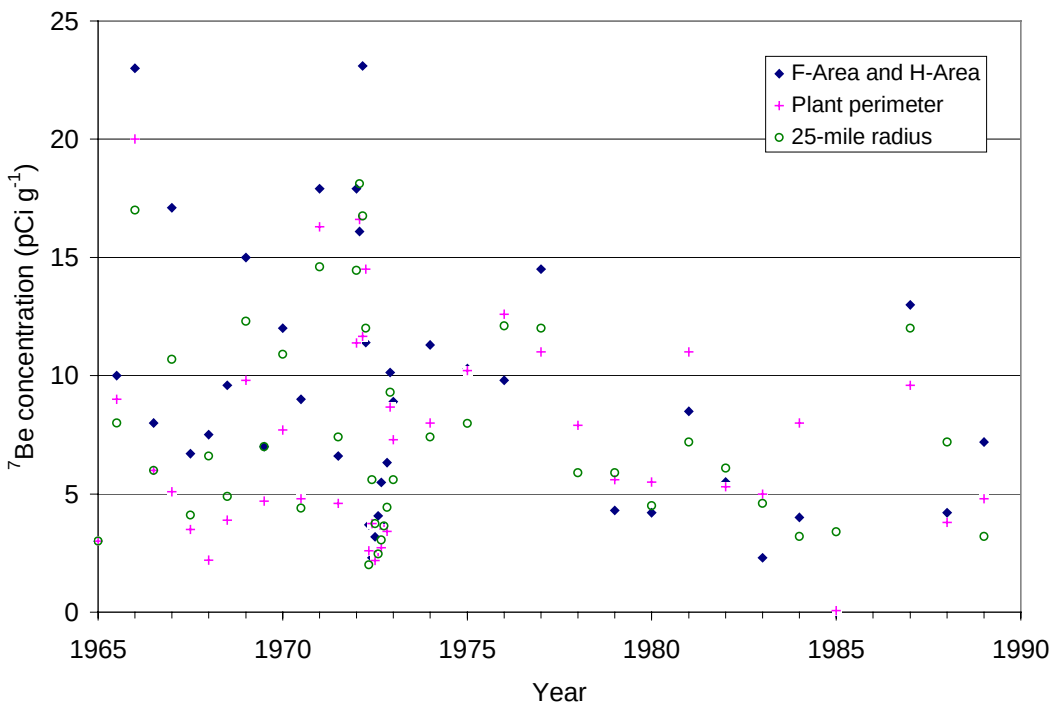

Figure 9-23. Beryllium-7 concentrations measured in vegetation (dry weight) at the F-Area and H-Area, plant perimeter, and 25-mi radius locations. Link to tabulated figure data.

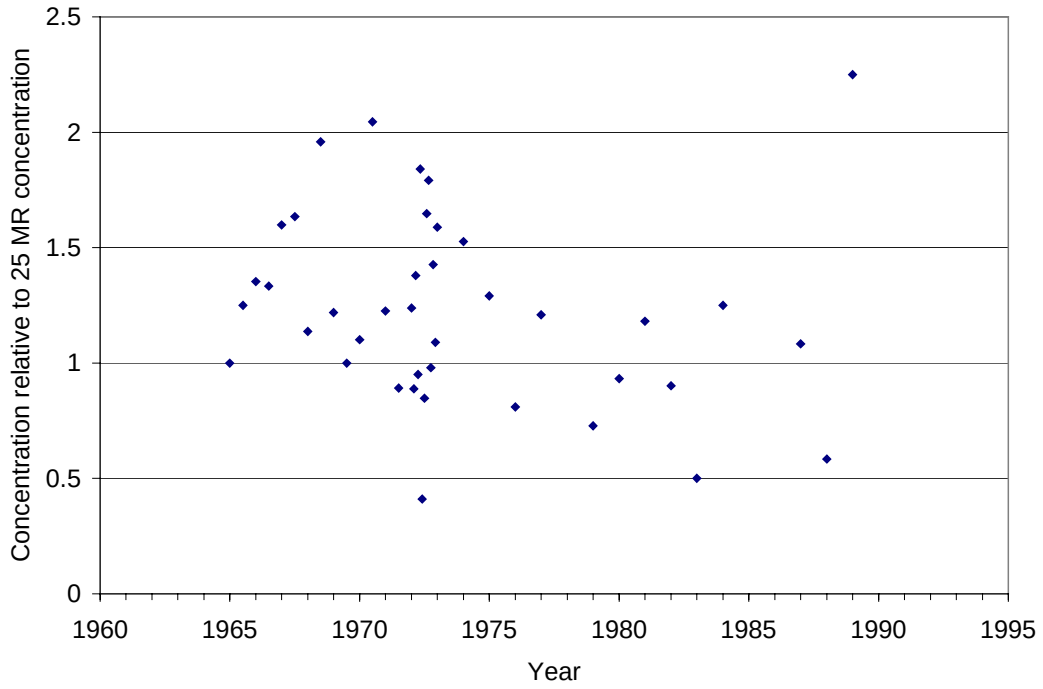

Figure 9-24. Ratio of ^{7}Be concentrations measured in vegetation (dry weight) at the F-Area and H-Area to those measured at 25-mi radius locations. Link to tabulated figure data.

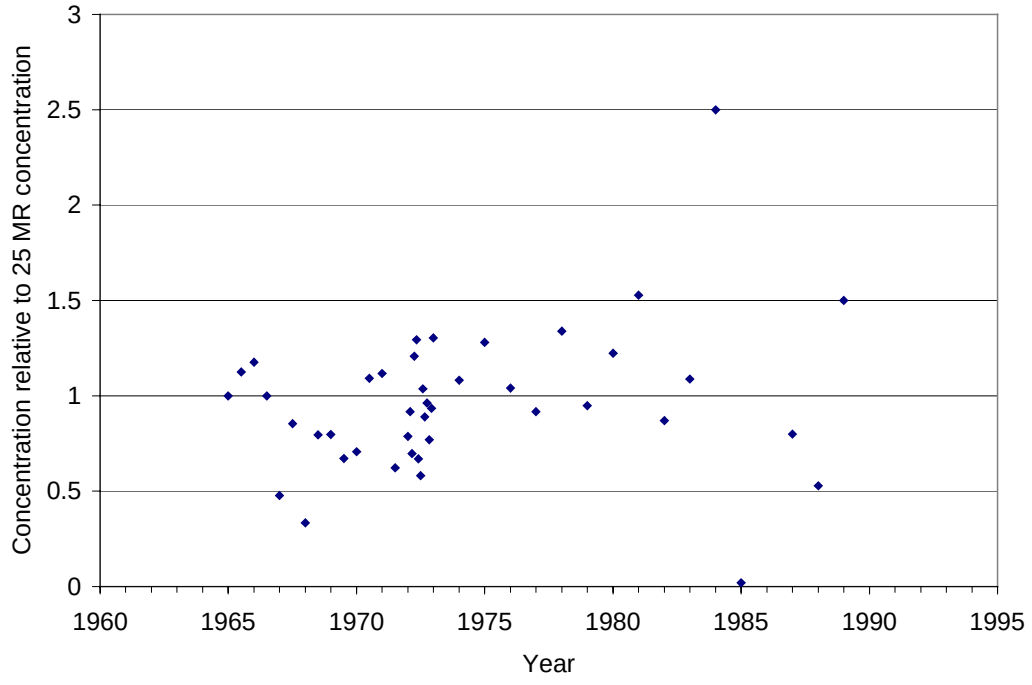

Figure 9-25. Ratio of ^{7}Be concentrations measured in vegetation (dry weight) at the plant perimeter to those measured at 25-mi radius locations. Link to tabulated figure data.

Naturally occurring ^{40}K concentrations have also been reported since 1979. Concentrations of ^{40}K have been similar for the plant perimeter and 25-mi radius locations and higher for the 100-mi radius locations (Figure 9-26). Concentrations also appear to have increased from 1985 through 1989. The variations noted for ^{40}K concentrations are possibly due to natural spatial heterogeneity. However, Horton (1954) attributed higher ^{40}K concentrations at the 25-mi radius locations to the use of commercial fertilizers in this region, which contain naturally occurring ^{40}K. The plant perimeter vegetation, on the other hand, was collected from unfertilized fields. Therefore, at least some of the variation noted for ^{40}K concentrations may be due to differences in fertilizer use.

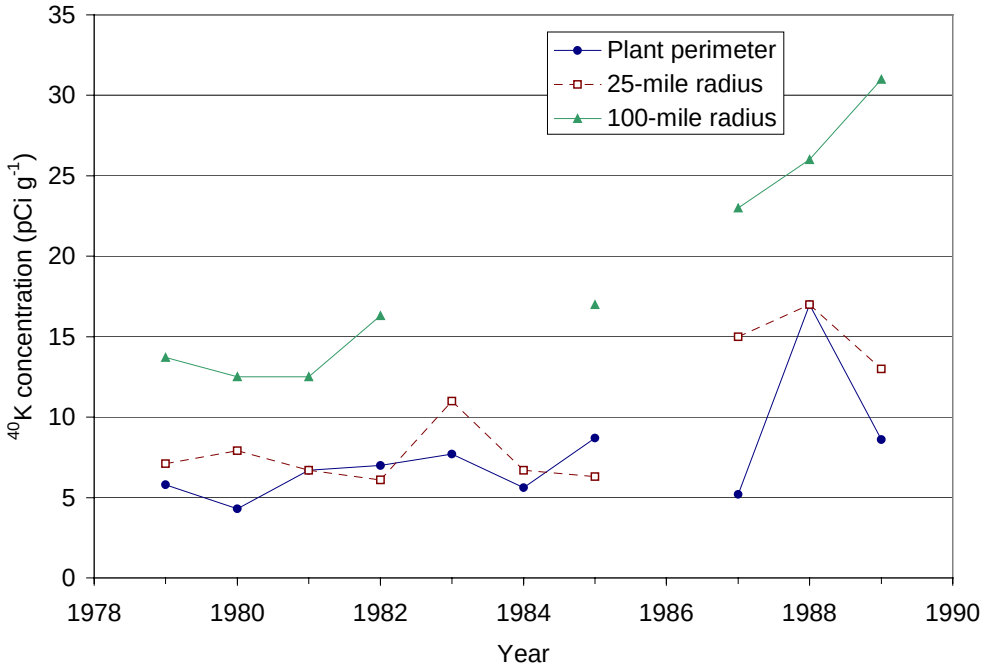

Figure 9-26. Potassium-40 concentrations measured in vegetation (dry weight) at the plant perimeter, 25-mi radius, and 100-mi radius locations. Link to tabulated figure data.

Summary

Close inspection of the gross alpha, nonvolatile beta, and gamma-emitting radionuclide data reveals consistently higher concentrations during the first half of the year for the F-Area and H-Area, plant perimeter, and 25-mi radius locations. This also implies deposition of atmospheric fallout because fallout rates are typically highest during late winter and spring because of accelerated sinking of stratospheric air and generally greater amounts of precipitation during the spring and early summer (Whicker and Schultz 1982).

There is little question that SRS activities have resulted in elevated tritium concentrations in vegetation outside the plant perimeter. The SRS has attributed all other radionuclides detected offsite to atmospheric fallout resulting from weapons testing. However, there is evidence to support elevated concentrations of ^{131}I in 1957 (and likely 1955 and 1956) extending at least to the plant perimeter locations. There is little evidence to suggest elevated concentrations for

radionuclides other than tritium and ^{131}I at the plant perimeter, 25-mi radius, or 100-mi radius locations.

Based on the data we examined, F-Area and H-Area concentrations appear elevated for all radionuclides. However, with the exception of tritium and ^{131}I, SRS activities do not appear to have resulted in elevated concentrations in vegetation at the plant perimeter or offsite locations. Elevated ^{131}I concentrations at the F-Area and H-Area locations are likely the result of releases from the F-Area and H-Area stacks. Elevated gross alpha, nonvolatile beta, and gamma-emitting radionuclide concentrations at the F-Area and H-Area locations, on the other hand, may be partially related to areal variations in atmospheric deposition rates. The concentrations are consistent with seasonal patterns of deposition at all locations, and the same trend of higher F-Area and H-Area concentrations was noted for naturally occurring ^{7}Be. Higher concentrations of fission products (also plutonium and uranium) in general would be expected at the F-Area and H-Area locations because of chronic releases of small amounts of radionuclides associated with the chemical separation processes carried out in these areas. However, it would be difficult to conclude with any confidence that SRS activities have resulted in elevated radionuclide (except tritium and ^{131}I) concentrations in vegetation at any location beyond the SRS plant perimeter.

A further complicating factor in assessing potential SRS contributions to radioactivity detected in vegetation is the fact that the exact type of vegetation sampled is not known with any certainty. Since 1971, the monitoring reports have stated that Bermuda grass has been the primary species sampled. However, little information is provided before 1971 regarding the specific types of vegetation that were sampled. Harvey et al. (1959a) indicated greater sampling of Bermuda and crab grass at the F-Area and H-Area and inner perimeter locations compared to the plant perimeter and 25-mi radius locations. Horton (1954) indicated onsite samples were comprised of trees and weeds while 25-mi radius samples were collected from grain fields. Reinig et al. (1953) and Albenesius (1954) provide nonvolatile beta concentrations for various vegetation species, including oats and oak and pine trees. Mealing and Horton (1957) provides nonvolatile beta concentrations for Bermuda grass and oak and pine trees. The concentrations vary by more than a factor of 2 between the various species.

Table 9-3 lists the calculated ratios based on concentrations for various radionuclides measured at the F-Area and H-Area, plant perimeter, and 100-mi radius locations compared to concentrations measured at 25-mi radius locations. The numbers represent mean and median relative concentrations for all years during which concentrations were reported at both locations. Mean or median ratios close to one suggest little difference from the 25-mi radius location concentrations while mean or median ratios significantly greater than one suggest consistently higher concentrations than those measured at the 25-mi radius locations. The F-Area and H-Area concentrations clearly appear elevated based on both mean and median ratios, and concentrations measured at the plant perimeter and 100-mi radius locations appear quite similar. In most instances, the mean and median ratios at all locations are within about a factor of 2, except F-Area and H-Area ^{131}I concentrations. The fact that the 100-mi radius and plant perimeter locations appear similarly elevated compared to 25-mi radius locations also lends credibility to the conclusion that general variability in areal deposition rates and a variety of sampled plant species may account for the variations noted for different locations. Cesium and ruthenium concentrations do appear slightly more elevated at the plant perimeter locations. However, considering the possible areal variations in deposition and the lack of knowledge regarding species of vegetation that have been sampled, it would be extremely difficult to attribute any plant

perimeter or offsite vegetation radionuclide burdens to SRS operations. Additionally, median values for all radionuclides at the plant perimeter and 100-mi radius locations suggest little difference from 25-mi radius locations.

Table 9-3. Mean and Median Ratios Calculated Based on Comparison of Radionuclide Concentrations at the F-Area and H-Area, Plant Perimeter, and 100-Mi Radius Locations to the 25-Mi Radius Locations

Radionuclide	F-Area and H-Area		Plant perimeter		100-mi radius	
	Mean	Median	Mean	Median	Mean	Median
Gross alpha	1.65	1.33	1.15	1.0	1.42	0.90
Nonvolatile beta	1.37	1.28	1.09	1.0	1.02	0.85
^{137}Cs	2.32	2.0	1.44	1.10	1.06	0.84
^{144}Ce	1.54	1.39	1.19	1.04	1.09	1.0
^{106}Ru	2.62	1.57	2.02	1.04	2.71	1.86
^{95}Zr/Nb	1.70	1.18	1.02	0.97	1.34	1.38
^{131}I	4.38	1.41	1.20	1.0	a	a
^{7}Be	1.23	1.21	0.96	0.94	1.01	0.96

[a]Insufficient data to calculate.

Special Studies

In response to periodic releases of radionuclides from the SRS, particularly tritium releases, several special studies have been initiated to quantify the extent and magnitude of dispersal. These studies have generally involved vegetation sampling at onsite, plant perimeter, and distant locations. The samples have been collected along the projected plume centerline and have assisted in confirming plume trajectory predictions. Table 9-4 summarizes information pertaining to reported releases of tritium since 1963 and resultant vegetation concentrations.

Detailed information regarding the environmental effects of the 1974 and 1975 releases can be found in Marter (1974, cited in Du Pont 1975) and Jacobsen (1976, cited in Du Pont 1976), respectively. Additional tabular information pertaining to episodic releases occurring in 1983–1987 and 1990 is compiled in the annual environmental monitoring reports for those years (Ashley et al. 1984; Zeigler et al. 1985, 1986b, 1987b; Mikol et al. 1988b, and Cummins et al. 1991).

Additional special monitoring of vegetation for tritium was conducted in 1973 and 1974. Samples of pine needles, oak leaves, and grass were collected at 1-mi intervals in north, south, east, and west directions from the H-Area. Concentrations generally decreased as a power function with increasing distance from H-Area for both years.

In January 1965, samples of Spanish moss and Bermuda grass were collected at several onsite locations to examine fallout radionuclide concentrations in each species. Ratios of total activity on moss to grass ranged from approximately 2 to 4, indicating that Spanish moss is a more effective collector of atmospheric fallout than Bermuda grass. This is likely due to a greater surface area to mass ratio for the Spanish moss.

On November 6, 1978, approximately 32 Ci of ^{106}Ru and 1 Ci of ^{103}Ru were released from a fuel separations facility. In response to this release, over 300 environmental samples, including

vegetation, were collected for analysis. Concentrations decreased rapidly with distance from the separations area, and only 1 of approximately 50 samples collected beyond the plant perimeter showed detectable ^{106}Ru activity (20 pCi g^{-1}). The maximum concentration in plant perimeter vegetation was 48 pCi g^{-1}. Both of these samples were collected in a northwest direction from the point of release.

In March 1982, approximately 44 mCi of ^{106}Ru and 3 mCi of ^{103}Ru were released from a fuel separations facility. Vegetation samples collected downwind from the stack indicated no measurable ^{103,106}Ru activity.

Table 9-4. Maximum Tritium Concentrations Measured in Vegetation Following Reported Episodic Releases[a]

Date	Release in curies (form of tritium)[b]	Maximum measured concentration (pCi mL^{-1})		
		Onsite	Plant perimeter	Offsite (miles from SRS)
5/2/74	479,000 (HT)	–[c]	–	–
12/31/75	182,000 (HT)	687	92	9
3/27/81	33,000 (HTO)	–	270	100
3/23/83	4,000 (HTO)	216	–	–
5/17/83	1,100 (HTO)	25	–	–
7/16/83	56,000 (HT)	–	110	80 (25), 27 (50)
8/31/83	2,000 (HTO)	–	2.8	2.3
9/16–19/83	11,500 (HTO)	–	119	17 (25)
11/7/83	150 (HT)			96 (15), 46 (25)
2/16/84	900 (HTO)	1,010	115	103 (25)
3/23/84	7500	95	560	1380 (25), 190 (50)
9/2-3/84	43,800 (HTO)	501	2,500	9859 (25), 240 (77)
1/24/85	9,000 (55% HTO)	1,100	210	70 (25)
1/31/85	9,300 (50% HTO)	7,600	450	380 (25)
3/27/85	19,400 (HTO)	61,890	970	589 (25), 143 (60)
11/4/85	6000 (97% HT)	240	13	–
5/29/86	5900 (95% HTO)	2,600	66	12 (25)
7/31/87	172,000	5,760	4,690	8 (25)
2/7/90	100	–	11	–

[a] Data are from annual environmental monitoring reports.
[b] HT = elemental tritium and HTO = tritium oxide.
[c] – = not reported.

ELECTRONICALLY COMPILED VEGETATION DATA

Vegetation sampling data were reported beginning in 1954 for the inner perimeter, plant perimeter, and 25-mi radius locations. Reporting of data began for the F-Area and H-Area locations in July 1955 and for 100-mi radius locations in 1962. Sampling, and consequently reporting, at inner perimeter locations was discontinued in 1959.

The various data summarized in this chapter are electronically compiled in two Microsoft Excel® workbooks. One workbook (Ch9-Figure_data.xls) contains the figures depicted in this chapter (including the addendum to this chapter) as well as the tabulated data that were used to produce the figures. In this workbook, there is a separate worksheet for each figure and one worksheet contains the tabulated data for all of the figures. The second workbook (Ch9-

All_data.xls) contains the data that have been tabulated from various environmental monitoring reports, aperture card printouts, and raw data sheets. The workbook contains several named worksheets that include brief summary of the compiled data.

Table 9-5 summarizes the data that have been electronically compiled for wild game collected as part of the routine environmental monitoring program maintained by the SRS. It also provides the names of the individual spreadsheets in which these data are compiled (including a brief description of the data). Monthly (and weekly for ^{131}I) data have been compiled from aperture card printouts for 1957 and 1959 through 1973. All other data have been compiled from semiannual and annual environmental monitoring reports.

Table 9-5. Description of Radionuclide Concentration Data Electronically Compiled for Vegetation

Workbook name	Worksheet name	Brief description of data
Ch9-Figure data.xls	Figures 9-1 through 9-26	Each worksheet contains a separate figure depicted in this chapter
	Figures 9A-1 through 9A-4	Each worksheet contains a separate figure depicted in the addendum to this chapter
	Data for figures	This worksheet contains the tabulated data for each of the figures
Ch9-All data.xls	F-Area	Concentrations measured in F-Area vegetation
	H-Area	Concentrations measured in H-Area vegetation
	Outer perimeter	Concentrations measured in vegetation collected from plant perimeter locations
	25-mi radius	Concentrations measured in vegetation collected from 25-mi radius locations
	100-mi radius	Concentrations measured in vegetation collected from 100-mi radius locations
	Inner perimeter	Concentrations measured in vegetation collected from inner perimeter locations
	Nonvolatile beta	Nonvolatile beta concentrations at all locations
	Alpha	Gross alpha concentrations at all locations
	I-131	^{131}I concentrations at all locations
	I-131 (weekly)	Weekly ^{131}I concentrations for 1957 and 1959—1961
	Cs, Ce, Ru, Zr	Cs, Ce, Ru, and Zr/Nb concentrations at all locations
	Be-7	^{7}Be concentrations at all locations
	K-40	^{40}K concentrations at all locations
	H-3 by location	Tritium concentrations at specific plant perimeter and 25-mi radius locations
	Average H-3	Average tritium concentrations at four distances (F-Area and H-Area, plant perimeter, 25-mi radius, and 100-mi radius locations)

RADIONUCLIDE CONCENTRATIONS MEASURED IN AGRICULTURAL PRODUCTS

Collection of local agricultural products began in May 1961. Samples have routinely been collected at 14 locations in the 6 counties surrounding the SRS, with 6 locations situated near the plant perimeter and 8 locations situated approximately 25 mi distant. Food products that have been sampled include collards, plums, peaches, oats, wheat, soybeans, rye, corn, and meat (including chicken and beef).

Food products were prepared as if they were to be eaten. Peelings, seeds, and other nonedible portions were removed. Wheat (containing the whole grains only) and oats (containing both grains and husks) were processed unwashed. In general, the majority of samples were collected during the harvest season.

Only annual average concentrations for all 14 locations were reported in the annual environmental monitoring reports. Since 1970, all radionuclide concentrations (except tritium) were reported as near or below the limits of detection. The annual environmental monitoring reports stated that the concentrations for all radionuclides (except tritium) were indistinguishable from weapons testing fallout. However, based on the vegetation data, there is evidence to suggest that SRS activities resulted in elevated offsite concentrations of radioiodine as well as tritium. Unfortunately, elevated radioiodine concentrations at offsite locations likely would have occurred primarily during years before 1962, and the agricultural product sampling program did not begin until May 1961. The environmental monitoring reports did not report concentrations for radioiodine, presumably because they were below the limits of detection, and aperture card printouts are not available for that time period. Tritium concentrations were reported from 1970 through 1991. In 1964, soybeans were noted to selectively concentrate ^{137}Cs, and collards contained the highest ^{90}Sr concentrations for the majority of years.

The fact that only average concentrations for all 14 locations were reported in the annual monitoring reports precludes any spatial or temporal analyses of these data. Ashley and Zeigler (1978a) stated that there was no significant difference between the plant perimeter and 25-mi distant locations for fallout radionuclides. Based on data provided for vegetation (primarily Bermuda grass), it seems unlikely that any significant differences would have been noted for any of the other years. The exception to this might be radioiodine concentrations during the early 1960s. In general, tritium concentrations were reported to be highest at the plant perimeter locations. This is consistent with data provided for vegetation. Because food products were generally collected during late summer and fall, it is not possible to examine the data for expected seasonal patterns of atmospheric deposition.

Concentrations for specific locations were provided in the aperture card printouts. This would allow for further spatial analysis of the data. However, based on vegetation concentrations, impact beyond the plant perimeter from SRS releases is not evident during the years for which aperture cards have been obtained.

USEFULNESS AND LIMITATIONS OF THE VEGETATION AND AGRICULTURAL PRODUCT DATA FOR DOSE RECONSTRUCTION

There are a number of factors that impact how the vegetation and agricultural product data may be used during subsequent phases of the dose reconstruction project. These factors include the availability of sufficient original monitoring data sets to verify reported summary data and evaluate spatial and temporal trends, as well as the ability to distinguish between Site releases of contaminants and other sources of the same contaminants in the environment (i.e., establish appropriate background concentrations).

Vegetation Data

Semiannual and annual vegetation concentrations in the monitoring reports are consistent with weekly and monthly concentrations reported in the aperture card printouts from 1957 and 1959 through 1973. This provides a good measure of the quality of the reported data and suggests that routinely reported summary data accurately reflect the original, more detailed data.

Concentrations of radionuclides at all plant perimeter and offsite locations have been consistent with periods of reported atmospheric testing and with expected patterns of seasonal deposition. Measured radionuclide concentrations at plant perimeter and offsite locations have generally been indistinguishable, with the exception of ^{131}I and tritium. Based on vegetation concentrations, resultant exposure to the general public from radionuclides other than ^{131}I and tritium appears almost entirely attributable to deposition from weapons testing fallout. Furthermore, it appears that 25-mile radius and 100-mile radius location concentrations are accurate predictors of expected regional background concentrations (i.e., concentrations related to sources other than SRS).

F-Area and H-Area concentrations have been consistently elevated compared to plant perimeter and offsite concentrations for all radionuclides. There is some evidence, based on measured concentrations of naturally occurring ^{7}Be, that these elevated concentrations may partially be the result of areal variations in deposition rates. However, elevated ^{131}I and tritium concentrations at the F-Area and H-Area locations are likely primarily the result of releases from the F-Area and H-Area stacks. The fact that concentrations have generally been reported for composite samples at each distance precludes further spatial analysis of the data to quantitatively determine what impact areal differences in deposition may have on higher onsite concentrations.

There are no apparent spatial trends for tritium, and reported concentrations have been similar in all directions and at all locations for each distance. Concentrations appear to decrease as a power function (i.e., concentrations decrease exponentially as distance increases exponentially) with increasing distance from the SRS. This may be useful for determining vegetation concentrations, as well as verifying air concentrations and deposition rates, at any given distance from the plant. The usefulness is limited, though, by the fact that only annual average concentrations are available for tritium.

The monitoring reports typically state that tritium is the only radionuclide of SRS-origin to be detected in offsite vegetation. However, based on the information in the documents we reviewed regarding vegetation concentrations, there is also evidence to suggest that SRS operations have resulted in offsite deposition of ^{131}I, particularly during the late 1950s and early 1960s. Measured concentrations of ^{131}I in vegetation indicate offsite deposition during periods of

high releases in the first half of 1957. It is likely that offsite deposition also occurred in 1955 and 1956 based on the magnitude of Site releases during those years, but data availability for those years (i.e., only monthly and semiannual averages) and the relatively short half-life of ^{131}I limits the comparisons that can be made. To more closely examine temporal trends and potential SRS contributions to ^{131}I concentrations in offsite vegetation during 1955 and 1956, monthly and, if possible, weekly measured concentrations would be necessary. Unfortunately, these data do not appear to be available. In addition, results were composited for each distance, which limits the usefulness of the data with respect to source term verification and model validation and may hinder quantifying elevated plant perimeter concentrations at specific locations.

It is assumed that results have been reported for Bermuda grass since 1971, but various species appear to have been sampled before then, particularly during the 1950s. Depending on the extent to which these data are used for future model validation, it may be necessary to make some assumptions and generalizations about vegetation species to select appropriate radionuclide uptake and concentration factors.

In summary, the information provided in the documents we reviewed is quite useful for quantifying radionuclide content in vegetation at the plant perimeter, 25-mi radius, and 100-mi radius locations. The information is particularly useful for establishing the primary source of atmospherically deposited radionuclides and may eventually be useful to some extent for source term and model validation. However, the fact that samples were composited and only average concentrations at each distance were reported during the major iodine releases limits the amount of spatial analyses that can be done. Tritium concentrations are available for individual locations at each distance, but only annual average concentrations were routinely reported. For some episodic tritium releases, more detailed information is available in the annual environmental monitoring reports that may be useful for source term verification and model validation (see Special Studies section).

Agricultural Product Data

The usefulness of the agricultural product data for dose reconstruction is limited based on the available data. The fact that only average concentrations for all locations have been reported in the annual environmental monitoring reports precludes spatial analyses of the data. Concentrations for individual locations are available in the aperture card printouts. However, based on vegetation data, it seems likely that the majority of radionuclides detected in food products (with the exception of tritium and ^{131}I) have resulted from atmospheric fallout and that SRS contributions have been minimal. Further analyses will depend on the identification of data reported for individual locations during the years for which specific episodic releases were important beyond the plant perimeter. At this point, food product data have not been compiled, and future compilation of the data will depend on the availability of useful data collected during identified episodic atmospheric releases.

There is little evidence to suggest that the patterns of deposition for agricultural products would be different than those for vegetation. Concentrations could vary between species because of differences in their ability to concentrate particular radionuclides. For example, collards appear to selectively concentrate ^{90}Sr to a greater extent than other vegetable products. Further analyses should be focused on tritium and ^{131}I concentrations or on available information during identified episodic releases.

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ADDENDUM 9A

TRITIUM CONCENTRATIONS IN VEGETATION AT PLANT PERIMETER LOCATIONS

DRAFT

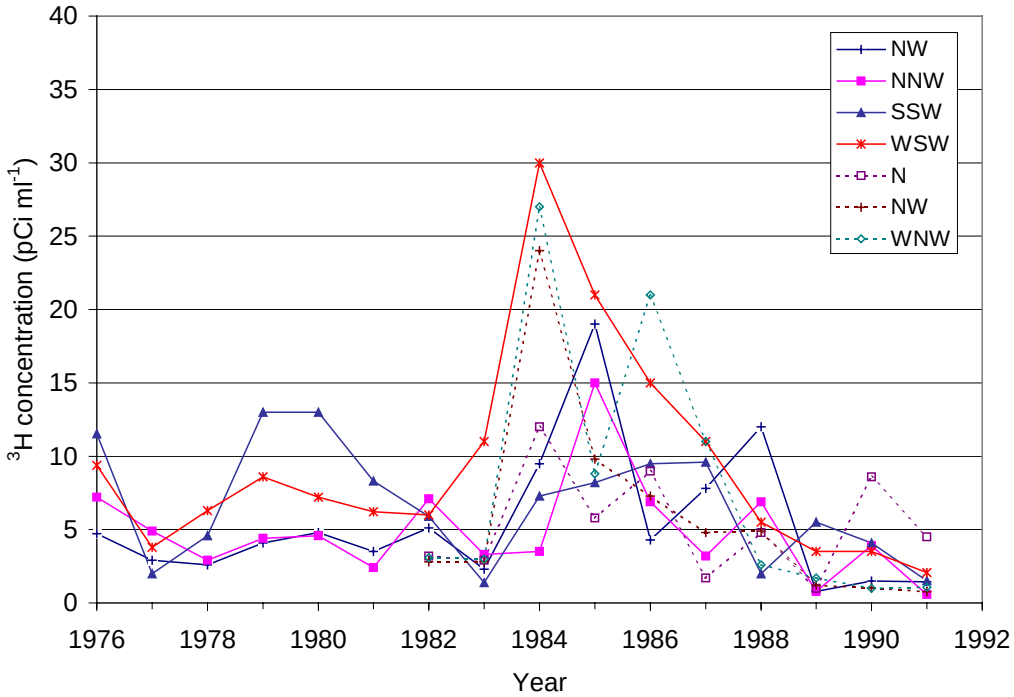

Figure 9A-1. Tritium concentrations in vegetation at individual plant perimeter locations since 1976. See Table 9-1 for location names. Link to tabulated figure data.

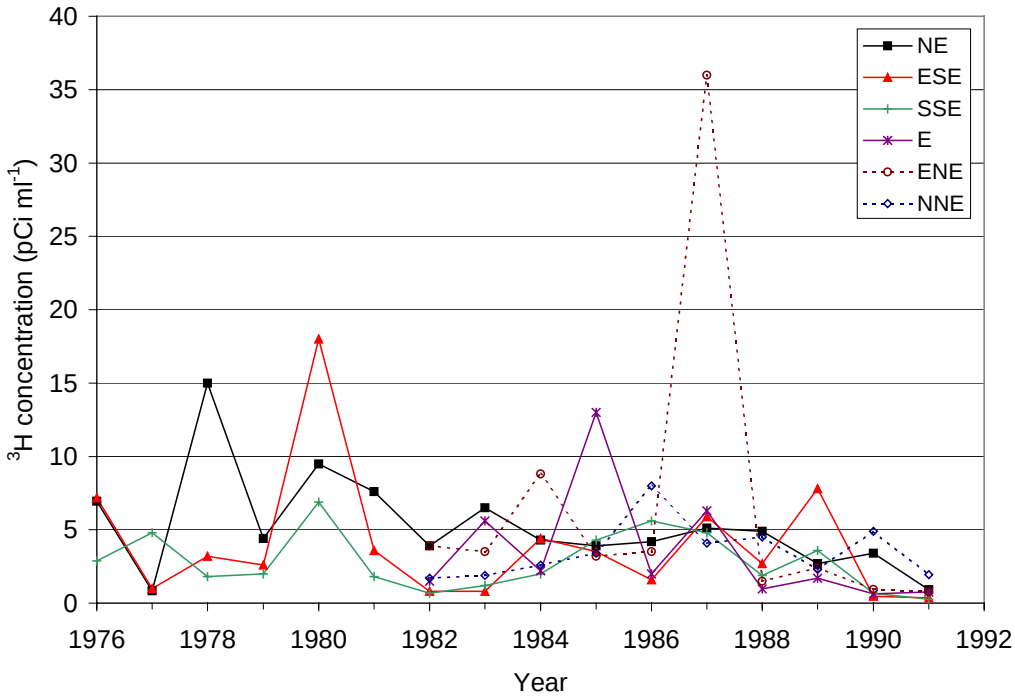

Figure 9A-2. Tritium concentrations in vegetation at individual plant perimeter locations since 1976. See Table 9-1 for location names. Link to tabulated figure data.

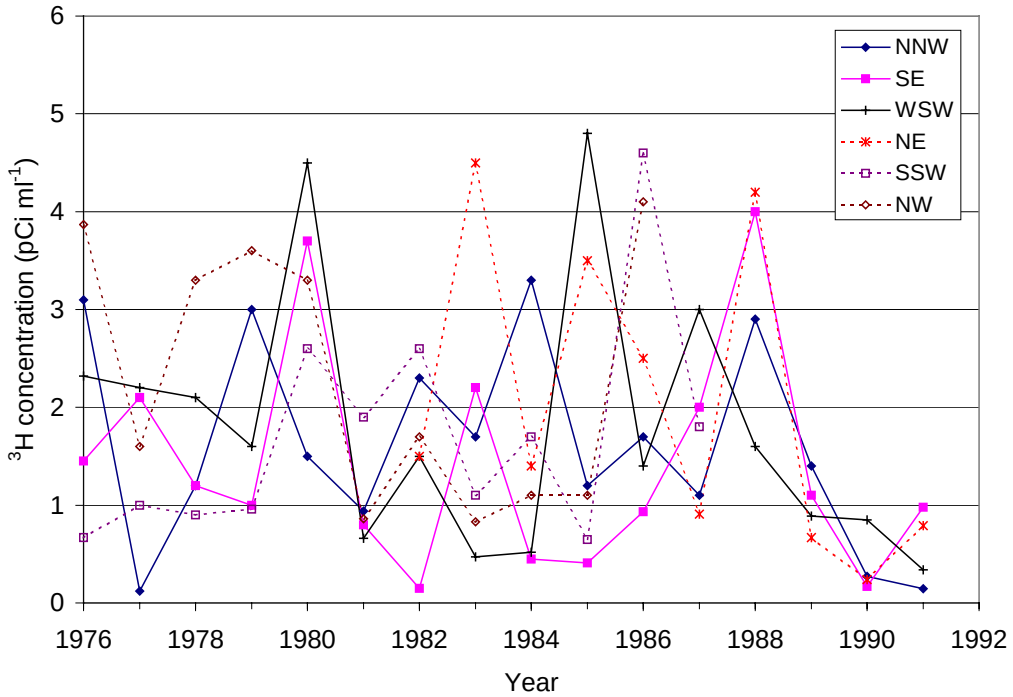

Figure 9A-3. Tritium concentrations in vegetation at individual 25-mi radius locations since 1976. See Table 9-1 for location names. Link to tabulated figure data.

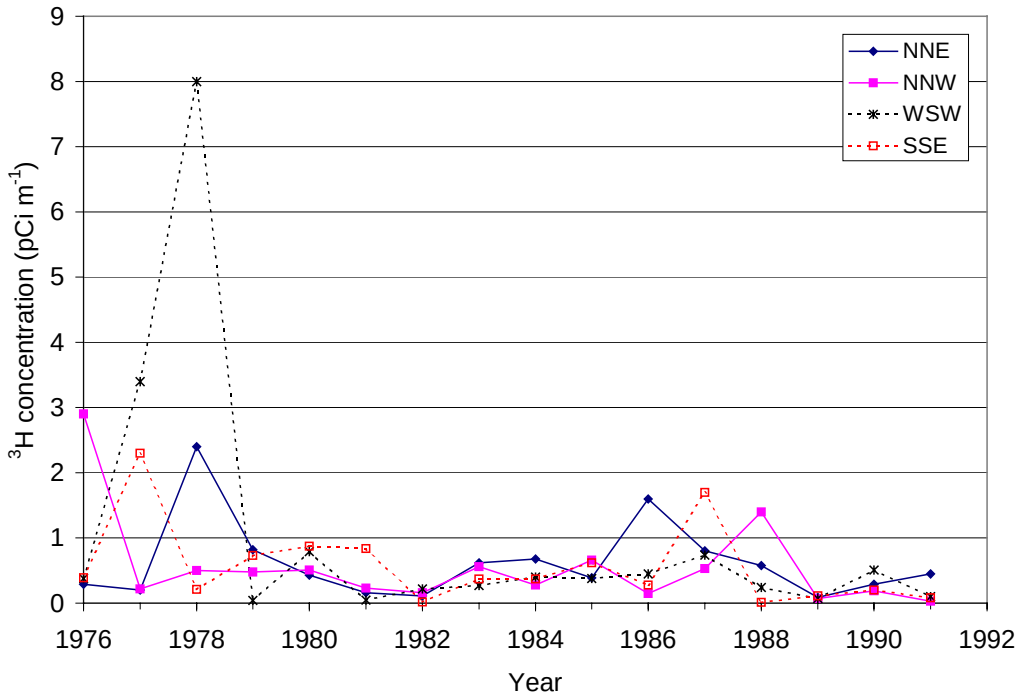

Figure 9A-4. Tritium concentrations in vegetation at individual 100-mi radius locations since 1976. See Table 9-1 for location names. Link to tabulated figure data.

CHAPTER 10

RADIONUCLIDES IN MILK

ABSTRACT

In this chapter we discuss radionuclide concentrations measured in milk samples collected in the vicinity of the Savannah River Site (SRS). The routine milk sampling data for ^{131}I and ^{90}Sr are potentially the most useful for dose reconstruction. This chapter compiles these data in an electronic form and discusses them in detail. Tritium and ^{137}Cs were also analyzed routinely at the SRS. However, we did not compile these data because many of the samples contained concentrations below the minimum detection limit. Furthermore, the tritium measurement techniques before 1971 had poor sensitivity.

We attempted to verify the data by comparing the reported concentrations from as many sources as possible. The primary source of the data is original handwritten compilations of effluent and environmental monitoring results. In general, we found these original data to be in good agreement with the data reported in monthly, quarterly, semiannual, and annual summary reports generated by SRS.

We also compiled milk monitoring data generated by the Public Health Service (PHS) (which later became the U.S. Environmental Protection Agency) for the three monitoring locations closest to the SRS. These data are very useful because they provided an independent source of information about the concentrations of atmospheric radionuclides in milk in the southeastern U.S. A comparison of the SRS and PHS data suggests that, in general, fallout from atmospheric weapons testing is the probable source of the radionuclide concentrations measured in milk around the SRS. However, there is evidence that SRS activities have contributed to offsite contamination in a few instances. Therefore, these data may be useful for source term verification and environmental transport model validation. These data also allow us to quantify the magnitude of exposures from consumption of contaminated milk. Appendix K further discusses potential uses for environmental monitoring data in dose reconstruction.

INTRODUCTION

Milk can be an important human exposure pathway for environmental releases of radionuclides, in particular, radioiodine. Following an atmospheric release, radionuclides may be deposited onto forage that is ingested by cows. A portion of the ingested radioactivity is secreted into the milk, which can be consumed by humans. Other mechanisms, such as irrigation or flooding of the pasture with contaminated water and root uptake from contaminated soil, may also contaminate forage. Milk monitoring data that are of high quality and above detection limits can be very useful for a dose reconstruction. They may allow the doses from this exposure pathway to be quantified directly or they may be used to test the performance of the environmental transport models used in the dose reconstruction or to quantify individual parameter values in the models.

Three different categories of milk samples were routinely collected and analyzed by the SRS:

1. Milk from a farm cow (individual family cow)
2. Milk from a local dairy
3. Milk collected from local dairies and sold by a major distributor.

Originally, the farms and dairies that were sampled were located within a 50-mile radius of the Site. Around early 1962, the sampling locations were selected to be within a 25-mile radius of the Site. The number of sampling locations and frequency of sampling varied over time according to the findings, the availability of suitable monitoring locations, and resources. Figure 10-1 shows the various milk sampling locations that have been used by the SRS for the radiological monitoring of milk. Table 10-1 summarizes the routine radiological monitoring for milk.

Radiological monitoring of milk was not included in the preoperational background survey (Reinig et al. 1953). That survey was designed to establish background levels of naturally occurring radionuclides before plant startup.

Table 10-1. Summary of Routine SRS Radiological Monitoring for Milk

Year	Radioiodine	Sr-90	Tritium	Cs-137
1953–55	—	—	—	—
1956	wk: July onward	—	—	—
1957	wk	—	—	—
1958	wk	qtr: Fall onward	—	—
1959	wk	qtr	—	—
1960	wk	qtr	wk: July onward	—
1961	wk	qtr	wk	qtr: Fall onward
1962	wk	mo[a]	wk	qtr
1963	wk	qtr	wk	qtr
1964	wk	qtr	wk	qtr
1965	wk	qtr	wk	qtr
1966	2wk:April onward	qtr	2wk	qtr
1967	2wk	qtr	2wk	qtr
1968	2wk	qtr	2wk	qtr
1969	2wk	qtr	2wk	qtr
1970	2wk	qtr	2wk	qtr
1971	2wk	qtr	2wk	qtr
1972–80	2wk	qtr	2wk	qtr
1981–86	2wk	qtr	2wk	mo
1987–90	2wk	qtr	2wk	2wk

wk=every week, 2wk=every 2 weeks, mo=monthly, qtr= every quarter (3 months)
[a] to ascertain arrival and amount of ^{90}Sr from recent atmospheric nuclear weapons tests in local milk

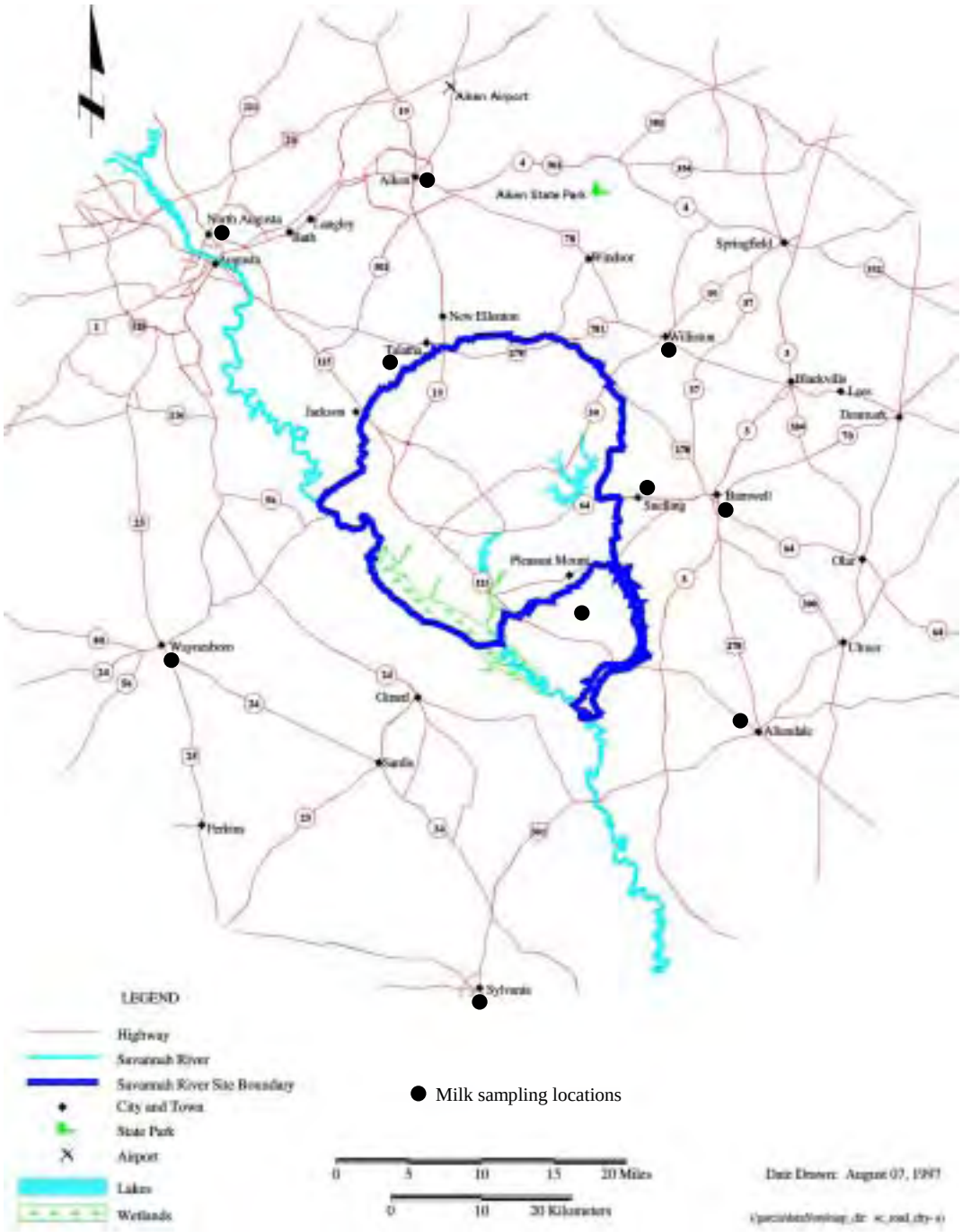

Figure 10-1. Milk sampling locations used by the Savannah River Site.

MEASUREMENTS OF BETA-GAMMA-EMITTERS

The beta-gamma-emitters that were routinely monitored by SRS were radioiodine, strontium, tritium, and ^{137}Cs (Table 10-1). The earliest sampling of milk is reported in the Control Weekly Report dated June 18–22, 1956 (Du Pont 1956a), which indicates that SRS was experimenting with methods for analyzing iodine and strontium. Routine sampling of milk began toward the end of 1956 for ^{131}I, in the fall of 1958 for ^{90}Sr, in July 1960 for tritium, and in September 1961 for ^{137}Cs.

In general, samples were analyzed weekly and later (beginning April 1966) every 2 weeks (biweekly) for radioiodine and tritium. Milk samples were analyzed quarterly for ^{90}Sr except during 1962 when the monitoring was increased to monthly to ascertain when and to what extent ^{90}Sr from recent atmospheric nuclear tests would appear in local milk (Du Pont 1962a). Milk samples were analyzed quarterly for ^{137}Cs until 1981 when the sample frequency was increased to monthly. This continued until 1987 when the sample frequency was increased to biweekly. Details of the analytical and counting procedures are presented in Appendix A.

We compiled the milk monitoring results for ^{131}I and ^{90}Sr electronically in a Microsoft Excel© workbook (filename: Ch10-All_data.xls). These data are described in detail in the following sections. The tritium measurements have not been compiled because many of the milk sample concentrations were below the minimum detection limit (4000 pCi L^{-1}). The sensitivity of the measurement techniques improved by an order of magnitude in 1971 allowing more reliable tritium concentrations in milk to be determined. Many of the ^{137}Cs measurements in milk samples were also below the sensitivity of the analysis and have not been compiled for this reason. The tritium and ^{137}Cs data are readily available for more detailed examination if necessary.

The data used to plot the figures that are presented later in this chapter are extracted from the Microsoft Excel© workbook Ch10alldata.xls that combines all the data from a separate Microsoft Excel© workbook (filename: Ch10-Figure_data.xls).

Radioiodine

A summary of the source and number of milk samples (e.g. farm cow, local dairy) that were routinely collected as part of the SRS milk-monitoring program is presented in Table 10-2. Details about the frequency of sampling these locations are given in Table 10-1. The SRS environmental monitoring program measurements for analyzing milk samples for radioiodine have been compiled electronically in a Microsoft Excel© workbook (filename: Ch10-All_data.xls) that has a separate spreadsheet for each year of data. The spreadsheets are named according to the year for which the data have been compiled, using the following format, I-131_19xx, where xx represents the last two digits of the year. For example, I-131_1960 contains the ^{131}I measurements in milk for 1960. The data are compiled for 1956 through 1970. As explained below, the data for 1956 through July 1959 are sparse and incomplete.

The primary source of these data is handwritten compilations of effluent and environmental monitoring results on ledger sheets. At some point in time, each sheet was photographed by the Site in a format similar to microfiche and mounted on a computer card with a hole cut in the center. These cards, called aperture cards, were found for August 1959 through December 1973 for ^{131}I concentrations in milk. The aperture card data were cross-checked with the data in the weekly, monthly, quarterly, semiannual, and annual environmental monitoring reports. This was accomplished by entering the average concentrations reported for a specific time period

separately in the Microsoft Excel© workbook and comparing these values with those obtained by summing the relevant data from the aperture cards. In general, we observed good agreement between the aperture card and report data. A note is made in the workbook where there is disagreement. For the time period before August 1959, the data are compiled from the weekly and semiannual monitoring reports. These data are incomplete. From 1974 onward, data were compiled only for those time periods when positive monitoring results were reported. These data are obtained from the monthly reports (if available) and the annual monitoring reports. See Chapter 7 for a summary of the different sources of environmental monitoring data.

Table 10-2. Summary of Routine Monitoring Sources for I-131 in Milk

Year	Farm cows	Local diaries	Milk distributor
1953–55	—	—	—
1956	3	—	—
1957	2	—	—
1958	2	2	—
1959	2	3	—
1960	2	3	—
1961	2	3	—
1962	4	6	1
1963	4	6	1
1964	2	4	1
1965	—	4	1
1966	—	4	1
1967	—	3	1
1968	—	3	1
1969	—	3	1
1970	—	3	1
1971–90	—*	4–7	1

* milk from farm cows was monitored in 1976, 1977, 1978, 1986 when measurable ^{131}I concentrations were detected in milk from dairies. Increases were attributed to atmospheric fallout from Chinese bomb tests and Chernobyl reactor accident.

The compiled ^{131}I data are organized in the following way in the Microsoft Excel© workbook (filename: Ch10-All_data.xls). In the top left hand corner of each spreadsheet the following information is recorded: the year that the sample was collected and analyzed, the type of sample analyzed (milk), the radionuclide analyzed (^{131}I), and the units in which the measurements are reported (pCi L^{-1}).

The source of the data used to compile the monitoring measurements is presented at the top of the spreadsheets by reference to its SRS Phase II Database number together with an indication of the type of document (e.g., aperture card, annual report, or weekly report).

A limited amount of color coding is also used in the spreadsheets, with a key provided at the top of each spreadsheet. For measurements that are reported as less than values, the cell is highlighted in pale blue. The value is entered into the cell.

A cell is highlighted in yellow if there is a discrepancy between documented sources of the value. This situation tends to occur when the reported average concentration for a specified time period (month, 6 months) is different than that calculated from the reported individual measurements.

A red dot placed in the upper right hand corner of a spreadsheet cell indicates that a note is attached to that cell. The note contains additional information about that measurement.

The first column of data reports the sample date. The subsequent columns report the measured values by sample location. The sample locations are arranged by sample type (e.g., farm cow, local dairy, or major distributor).

Additional columns or rows are inserted into the spreadsheets to cross-check the reported data if sufficient information is available. For example, the average concentration for a sample type that is reported for a specific time period is compared with the value obtained by summing the relevant data from the aperture cards. Similarly, the average monthly or 6-month concentrations that are calculated from the individual measurements are compared with those reported in the monthly, semiannual, or annual reports.

A general overview of ^{131}I concentrations in milk can be gained from Figure 10-2. Measurements for ^{131}I concentrations in milk from farm cows at two locations, and milk from dairies at two different locations are presented. To maintain the legibility of the figure not all of the available data are plotted, however, the plotted data are representative of the measurements in general.

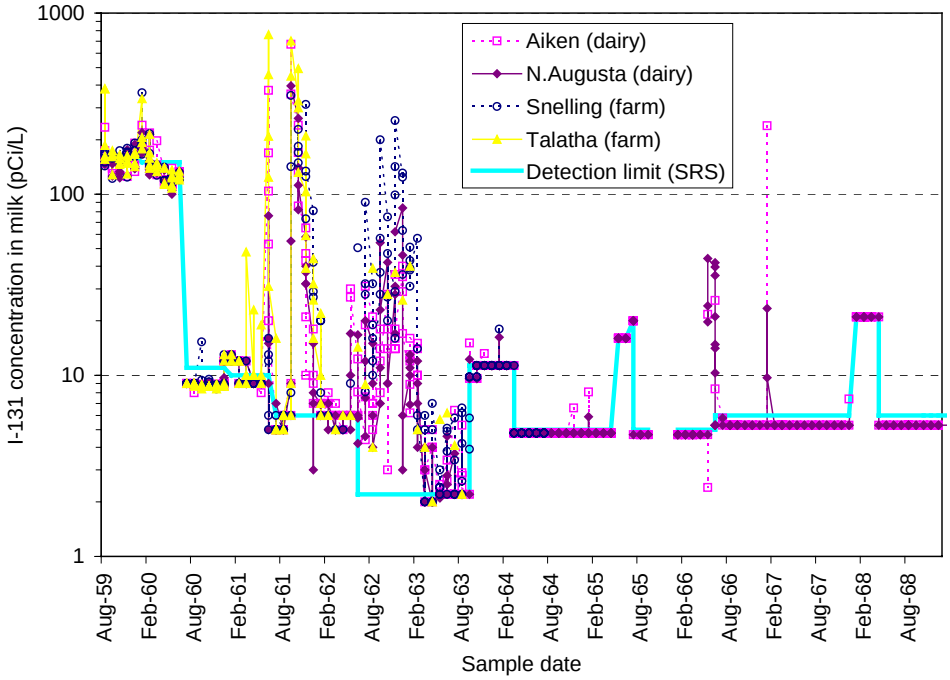

Figure 10-2. Overview of the temporal variation in ^{131}I concentrations measured in milk from farm cows and dairy cows in the vicinity of SRS. Link to tabulated figure data.

Description and History of the Milk Monitoring Program for Radioiodine

According to Kantelo et al. (1993), the SRS began local milk analyses for ^{131}I in 1957. Based on the weekly report dated June 18–22, 1956, (Du Pont 1956a) it appears that SRS was experimenting with methods for analyzing iodine and strontium in the first half of 1956. The earliest recorded measurements of radioiodine concentrations in weekly milk samples are reported in the August 6–10, 1956, weekly report (Du Pont 1956b) for three consecutive weeks commencing July 23, 1956. The milk was collected from three separate locations: Talatha Gate, Snelling, and Allendale. The weekly reports indicate that routine weekly samples of milk were collected from September 17, 1956, onward (Du Pont 1956c). The results are presented as the average and maximum concentration for all sample locations. Consequently, it is not possible to determine the concentration measured at each location. However, in some cases the location with the maximum concentration is noted. Furthermore, the number of locations sampled varied. The absence of reported concentrations from late October to the end of the year suggests they were below the sensitivity of the analysis (200 pCi L^{-1}) rather than not sampled.

In the January through June 1957 semiannual report (Horton and Mealing 1957), no mention is made of milk monitoring, but a detection limit for radioiodine in milk is listed as 120 pCi L^{-1} (Appendix A; Table A-5). In the subsequent semiannual report (Horton et al. 1958), the detection limit for radioiodine in milk is not listed and there is no mention of milk monitoring. Despite this, the monthly reports do provide results of milk monitoring for radioiodine. Weekly milk samples appear to have been collected routinely from Talatha and Snelling. The results presented in the weekly and monthly reports have been compiled and compared in the Microsoft Excel© workbook. The values given in the monthly reports generally agree with the weekly values. The monthly values for 1957 include the last week of the previous month and do not include the last week of the current month. It is a little more difficult to reconcile the values for the first 6 months of 1957. Gaps in the data may indicate that the measured concentrations were below the sensitivity of the analysis.

It is difficult to accurately reconstruct the measured concentrations of radioiodine in milk for 1958 based on the two sources of available information: weekly and semiannual reports. The monthly reports provide no information on this topic. The weekly reports for 1958 indicate that two routine milk samples were collected weekly from Talatha and Snelling until April when only one sample was collected (from Talatha). This was restored to two weekly samples at the end of May. The first positive analysis for radioiodine was reported on April 21, 1958, (170 pCi L^{-1}) for milk samples from Talatha (Du Pont 1958). This is consistent with the January through June 1958 semiannual report (Harvey et al. 1959a), which states that the maximum concentration observed was 270 pCi L^{-1} at Snelling on January 20. The semiannual report also states that 55 milk samples were collected during the first 6 months, of which four were positive samples, with an average concentration of 190 pCi L^{-1}. The semiannual report states that the samples were taken from four locations (Talatha, Snelling, Aiken, and N. Augusta) rather than two. The weekly reports indicate that from April 21 onward, milk samples were also collected on weekly for a special study and that generally two or fewer locations were sampled. This suggests that the radioiodine results from Aiken and N. Augusta were included in the monitoring results presented in the semiannual reports. The July through December 1958 semiannual report (Harvey et al. 1959b) states that an additional sample location (Langley) was added to the program after November 10. The report goes on to state that 107 milk samples were analyzed for radioiodine

and that 24 contained detectable concentrations. The average concentration of the 24 samples was 270 pCi L^{-1}, and the maximum concentration of 1100 pCi L^{-1} was in a sample collected from Talatha on July 21. All detectable concentrations (100 ±6 pCi L^{-1} detection limit) occurred in samples collected during July 14 to August 18 and October 6 to November 18, periods during and immediately after weapons tests. See Chapter 6 for more details regarding other sources of radionuclide contamination.

Throughout 1959, 1960, and 1961, milk samples were collected weekly from the same five locations (Talatha, Snelling, Aiken, N. Augusta, and Langley). These consisted of three dairies located in Aiken (Holley's), Langley (Key's), and North Augusta (Haskell's) and two farm cows at farms located in Talatha and Snelling. From July 1961 onward, a fourth dairy (Simmond's) in Pleasant Mount was also sampled routinely.

According to the 1959 and 1960 semiannual reports (Du Pont 1959a, 1960a, 1960b, 1961a), the average concentration of ^{131}I in the milk samples (collected weekly) during 1959 and 1960 was below the reported 6-month average detection limits. The detection limit decreased from 170 to 160 pCi L^{-1} for the second 6-month period and decreased from 150 to 9 pCi L^{-1} in July 1960. These summary data agree with the handwritten values reported by location on aperture cards.

Radioiodine was first detected (19 pCi L^{-1}) in a milk sample collected from Talatha, South Carolina, on May 10, 1961. This increase in concentration was attributed to increased releases of ^{131}I from the 292-F stack because processing of shorter cooled uranium had commenced on April 28, 1961 (Du Pont 1961b). This value is not reported in the quarterly or semiannual reports that cover this time period. Instead, these reports indicate that no routine milk sample was collected on the following day, May 11, 1961, from Talatha, South Carolina.

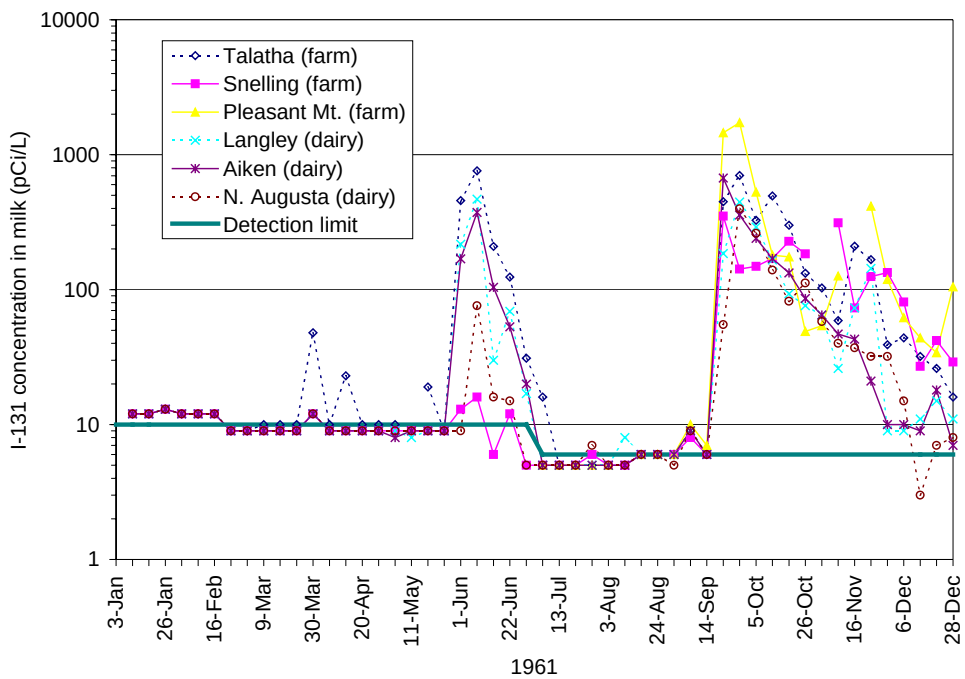

Figure 10-3. Comparison of ^{131}I concentrations in milk samples taken from farm cows and local dairies in the vicinity of the SRS weekly throughout 1961. Link to tabulated figure data.

Figure 10-3 plots ^{131}I concentrations measured in weekly milk samples collected throughout 1961 from both individual farm cows and local dairies. The figure indicates an earlier increase in ^{131}I concentrations in milk from Talatha, South Carolina, at the end of March and early April. However, this may not be real because these concentrations were reported as less than values on the aperture cards, and they are not discussed in any reports. Concentrations of ^{131}I in milk sampled weekly increased dramatically at most locations throughout June (Figure 10-3). This was associated with a substantial increase in the daily rate of release of ^{131}I from the 291-F stack beginning May 29 (Du Pont 1961c). Releases continued through June 23, with the largest releases occurring during the first week of these releases (Du Pont 1961c). In response, the Site conducted extensive milk sampling in the vicinity of SRS starting on June 5. One hundred and twenty-two milk samples were collected from 48 individual farms and dairies in this special study. The maximum concentration of ^{131}I in milk (5451 pCi L^{-1}) was reported for a milk sample obtained from a farm 1 mile west of New Ellenton. Unfortunately, apart from this single value, the results of the individual sample analyses are not reported in any of the documentation. The documentation presents only the percentage of milk samples in the various concentration ranges, with the exception of a farm cow located at Pleasant Mount. These daily measurements are compiled separately on the spreadsheet I-131_1961 and plotted in Figure 10-4. Throughout July and August, ^{131}I concentrations in milk decreased to at or below the detection limit before increasing again during the last 2 weeks in September (Figure 10-3). These increases were attributed to fallout from Russian detonations that began September 1, 1961 (Du Pont 1961d). Detectable concentrations of ^{131}I were measured in milk samples through the end of the year. The minimum detection limit for ^{131}I in milk decreased from 11 pCi L^{-1} to 6 pCi L^{-1} in July 1961 (Figure 10-3).

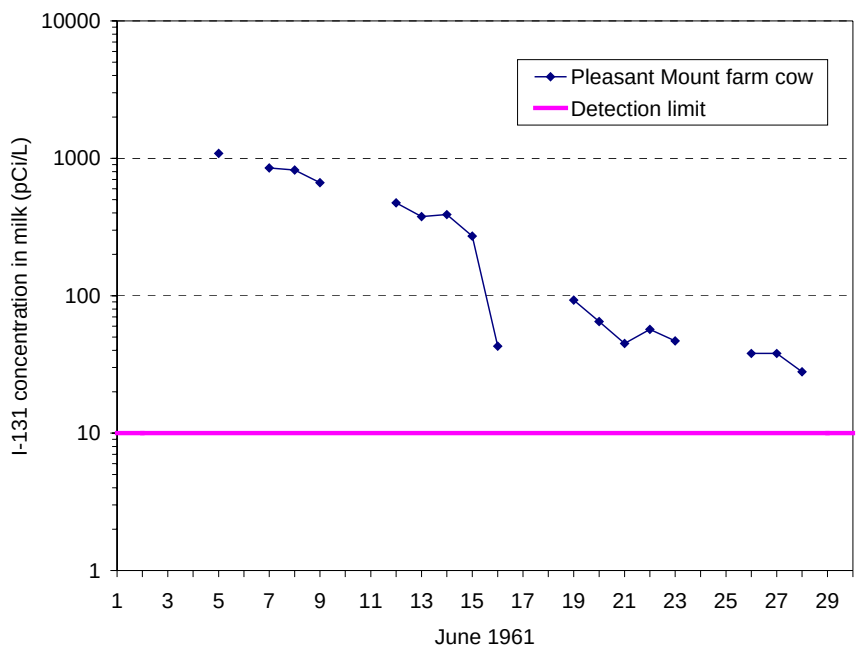

Figure 10-4. Daily ^{131}I concentrations in milk sampled from a farm cow located at Pleasant Mount, South Carolina, in a special study following increased releases from 291-F stack. Link to tabulated figure data.

Sample collection from the small dairy in Langley, South Carolina, was discontinued in January 1962. Barnwell and Allendale were added on January 25, 1962 (Du Pont 1962b). Although the frequency of milk sampling for radioiodine was reported as weekly, there were exceptions to this. In January 1962, milk was sampled weekly from three sites with farm cows and monthly from five dairies and one milk distributor. From February through June 1962, the sampling schedule changed somewhat and milk was collected just monthly from the same three sites with farm cows and weekly from the five dairies and one milk distributor. During this time period, sampling of the Sylvania, Georgia, farm cow was discontinued. However, it appears that a replacement farm cow was identified in Waynesboro, Georgia, and sampling began again in July. The detection of ^{131}I in milk samples in May and June 1962 was attributed to fallout from U.S. nuclear tests (Du Pont 1963a).

From July 1962 until late in 1963, of the four locations with farm cows, two were sampled weekly and two monthly. The six dairies and one distributor were all sampled weekly. In late 1963, sampling was discontinued at two farm cow sites (Talatha and Pleasant Mount) and two local dairies (Barnwell and Williston), presumably because previous milk samples consistently contained concentrations below the minimum detection limit at that time (2.2 pCi L^{-1}). Sampling of the two farm cows at Snelling, South Carolina, and Waynesboro, Georgia, continued weekly until August 1964, when sampling at those locations was discontinued. Weekly sampling of the four dairies and one distributor continued throughout 1964. In August 1963, the sample size was reduced from 1 to ½-gal for all locations because fallout radioiodine was no longer detectable in milk (Johnson 1963).

Du Pont (1963b) reports the sampling schedule as of 7/15/63. This report indicates that the weekly milk samples were collected on Thursdays in the first 4 weeks of every month, and that the monthly samples were collected on the Thursday of the second week of every month. These were 1-gal samples with the exception of Borden (distributor), from which a 2-gallon sample was obtained. There is no explanation why the minimum detection limit reported for the analysis of radioiodine in Borden milk samples is 12 pCi L^{-1} versus 6 pCi L^{-1} for other samples.

In August 1964, the number of locations where milk samples were collected weekly was reduced to four (Aiken, N. Augusta, Waynesboro, and Allendale) (Johnson 1964). Slight increases in ^{131}I concentrations in milk samples collected between October 29, 1964, and November 11, 1964, were attributed to fallout from a Chinese nuclear weapons test (Du Pont 1965a). The monthly report for December 1964 (Du Pont 1964) states that 350 mCi of radioiodine was released from two sources in the Savannah River Laboratory during the period December 7–21, but that milk samples from Jackson and Talatha contained no measurable ^{131}I.

Milk from four dairies (Aiken, N. Augusta, Waynesboro, and Allendale) was sampled weekly during 1965. The ^{131}I concentration was reported to be less than the sensitivity of the analysis (5 pCi L^{-1}) throughout the entire year (Du Pont 1965b, 1966a).

During the first quarter of 1966, (Du Pont 1966b) no radioiodine was detected in milk samples despite reported iodine releases from F-Area during January and February (associated with iodine suppression tests). Again, the sensitivity of the analysis was reported as 5 pCi L^{-1}.

In 1966, it was proposed that the sampling frequency should be reduced to biweekly. This was recommended because an 8-month test had shown that the air pumps for the air samples in 614 Buildings could be operated on a 2-week cycle without excessive wear or maintenance problems (Marter and Johnson 1966). It was noted that a longer sampling period was a

disadvantage for monitoring low-level releases of ^{131}I from the plant because of its short half-life. However, it was considered likely that the stack air monitors would alert the Site to any unusual iodine releases that would require shortened sampling periods (Marter and Johnson 1966). The milk monitoring results summarized on the aperture cards indicate that biweekly sampling started in April, but that more frequent sampling of the small dairy in North Augusta did occur in June following a Chinese nuclear bomb test.

The routine milk monitoring results reported in the semiannual report for the first 6 months of 1966 (Du Pont 1966c) do not agree exactly with the information recorded on the aperture cards (Du Pont 1966d). The aperture card for 1966 shows that more frequent sampling of the small dairy in N. Augusta was conducted in late May and June. The analyses that fell outside the normal sampling schedule were not included in the calculation of the 6-month average value. Consequently, an average ^{131}I concentration of 6 pCi L^{-1} is reported rather than 12 pCi L^{-1}. There is also a small discrepancy in the 6-month average value reported for the dairy in Allendale. The semiannual report gives an average ^{131}I concentration of 11 pCi L^{-1} compared to the value of 8 pCi L^{-1} that is calculated from the aperture card data. No explanation for this can be found. The August 1966 environmental monitoring monthly report (Du Pont 1966e) states that milk samples collected from a family cow in Snelling on August 24 and 26 contained 12 pCi L^{-1} and 25 pCi L^{-1}, respectively, of ^{131}I. The samples were collected following an increased release of ^{131}I that resulted from routine processing of short-cooled material. The monthly report states that increased concentrations of ^{131}I were not detected in any of the routine dairy milk samples. This corresponds to the results shown on the aperture card and reported in the semiannual report.

Sampling at the Allendale dairy was discontinued in 1967. The remaining three dairies (N. Augusta, Aiken, and Waynesboro) and a major milk distributor of locally produced milk (Augusta/Borden) were sampled biweekly. The semiannual and annual reports give lower average values for the first 6 months than those that are calculated from the aperture cards. The discrepancy arises in January where the average January values recorded on the aperture cards cannot be reproduced based on the reported weekly measurements. The reported weekly measurements yield a January average for each sample location that is up to a factor of 2 larger than the January average that is written on and calculated from data on the aperture card. Also in March, a small increase (22 pCi L^{-1}) in ^{131}I concentrations in milk samples (with maximum concentration on March 6) from farm cows at Snelling, South Carolina, and Waynesboro, Georgia, was noted in the 1967 environmental monitoring annual report (Ashley 1968) in the section entitled "Radioactive Waste Releases: Separations Areas." This was associated with special tests in March. The farm cows were no longer part of the routine monitoring program that sampled small dairies. The April 1968 monthly report prepared by the Environmental Monitoring Group (Du Pont 1968) also notes increased concentrations of ^{131}I in composite thyroid samples of white tailed deer collected on the SRS during 1967. The maximum concentration was found in deer collected March 8 (see Chapter 11 for more details concerning radionuclide concentrations in deer and other wild game).

From 1968 onward, milk monitoring results ceased to be reported as a separate section in the annual environmental reports. The monitoring results for ^{131}I (tritium and ^{90}Sr) are tabulated in Appendix A, Table 5, of the 1968 annual environmental monitoring report (Ashley 1969). The semiannual reports continued to devote a section to milk monitoring results. Throughout the year, the concentration of ^{131}I in milk was less than the minimum detection limit (2.2 pCi L^{-1}).

The concentration of ^{131}I in milk continued to be less than the minimum detection limit throughout 1970 and 1971. During April and the first week of May 1972, low concentrations of ^{131}I (≤25 pCi L^{-1}) were detected in milk at all four dairy sample locations (N. Augusta, Aiken, Waynesboro, and Barnwell) and the milk distributor (Borden) in Augusta. This increase was attributed to fallout from atmospheric weapons testing as a second peak of activity in air, possibly after travel around the earth (Ashley and Zeigler 1973). No mention is made of positive measurements of ^{131}I in milk during January and March. According to the aperture cards, low concentrations of ^{131}I (≤5 pCi L^{-1}) were measured in milk from dairies in N. Augusta, Aiken, and Waynesboro during the first week of September and at Barnwell during the third week of September (biweekly sampling). These measurements are not discussed in the annual reports or the monthly reports.

Milk samples from small local dairies (anywhere between four and seven) and one major distributor continued to be analyzed for ^{131}I up through 1990 (more recent annual environmental monitoring reports have not been examined). The concentration of ^{131}I in milk was below the sensitivity of the analysis at all times with the exception of three time periods in 1976, 1977, and 1978 when ^{131}I was detected in the milk samples. At these times, farm cows were also sampled in addition to the routine monitoring of the milk from small diaries. The increases were attributed to atmospheric fallout from Chinese bomb tests. A single milk sample from Gracewood, Georgia, (near Augusta, Georgia) in May 1986 also contained a measurable concentration of ^{131}I (11 pCi L^{-1}). This was attributed to fallout following the Chernobyl nuclear accident.

Strontium

A general overview of the time period and frequency that the SRS has sampled milk on a routine basis for ^{90}Sr is presented in Table 10-1. The SRS environmental monitoring program results for ^{90}Sr analyzes of milk samples are compiled as two spreadsheets in the same Microsoft Excel© workbook that contains the ^{131}I measurements (filename: Ch10-All_data.xls). One spreadsheet covers the data for 1960 through 1961 and is labeled Sr-90_60-61. The other spreadsheet covers the data for 1962 through 1973 and is labeled Sr-90_62-73. These data are taken from handwritten compilations of effluent and environmental monitoring results on ledger sheets (the aperture cards) and from the various environmental monitoring reports the Site produced. The aperture card data are available from July 1962 through December 1973. For the time period from March 1960 up to July 1962, the data are compiled from the weekly and biweekly monitoring reports and cross-checked with the semiannual reports.

From 1974 through 1990, the ^{90}Sr (also ^{137}Cs and tritium) concentrations measured in milk are reported in the annual reports as the minimum and maximum annual concentrations by location. The annual average concentration was also reported for the earlier years in this time period. It is not possible to discern the actual concentration measured each quarter at a specific location. For this reason, these data have not been compiled in the Microsoft Excel© workbook.

The general format and layout of the spreadsheets is similar to that used for the ^{131}I data. However, there are fewer measurements because milk samples for ^{90}Sr were taken quarterly rather than weekly.

In the top left hand corner of each spreadsheet the following information is recorded: the years that the samples were collected and analyzed, the type of sample analyzed (milk), the radionuclide analyzed (^{90}Sr), and the units in which the measurements are reported (pCi L^{-1}).

The source of the data used to compile the monitoring measurements is presented at the top of the spreadsheets by reference to its SRS Phase II Database number together with an indication of the type of document (e.g., aperture card, annual report, or weekly report).

A red dot placed in the upper right hand corner of a spreadsheet cell indicates that a note is attached to that cell. The note contains additional information about that measurement or time period.

The first column of data reports the sample date. The subsequent columns report the measured values by sample location. The sample locations are arranged by sample type (e.g., farm cow, local dairy, or major distributor).

Additional columns or rows are inserted into the spreadsheets to cross-check the reported data if sufficient information is available. For example, the average concentration for a sample type that is reported for a specific time period is compared with the value obtained by summing the relevant data from the aperture cards. Similarly, the average monthly or 6-month concentrations that are calculated from the individual measurements are compared with those reported in the monthly, semiannual, or annual reports.

Description and History of the Milk Monitoring Program for Radiostrontium

Sampling of milk for ^{90}Sr began during the fall of 1958 (Du Pont 1959b) (Table 10-1). Samples were taken from three small farms (family-owned cows) and from two small dairies in the vicinity of the SRS; however, the sample locations are not identified in any of the reports. Although it may be assumed that these are the same five locations that were used for ^{131}I sampling, there is no way to correlate each sample to a specific location. A sample was also taken from a small farm 100 mile from SRS. The July through December 1959 environmental monitoring semiannual report (Du Pont 1960a) only reports the monthly average concentrations of ^{90}Sr for all the milk samples. Values provided for January, April, June, July, and December varied between 14.3 pCi L^{-1} and 23.3 pCi L^{-1}.

From March 1960 onward, the concentration of ^{90}Sr in milk was reported quarterly by location in the weekly (later biweekly) environmental monitoring reports. The measured ^{90}Sr concentration in milk was reported by location and can be cross-checked with the summary data presented in the semiannual reports. Milk was sampled quarterly on a routine basis from three farm cows (located in Talatha, South Carolina, Snelling, South Carolina, and Sylvania, Georgia). The ^{90}Sr content of milk from the farm in Snelling, South Carolina, increased significantly in May 1961 and remained higher than normal through August 1961 (Du Pont 1961e). Extensive sampling of soil and vegetation on pastureland where the cow grazed indicated an accumulation of fallout ^{90}Sr in low lying “sinks” because of drainage and/or windbreaks caused by hedgerows surrounding the grazing area (Du Pont 1961e). The sampling location of Pleasant Mount, South Carolina (farm cow) was added in September 1961. Interestingly, milk from this cow assayed highest in both ^{90}Sr and ^{137}Cs. The farm was located approximately 2 mile southeast of the Snelling, South Carolina, farm, which was located 1.5 mile east of the SRS perimeter.

Six local diaries (Waynesboro, N. Augusta, Langley, New Ellenton, Williston, and Sylvania) were also sampled quarterly on a routine basis. Sampling at New Ellenton ceased after March 1961, but Aiken was included in the program in March 1961.

In the previous year, farm cows at Cope, South Carolina, and Aiken, South Carolina, were sampled in a special study from June through December 1960. The study was carried out to

explain the higher ^{90}Sr concentrations found in milk from single-cow farms compared with milk from large dairies (see Figure 10-6). Morning and evening milk samples and samples of the diet were analyzed for various factors including calcium content.

Throughout 1962 and 1963, milk from six farm cow locations (Talatha, Snelling, Pleasant Mt., Sylvania, and two in Waynesboro) was analyzed for ^{90}Sr. The frequency of ^{90}Sr determinations in milk was increased from quarterly to monthly in 1962 (Du Pont 1962a). The purpose of the accelerated analysis schedule was to determine when and to what extent ^{90}Sr from recent atmospheric nuclear tests would appear in local milk.

The ^{90}Sr data are not reported by location on the aperture cards after March 1963; only average concentrations are reported for farm milk, dairy milk, and distributor milk. Fortunately, June 1963 site-specific data were reported in the September 1963 monthly report (Du Pont 1963c). However, site-specific data for the rest of the year are not reported anywhere. Du Pont (1963b) reports the sampling schedule as of 7/15/63 and indicates that quarterly samples were collected on the Thursday of the second week of March, June, September, and December. The quarterly samples were ½-gal in size.

From 1964 onward, only two farm cows were sampled quarterly on a routine basis, at Snelling and at Waynesboro (McLeroy). Sampling of Waynesboro was discontinued after June 1967, and sampling at Snelling was discontinued after December 1969.

Milk from six dairies (Aiken, N. Augusta, Williston, Waynesboro, Allendale, and Barnwell) was sampled for ^{90}Sr from 1962 onward. The dairies at Barnwell and Aiken were added to the routine sampling schedule on January 25, 1962. Sampling was discontinued at Williston and Barnwell after August 1963 and at Allendale after August 1966. From March 1973 through 1976, only two dairies were sampled routinely (quarterly) (N. Augusta [Keys] and Waynesboro [Boyce]). In 1977, a third dairy in Denmark, South Carolina, was also sampled. Three dairies were sampled in 1978 through 1980, but the sample locations changed. The number of dairies sampled increased after 1980; the exact number fluctuated from year to year. For example, six dairies were sampled from 1981 through 1984, seven in 1986 (two of the dairies ceased operations early during 1986, effectively leaving only five dairies from which to sample), and five in 1987 through 1990.

Milk from three major distributors that was produced in the local milkshed was sampled routinely from 1960 through 1973. From March 1973 through 1990, milk from just one major distributor (Borden) continued to be sampled quarterly and analyzed for ^{90}Sr.

USEFULNESS AND LIMITATIONS OF THE MILK DATA FOR DOSE RECONSTRUCTION

Factors that determine how useful the milk monitoring data may be for reconstructing historical releases from the SRS include the

- Ability to reconstruct the historical measurements at individual locations
- Availability of sufficient datasets to allow spatial and/or temporal trends in the data to be identified
- Ability to differentiate between Site releases of contaminants and other sources of the same contaminants in the environment.

The following sections discuss these factors and also address some other issues that may limit the usefulness of the data.

Availability of Original Monitoring Measurements

One of the problems with the milk monitoring data that are reported in the quarterly, semiannual, and annual reports is that results are often averaged over large spatial areas or over more than one sample period for a given location. For example, for the late 1950s and much of the 1960s, the ^{131}I and ^{90}Sr results for a given sample date are combined and reported as an average value; the maximum value is also reported. This applies to the analyses for farm cow milk, dairy milk, and large distributors' milk. Similarly, from 1974 onward, even though the ^{131}I and ^{90}Sr concentrations are reported by location, they are reported annually, normally as the maximum and minimum concentration measured throughout the year. The annual average is also reported occasionally. Based on these data, it is difficult to infer a great deal about releases from the Site. Spatial and temporal trends cannot be assessed without knowing individual results. Fortunately, the compilations of data on the aperture cards are reported for each sampling location. Summing these data by sample type (e.g., single cow or dairy) and comparing the results with the data presented in the Site reports represents a type of data verification. Also, for the very early time periods before the aperture card data are available, the weekly and monthly reports generally contain sufficient detail to reconstruct the measured concentrations. The individual datasets that have been compiled electronically for the separate milk sampling locations may allow meaningful conclusions to be reached about releases of iodine and strontium from the SRS.

The aperture cards provide data through 1973. For the time period after 1973, a mechanism to determine the concentrations measured in milk at each sample time has not been established. This is not of concern for ^{131}I data because the measured concentrations were invariably below the sensitivity of the analysis. For those rare occasions when a detectable concentration was measured, individual results are reported. This is not the case for ^{90}Sr, which was consistently detected in all milk samples throughout the entire time period but only reported annually. The ^{90}Sr concentrations measured in milk were invariably attributed to worldwide fallout from weapons tests.

Availability of Spatial and Temporal Datasets

Although the number and location of milk sampling stations has fluctuated considerably over the history of the SRS operations, a comprehensive dataset of ^{131}I and ^{90}Sr measurements in milk has been compiled. In the early days, most of the milk samples were from single milk cows on small farms. However, these farms gradually disappeared with time, and the number of samples from small dairies increased (Table 10-2). The milk sampling stations were located in most directions around the SRS and provide reasonable spatial coverage (Figure 10-1). This may help to distinguish between increases in contaminant concentrations because of fallout from atmospheric weapons testing and releases from the SRS. The following section discusses this point further.

For the ^{131}I measurements in milk, the sensitivity of the analyses has allowed measurable concentrations to be detected in milk samples from the late 1950s through the mid-1960s. The most significant increase in sensitivity occurred in June 1960 with the introduction of an ion exchange-gamma spectrometric technique (Figure 10-2). This technique, which replaced a radiochemical separations technique that used a Geiger-Mueller counter to count the sample,

reduced the minimum detection limit by over an order of magnitude (~10 pCi L^{-1} versus ~160 pCi L^{-1}). For further details see Appendix A. Beginning in 1966, the concentration of ^{131}I measured in milk samples was usually below the sensitivity of the analyses. Milk samples were generally sampled weekly through 1966, after which the frequency decreased to every 2 weeks. Decreasing the frequency of sampling increased the likelihood that releases of ^{131}I would go undetected because of the short half-life of ^{131}I.

Concentrations of ^{90}Sr have been measured in milk samples from around the SRS throughout the entire monitoring program for ^{90}Sr (1959–1990) (Table 10-1). However, the milk was sampled just four times a year at each location, with the exception of 1962, when monthly samples were analyzed.

Other Sources of Contamination

The SRS was not the only potential source of ^{131}I and ^{90}Sr in the environment. Fallout from atmospheric weapons testing was a source of radionuclides in the environment and, hence, in milk samples. Fallout concentrations of these radionuclides were greatest in the early 1960s. The concentrations in fallout decreased significantly from 1963 onward following the bomb testing moratorium.

The PHS started a milk monitoring program in 1957. Milk was sampled from locations throughout the U.S. The three monitoring locations closest to the SRS were Atlanta, Georgia, Charleston, South Carolina, and Charlotte, North Carolina. The milk monitoring results for all three locations have been compiled electronically in a Microsoft Excel© workbook (filename: Ch10-All_data.xls). These data are located in the sheets labeled PHS and Raw. The data are compiled from the series of reports titled Radiological Health Data that the PHS produced from 1960 through 1970. From 1971, these reports were produced by the U.S. Environmental Protection Agency and were titled *Radiological Health Data and Reports* in 1971, *Radiation Data and Reports* from 1972 through 1975, and *Environmental Radiation Data* from 1975 through 1994. The results are compiled for the following radionuclides: ^{137}Cs, ^{140}Ba, ^{131}I, ^{89}Sr-89, and ^{90}Sr. The mean monthly concentration of each radionuclide is reported in picocuries per liter, together with the counting error and number of samples if such data were available.

If no entry was made for the number of samples collected, then a single sample was collected for that month; the exact dates of collection are not included (they were reported beginning in 1979). In some cases, more than one sample was collected in a given month. Beginning in 1979, the dates of collection are included in those instances. Dates of collection are also included for samples collected in response to Chinese weapons testing on September 26 and November 11, 1976, September 17, 1977, and March 14, 1978, and in response to the accident in Chernobyl on April 26, 1981.

Before July 1974, a zero entry or no entry at all for a given radionuclide indicates that the measurement was below the minimum detectable concentration (MDC) or practical reporting level. This was quite common for ^{140}Ba, ^{131}I, and ^{89}Sr. Since July 1994, we included everything reported in the spreadsheet. The spreadsheet also indicates no analysis (na), no sampled collected (ns), not reported (nr), and not detected (nd).

From 1957 through June 1961, raw milk samples were analyzed. From 1960 through 1993, pasteurized milk was analyzed; thus, there was a brief period when both milk types were analyzed.

The PHS milk monitoring data provide an independent source of information about the concentrations of fallout radionuclides found in milk in the southeastern U.S. and allow a comparison to be made with the concentrations measured in milk samples from around the SRS. Average monthly concentrations of ^{131}I measured in milk samples taken by the PHS from Atlanta, Georgia; Charlotte, North Carolina; and Charleston, South Carolina, are compared with the average monthly concentration of ^{131}I measured in milk samples taken by the SRS from small dairies located near the SRS (Figure 10-5). The ^{131}I concentration in milk from small dairies increases at all locations during the fall of 1961 and 1962. This increase is reflected in the PHS samples at locations remote from SRS, indicating that fallout from atmospheric weapons testing is the probable source of contamination. Another peak in ^{131}I concentrations in milk was observed at some dairy and farm cow locations during June 1961 (Figures 10-5 and 10-6). This increase may indicate a release of ^{131}I from the Site. There was a substantial increase in daily ^{131}I release rates from F-Area at this time, and a similar increase is seen for vegetation concentrations (see Chapter 9, Figure 9-10). A further complicating factor is the different feeding regimes for the farm cows, discussed in the next section. The Talatha, Snelling, and Pleasant Mount locations are expected to have the greatest impact from Site releases because they are closest to the Site boundary. Unfortunately, they are all farm cow locations and are also expected to have higher concentrations because of their greater dependence on pasture grass.

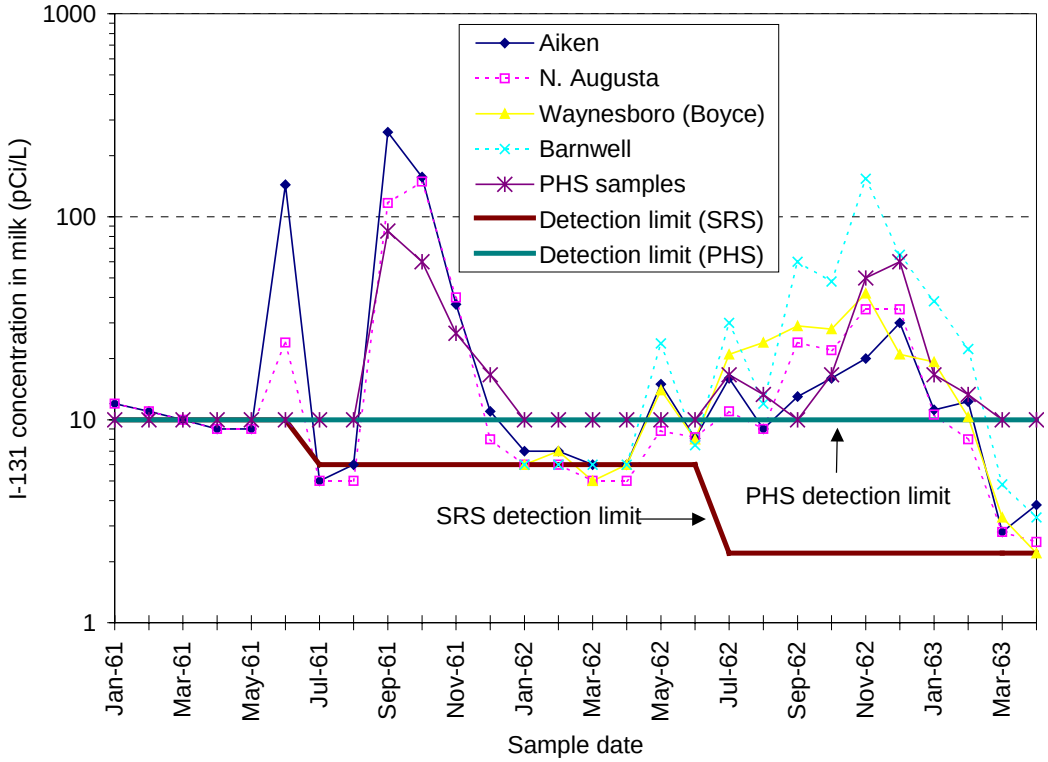

Figure 10-5. Monthly average ^{131}I concentrations in milk sampled from local dairies around the SRS and in milk sampled by the Public Health Service (PHS) at Atlanta, Georgia; Charlotte, North Carolina; and Charleston, South Carolina. Link to tabulated figure data.

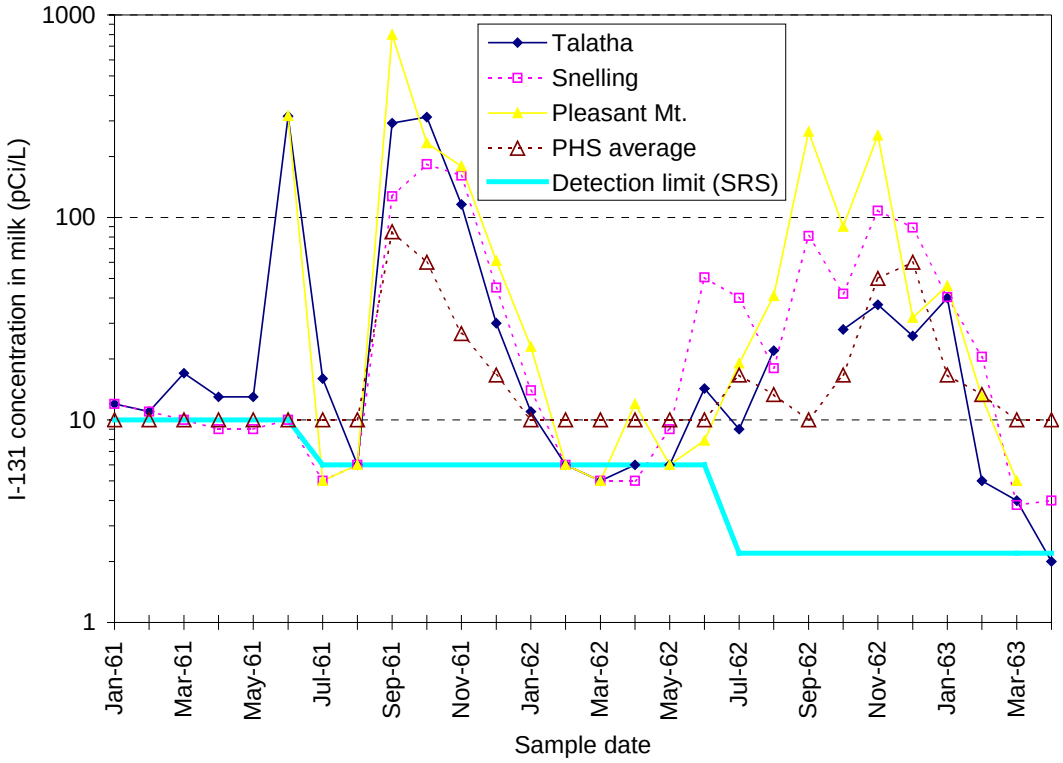

Figure 10-6. Monthly average ^{131}I concentrations in milk sampled from farm cows around the SRS. Link to tabulated figure data.

Limitations to the Data

It was observed that the farm cows consistently produced milk with higher radionuclide concentrations than the dairy cows. A number of investigations were made on this topic for strontium (Du Pont 1962c; Butler 1960a, 1960b) and it was determined that the effect occurred as a result of the different feeding regimes of the two types of cows. Farm cows (individual family-owned) generally received little commercial feed and primarily grazed on topshoots or shallow-rooted grasses, both of which are susceptible to fallout contamination. In contrast, dairy cows received less pasture grass and more dried silage and commercial feed (Du Pont 1962c, 1963d). This trend can be seen in Figure 10-7 for the measured concentrations of ^{90}Sr in milk.

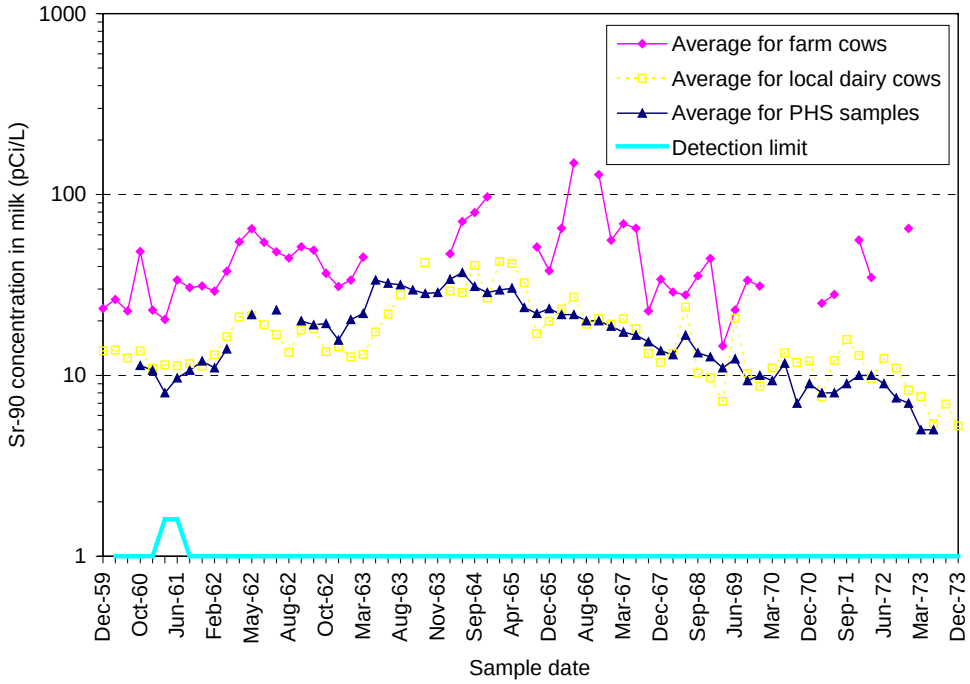

Figure 10-7. Comparison of average ^{90}Sr concentrations in milk from farm cows and local dairies in the vicinity of the SRS and milk samples taken by the Public Health Service in Atlanta, Georgia; Charlotte, North Carolina; and Charleston, South Carolina. Link to tabulated figure data.

Comparison of Figures 10-5 (local dairies) and 10-6 (farm cows) shows that the same trend was seen for ^{131}I concentrations in milk. As early as 1957, a statement was made in the weekly report (October 28–November 1, 1957) (Du Pont 1957) that cows feeding on pasture grass would yield ^{131}I concentrations in milk 20 times greater than those observed in the routine analysis. The single farm cows that provided the routine milk samples at Talatha and Snelling were fed on hay in the barn. Likewise, Holley's cows (Aiken dairy) were stall fed large quantities of roughage and concentrates. Based on this information, it would seem that the feeding regimes of the two types of cow were not consistently different, which may account for the often less than a factor of 20 difference in the two concentrations. Hay from the barn was probably cut from local pastures and could still contribute to higher concentrations.

Early in the milk monitoring program, a transition was made from sampling raw milk to sampling pasteurized milk. A number of studies were carried out to determine if processing milk affected the radionuclide concentrations. A comparison was made of the radioiodine concentration in samples of raw and pasteurized milk originating in a common batch from Holley's Dairy, Aiken on November 10, 1961. The Environmental Monitoring and Allied Studies October 28–November 17, 1961, report (Du Pont 1961f) states that the ^{131}I concentration in raw milk was 37 pCi L^{-1} compared with 44 pCi L^{-1} in processed milk. It was concluded that processing the milk had no effect on the ^{131}I content of pasteurized milk. A similar conclusion was reached concerning the ^{90}Sr content of milk compared with buttermilk. Both normal milk and buttermilk samples were routinely collected from the large milk distributor in Aiken (Borden) and analyzed for ^{90}Sr. The concentrations were not significantly different.

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CHAPTER 11

RADIONUCLIDES IN WILD GAME

ABSTRACT

This chapter discusses radionuclide concentrations measured in wild game collected at the Savannah River Site (SRS). We evaluated these data with regard to their potential usefulness for dose reconstruction. We also attempted to validate the data by making comparisons of concentrations among as many sources as possible. This included original, hand-written compilations for many years. We found these original data to consistently correspond to the data reported in monthly, semi-annual, and annual summary reports.

We compiled and examined wild game data and other environmental data to determine their usefulness for source term verification, model validation, and direct exposure assessment. In general, the wild game data are most valuable for direct exposure assessment. However, because range and eating habits for the various animals are not well defined, the data are not particularly useful for source term verification or model validation. Appendix K further discusses potential uses for environmental monitoring data.

INTRODUCTION

Before the Atomic Energy Commission acquired the SRS in 1950 and 1951, the white-tailed deer (*Odocoileus virginianus*) population was estimated at several dozen animals. Under conditions of virtually absolute protection, the population increased to approximately 1400 animals in 1963 and to approximately 5000 animals in the spring of 1965 when managed public hunts were initiated to control the expanding population (Rabon 1976). Deer at the SRS represent a potentially important exposure pathway for historical exposures of the public from radionuclides because large numbers of deer have been harvested during these annual hunts. In addition, smaller numbers of feral hogs have been harvested from the SRS during the public hunts.

Other fur-bearing animals, including cats, mice, rats, foxes, raccoons, dogs, bobcats, squirrels, and opossums, have also been trapped and analyzed by the SRS to determine radionuclide concentrations. Additionally, waterfowl, including gulls, grebes, coots, and several species of duck, have been collected, primarily from Par Pond, and analyzed to determine radionuclide concentrations. The following discussion focuses on deer concentrations because they represent the most significant pathway for potential exposure to the general public. However, this chapter also examines concentrations in other wild game species.

This section summarizes reported information regarding radionuclide concentrations in wild game collected on or in the vicinity of the SRS. We examined several sets of routine semiannual and annual environmental monitoring reports, prepared by the SRS contractor and spanning the years 1953 through 1992. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series.

Several additional documents have been reviewed for information pertaining to deer monitoring at the SRS. These include monthly monitoring reports from 1962 through 1965, weekly monitoring reports from 1959 through 1962, and aperture card printouts (handwritten data

entry sheets) from 1970 through 1981. Although these printouts were sometimes not legible, they provided general verification of data presented in the monitoring reports. The results were not summarized, but reported concentrations are consistent with those given in the monitoring reports. Original field data from 1965, 1966, 1982, 1984, and 1985 were also reviewed. In general, the data provided in these documents are also consistent with the data provided in the monitoring reports. Additionally, aperture card printouts from 1970 through 1981 provided general verification of the data provided in the monitoring reports for waterfowl and other animals.

The radionuclides for which data are available for wild game include nonvolatile beta, ^{137}Cs, ^{129,131}I, ^{89,90}Sr, ^{238,239}Pu, ^{65}Zn, and ^{3}H. Appendix A details analytical and counting procedures for wild game. Concentrations of heavy metals (chromium, cadmium, lead, and mercury) in deer tissue were also reported in 1987 and 1989 (Mikol et al. 1988b and Cummins et al. 1990), and most concentrations were below the detection limit.

Figure 11-1 shows the general time periods for which individual radionuclide concentrations were reported for deer in the various environmental monitoring reports and data summaries.

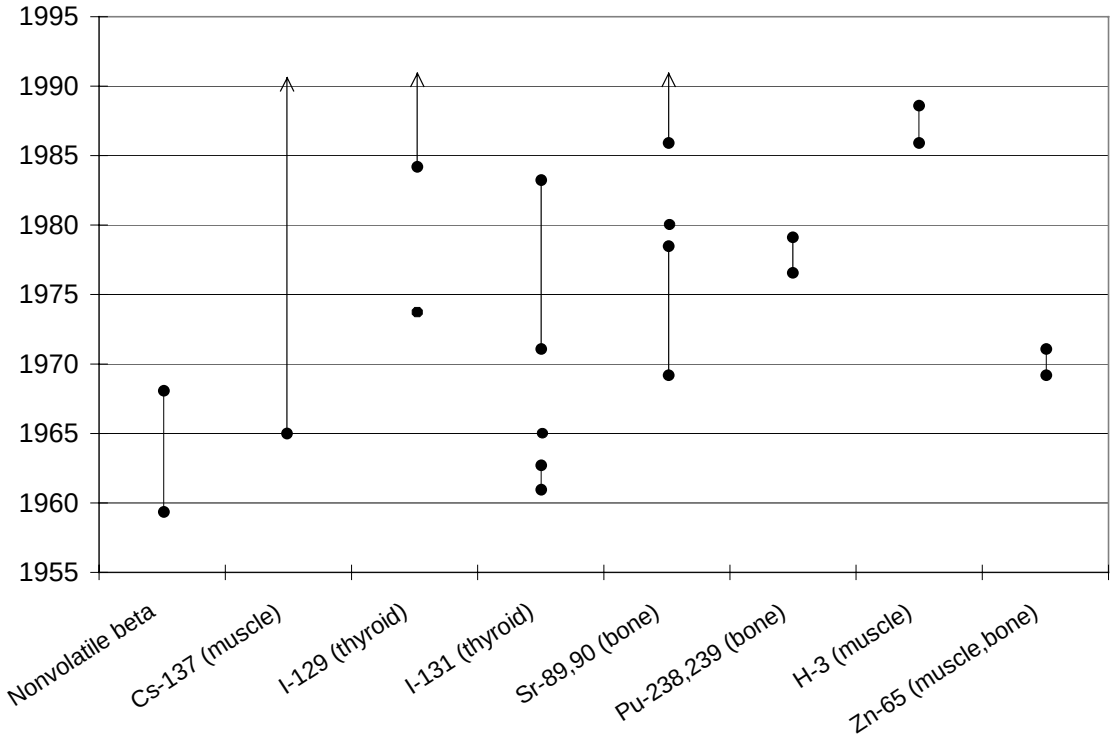

Figure 11-1. General time periods for which individual radionuclide concentrations were reported for deer. Link to tabulated figure data.

RADIONUCLIDE CONCENTRATIONS MEASURED IN DEER

Nonvolatile Beta and Cesium-137

Cesium-137 distributes in body tissues similarly to potassium, residing primarily in soft tissues such as muscle. Consequently, the majority of analyses have focused on ^{137}Cs concentrations in muscle tissue. The SRS has made field measurements on deer killed during annual public hunts since 1970. The fraction of deer analyzed in the laboratory for field measurement verification from 1970 through 1992 ranged from 2 to 20%, with an annual average of 7 %. An average of 1064 animals has been harvested annually from 1965 through 1992.

Before 1965, only nonvolatile beta concentrations were reported for muscle tissue. Nonvolatile beta activity refers to activity resulting from the presence of radionuclides that do not escape during sample preparation, such as evaporation or wet-ashing. The primary radionuclides that could potentially contribute to nonvolatile beta activity at the SRS include ^{137}Cs, ^{65}Zn, ^{90}Sr, ^{32}P, and naturally occurring ^{40}K. In general, ^{137}Cs is the radionuclide of greatest concern because of its relatively long half-life (30 years) and its tendency to accumulate in edible (muscle) tissues.

Nonvolatile beta and ^{137}Cs concentrations were reported from 1965 through 1968. Table 11-1 summarizes the information provided in the environmental monitoring reports for nonvolatile beta and ^{137}Cs concentrations in muscle during the years both were reported. The nonvolatile beta concentrations are very similar to the ^{137}Cs concentrations from 1965 through 1969. It is not known whether the ^{137}Cs concentrations for those years were derived from the nonvolatile beta concentrations, but based on their similarity it appears possible. Regardless, the nonvolatile beta concentrations reported for the few deer collected before 1965 appear similar to or less than the concentrations reported since 1965. Unfortunately, very few deer were analyzed, and it is difficult to make many conclusions based on the values reported before 1965. Public exposure to deer from the SRS would likely have been minimal before 1965, when public hunts on the SRS were initiated. However, since the precise range and mobility of the deer residing on the SRS is not known, it is certainly possible that some animals spending the majority of time on the SRS migrated offsite and were harvested by hunters.

Since 1987, several deer muscle samples have been analyzed annually by the Department of Physiology and Biophysics at the University of Tennessee in Memphis. In general, the laboratory analyses conducted by the University of Tennessee have been in good agreement with the laboratory analyses conducted by the SRS and they provide some measure of validation for the measurements reported by the SRS (Table 11-2). While not explicitly stated in the monitoring reports, it is assumed that the muscle tissue samples sent to the University of Tennessee for analysis represented a portion or subset of the tissue samples routinely collected for field measurement verification. Without information regarding the specific concentrations measured for individual deer by the two laboratories, this information is admittedly of limited value.

Table 11-1. Mean Nonvolatile Beta[a] and ^{137}Cs[b] Concentrations (pCi g^{-1}) in Deer Muscle

Year	Nonvolatile beta (number of deer)	^{137}Cs (number of deer[c])
1958[d]	9 (1)	nr[e]
1959[d]	3 (1)	nr
1960	3 (2)	nr
1961	5 (3)	nr
1962	4 (2)	nr
1963	(0)	(0)
1964	8 (6)	nr
1965	11 (146)	10 (198)
1966	6 (212)	6 (541)
1967	10 (126)	9 (1032)
1968	11 (58)	11 (699)

[a] Reported in Du Pont (1959, 1960a, 1960b, 1961, 1962c, 1962d, 1963d, 1963e, 1964c) and Ashley (1965, 1966, 1967, 1968, 1969).
[b] Reported in Ashley and Zeigler (1976).
[c] This likely represents the total number of deer harvested and not the number of deer analyzed.
[d] Reported in Harvey and Rabon (1965).
[e] nr = not reported.

Table 11-2. Comparison of ^{137}Cs Concentrations (pCi g^{-1}) Measured by the SRS and the University of Tennessee[a]

	SRS analysis		University of Tennessee analysis	
Year	Number of samples	Mean ± 2 SD[b]	Number of samples	Mean ± 2 SD
1987	35	8.3 ± 21.7	24	3.8 ± 5.0
1988	67	9.8 ± 19.5	6	10.4 ± 10.0
1989	96	8.9 ± 10.2	41	6.0 ± 11.0
1990	92	8.1 ± 8.8	10	11.1 ± 15.6
1991	124[c]	3.3 ± 5.1	28	2.9 ± 5.1

[a] Data are from Mikol et al. (1988b), Davis et al. (1989b), Cummins et al. (1990, 1991), and Arnett et al. (1992).
[b] SD = standard deviation.
[c] Includes measurements for both deer and hogs.

Since 1968, ^{137}Cs concentrations have also been reported for South Carolina Coastal Plain (SCCP) deer. The SCCP is an 18,000-acre controlled hunting camp located approximately 65 mi southeast of the SRS in Beaufort County. In 1974, 10 deer from Beaufort, South Carolina, (approximately 100 mi southeast of the SRS) were analyzed, and in 1979, four deer from the Fort Jackson military reservation (approximately 15 mi southeast of Augusta, Georgia) were analyzed. The SCCP deer and the additional deer sampled in 1974 and 1979 have served as indicators of background ^{137}Cs burdens from weapons testing fallout that would be found in deer living in

similar environments. The SRS deer concentrations are lower than the concentrations in deer taken from offsite; therefore, they do not appear to be elevated above expected background concentrations. Table 11-3 summarizes the measured concentrations in 1974 and 1979.

Table 11-3. Comparison of Mean ^{137}Cs Concentrations[a] (pCi g^{-1}) in Deer from the SRS and Vicinity

Year	SRS	SCCP	Beaufort	Ft. Jackson
1974	5 (n=1551)	9 (n=89)	10 (n=10)	
1979	10 (n=1079)	12 (n=57)		15 (n=4)

[a] Data are from Du Pont (1975a, 1980a, and 1981a).

The authors of the monitoring reports have consistently maintained that the ^{137}Cs concentrations measured in deer from the SRS are due almost entirely to worldwide fallout resulting from weapons testing. Soils from the lower coastal plain of the southeastern U.S. are typically sandy and low in clay content, organic matter, and available potassium. This leads to increased amounts of ^{137}Cs available for plant uptake and subsequent deer ingestion relative to other areas of the continental U.S., which makes it difficult to distinguish SRS-derived ^{137}Cs from global fallout resulting from weapons testing. However, concentrations measured in deer collected from offsite locations give no indication that mean ^{137}Cs burdens in SRS deer are elevated above expected background concentrations. Van Middlesworth (1993) concluded that most of the ^{137}Cs in deer from the SRS, SCCP, and other parts of the southeastern U.S. can be attributed to worldwide fallout.

The mean concentrations have consistently been lower for SRS deer than for deer collected from the SCCP. This is likely the result of different soil characteristics for the two areas. The SRS is situated on the boundary of the Piedmont and Coastal Plains. The SCCP is, as the name implies, located in the Coastal Plain area. Soils in the Coastal Plain area are characterized by more sand and organic matter, less clay, more ammonia, and a higher water table, all of which contribute to increased ^{137}Cs mobility (Whicker 1997). Therefore, higher mean concentrations would be expected for deer taken from this area. Consequently, the fact that mean concentrations have been consistently lower for SRS deer than for SCCP deer does not necessarily indicate that SRS activities have not affected ^{137}Cs concentrations in the resident deer population. To estimate exposure to hunters who harvested deer from the SRS, it may be necessary to quantify the extent of SRS-derived ^{137}Cs in onsite deer.

In 1989, Winn (1990) attempted to distinguish whether ^{137}Cs measured in individual deer originated from the SRS or global fallout. Cesium releases from the SRS have an identifiable ^{134}Cs/^{137}Cs fingerprint that is different from that of global fallout. This method alone was considered insufficient to identify SRS deer, but the calculated isotopic ratios (0.001–0.003) were consistent with global fallout and were comparable to those calculated for offsite deer. The results of the study also implied that deer feed primarily on land areas, as opposed to the banks of effluent streams. However, the scope of the study was limited because only six animals were analyzed.

The annual maximum concentrations for SRS deer have consistently been higher than those measured in offsite deer. These higher concentrations likely reflects uptake of ^{137}Cs from contaminated streams or ponds for a few individual deer, but based on the annual means, not a significant portion of the population. The maximum concentration in 1966 was for an animal

taken from along Four Mile Creek, and the maximum concentrations for 1967 through 1970 and 1979 were for animals taken from near Steel Creek. This suggests that the maximum concentrations in deer from the SRS are likely to be found in animals residing and feeding near the most contaminated areas (e.g., on the banks of contaminated onsite streams).

Since 1965, during the annual public hunts, there has been only one deer confiscated (204 pCi g^{-1} in 1969) and only three additional deer with a measured ^{137}Cs concentration greater than 100 pCi g^{-1} (104 pCi g^{-1} in 1967, 103 pCi g^{-1} in 1969, and 117 pCi g^{-1} in 1970). In addition, several deer collected during special sampling along Four Mile Creek in July 1968 had concentrations greater than 100 pCi g^{-1}, and one had a concentration of 204 pCi g^{-1} (Hagelston 1970). Consequently, that location was not included in the public hunt plans for 1968.

As previously discussed, many of the deer with the highest concentrations were collected from locations near Steel Creek. Following shutdown of the L-Reactor in February 1968, Steel Creek was narrowed significantly. This allowed for growth of vegetation on the exposed contaminated sediment. The new vegetation was palatable, available to deer, and had higher radionuclide concentrations (Hagelston 1970). Consequently, deer feeding on this vegetation may have accumulated higher levels of ^{137}Cs relative to years before the L-Reactor shutdown.

The maximum measured concentrations for deer from the SRS are not significantly elevated above concentrations that have been measured at other locations in the southeastern United States. Jenkins and Fendley (1969, cited in Rabon 1976) reported concentrations, attributed to global fallout, up to 153 pCi g^{-1} in deer from south Georgia and northern Florida in 1968. For comparison, the maximum measured concentration for SCCP deer was 80 pCi g^{-1} in 1968. Five deer collected from Eglin Air Force Base near Pensacola, FL in February 1968 had a mean ^{137}Cs concentration of 78 pCi g^{-1}, and eight deer collected from the same area in March had a mean ^{137}Cs concentration of 43 pCi g^{-1} (Du Pont 1968c). For comparison, the mean concentration measured in SRS deer during February was 8 pCi g^{-1} and during March was 7 pCi g^{-1}.

The reported measured concentrations in SRS deer since 1965 also likely reflect the maximum seasonal concentrations because most deer have been harvested in the fall. Several researchers have attributed increased ^{137}Cs body burdens in ruminants to seasonal changes in diet (Rabon 1968, 1976; Jenkins and Fendley 1968, 1969, cited in Rabon 1976; Svenson and Liden 1965; Hanson and Palmer 1965; Whicker et al. 1965). The seasonal changes in diet for deer at the SRS result from the abundance of autumnal food items such as acorns and fungi, which are tenacious concentrators of cesium. The range of measured concentrations in deer has been attributed by the SRS to concentration variability in the diets of these deer.

Figure 11-2 shows mean and maximum concentrations provided in the annual environmental monitoring reports for SRS and SCCP deer from 1968 through 1991 (note the logarithmic scale for the y-axis). Watt et al. (1983) reported the same concentrations. The concentrations appear to have declined during this period, and the data were best described by exponential functions. Linear regression of the log-transformed data confirmed slopes statistically different from zero ($p<0.01$ for both SRS and SCCP deer). The effective half-times for ^{137}Cs in the SRS and SCCP deer during this time period have been about 16.6 and 12.5 years, respectively. Although the effective half-times are not the same, they are statistically indistinguishable at the 95% confidence level. There are no apparent reasons for differing effective half-times for the two populations of deer. The noted differences are likely due to spatial heterogeneity, differences in diet, accumulation of some SRS-derived ^{137}Cs in the SRS deer, additional fallout resulting from Chinese atmospheric weapons tests, and general variability in the data.

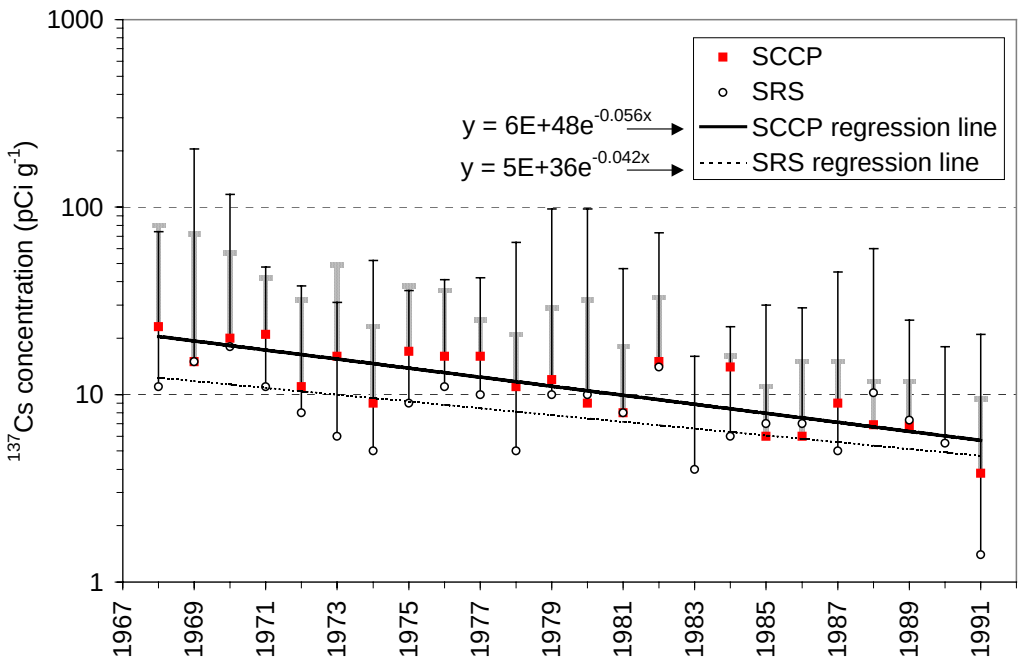

Figure 11-2. Annual mean ^{137}Cs concentrations in deer collected from the SRS and the SCCP. Thick and thin error bars represent maximum concentrations measured each year for the SCCP and SRS deer, respectively. The lower limit of detection (LLD) was 1 pCi g^{-1}. Link to tabulated figure data.

Figure 11-3 shows the maximum measured ^{137}Cs concentrations since 1965 for SRS and SCCP deer. It is clear that annual maximum concentrations have been consistently higher for SRS deer. It also appears that maximum measured concentrations are decreasing as a function of time for both locations. In addition, the maximum measured concentrations in SRS appear to peak during the late 1960s, during the late 1970s through the early 1980s, and during the late 1980s.

Frequency distributions of ^{137}Cs concentrations measured in the field indicate log-normally distributed data (Addendum 11A). For the 1985 data in particular, it is clear that including concentrations reported as 1 pCi g^{-1}, which was the apparent detection limit[1], results in a large number of animals in the 0–1 pCi g^{-1} category. These data were compiled from raw data sheets for the years 1965, 1966, 1982, 1984, and 1985. The data sets from 1982, 1984, and 1985 appeared to provide data for all deer analyzed in those years. The data sets from 1965 and 1966 provided data for only a subset of the total number of deer harvested in those years (approximately 20% of forelegs were retained for laboratory analysis). Watts et al. (1983) indicated ^{137}Cs concentrations measured in SCCP deer between 1975 and 1979 to also be qualitatively described as log-normally distributed.

[1] The lower limit of detection (LLD) values are provided in various figures throughout this chapter whenever they were reported in the environmental monitoring reports. In many cases, however, the reports state "...the sensitivity of analysis varied due to differing sample size..." and no LLD values were provided.

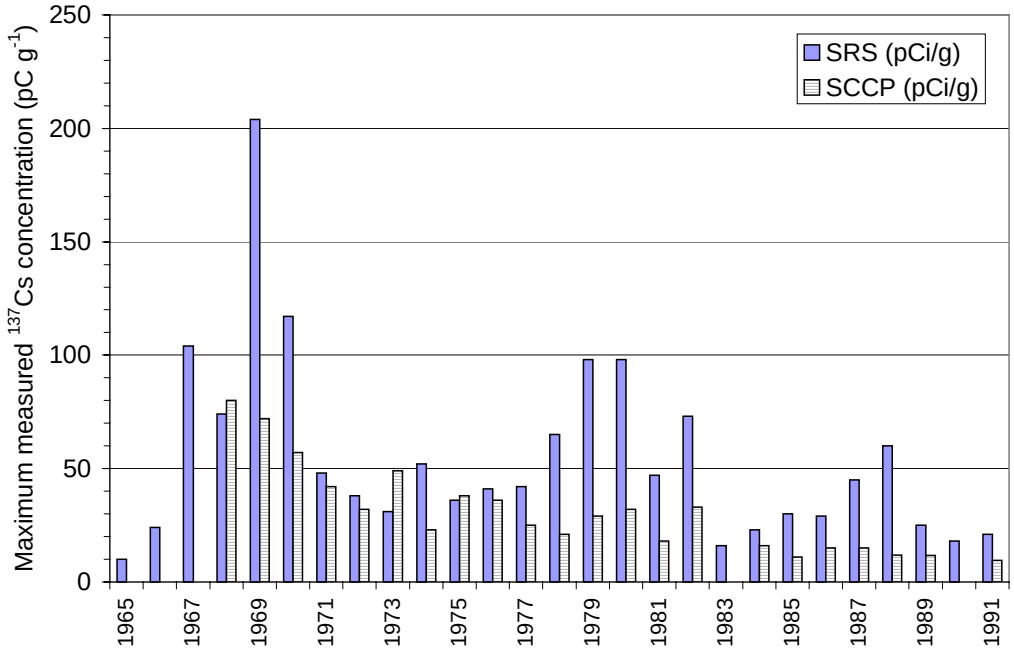

Figure 11-3. Maximum ^{137}Cs concentrations measured in deer collected from the SRS and the SCCP since 1965. SCCP deer were not collected until 1968. Link to tabulated figure data.

To complete the deer exposure pathway, it may ultimately be necessary to further examine the deer data because routinely reported arithmetic mean concentrations are likely not the best descriptors for central tendencies in deer concentrations. Estimates of live weight-dressed weight and live weight-edible meat relationships may also be necessary to fully evaluate the pathway; this information is provided in Severinghaus (1949) for white-tailed deer.

Radioiodine

Radioisotopes of iodine are released from SRS facilities and are also an important component of weapons fallout (see Chapter 6 for more details regarding other sources of contamination). With a short half-life of about 8 days, ^{131}I does not persist in the environment. Therefore, concentrations measured in the environment reflect recent releases. Iodine-129, in contrast, has a very long half-life and does persist in the environment. Radioiodine concentrations were reported for deer thyroid samples beginning in 1961. The thyroids were analyzed because of their ability to concentrate iodine; therefore, they are a sensitive indicator of the impact of SRS releases. Consumption of deer thyroid, however, is not considered a human exposure pathway.

Mean annual radioiodine concentrations measured in thyroid tissue are shown in Figure 11-4. Iodine-131 concentrations were measured in thyroid tissue in 1961, 1962, 1965, 1967, and from 1971 through 1983. Elevated concentrations were reported in 1961, 1962, 1976, and 1977. SRS researchers attributed these increases to fallout from Soviet and Nevada Test Site testing in the early 1960s and Chinese weapons testing in 1976 and 1977. There was also a significant release of ^{131}I from an F-Area stack during May and June 1961, which may have contributed to

the elevated concentrations measured during that year. Thirty-three deer thyroids were analyzed in 1967, and the maximum measured concentration (160 pCi g^{-1}) was attributed to a 6 Ci release of ^{131}I from the F-Area dissolver off-gas stack on March 1, 1967 (Du Pont 1968c). Concentrations for the remaining years were near the limits of detection (6 pCi g^{-1} in 1965 and 1 pCi g^{-1} since 1976). Similar concentrations of ^{131}I have been reported for North American deer collected in Alaska, California, Colorado, Maryland, Washington, and Wyoming during 1961 and 1962 (Hanson et al. 1963).

Iodine-129 concentrations were reported for thyroid tissue in 1974 and from 1984 through 1991 (Zeigler et al. 1985, 1986a, 1986b, 1987a, 1987b; Mikol et al. 1988a, 1988b; Davis et al. 1989a, 1989b; Cummins et al. 1990, 1991; Arnett et al. 1992). Four thyroids sampled in 1974 had ^{129}I concentrations ranging from 2 to 18 pCi g^{-1} and a mean of 9 pCi g^{-1}. The mean concentration reported in 1984 was calculated excluding a 210 pCi g^{-1} sample, and the mean concentration reported in 1989 was calculated excluding a 2145 pCi g^{-1} sample (Zeigler et al. 1985; Cummins et al. 1990). The lower limit of detection (LLD) was not given, but concentrations appear low through 1988, after which concentrations increased somewhat. However, Van Middlesworth (1993) reported no distinguishable temporal trend between 1985 and 1991 for ^{129}I in thyroids taken from SRS deer. In 1991, the average concentration was 49 pCi g^{-1}, but 90% of the samples were less than 50 pCi g^{-1} and 50% were less than 1.5 pCi g^{-1}. In general, ^{129}I concentrations have been significantly elevated only in a few isolated individuals. Because thyroids are not typically consumed, this is unlikely to represent a significant exposure pathway to hunters or their families who consumed the deer meat.

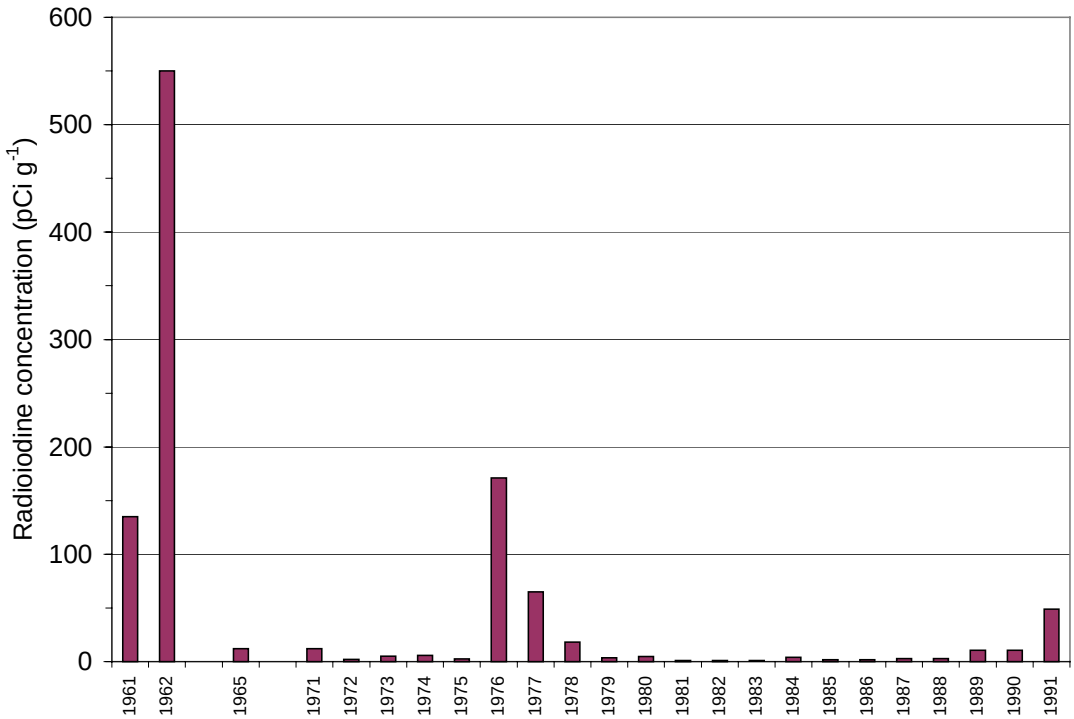

Figure 11-4. Mean radioiodine concentrations reported for deer thyroids. Iodine-131 concentrations were reported in 1961, 1962, 1965, and 1971 through 1983. Iodine-129 concentrations were reported from 1984 through 1991. Link to tabulated figure data.

Other Radionuclides

Strontium-89,90 concentrations were reported for bone tissue sampled from 1969 through 1980 and from 1986 through 1991. Radionuclides of the element strontium are metabolically similar to calcium and, thus, are found primarily in the bone of vertebrate animals. With a half-life of about 28 years, the fission product strontium-90 is an important relatively long-lived component of weapons fallout. It has also been released from SRS facilities. As with thyroid samples, bone samples were analyzed to assess SRS impact and not as a human exposure pathway.

Mean ^{89,90}Sr concentrations ranged from 2 to 54 pCi g^{-1}, with a maximum value of 207 pCi g^{-1} in 1974. Concentrations measured in 1986 and 1987 (26.7 and 54.2 pCi g^{-1}, respectively) were higher than other years, but relatively few samples were analyzed (six and five, respectively). Rabon (1968) reported a negative correlation of radiostrontium concentration with deer age (that is, younger deer had higher concentrations), and no correlation between radiostrontium concentration and deer size and sex or season.

Zinc-65 is a relatively short-lived (half-life 245 days) activation product produced in nuclear reactors and in the fallout from nuclear weapons detonations. Zinc-65 concentrations were reported for 1969 through 1971 for a few animals, but the mean concentrations were at or below the detection limit. Aside from ^{65}Zn and naturally occurring ^{40}K, ^{137}Cs has been the only gamma-emitting radionuclide reported as detected in deer taken from the SRS.

Plutonium-238,239 concentrations, alpha-emitting radionuclides, were reported for bone tissue sampled in 1977, 1978, and 1979. Concentrations of ^{238}Pu ranged from 0.6 to 5.6 fCi g^{-1}, and concentrations of ^{239}Pu ranged from <0.18 to 3.4 fCi g^{-1} with a mean of 1.7 fCi g^{-1} for both isotopes (1 femtocurie [fCi] is equal to 10^{-15} Ci).

Tritium concentrations were reported from 1986 through 1988. This beta-emitting radionuclide is incorporated into the water molecule; consequently, it is present in all tissues. In 1986, nine muscle tissue samples were analyzed and they had a mean concentration of 17 pCi mL^{-1}. In 1987, three samples were analyzed and they had a mean concentration of 250 pCi mL^{-1}, which was largely the result of one 645 pCi mL^{-1} sample. In 1988, 98 samples were analyzed and most had concentrations less than 1 pCi mL^{-1}.

RADIONUCLIDE CONCENTRATIONS MEASURED IN FERAL HOGS AND OTHER TERRESTRIAL ANIMALS

Smaller numbers of feral hogs have also been collected from the SRS during the annual public hunts. In general, mean ^{137}Cs concentrations have been similar to or less than concentrations reported for deer (Figure 11-5). Little information was available with regard to radionuclide concentrations in hogs before 1971. Nonvolatile beta concentrations reported for a few hogs collected in 1963 and 1964, though, were similar to those reported for deer.

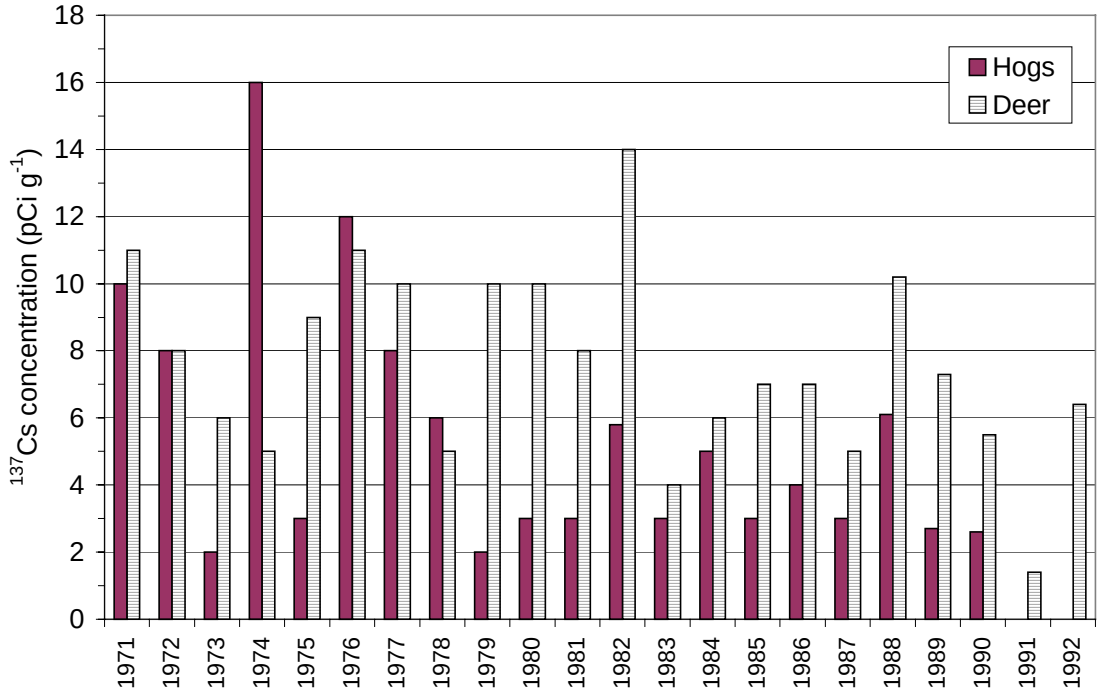

Figure 11-5. Mean ^{137}Cs concentrations reported for feral hogs and deer from 1971 through 1992. Link to tabulated figure data.

Other terrestrial animals have also been routinely trapped, including cats, mice, rats, foxes, raccoons, dogs, bobcats, squirrels, and opossums. Routine trapping locations were not clearly specified, but maximum concentrations were generally found in animals collected from locations in the vicinity of Four Mile Creek, Lower Three Runs, Steel Creek, Burial Grounds, H-Area and F-Area seepage/retention basins, R-Area seepage basins, and K-Area containment/seepage basins.

Figure 11-6 shows mean radionuclide concentrations for rabbits, raccoons, and opossums from 1951 through 1989 (note the logarithmic scale). Nonvolatile beta concentrations are shown from 1951 through 1970, and ^{137}Cs concentrations are shown from 1971 through 1989. Concentrations appear to have decreased significantly since the late 1960s. In general, concentrations measured in these animals are similar and within the ranges of concentrations reported for other fur-bearing animals. However, concentrations for carnivorous animals (raccoons and opossums) appear slightly higher than concentrations for herbivorous animals (rabbits). A special animal trapping program in 1974 concluded that concentrations in 37 trapped animals were within the range of concentrations detected in deer for that year (Ashley and Zeigler 1975). Mean concentrations for such animals were, however, often heavily influenced by significantly elevated concentrations measured in a few individual animals. This is evident in 1956 and for several years during the 1960s and early 1970s. As mentioned previously, the highest concentrations were measured in animals collected from locations in the vicinity of contaminated areas.

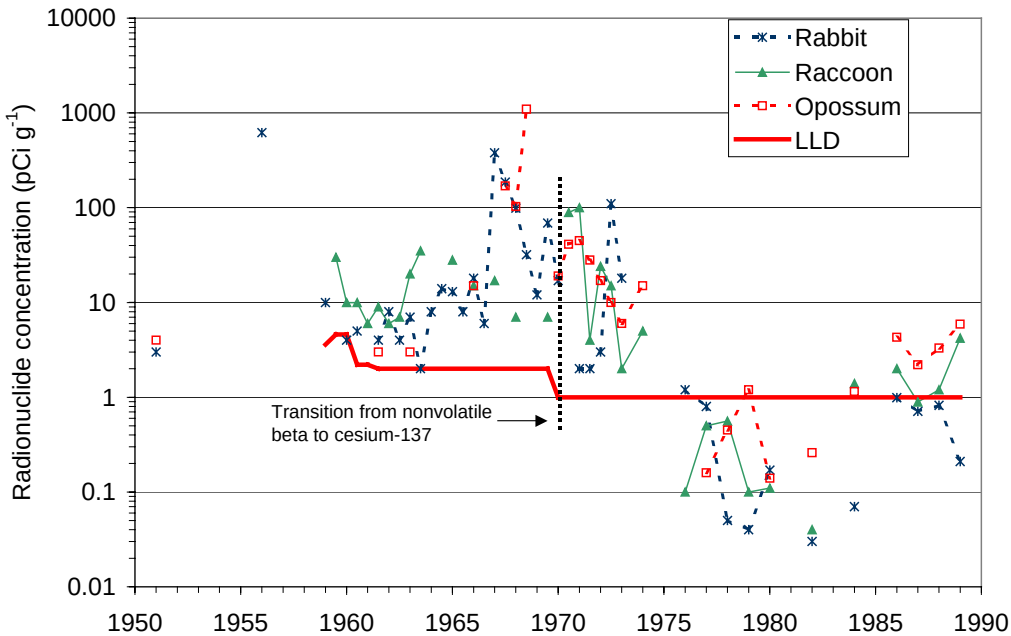

Figure 11-6. Mean radionuclide concentrations reported for rabbits, raccoons, and opossums. Nonvolatile beta (1951 through 1970) and ^{137}Cs (1971 through 1989) concentrations are shown. Link to tabulated figure data.

Significantly elevated concentrations have been measured in animals collected from the vicinity of contaminated areas such as the R-Area seepage basins, which were back-filled in late 1960. Table 11-4 shows mean nonvolatile beta concentrations that were measured in domestic cats and raccoons collected from the vicinity of the R-Area seepage basins and from random onsite locations in 1960. Concentrations in cats are clearly elevated for R-Area locations, and concentrations were also significantly elevated for mice collected from this area. The same trends are not apparent for raccoons. A maximum concentration of 20,400 pCi g^{-1} nonvolatile beta activity was measured in a domestic cat collected from this area during the first half of 1960. Slightly elevated concentrations (significantly lower than in 1960) were also noted for animals collected from locations in the vicinity of the R-Area seepage basins in 1961 after they were back-filled (Du Pont 1962c, 1962d). Though the reports did not clearly specify this, it is presumed that feral cats were occasionally caught during the routine trapping program and referred to as domestic cats.

Table 11-4. Mean Nonvolatile Beta Concentrations[a] (pCi g^{-1}) Measured in Raccoons and Domestic Cats Collected from Locations in the Vicinity of the R-Area Seepage Basins and from Random Onsite Locations in 1960

	January through June		July through December	
Location	Cats	Raccoons	Cats	Raccoons
R-Area	4900	10	4800	8
Random onsite locations	3	10	4	10

[a] Data from Du Pont (1960b and 1961).

Figure 11-7 (note the logarithmic scale) shows the maximum reported radionuclide concentrations documented for trapped terrestrial animals from 1956 through 1989. Nonvolatile beta concentrations are shown from 1956 through 1970, and ^{137}Cs concentrations are shown from 1971 through 1989. Maximum concentrations have been measured in a number of species of animals, including rats, domestic cats, squirrels, opossums, raccoons, rabbits, and bobcats. Almost invariably, the highest concentrations measured in terrestrial animals have been in those animals collected from the vicinity of waste disposal areas.

In 1958, ^{134} Cs and ^{137}Cs were reportedly the radionuclides that comprised the majority of nonvolatile beta activity (75 pCi g^{-1}) in a raccoon (Harvey et al. 1959a). Cesium-137 was reportedly the radionuclide that comprised the majority of activity in muscle tissue for most fur-bearing animals in 1961 (Du Pont 1962d). Raccoons were an exception because ^{65}Zn comprised the majority of activity in muscle tissue. This may be related to the fact that raccoons are aquatic feeders, and ^{65}Zn concentrations were typically higher for aquatic animals than for terrestrial animals. The terrestrial animal trapping program was discontinued in 1990.

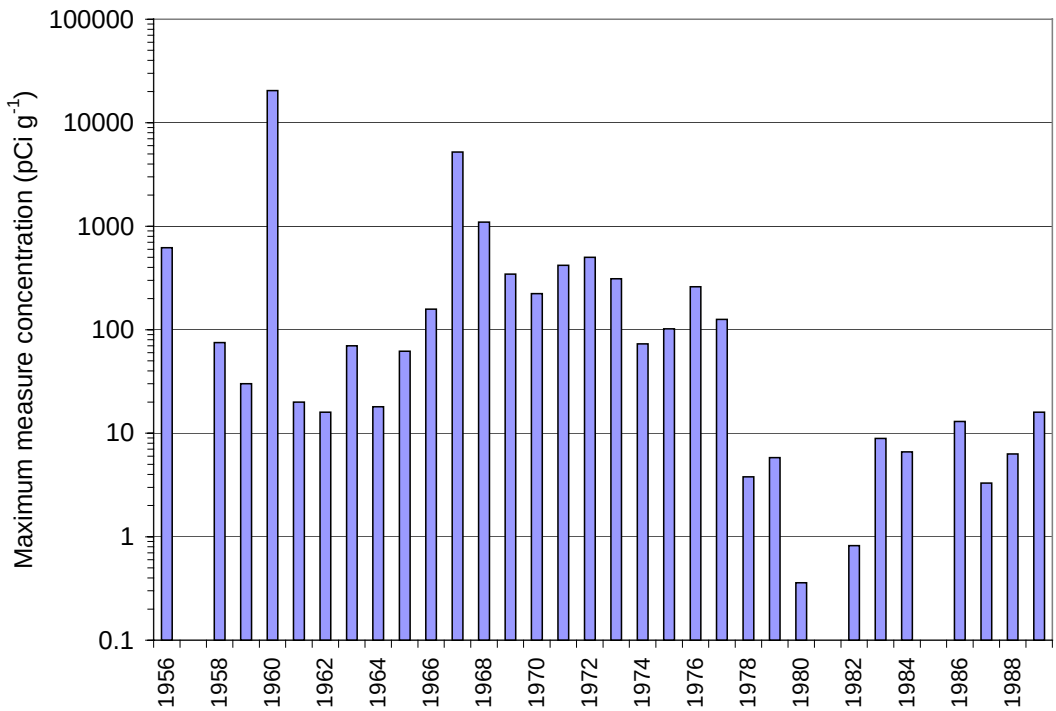

Figure 11-7. Maximum radionuclide concentrations measured in trapped fur-bearing animals. Nonvolatile beta (1956 through 1970) and ^{137}Cs (1971 through 1989) concentrations are shown. Link to tabulated figure data.

RADIONUCLIDE CONCENTRATIONS MEASURED IN WATERFOWL

Waterfowl were also routinely collected by the SRS, primarily from Par Pond. In 1972, the program was expanded to include Steel Creek, and concentrations were reported for waterfowl collected from Pond B beginning in 1976. The types of waterfowl that were collected included carnivorous (animal-eating) species, such as gulls and grebes, omnivorous (plant- and animal-eating) species, such as coots, and herbivorous (plant-eating) species, such as ring-neck, mallard,

ruddy, wood, scaup, teal, and bufflehead ducks. We have compiled data reported for ring-neck ducks and coots collected from Par Pond that have been reported in the various annual environmental monitoring reports.

Nonvolatile beta concentrations were sporadically reported before 1962. Maximum concentrations ranged from 15 to 80 pCi g^{-1}. Cesium-137 and ^{65}Zn were the primary radionuclides measured in waterfowl flesh in 1962 (Du Pont 1963d). Elevated ^{65}Zn concentrations were reported from 1962 through 1964 (maximum of 210 pCi g^{-1} in a coot), after which concentrations were reported as below the LLD. In general, ^{137}Cs concentrations appear higher in carnivorous and omnivorous species than in herbivorous species. Brisbin (1982) reported higher ^{137}Cs concentrations for coots than for any of the several species inhabiting Par Pond during studies conducted in 1971 and 1972. Figure 11-8 shows average ^{137}Cs concentrations reported for coots (omnivorous) and ducks (herbivorous) collected from Par Pond from 1962 through 1988. Concentrations are consistently higher for coots. Significantly elevated concentrations were reported for coots in 1963 and 1964, and concentrations appear to have generally decreased during this time period. Concentrations similar to those shown in Figure 11-8 were reported for coots collected from Par Pond in 1971–1972 and 1975–1976 (Brisbin 1982).

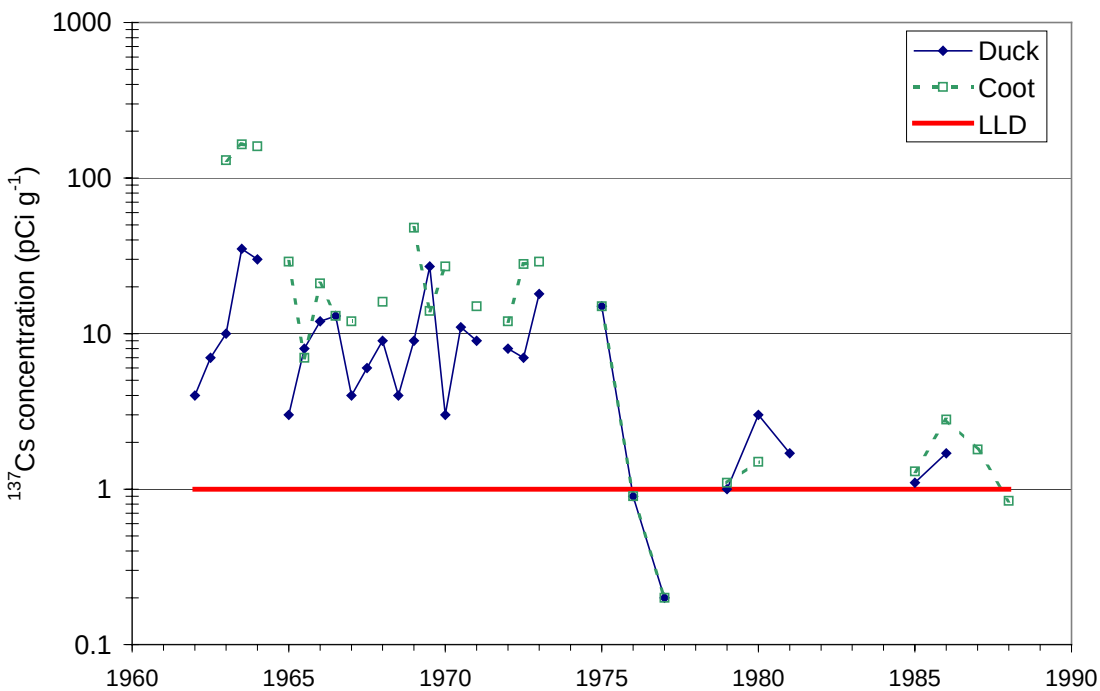

Figure 11-8. Mean ^{137}Cs concentrations measured in ducks and coots collected from Par Pond from 1962 through 1988. Link to tabulated figure data.

There is evidence to suggest that waterfowl residing on more highly contaminated ponds or streams (such as Pond B and Steel Creek) may have accumulated greater burdens of ^{137}Cs than waterfowl residing on Par Pond. Table 11-5 shows concentrations measured in waterfowl collected from Par Pond as well as concentrations measured in waterfowl collected from Pond B and Steel Creek.

Table 11-5. Mean ^{137}Cs Concentrations[a] (pCi g^{-1}) Measured in Ducks and Coots Collected from Par Pond, Steel Creek, and Pond B

	Ducks		Coots	
Year	Par Pond	Steel Creek	Par Pond	Pond B
Jan–June 1972	8	130		
July–Dec 1972	7	49		
1973	18	67		
1974		74		
1975	15	20		
1987			1.8	73.8
1988			0.8	54

[a] Data from Ashley and Zeigler (1974, 1975, 1976, 1978a), Mikol et al. (1988b), and Davis et al. (1989b).

Fendley et al. (1977) reported ^{137}Cs concentrations for wood ducks kept in an enclosure on Steel Creek in 1974. Twenty male and female ducks accumulated an average concentration of 16.6 pCi g^{-1} after 30 days. The time to reach equilibrium was estimated at 17 days, with a biological half-time estimate of 5.6 days.

ELECTRONICALLY COMPILED WILD GAME DATA

The various data summarized in this chapter are electronically compiled in two Microsoft Excel® workbooks. One workbook (Ch11-Figure_data.xls) contains the figures depicted in this chapter (including the addendum to this chapter) as well as the tabulated data that were used to produce the figures. In this workbook, there is a separate worksheet for each figure and one worksheet that contains the tabulated data for all of the figures. The second workbook (Ch11-All_data.xls) contains the data that have been tabulated from various environmental monitoring reports, aperture card printouts, and raw data sheets. The workbook contains several named worksheets that include brief summary of the compiled data.

Table 11-6 summarizes the data that have been electronically compiled for wild game collected as part of the routine environmental monitoring program maintained by the SRS. It also provides the names of the individual spreadsheets in which these data are compiled (including a brief description of the data).

Table 11-6. Description of Data That Have Been Electronically Compiled for Wild Game

Workbook name	Worksheet name	Brief description of data
Ch11-Figure data.xls	Figures 11-1 through 11-9	Each worksheet contains a separate figure depicted in this chapter
	Figures 11A-1 through 11A-4	Each worksheet contains a separate figure depicted in the addendum to this chapter
	Data for figures	This worksheet contains the tabulated data for each of the figures
Ch11-All data.xls	Cs-137 (SRS)	Summary of ^{137}Cs concentrations in deer collected from the Savannah River Plant
	Cs-137 (SCCP)	Summary of ^{137}Cs concentrations in deer collected from the South Carolina Coastal Plain
	Radioiodine	Summary of Iodine concentrations measured in SRP deer
	Sr-89,90	Summary of ^{89,90}Sr concentrations measured in SRP deer
	Pu-238,239	Summary of ^{238,239}Pu concentrations measured in SRP deer
	H-3	Summary of tritium concentrations measured in SRP deer
	Annual report, 1964—1976	Data from Environmental Monitoring at the Savannah River Plant annual reports, 1964—1976
	Annual report, 1971—1983	Data from Environmental Monitoring in the Vicinity of the Savannah River Plant, 1971—1983
	Field-lab compare	Comparison of field and laboratory ^{137}Cs measurements
	% verified	Percent of field measurements verified by laboratory measurements
	U. of Tennessee	Results of analysis of deer muscle samples analyzed by the University of Tennessee
	A.C. 1970–1981	Summary of aperture card printouts for 1970–1981
	Raw field data	Raw field data for 5 years
	Furbearers	Summary of concentrations reported for other wild game (hog, rabbit, opossum, raccoon)
	Waterfowl	Summary of concentrations reported for waterfowl collected from Par Pond

USEFULNESS AND LIMITATIONS OF THE WILD GAME DATA FOR DOSE RECONSTRUCTION

There are a number of factors that impact how the wild game data may be used during subsequent phases of the dose reconstruction project. These factors include the availability of sufficient original monitoring data sets to verify reported summary data and evaluate spatial and temporal trends, as well as the ability to distinguish between Site releases of contaminants and other sources of the same contaminants in the environment (i.e., establish appropriate background concentrations).

With few exceptions, ^{137}Cs, ^{89,90}Sr, and ^{131}I concentrations reported for deer and other wild game in the monitoring reports have been consistent with concentrations provided in aperture card printouts (1970–1981) and raw data sheets (1965, 1966, 1982, 1984, and 1985). Some slight differences were apparent, but this appears to be the result of incomplete original data for some years (e.g., 1965 and 1966) and occasionally illegible aperture card printouts. In addition, it was not always clear how reported average concentrations were calculated with respect to "background" and "less than" concentrations. In general, though, the aperture card printouts and raw data sheets provided consistent verification of the values reported in the monitoring reports.

Comparisons of field and lab data have shown that field estimates using a portable NaI(Tl) detector provided accurate measurements. Additionally, measurements made by the SRS are generally consistent with measurements made by the University of Tennessee, although information enabling a comparison of measurements for individual deer is unavailable. Some differences were evident, but this appears to have resulted from analysis by the University of Tennessee of only a subset of the total number of samples analyzed by the SRS. There is also no evident disparity between concentrations measured in South Carolina Coastal Plain (SCCP) deer by the University of Georgia from 1968 through 1983 and concentrations measured in SCCP deer by the SRS from 1985 through 1991.

The reported ^{137}Cs concentrations provide a good estimate of concentrations in the SRS deer population because large numbers of deer have been monitored annually. However, an arithmetic average is likely inappropriate for describing the central tendency of ^{137}Cs concentrations in the deer (and other wild game) population because concentrations appear log-normally distributed. Geometric means and geometric standard deviations or medians and percentiles may be more appropriate descriptors of the data. However, arithmetic mean concentrations likely error on the conservative side (i.e., provide an overestimate) of the central tendency for cesium concentrations in deer because the distributions are generally skewed to the right (e.g., log-normal). Examination of entire data sets (i.e., values for individual deer) has only been completed for 1982, 1984, and 1985 at this time; entire data sets have not been located for other years.

The maximum measured ^{137}Cs concentrations from 1965 through 1991 may be most useful for estimating maximum potential exposure to hunters or their families who consumed the deer or hog meat. The maximum ^{137}Cs concentrations measured in SRS deer (204 pCi g^{-1}) are not significantly elevated above concentrations measured at other locations in the southeastern U.S. (153 pCi g^{-1}), though this comparison is of limited use because different soil characteristics can result in different rates of ^{137}Cs uptake.

There is also some question about the appropriateness of using SCCP deer as background indicators because the soil characteristics for the SCCP likely result in greater uptake for these deer than for SRS deer. It is also difficult to establish with certainty the number of deer that may

have been impacted by Site operations. To completely assess this exposure pathway, it may be necessary to establish an acceptable method for quantifying appropriate background concentrations as well as the number of deer that may have accumulated excess concentrations of ^{137}Cs resulting from SRS operations.

Furthermore, very limited data are available for wild game and only nonvolatile beta concentrations were reported before 1965. Public exposure to deer from the SRS would likely have been minimal before 1965, when public hunts on the SRS were initiated. However, since the precise range and mobility of the deer residing on the SRS is not known, it is certainly possible that some animals spending the majority of time on the SRS migrated offsite and were harvested by hunters. To assess this exposure pathway, it would be necessary to estimate ^{137}Cs concentrations based on a very small number of measured nonvolatile beta concentrations.

Completely evaluating exposure pathways involving wild game from the SRS may also require acquiring additional data. Major hunting areas and waterfowl migration patterns are examples of additional information that might assist with future phases of the SRS Dose Reconstruction Project. Because the range and eating habits of wild game residing at the SRS are not known with any certainty, though, it will be quite difficult to use the wild game data for definitive source term verification or model validation.

In summary, the information provided in the documents that have been reviewed is potentially quite useful for quantifying radionuclide concentrations, particularly ^{137}Cs, in deer and other wild game at the SRS. In general, the mean concentrations measured in SRS deer have been lower than mean concentrations measured in deer from locations thought to be representative of background concentrations. In addition, with a few exceptions, elevated radioiodine concentrations measured in deer thyroids have consistently corresponded to periods of weapons testing. It can then be concluded that the majority of deer from the SRS reflect radionuclide concentrations that have resulted from atmospheric weapons tests.

However, there is also evidence to suggest that a few individual animals have accumulated SRS-derived radionuclides. This is reflected by the consistently higher maximum ^{137}Cs concentrations measured in SRS deer relative to SCCP deer since 1968. It is also reflected by the significantly elevated radionuclide concentrations reported for other fur-bearing animals collected from areas of known contamination. Elevated ^{129}I concentrations have also been measured in individual deer for years during which weapons tests were not conducted (e.g., 1984, 1989, and 1991). The wild game data are likely most useful for establishing the maximum concentrations of radionuclides, ^{137}Cs in particular, to which hunters may have been exposed, but it will be necessary to establish an acceptable methodology for estimating appropriate background concentrations.

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ADDENDUM 11A

FREQUENCY DISTRIBUTIONS FOR FIELD ANALYSES OF DEER

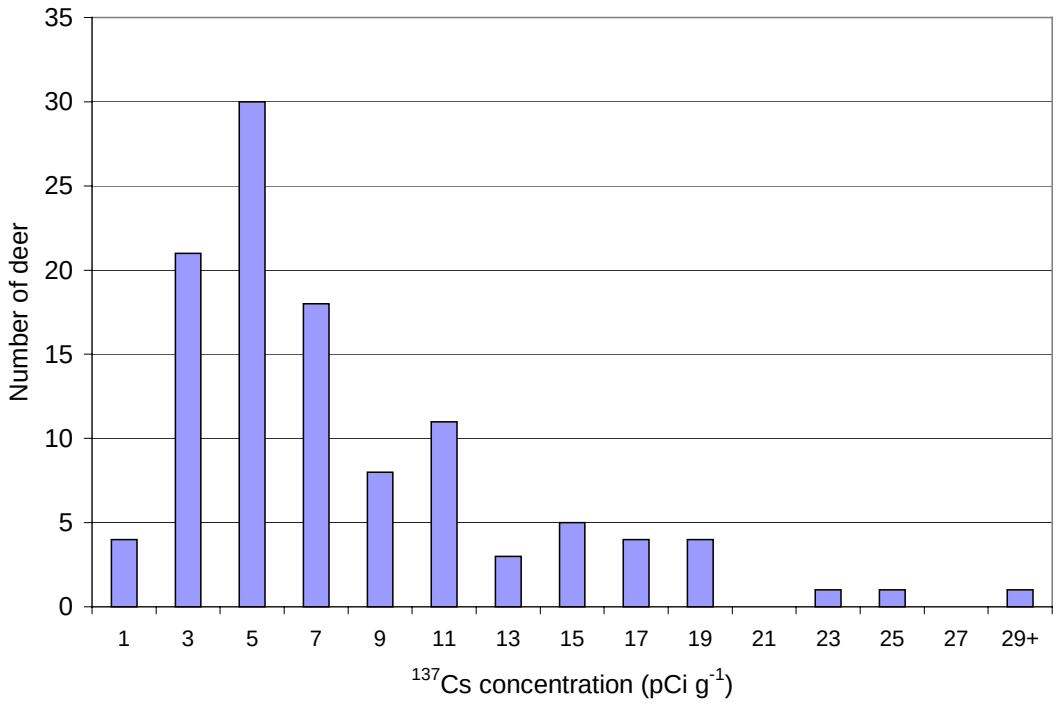

Figure 11A-1. Frequency distribution for 111 field analyses of deer collected from the SRS in 1965 (geometric mean = 5.7, geometric standard deviation = 2.1). Link to tabulated figure data.

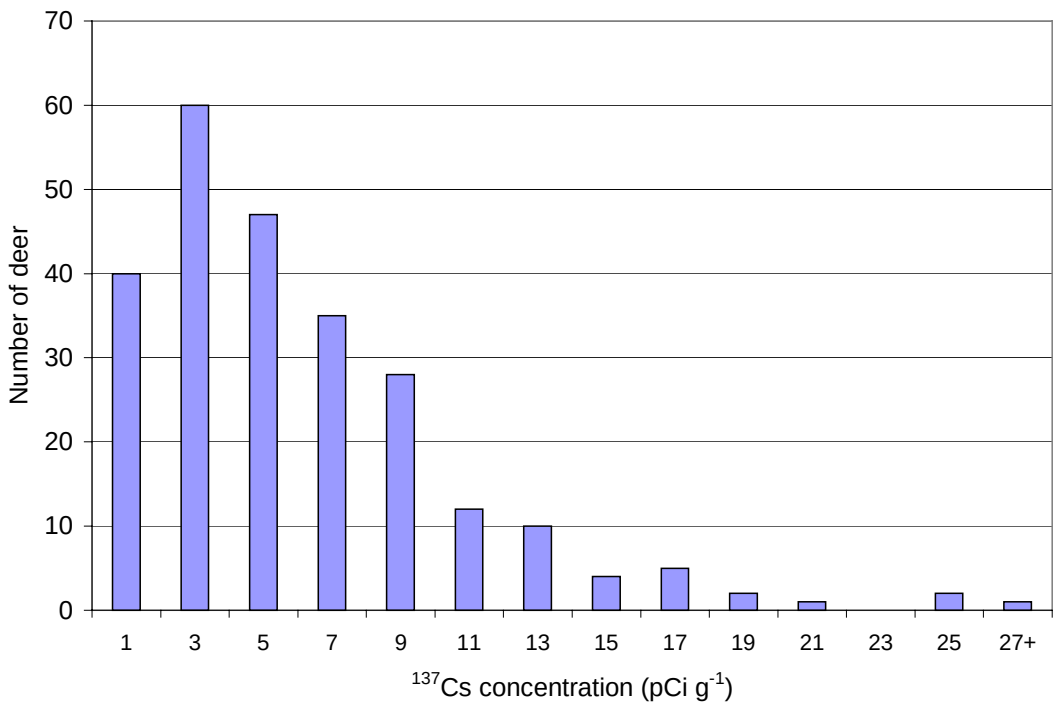

Figure 11A-2. Frequency distribution for 247 field analyses of deer collected from the SRS in 1966 (geometric mean = 3.7, geometric standard deviation = 2.4). Link to tabulated figure data.

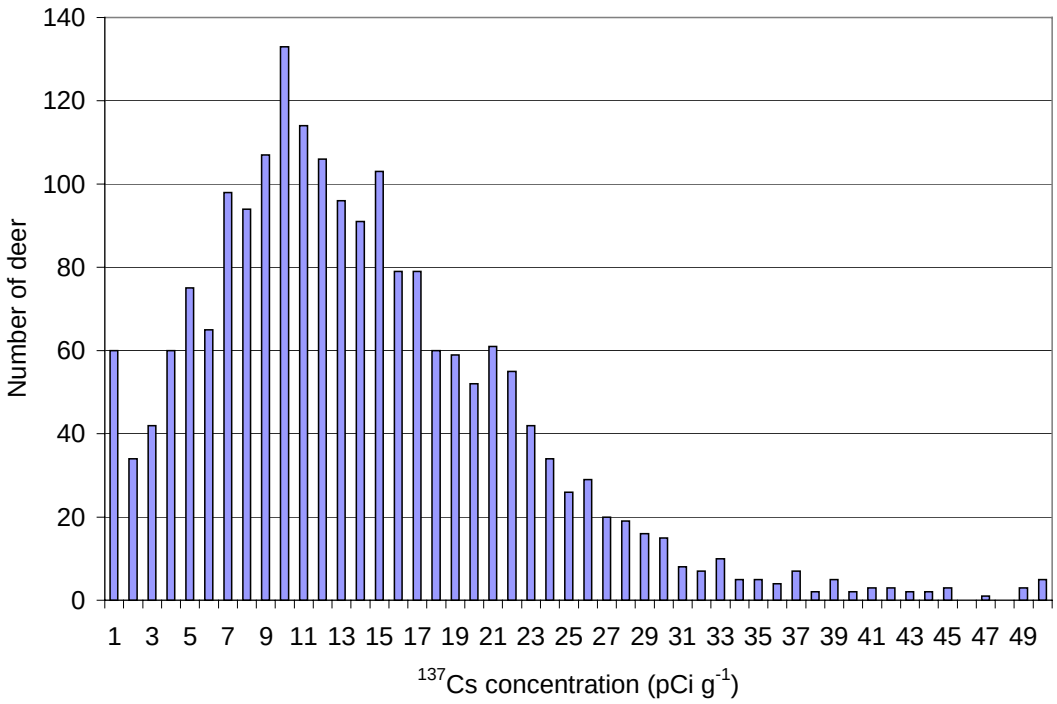

Figure 11A-3. Frequency distribution for 2001 field analyses of deer collected from the SRS in 1982 (geometric mean = 11.3, geometric standard deviation = 2.1). Link to tabulated figure data.

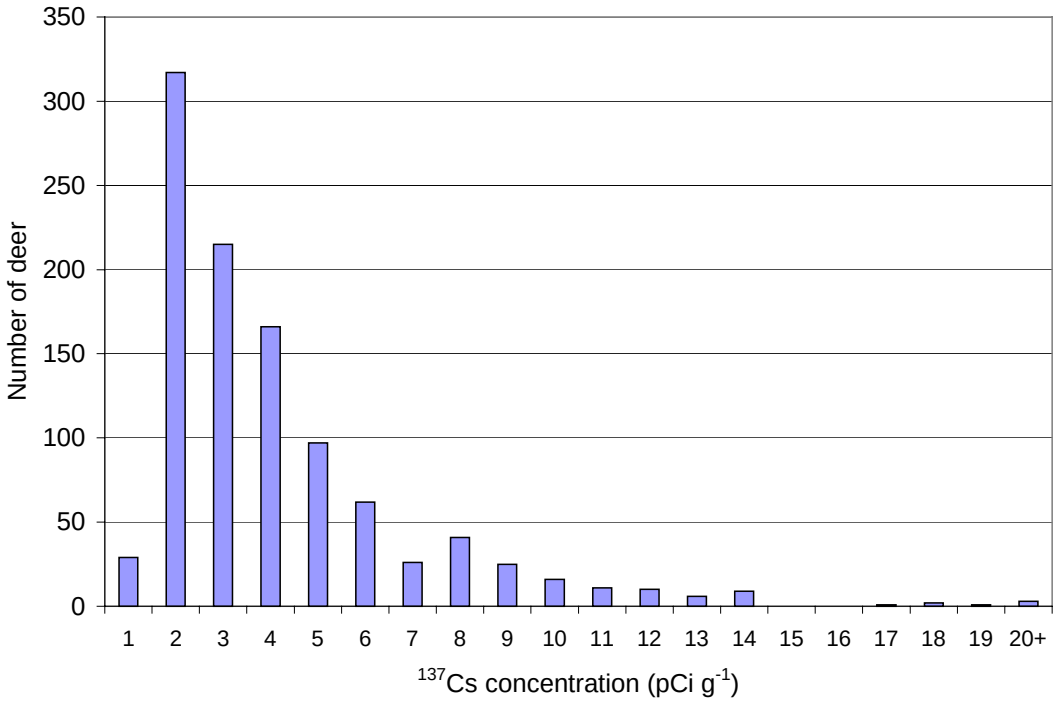

Figure 11A-4. Frequency distribution for 1037 field analyses of deer collected from the SRS in 1984 (geometric mean = 3.5, geometric standard deviation = 1.8). Link to tabulated figure data.

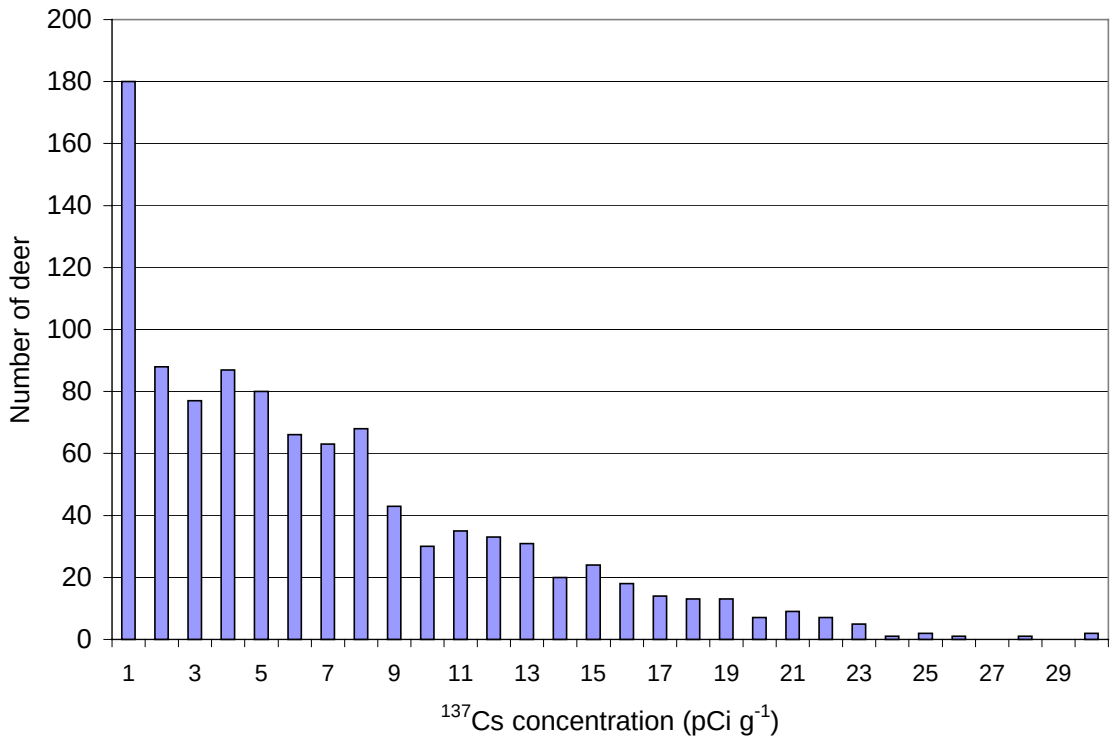

Figure 11A-5. Frequency distribution for 1018 field analyses of deer collected from the SRS in 1985 (geometric mean = 4.7, geometric standard deviation = 2.6). Link to tabulated figure data.

CHAPTER 12.1

RADIONUCLIDES IN SEDIMENTS

ABSTRACT

This chapter discusses radionuclide concentrations measured in sediment samples collected on or in the vicinity of the Savannah River Site (SRS). We reviewed the history of sediment monitoring at the SRS and evaluated these data with regard to their potential usefulness for dose reconstruction.

We compiled and examined sediment data to determine their potential value in source term verification, model validation, and direct exposure assessment. Uranium data collected from Steed's Pond, which received liquid effluents from the M-Area via Tim's Branch, may be useful for source term verification and model validation of uranium releases from the M-Area. However, the potential usefulness of the Steed's Pond data, which are limited in number, is bounded by spatial resolution. Results of measurements of long-lived radionuclides (uranium, plutonium, and ^{137}Cs) in streambeds do not appear useful because samples do not represent the entire streambed and demonstrate no spatial trends because of the complex sediment deposition processes involved. Appendix K further discusses potential uses for environmental monitoring data.

INTRODUCTION

Long-lived contaminants, deposited by releases to air and SRS streams, tend to accumulate in bottom sediments. Deposits may be in distinct layers, for example, after discrete release events; thus, they preserve the temporal history of liquid releases to a stream, pond, or river. Deposition patterns often show concentrations decreasing as a function of distance from the source of contamination, thus, revealing spatial trends and defining the extent of contamination.

This chapter summarizes reported information regarding radionuclide concentrations in sediments on or in the vicinity of the SRS. We examined several sets of routine semiannual and annual environmental monitoring reports, prepared by the SRS contractor and spanning the years 1953 through 1992. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series.

Data usefulness was considered in terms of the following:

- The contaminant is a key contaminant (i.e., screened in Task 3 of Phase I of the dose reconstruction to potentially contribute significantly to dose)
- The contaminant was monitored during the period of interest (i.e., 1951 through 1990)
- Contaminant concentrations were above detection limits
- The results demonstrate expected trends (spatially and temporally)
  - Concentrations were generally higher close to the source
  - Concentrations in sediment cores are often present in discrete layers.

Data that are available for sediments include gross alpha activity; nonvolatile beta activity; and concentrations of gamma-emitting radionuclides, uranium and plutonium. Appendix A details analytical and counting procedures for sediment samples. We provide the summary results for the onsite streams and Savannah River locations in this chapter.

MEASUREMENT OF CONTAMINANTS IN SEDIMENTS

Routine sediment monitoring began in 1951 with the collection of stream sediments for use in establishing background levels of radioactivity at 13 locations along the inner perimeter and 17 locations along the outer perimeter of the SRS (Reinig et al. 1953; Reinig 1952). The sampling locations included stations on the Upper Three Runs Effluent System (including Tim's Branch), Four Mile Creek, Steel Creek, Pen Branch, Indian Graves Branch, Lower Three Runs, and the Savannah River. Sample collection methods are not detailed, although Reinig (1952) states that "approximately 1000 boring samples were analyzed," implying that a soil-coring device of unspecified diameter was used. Samples from the 1951 survey varied in weight from 0.7 to 2.5 grams (g). The samples were analyzed for gross alpha and gross beta activity and plotted as activity versus location. This sampling, analysis, and reporting regime continued in 1953 following the start up of the plant (Albenesius 1954). Table 12.1-1 summarizes general features about the sediment sampling program at the SRS through 1991, including the cessation of a routine sediment sampling program from 1963 through 1977.

Table 12.1-1. Sediment Sampling Summary at SRS[a]

Years	Site monitoring areas	No. of samples	Measured
1951-1953	At mouth of Upper Three Runs, Four Mile Creek, Steel Creek, Pen Branch, Indian Grove Branch. Lower Three Runs	30	Gross alpha; gross beta
1954	Same as above but sampling along stream length	46	Alpha (U+Pu); nonvolatile beta
1955-1961	TNX effluent ditch added to above; Table 12.1-2 and Figures 12.1-1, 12.1-2, 12.1-3 summarize locations and data	48	Alpha (U+Pu); nonvolatile beta
1963-1977	*No routine sediment monitoring program; resumed in 1977*		
1977-1991	Routine sediment monitoring resumed in site streams and in Savannah River	15	Gamma-emitting radionuclides, ^{238}Pu, ^{239}Pu, ^{90}Sr
1967, 1981, 1984	Special study: Steed's Pond receiving M-Area effluent	6-15	Uranium
1974-1976	Special study: Savannah River swamp		^{137}Cs, ^{238}Pu, ^{239}Pu, ^{90}Sr

[a] From annual environmental monitoring reports (see Table 7-1).

In 1954, the number of locations sampled for sediment increased to 46 along the five major stream systems that carry waste into the Savannah River (Upper Three Runs, Four Mile Creek, Steel Creek, Pen Branch, and Lower Three Runs); Tim's Branch (which carries plant waste into Upper Three Runs); and the Savannah River. The sampling design was changed from perimeter sampling to sampling along the length of each stream system (Horton 1954, 1955). Approximately 80% of the weekly samples were analyzed for alpha activity (uranium and plutonium) using tri-n-butyl phosphate (TBP) extraction and scintillation counting (see

Appendix A). All of the samples were analyzed for nonvolatile beta activity. The maximum and average uranium or plutonium activity for each 6-month period and stream location was reported in the semiannual environmental monitoring reports as disintegrations per minute per gram.

In 1955, the sampling network was changed to include the TNX effluent ditch, which empties effluent directly into the Savannah River (Alexander and Horton 1956; Horton and Mealing 1956). Approximately 70% of the weekly samples were analyzed for uranium or plutonium activity. All were analyzed for nonvolatile gross beta activity. A similar sampling strategy continued through 1960 (Mealing et al. 1958, Harvey et al. 1959a, 1959b, and Du Pont 1959, 1960), but results of TBP extraction and scintillation counting were reported as gross alpha activity (curies per gram) instead of uranium and plutonium activity. The sampling designs for 1955 through 1960 are outlined in Table 12.1-2, Figures 12.1-1 and 12.1-2 show water and sediment sampling locations from 1954 to 1958 and from 1959 to 1960, respectively.

Marter and Boulogne (1961) provide some information on sediment sample collection methods. This document states that stream mud samples, taken at all river and stream locations, were generally collected close to the bank and "bear no relation to localized 'silt out'."

In 1961, the sediment sampling program consisted of weekly samples collected from 16 stream locations and weekly and monthly samples collected from 7 river locations (as shown in Figure 12.1-3). Monthly sediment samples from five river locations (2, 3, 5, 8, and 9) and weekly samples from two river locations (10 and 11) and two stream locations (3 and 4) were analyzed for TBP extractable alpha activity.

Table 12.1-2. Sediment Sampling Design (1955–1960)

Sampling area	Number of sampling locations					
	1955	1956	1957	1958	1959	1960
Tim's Branch	5	4	3	4	4	3
Upper Three Runs	4	4	4	4	4	4
Four Mile Creek	7	7	7	7	5	5
Pen Branch	3	3	3	3	2	2
Steel Creek	5	4	4	4	3	3
Lower Three Runs	3	3	3	4	5 [c]	5[c]
TNX Effluent	1	1	1	–	–	–
Savannah River	10	10	10	11	11	11
Collection frequency	Weekly[a]	Weekly	Monthly[b]	Monthly[b]	Monthly[b]	Weekly[d]
Total number of samples analyzed for nonvolatile beta	2209	1350	475	520	520	1166
Total number of samples analyzed for uranium and plutonium	1446	1036	339	454	454	1166

[a] Sediments collected biweekly at 8 of 10 locations along the Savannah River beginning in July 1955.

[b] Sediments collected monthly, except samples were collected weekly at one location on the Savannah River, where it intersects Highway 301.

[c] Includes three sample locations at Par Pond.

[d] Sediments collected monthly on the Savannah River, except at two locations, where samples collected weekly.

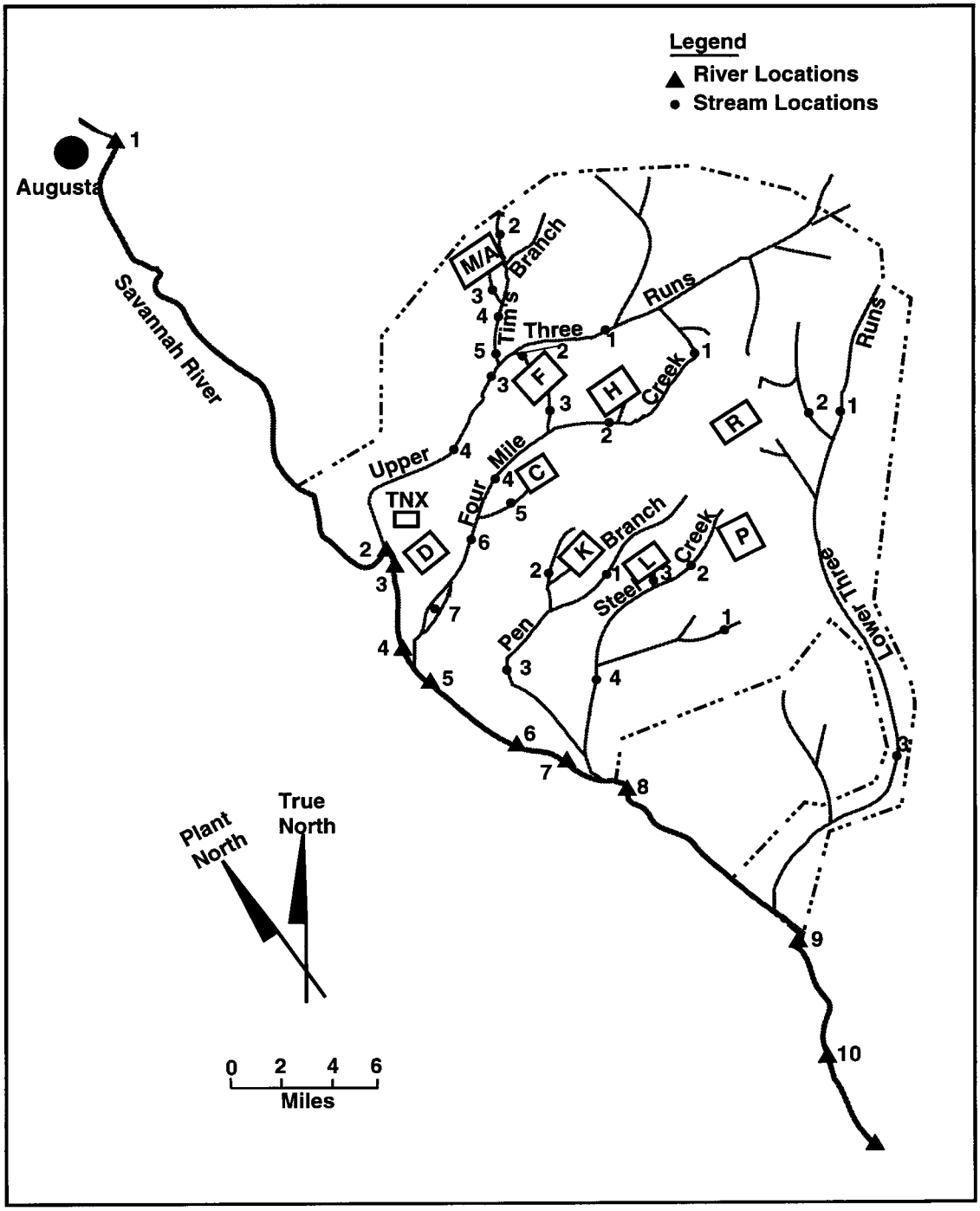

Figure 12.1-1. Map of SRS routine stream and river sampling locations used in the sediment sampling programs from 1954 through 1958.

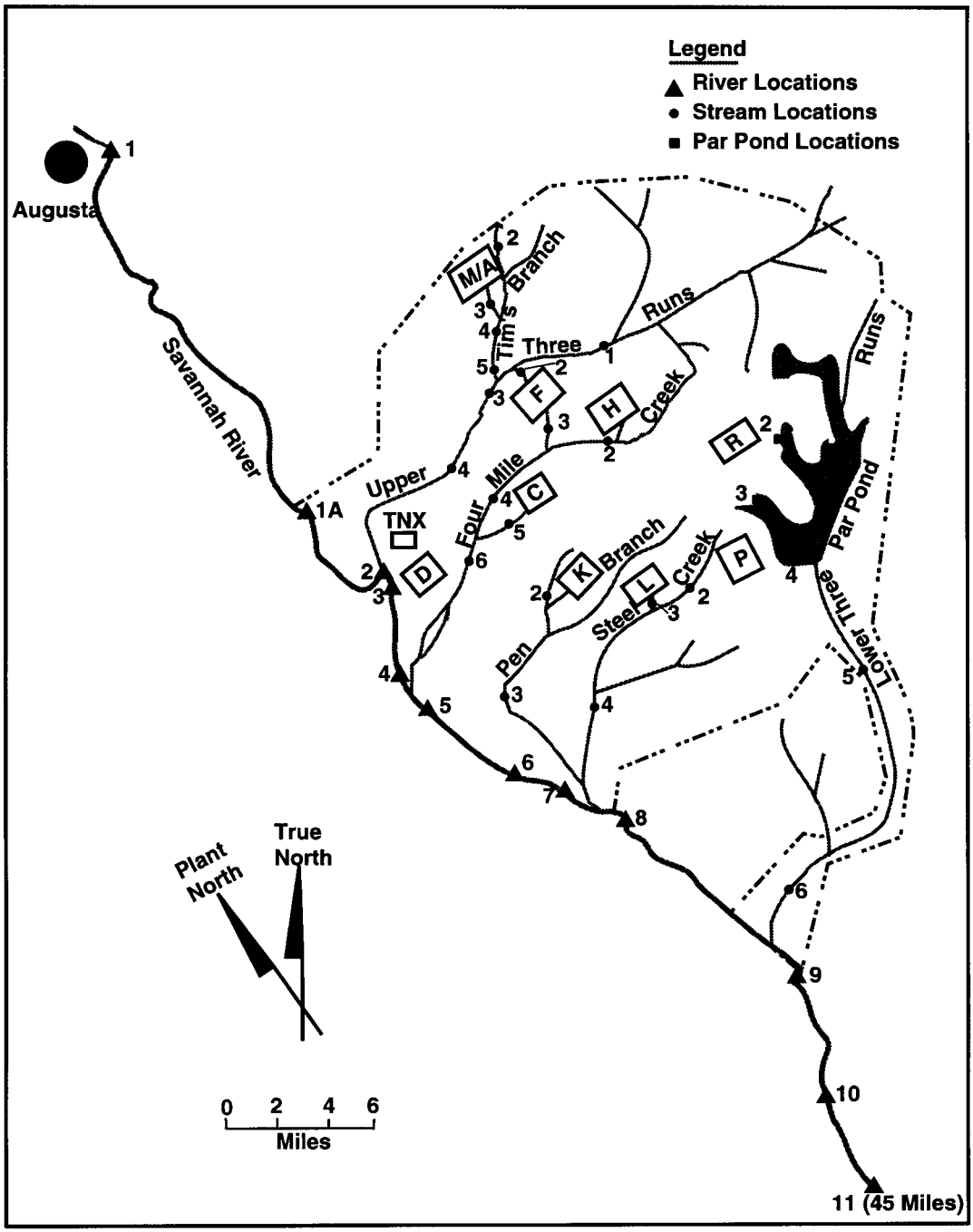

Figure 12.1-2. Map of SRS routine stream and river sampling locations used in 1959 and 1960 sediment sampling programs.

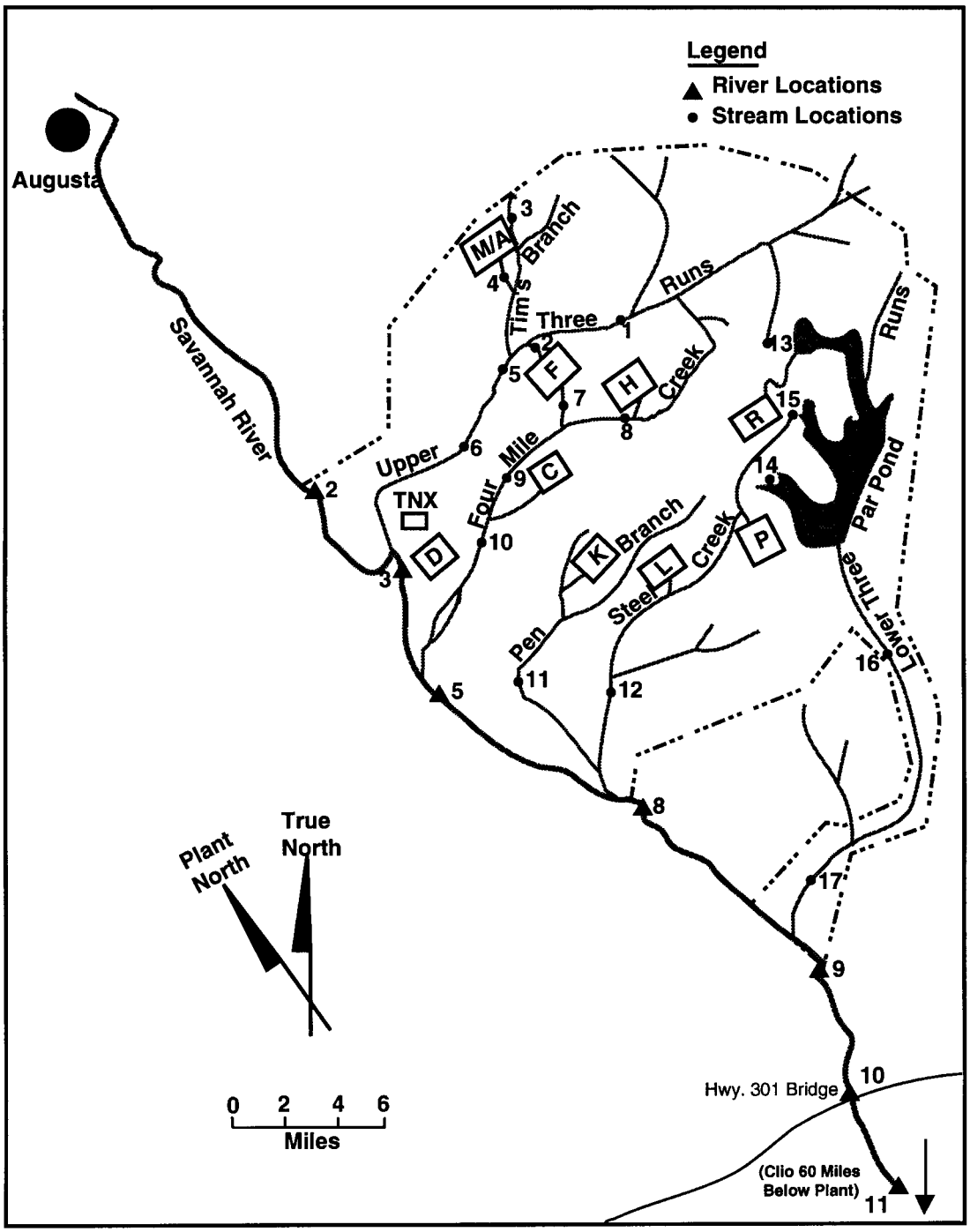

Figure 12.1-3. Map of SRS routine stream and river sampling locations used in 1961 and 1962 sediment sampling programs.

In 1963, it was proposed that all routine sediment sampling be eliminated on the basis that "the information gained with present methods is meaningless" (Johnson 1963). Routine sediment sampling was discontinued from 1963 through the mid-1970s. Collection of sediments was resumed in 1974 (Ashley and Zeigler 1976). Ten sediments cores, 25 cm deep, were collected from each of 10 trails transecting the Savannah River swamp, where radioactive materials from

Savannah River Plant (SRP) releases in 1960s were deposited in about 1.7 mi^2 of offsite swamp downstream from the SRP. The cores were divided into two increments (from 0 to 7.6 cm and from 7.6 to 25 cm), which were composited by increment from each location (trail) for plutonium analysis. In addition, 12 sediment samples were collected in the Savannah River, starting above the Site and continuing to the Savannah, Georgia harbor. Samples were analyzed for gamma-emitting radionuclides (^{137}Cs in particular), ^{238}Pu, and ^{239}Pu. In 1976, samples were also analyzed for ^{90}Sr (Ashley and Zeigler 1977).

Sediment collections in the Site streams resumed in 1977 to obtain an estimate of the maximum accumulation of radioactivity released from the Site (Ashley and Zeigler 1978). The 1977 and subsequent annual reports state that stream sediment sampling locations were designed to obtain an estimate of the maximum accumulation and are, therefore, not representative of the entire stream bed. The sampling approach called for collecting approximately the top 8 cm of sediment in areas where fine sediment accumulates. The sediments in the Savannah River continued to be sampled, although sediment sampling in the Savannah River swamp was reduced in frequency in 1978 because the results of these surveys showed no significant changes in radiological conditions. At that time it was decided to conduct comprehensive surveys of the swamp every 5 years, including sediment, vegetation, animals, fish, and thermoluminescent dosimeter (TLD) measurements unless additional surveys were warranted by changes in the annual TLD radiation measurements (Ashley and Zeigler 1979).

Several nonroutine sediment sampling studies have been conducted over the years. Three studies that may be of particular importance to the dose reconstruction are the surveys of Steed's Pond sediments for uranium conducted in 1967, 1981, and 1984 (Ashley and Zeigler 1982, Zeigler et al. 1986). Before 1985, when an advanced liquid waste treatment facility was put into operation, untreated liquid effluents containing uranium were released to Tim's Branch via a drainage ditch. Tim's Branch flowed into Steed's Pond, which acted as a settling basin and accumulated uranium in the sediments. Steed's Pond originally had an area of approximately 14 acres and had a spillway that partially collapsed in the early 1960s. The spillway was only partially removed, as evidenced by the retention of a few acres of water. The first survey was conducted on the sediments that were exposed. At that time, six 18-in. sediment cores were collected, separated into 6-in. segments, and analyzed for uranium. In the early 1970s the spillway was repaired and the pond returned to a surface area of about 11 acres.

The spillway was opened in January 1981, and the pond was drained for turtle studies. While the pond was drained, six 6-in. sediment cores were collected and analyzed for uranium. In August 1984, the spillway collapsed and the pond was drained completely. Fifteen 24-in. sediment cores were collected. In addition, 15 samples were taken between Steed Pond and the M-Area effluent ditch. The samples were divided into increments of 0–6 in., 6–12 in., and 12–24 in., and they were analyzed for uranium as well as for chromium, copper, lead, nickel, sodium, aluminum, ammonia, chlorine, nitrate, and thorium. The uranium concentrations in the 1985 sediment samples were analyzed using more sophisticated radiochemical techniques than TBP extraction.

The Savannah River Environmental Laboratory (SREL) did extensive research on the behavior of ^{137}Cs within the Steel Creek flood plain. This research, which is summarized in Carlton et al. (1992), addresses the availability of cesium in the sediments for plant uptake and its subsequent transfer through the food chain.

Measurements of Uranium in Sediments

Greater than 97% of the gross alpha activity released from SRS facilities occurred from the M-Area (Evans et al. 1992). Essentially all of the alpha activity in the process effluent in the M-Area has been identified as depleted uranium. Most of this activity was released between 1966 and 1969 during the development of a new uranium/aluminum bonding process for fuel and target assemblies. Because the effluent from the M-Area was released into Tim's Branch, the dose reconstruction should focus on uranium data collected from sediments of this stream.

Steed's Pond acted as a settling basin for Tim's Branch, as evidenced by the accumulation of sediment of about 3 ft of sediment in the vicinity of the Steed's Pond spillway. Water entering the pond had a residence time of about 3 days (Evans et al. 1992). The first survey for uranium, conducted in February 1967, indicated that the top 6 in. of sediment contained greater than 90% of the uranium activity detected in the cores (Ashley and Zeigler 1982). The uranium concentrations in the upper 6 in. ranged from 20 to 531 pCi g^{-1}. Concentrations in the second 6-in. increment had decreased to about one-eighth the surface value. The uranium concentration was near background concentrations (approximately 5 pCi g^{-1}) between 12 and 24 in.

In 1981, the six 6-in. cores collected from Steed's Pond had an average uranium concentration of 170 to 700 pCi g^{-1}, about twice as great as the 1967 results. Evans et al. (1992) attributes this increase to additional uranium released to the effluent since 1966. Approximately 10 Ci of uranium was released to Tim's Branch from 1954 through 1966. An additional 15 Ci was released from 1966 through 1981.

The 1984 sediment sampling of Steed's Pond was more extensive than previous studies. Figures 12.1-4 and 12.1-5 show that uranium activity was generally higher in the sediments of Steed's Pond than in sediments above the pond, and that most of the uranium is in the upper 6 in. of sediment. However, in the 1984 survey, it was estimated that 54% of the uranium was in the upper 6 in., compared to 90% in the 1967 survey (Evans et al. 1992). This decrease may be attributed to an increase in sediment deposition in the pond after the spillway was repaired in the early 1970s. Most of the cores show a pattern of highest concentration in the first 6 in. and decreasing concentrations as a function of sediment depth. However in four of the cores, the highest uranium concentration occurred in the segment below 6 in. Evans et al. (1992) states that some of the processes that could be involved in determining depth distributions include deposition, erosion, and redeposition; size sorting during deposition; changes in the flow regime; flood events; manmade changes; changes in water level in a pond; biological processes; alterations in water chemistry; and unplanned releases.

There also appears to be a pattern of increasing uranium concentrations in cores as a function of distance from the pond inlet (Figure 12.1-4). That is, the lowest numbered samples, collected near the inlet, have the lowest concentrations of uranium. In contrast, the highest numbered samples, collected near the spillway, have the highest concentrations of uranium. This makes sense since the deepest part of the pond, where the heaviest deposition of sediments is most likely to occur, is near the spillway.

Comparisons of the 1967 and 1981 results from Steed's Pond sediment studies cannot be made with the 1984 results because more sensitive and more accurate nonradiological methods were used to measure the uranium concentrations in sediments in the 1984 study.

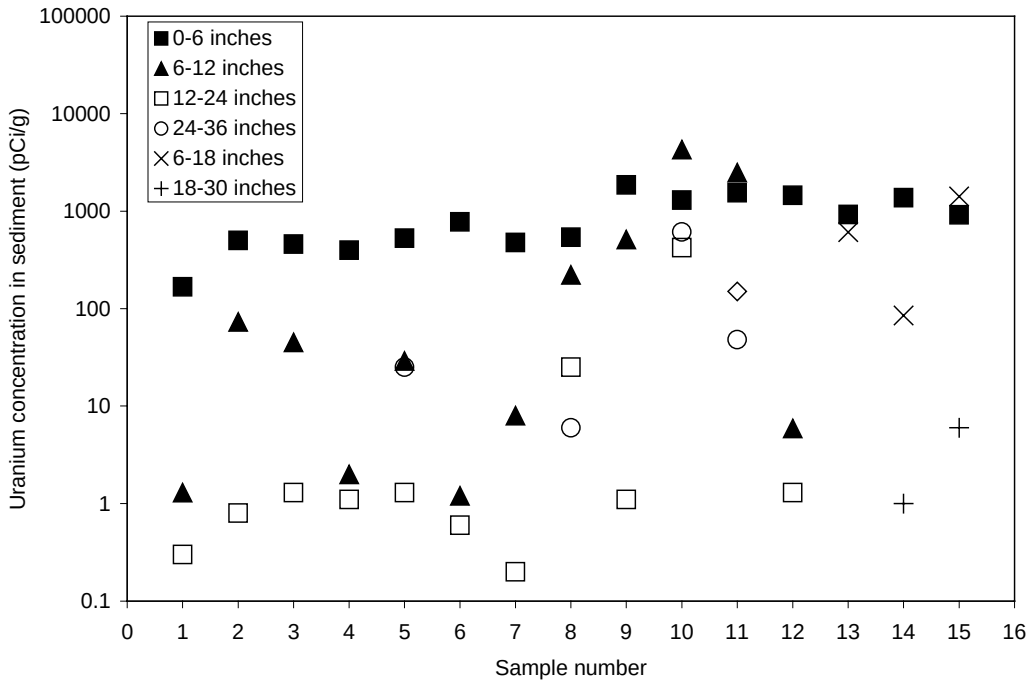

Figure 12.1-4. Uranium in Steed's Pond sediment cores collected in 1984. Link to tabulated figure data

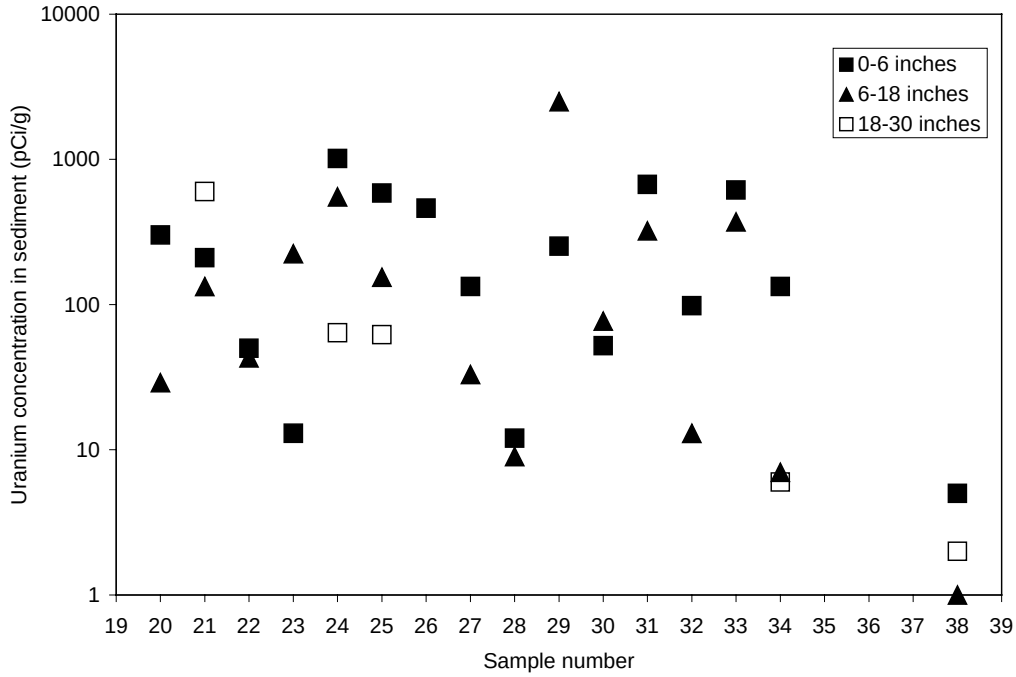

Figure 12.1-5. Uranium in Tim's Branch sediment cores, collected above Steed's Pond in 1984. Link to tabulated figure data

Measurements of Plutonium in Sediments

Carlton et al. (1992) states that fallout deposited on the Savannah River watershed has been estimated at 55 Ci with a $^{238}Pu/^{239,240}Pu$ ratio of 0.04 to 0.18. Approximately 1.5 Ci was deposited directly on water impoundments and 1.2 Ci was deposited within the boundaries of the SRS. Atmospheric plutonium releases from SRS operations, including unidentified alpha, have totaled about 3.8 Ci, and direct releases to streams have totaled 0.6 Ci. About 90% of the liquid releases containing plutonium and unidentified alpha has been released from F-Area and H-Areas into the Four Mile Creek watershed. For this reason, this stream has received more study. An examination of $^{238}Pu/^{239,240}Pu$ ratios in sediments (Table 12.1-3) collected from Site streams shows that Four Mile Creek has an elevated ratio, indicating that the source of plutonium in this stream is plutonium effluent from Site operations (Alberts et al. 1986). Steel Creek, which received cooling water from L- and P-Reactors, and Pen Branch, which received cooling water from K-Reactor, also showed elevated $^{238}Pu/^{239,240}Pu$ ratios in sediments. Upper Three Runs and Lower Three Runs show ratios similar to that of the Savannah River upstream of the SRS, indicating little contribution from Site operations. The isotopic ratios do not indicate whether the source of the plutonium is from atmospheric or liquid releases; however, it is most likely that the source of plutonium in Four Mile Creek is liquid effluent.

Table 12.1-3. Sediment Sample Plutonium Isotopic Ratios[a]

Sampling location	$^{238}Pu/^{239,240}Pu$
Upper Three Runs (Treadway Bridge)	0.141
Four Mile Creek (Road A)	3.29
Steel Creek (Road A)	2.64
Pen Branch (Road A)	1.84
Lower Three Runs (Road S-3-17	0.167
Savannah River (Jackson)	0.189

[a] Source: Alberts et al. (1986).

Tables 12.1-4 and 12.1-5 present a summary of routine monitoring for plutonium in stream sediments from 1977 through 1986. Previous data (1955 through 1962) are not presented because the sampling and analysis design for that period is not comparable to the design used since 1977. The swamp locations represent discharge areas for the streams. The data shown indicate no spatial or temporal trends. Many of the results are near or below the detection limit. Only concentrations measured at Four Mile Creek at Road A are consistently above the detection limit. Thus, it appears from routine measurements that, in general, plutonium releases from the SRS were small, are subject to various deposition processes, and result in concentrations in sediments that are, with a few exceptions, indistinguishable from background concentrations. In addition, the samples collected are probably not representative of the entire streambed making temporal and spatial trend analysis improbable. Concentrations in sediment of the Savannah River collected above the SRS during the same time period ranged from 0.0001 to 0.008 pCi g^{-1}.

Table 12.1-4. Concentration of Plutonium (pCi g^{-1}) in Sediments at Stream Locations at the SRS (1977 through 1991)

Year	Four Mile Creek at Road A7	Steel Creek at Road B	Steel Creek-Pen Branch Mouth
^{238}Pu			
1977	1.17	0.016	<0.001
1978	0.39	0.04	<0.003
1979	0.2	0.053	0.002
1980	0.31	0.017	<.001
1981	.008	0.043	0.002
1982	NA[a]	NA	NA
1983	0.3	0.05	0.003
1984	0.2	0.006	0.0006
1985	1.3	0.006	0.0004
1986	0.36	0.0008	0.0008
1987	0.074	0.043	0.05
1988	0.4	0.009	0.035
1989	0.02	0.021	0.049
1990	0.78	0.09	0.05
1991	0.078	0.0015	0.013
$^{239,240}Pu$			
1977	0.97	0.007	0.001
1978	0.29	0.03	<0.003
1979	0.074	0.044	0.004
1980	0.092	0.001	<0.001
1981	0.004	0.039	0.001
1982	NA	NA	NA
1983	0.11	0.15	0.002
1984	0.07	0.007	0.003
1985	0.38	0.008	0.0008
1986	0.016	0.004	0.002
1987	0.046	0.046	0.04
1988	0.15	0.012	0.036
1989	0.054	0.093	0.014
1990	0.23	0.007	0.004
1991	0.032	0.0043	0.026

[a] NA = No analysis.

Table 12.1-5. Concentration of Plutonium (pCi g^{-1}) in Sediments at Stream Discharge Locations at the SRS (1977 through 1991)

Year	Upper Three Runs Mouth	Four Mile Creek Swamp	Pen Branch Swamp	Steel Creek Swamp	Lower Three Runs Mouth
^{238}Pu					
1977	0.003	0.24	0.003	0.132	0.007
1978	0.006	0.12	0.02	0.09	0.04
1979	<0.001	0.02	0.022	0.006	0.001
1980	0.004	0.085	0.02	0.004	0.002
1981	0.001	0.003	<0.001	NA[a]	0.001
1982	NA	NA	NA	NA	NA
1983	0.002	0.08	0.003	0.03	0.005
1984	0.004	0.003	0.005	0.01	0.0005
1985	0.0003	0.006	0.019	0.2	0.0007
1986	0.003	0.002	0.0002	0.022	0.0005
1987	<0.003	0.003	0.024	0.012	0.02
1988	0.034	0.007	0.021	0.079	0.264
1989	0.061	0.015	0.07	0.005	<MD[b]
1990	0.006	0.007	0.02	0.002	0.04
1991	0.0052	0.0019	0.01	0.0029	0.0011
$^{239,240}Pu$					
1977	0.014	0.092	0.004	0.04	0.011
1978	0.02	0.07	0.04	0.05	0.04
1979	<0.001	0.012	0.021	0.046	0.003
1980	0.029	0.035	0.044	<0.001	0.008
1981	0.004	0.002	<0.001	NA	0.003
1982	NA	NA	NA	NA	NA
1983	0.007	0.11	0.02	0.04	0.001
1984	0.002	0.002	0.008	0.01	0.002
1985	0.001	0.006	0.032	0.12	0.001
1986	0.009	0.001	0.0004	0.015	0.0012
1987	0.012	0.002	0.018	0.01	0.02
1988	0.052	0.007	0.024	0.18	0.014
1989	0.02	0.006	0.019	0.003	<MD[b]
1990	0.007	0.003	0.002	0.0002	0.001
1991	0.0065	0.003	0.028	0.0071	0.0017

[a] NA = No analysis.
[b] <MD = Less than minimum detectable concentration.

Measurements of Gamma-Emitting and Other Radionuclides in Sediments

Stream and river samples collected routinely after 1975 were analyzed for gamma-emitting radionuclides and ^{90}Sr. Concentrations in river sediments were within the range typically detected from worldwide fallout. However, concentrations of ^{137}Cs often reflected contributions from Site releases, particularly along Four Mile Creek. However, no temporal or spatial trends were observed because of the complex sediment deposition processes involved and the fact that samples are probably not representative of the entire streambed.

ELECTRONICALLY COMPILED SEDIMENT DATA

Sediment sampling data were reported beginning in 1951 for at the inner and outer perimeter of the SRS along streams and the Savannah River. Samples were analyzed for gross alpha and gross beta activities. Later, samples were analyzed for specific radionuclides. The long-lived radionuclides, especially uranium and plutonium, are of particular interest to the dose reconstruction because they persist in sediments over long periods of time.

The various data summarized in this chapter are electronically compiled in a Microsoft Excel® workbook. This workbook contains the figures depicted in this chapter as well as the tabulated data that were used to produce the figures. In the workbook, each figure is represented in a chart. There is a separate worksheet that contains the tabulated data for each chart, or in some cases, for each group of charts. The following hyperlink allows readers of the electronic version of this document to directly access the workbook described above if the source application (Microsoft Excel®) is installed on the reader's computer: Ch12-Figure_data.xls.

USEFULNESS AND LIMITATIONS OF THE SEDIMENT DATA FOR DOSE RECONSTRUCTION

There are a number of factors that impact how the sediment data may be used during subsequent phases of the dose reconstruction project. These factors include the availability of sufficient original monitoring data sets to verify reported summary data and to evaluate spatial and temporal trends, as well as the ability to distinguish between Site releases of contaminants and other sources of the same contaminants in the environment (i.e., establish appropriate background concentrations).

Based on the information provided in the semiannual monitoring reports, it appears that surface water and sediment monitoring before 1958 was limited primarily to gross analyses. Because fallout was relatively high during this time period, nonvolatile beta data, especially for the Savannah River where concentrations were low, may be of limited value (i.e., SRS contributions may be masked by the presence of fallout).

The majority of alpha activity released via surface water effluents from the Site occurred from the M-Area. Essentially all of the alpha activity in the process effluent in the M-Area has been identified as depleted uranium. Steed's Pond received surface water from Tim's Branch, which received effluent from the M-Area. Thus, the sediment cores collected from Steed's Pond in 1967, 1981, and 1984 may provide uranium data useful for source term verification or model

validation. The small number of samples collected limits the usefulness of the data and may be insufficient to estimate the total activity of uranium in the pond.

Plutonium is a radioactive material that is typically of great concern to the public. However, the SRS did not conduct routine plutonium monitoring in surface water in the 1950s, 1960s, or 1970s. Uranium or plutonium alpha measurements were made in surface water from roughly 1954 to 1957. Also in the 1950s and 1960s, sediments from some of the Site streams and the river were analyzed for uranium/plutonium alpha. Routine plutonium measurements were initiated in Savannah River sediments in 1975. However, the sampling method as explained in Ashley and Zeigler (1978) and described above would not provide insight into historical releases of plutonium. As noted previously, routine sediment samples are probably unrepresentative of the entire streambed. The data may only be useful in that they confirm the sources of plutonium in specific streambeds. Routine ^{137}Cs measurements are, like those of plutonium, probably only useful for confirming the sources of the ^{137}Cs in specific streambeds.

In general, sediment sampling variability arises from the nature of the sample, the adequacy of the sampling population in representing the total population, and the bias resulting from the methods of sampling and analysis. The variations because of the heterogeneity of the sampled material (e.g., differing amounts of solid and suspended materials) frequently exceed the deviations that result from the analytical method. Characterizing variability because of sample heterogeneity and sampling methodology is extremely difficult, and it was not assessed as part of the historical SRS environmental monitoring program. Statistically speaking, increasing the number of samples collected can serve to increase the confidence that one has in the data, as the mean and range of values are generally better defined. As discussed previously, the number and locations of samples collected do not provide enough information to adequately characterize the streambeds at the SRS. At best, the data can serve to confirm that specific radionuclides were released into the streams and the Savannah River.

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CHAPTER 12.2

RADIONUCLIDES IN SOIL

ABSTRACT

During the period of greatest atmospheric releases at the Savannah River Site (SRS), from 1955 through the late 1960s, soil sampling and analysis were not routinely performed. A few special surveys performed between 1958 and 1970 are reported in this section, but methods for soil sampling and analysis were not standardized throughout the weapons complex until the early 1970s. *Radiological Assessments Corporation (RAC)* concludes that detailed analysis of soil radionuclide concentrations is not warranted because

(a) Data are not available for the early years
(b) It is not feasible to distinguish, using soil concentration data from the early years between radionuclides released from the Site and atmospheric weapons test fallout.
(c) The uncertainties involved in the use of soil concentration data to reconstruct doses to individuals living offsite are too large to allow useful dose reconstruction.

This section of the Phase II report summarizes the evidence indicating that soils data were not regularly collected during the years before 1970 and presents the limited special soil survey data *RAC* discovered for that period. We present summaries of documents found during the document search and extract and tabulate data of value. The summaries are organized by publication or memo date, beginning with the earliest reports found.

SUMMARIES OF DOCUMENTS RELATED TO SRS SOIL SAMPLING AND RADIONUCLIDE ANALYSIS

An internal SRS paper, Evans and Fenimore (1960), is one of the earliest *RAC*-discovered papers noting soil contamination. The contamination reported was entirely onsite and is not of value for dose reconstruction. Evans and Fenimore (1960) note that in November 1957, an experimental fuel element failed during calorimeter tests; as a result, an estimated 15,000 to 20,000 Ci of fission products was released to the emergency basin in Building 105-R. To minimize the release of activity to the Lower Three Runs Creek-Savannah River System, seepage basins were excavated in an abandoned construction area north of the reactor building, and the emergency basin water was pumped into these basins. About 2 years later, in December 1959, radioactive vegetation was discovered growing in the seepage basin area. The routine survey program, initiated by the Health Physics Section in January 1958, was expanded to determine the extent of spread of radioactivity around the basin system and to establish the cause of migration of radioactivity from the basins into surrounding soil strata and groundwater.

Analysis of vegetation from the seepage basin area indicated that deep-rooted plants had assimilated radioactivity from the soil. Highly radioactive vegetation was confined to the immediate vicinity of backfilled Basin 1 and an abandoned construction sewer line. Lower levels of radioactivity in vegetation were detected over a wider area south of Basin 1. The major soil contamination was confined to the Basin 1 and sewer line area. The movement of radioactivity in groundwater was more rapid in the sandy soils surrounding Basins 1 and 3 than in the clay strata

enveloping the remainder of the basins. Animal specimens collected near the basin system and in R-Area showed an uptake of radioactivity (Evans and Fenimore 1960).

Subsurface surveys were conducted by hand auguring 49 test wells. Soil samples were collected for laboratory analysis at 1-ft intervals during the drilling, and the radiation levels in the wells were determined with a gamma scintillometer. Samples less than 150 counts per minute, as surveyed with a Thyac, were discarded (laboratory analysis capability was limited at the time). The maximum concentration detected in soil from the 26 wells and samples laboratory-analyzed was 1100×10^{-9} Ci g^{-1} nonvolatile beta, dry weight analysis. The range was from 0.05 to 1100×10^{-9} Ci g^{-1}. The results indicate (per the SRS investigators) that the migration of gross quantities of radioactivity from the seepage basin system was limited. The areas of maximum contamination were confined to the soil immediately adjacent to Basin 1 and to the soil surrounding the abandoned construction sewer line (Evans and Fenimore 1960).

For the purposes of dose reconstruction based on soil contamination, these data are not of value because there is no indication in this report of soil concentrations offsite potentially exposing the public. The measurements presented are all associated with soil (and other) contamination well inside the SRS boundary, near the R-Reactor area.

H.A. McClearen (1974b) notes that the program to measure plutonium in soils was "intensified" at the Savannah River Plant (SRP) in 1972. It indicated Site perimeter and offsite soil plutonium levels of approximately 2 mCi km^{-2}. This value is the result of a 1973 soil monitoring survey. McClearen states that this average is "well within the range of deposition noted in the southeastern United States…," from atmospheric weapons testing. Health and Safety Laboratory measurements elsewhere in the country are cited to support this conclusion. The paper notes that "Measurement of releases at the emission point shows that 0.6 Ci of ^{238}Pu and 2.9 Ci of ^{239}Pu have been released since startup in 1955."

The study reported by McClearen (1974a) involved an initial onsite/offsite survey of seven, 10-core samples driven to 30-cm depth and analyzed in increments to determine depth and locations to sample in later studies. Most plutonium was found in the top 15 cm of the soil column. These results are listed in Table 12.2-1.

Table 12.2-1. Results of Initial 1973 Onsite and Offsite Soil Analyses for Plutonium[a]

Sample location	Plutonium deposition (mCi km^{-2})	
	^{239}Pu	^{238}Pu
Plant perimeter:		
NW quadrant	1.61	0.21
NE quadrant	1.37	0.08
SE quadrant	1.09	0.07
SW quadrant	1.28	0.07
Distant locations:		
Clinton, SC	1.81	0.08
Athens, GA	1.72	0.21
Savannah, GA	1.70	0.05

[a] Source: McClearen (1974a).

The study's second phase involved additional samples, all taken onsite, and located to characterize soils contaminated during two known plutonium release periods. The first of these

periods involved ^{239}Pu releases during reprocessing facility startup in 1955; the second involved ^{238}Pu releases in April 1969, after failure of the sand filter in H-Area. Wind data for those time periods were used to select sampling locations.

The 1973 results showed the influence of F-Area (10 samples) and H-Area (8 samples) on ^{239}Pu levels close to the facilities (F-Area data range: 3.2–26.2 mCi km^{-2} to 15-cm depth; H-Area range: 1.99–46.4 mCi km^{-2}). Close-in influence of the reprocessing areas was also evident for ^{238}Pu (F-Area range: 0.23–22.2 mCi km^{-2}; H-Area range: 0.59–91 mCi km^{-2}) (McClearen 1974a). For 26 other onsite samples taken more distant from the reprocessing facilities but inside the plant perimeter, results ranged from 1.26–2.99 mCi km^{-2} for ^{239}Pu and from <0.09–0.09 mCi km^{-2} for ^{238}Pu. These values approach those seen at the perimeter and offsite in the initial study and as reported by the Health and Safety Laboratory. McClearen (1974a) indicated minimal Site influence on offsite soil plutonium concentrations; however, releases from the two reprocessing facilities were clearly evident near the two facilities onsite.

Marter (1970) states, "Early in 1970, probably as a result of the fire at Rocky Flats, the AEC requested that soil samples from the environs of the Savannah River Plant be analyzed for plutonium. Soil analysis is not normally part of the routine environmental monitoring program at SRP, because the sampling and analysis of soils is considered to be an unreliable technique for determining deposited radioactivity." The Atomic Energy Commission (AEC) request prompted the development of sampling and analysis techniques and a series of SRS soil analyses. Marter (1970) summarized the results as follows: "No plutonium of SRP origin was detectable in soil samples from the plant perimeter and beyond. The measured plutonium activity in these samples, i.e., 0.07 d/m/g at plant perimeter, 0.05 d/m/g at 49 km, and 0.13 d/m/g at 60 km, is well below the range of 0.20 to 0.42 d/m/g predicted from global fallout. Dispersion calculations of deposition from SRP stacks at the plant perimeter (0.01 d/m/g) and at 30 km (0.001 dpm/g) confirm that the plant deposition off-site will be obscured and indistinguishable from global fallout because of the variability of global fallout and uncontrolled variables in obtaining representative soil samples."

Marter (1970) notes that deposition pans were installed in 1962 and were located at all onplant and offplant air monitoring stations as far as 25 mi away. Marter (1970) states that, "Deposition data are not available for the period from startup through 1962, a period during which 2.8 Ci of the total of 3.5 Ci (of plutonium) was released." Table 12.2-2 presents a summary of deposition pan data for the period 1963–1969, extracted from Marter (1970). Isotopic analyses were insufficient to make conclusions concerning plutonium versus other alpha emitter concentrations or perimeter versus offsite comparisons.

Table 12.2-2. Alpha Activity Deposited on 2 × 2-ft Pans Located at All SRS Air Monitoring Stations[a] Total alpha deposited, 1963–1969

Location	pCi m^{-2}	d/m/g of soil
F-Area	2310	0.40
H-Area	675	0.12
Plant perimeter stations	475	0.08
25-mi radius stations	390	0.07

[a] Source: Marter (1970).

During January 1970, soil was sampled from 12 onsite and two offsite locations. Results of the analyses are presented in Table 12.2-3, extracted from (Marter 1970).

Table 12.2-3. January 1970 Soil Analysis Results, SRS Onsite and Offsite[a]

Sample location	Plutonium in top centimeter of soil (d/m/g)
F-Area	
North	0.06 +/- 0.009
East	0.13 +/- 0.004
South	0.64 +/- 0.18
West	0.53 +/- 0.18
H-Area	
North	2.64 +/- 0.40
East	0.32 +/- 0.10
South	0.06 +/- 0.004
West	0.47 +/- 0.018
P-Area	
North	0.06 +/- 0.005
East	0.09 +/- 0.007
South	0.10 +/- 0.007
West	0.05 +/- 0.002
Fort Gordon, Georgia	0.05 +/-0.002
Bamberg, South Carolina	0.13 +/- 0.003

[a] Source: Marter (1970).

Marter concludes that SRS-released plutonium was indistinguishable offsite, obscured by fallout-deposited plutonium. The same SRS Phase II Database document also contains draft and handwritten versions of the reports noted above, including handwritten memos, data sheets, and data analysis sheets supporting the summaries in the previous tables. A handwritten note, probably from Ben Rusche to C. Patterson (Rusche 1973), mentions that "The variation of $^{238}Pu/^{239}Pu$ with distance suggest(s) that some of the Pu originated form SRP operations." Rusche suggests further analysis of samples to examine this question.

Another memo was written by J. E. Johnson (Johnson 1973). It confirms that "Soil sampling and analyses have not been a part of the routine SRP environmental monitoring program." Johnson presents the results of a few special soil surveys at SRS from 1958 through 1972. Some of these data are already reported previously in this section. The first survey noted by Johnson analyzed soil for ^{90}Sr in a number of onsite and offsite locations. Johnson (1973) states that "No evidence was found of plant [SRS] contribution." Table 12.2-4 presents those results.

Johnson also describes a 1967 study in which soils were collected along six traverses from the separations areas out to about 50 mi and analyzed for natural radioactivity, ^{90}Sr, and gamma-emitting radionuclides. He notes that "A possible slight contribution of cerium-144 was noted near the Separations Areas." Table 12.2-5 presents the results of that study.

Table 12.2-4. Comparison of ^{90}Sr in Top 20 cm of Soil (1958 versus 1967)[a]

Location	$\times 10^{-6}$ μCi g^{-1} 1958	1967
F-Area	0.124	0.215
H-Area	-	0.230
Plant inner	0.105	0.268
Plant outer	0.150	0.268
25-mi radius	0.171	0.310

[a] Source: Johnson (1973).

Table 12.2-5. Soil Radioactivity Levels per 1967 SRP Study; Transects Out to 50 Miles[a]

Element	Month	Depth (cm)	Radioactivity in soil ($\times 10^{-6}$ μCi g^{-1}) F-Area	H-Area	Plant inner	Plant outer	25-mi radius	50-mi radius
^{90}Sr	January	0–5	0.300	0.350	0.300	0.410		
		5–10	0.130	0.110	0.130	0.110		
	September	0–5				0.390	0.450	0.720
		5–10				0.160	0.170	0.250
^{137}Cs	January	0–5	1.000	0.820	1.030	0.800		
		5–10	0.230	0.090	0.280	0.220		
	September	0–5				1.070	1.120	1.610
		5–10				0.170	0.150	0.220
^{144}Ce	January	0–5	0.820	0.630	0.580	0.540		
		5–10	0.370	0.370	0.370	0.310		
	September	0–5				0.500	0.560	0.510
		5–10				0.240	0.220	0.190
Thorium	January	0–5	0.480	0.590	0.480	0.380		
		5–10	0.460	0.570	0.460	0.390		
	September	0–5				0.380	0.320	0.310
		5–10				0.360	0.300	0.270
Uranium	January	0–5	1.530	1.800	1.560	1.400		
		5–10	1.410	1.850	1.450	1.400		
	September	0–5				1.040	1.320	1.660
		5–10				1.120	1.190	1.500

[a] Source: Johnson (1973).

Johnson notes that, as of 1973, a laboratory had been dedicated to soils analysis, and the necessary field and laboratory equipment had been purchased or fabricated. He states that "Adequate cores will be collected on-plant and off-plant to determine the distribution of plutonium with soil depth and to inventory the ^{238}Pu and ^{239}Pu in the SRP environment… This work will be continued until we have a satisfactory inventory of plutonium in the plant and environs. Following this or toward the end of routine soil assays we will inaugurate a fallout collection program to assess and audit plutonium additions to the environs."

Johnson (1973) also reports a more detailed version of the 1970 plutonium-in-soil results presented earlier, shown in Table 12.2-6.

Table 12.2-6. Onsite and Offsite Soil Survey of Plutonium in Top 1 cm of Soil (1970)[a]

Location	Total plutonium ($\times 10^{-6}$ µCi g^{-1})	^{239,240}Pu ($\times 10^{-6}$ µCi g^{-1})	^{238}Pu ($\times 10^{-6}$µCi g^{-1})	Alpha % ^{239,240}Pu	Alpha % ^{238}Pu
Fort Gordon	0.0214	0.0197	0.0017	92.09	7.91
Bamberg	0.0603	0.0571	0.0032	94.64	5.36
F-Area					
North	0.0245	0.0163	0.0082	69.57	30.43
East	0.0581	0.0448	0.0131	77.36	22.64
South	0.2888	0,2600	0.0287	90.04	9.96
West	0.2392	0.1588	0.0804	66.60	33.40
H-Area					
North	1.1882	0.8193	0.3689	68.97	31.03
East	0.1454	0.0622	0.0833	42.78	57.22
South	0.0262	0.0136	0.0127	51.92	48.08
West	0.2107	0.0734	0.1374	34.78	65.22
P-Area					
North	0.0260	0.0195	0.0065	76.71	23.29
East	0.0394	0.0364	0.0031	92.49	7.51
South	0.0450	0.0407	0.0040	90.82	9.18
West	0.0203	0.0174	0.0033	84.71	15.29

[a] Source: Johnson (1973).

Johnson (1973) presents the results of a 1972 survey of ^{238}Pu, ^{239}Pu, ^{137}Cs, and ^{40}K in soil, at the plant perimeter, 25-mi radius, and 50-mi radius. Table 12.2-7 summarizes the maximum, minimum, and average values for the seven sets of soil samples analyzed at seven locations around the Site perimeter and each of the two circles at 25 and 50 mi. An attached note from H. McLendon (1973) states that five cores were extracted and composited at each location then dried and analyzed in the SRS laboratory.

Table 12.2-7. SRS Radionuclide Survey at the Perimeter and Two Radii Offsite (1972)[a]

Nuclide	Radioactivity in soil at 9-cm depth ($\times 10^{-6}$ µCi g^{-1} [dry weight])								
	Plant perimeter			25-mile radius			50-mile radius		
	Max	Min	Average	Max	Min	Average	Max	Min	Average
^{238}Pu	0.046	ND[b]	0.0068	0.0090	0.0004	0.0018	0.0036	0.0001	0.0009
^{239}Pu	0.035	0.002	0.0131	0.0189	0.0054	0.0120	0.0490	0.0027	0.0155
^{137}Cs	3.50	<0.05	0.79	3.01	<0.03	0.07	1.66	<0.03	0.54
^{40}K	6.5	<0.5	1.8	0.8	<0.6	<0.6	<0.7	<0.5	<0.6

[a] Source: Johnson (1973).
[b] ND = not detectable.

A memo written by S. L. Hoeffner evaluates the effect of water and soil quality variables on the sorption of ^{60}Co on SRP soils (Hoeffner 1984). The pH was a major factor influencing cobalt sorption, as indicated by the ^{60}Co distribution coefficient, Kd. The Kd ranged from 2 to more than 10,000 mL g^{-1} over a pH range of 2 to 9. Decreases in cobalt sorption that occur with elevated levels of Mg^{2+}, Ca^{2+}, or K^{+} are the result of accompanying small decreases in pH. The ions Na^{+}, Cl^{-}, and NO_3^{-} had no effect on cobalt sorption. Changes in cobalt sorption with soil clay content also were caused by changes in pH.

Hoeffner (1984) notes that ^{60}Co is a major radionuclide in the SRP burial ground. As of 1982, about 540,000 Ci of ^{60}Co were buried in the trenches. Hoeffner (1984) notes that low concentrations of ^{60}Co have been detected in groundwater monitoring wells and that laboratory studies that give a basic understanding of the factors influencing cobalt migration in the SRP burial ground have not been available before this work. The paper notes that ^{60}Co was found in 1 of 20 groundwater wells, at 13 pCi L^{-1}, and that most ^{60}Co remains at the bottom of the burial trenches.

An SRS internal paper (Arnett 1993) summarizes radioactivity in soil data collected by SRS. Table 12.2-8 summarizes these data.

A large set (6 ft^3) of environmental monitoring database printouts was found during the *RAC* Phase II dose reconstruction record search (Du Pont XX). We also found a computer data tape possibly containing the same information found on the printouts. SRS staff were unable to decode the data tape over several months of intermittent effort by various groups. The information from the printed monthly report tables has been used by *RAC* to provide SRS environmental monitoring and release data for a number of locations and years. Only a small quantity of soil concentration data is available on the printouts, however, and only a fraction of that information pertains to offsite monitoring. Onsite soil concentration data were often presented for F-Area, H-Area, and other onsite areas, but the onsite soil data are not of value for offsite dose reconstruction. Table 12.2-9 summarizes the offsite data from the printouts.

Carlton et al. (1992) collects and summarizes available data related to plutonium in the SRS environment onsite and offsite. While the sections related to releases and environmental monitoring are generally large and varied, only one-half a page of information is provided concerning plutonium soil sampling. Carlton et al. (1992) states that "The amount of ^{238}Pu and ^{239}Pu in the top 8 cm of soil has been measured at the same sampling sites since 1974…. Since the great majority of ^{239}Pu was released in a single year, 1955, and the majority of ^{238}Pu was released in 1969, the soil deposition actually is almost entirely a measure of deposition during the years of the greatest releases…. Therefore, the measurements do not show the steady accumulation of plutonium isotopes in the soil that would be expected if a steady accumulation had taken place over the period of measurement." Three figures summarize the influence of the SRS reprocessing canyons on plutonium in the environment and clearly show decreasing concentration in soil with distance from the canyons. Carlton et al. (1992) presents no data on offsite concentrations of plutonium.

Table 12.2-8. Radioactivity in Soil pCi g^{-1} (± 1 sigma) Dry Weight (0–8 cm depth)[a]

Location	^{90}Sr	^{137}Cs	^{238}Pu	^{239}Pu
F-Area				
2000 ft east	(1.21 ± 1.84)E-02	(5.54±0.37)E-01	(2.70 ± 0.10)E-01	(3.27±0.11)E-01
2000 ft west	(1.54 ± 1.75)E-02	(7.19±0.40)E-01	(4.03 ± 0.14)E-01	(2.92±0.11)E-01
2000 ft north	(1.99 ± 1.82)E-02	(1.05±0.05)E+00	(3.55 ± 0.29)E-02	(5.31±0.14)E-01
2000 ft south	(2.16 ± 1.82)E-02	(2.4±20.42)E-01	(6.45 ± 0.82)E-03	(8.11±0.92)E-03
H-Area				
2000 ft east	(0.09 ±1.63)E-02	(8.22 ±0.42)E-01	(1.42 ± 0.04)E-02	(5.54 ± 0.23)E-02
2000 ft west	(1.87 ±1.67)E-02	(2.31 ± 0.27)E-01	(5.33 ± 1.30)E-03	(1.65 ± 0.22)E-02
2000 ft north	(0.72 ±1.63)E-02	(7.72 ± 2.42)E-02	(1.97 ± 0.50)E-03	(4.85 ± 0.75)E-03
2000 ft south	(2.89 ±1.83)E-02	(1.32 ±0.05)E+00	(2.13 ± 0.16)E-02	(4.09 ± 0.21)E-02
S-Area				
#1	(0.02±1.77)E-02	(7.60 ± 0.42)E-01	(1.54 ± 0.18)E-02	(7.32 ± 0.41)E-02
#2	(0.25±1.14)E-02		(1.33 ± 0.44)E-03	(3.53 ± 0.63)E-03
#3	(1.27±1.87)E-02	(2.58 ± 0.27)E-01	(3.84 ± 0.19)E-02	(3.44 ± 0.18)E-02
#4	(2.21±1.81)E-02	(1.35 ± 0.28)E-01	(5.17 ± 2.07)E-04	(1.37 ± 0.29)E-03
Z-Area				
#1	(0.89±1.44)E-02	(6.15 ±1.95)E-02	(1.14 ± 0.51)E-03	(3.84 ± 1.04)E-03
#3	(0.78±1.45)E-02	(3.24 ± 0.36)E-01	(2.66 ± 0.45)E-03	(1.56 ± 0.11)E-02
#5	(2.44±1.63)E-02	(6.65 ± 0.44)E-01	(8.81 ± 0.88)E-03	(5.36 ± 0.24)E-02
#7	(1.01±1.65)E-02	(4.76 ± 0.36)E-01	(8.77 ± 1.42)E-03	(8.49 ± 0.47)E-02
Site perimeter				
NE quadrant	(2.88 ± 2.00)E-02	(4.31 ± 0.36)E-01	(4.75±3.39)E-04	(1.36 ± 0.12)E-02
NW quadrant	(1.52 ± 1.90)E-02	(4.84 ± 0.38)E-01	(6.08±3.48)E-04	(4.46 ± 0.85)E-03
SE quadrant	(0.34 ± 1.89)E-02	(3.01 ± 0.24)E-01	(1.61±0.70)E-03	(9.30 ± 1.44)E-03
SW quadrant	(1.50 ± 1.89)E-02	(4.65 ± 0.38)E-01	(2.19±1.01)E-03	(1.78 ± 0.25)E-02
100-mi radius				
Clinton, SC	(1.46 ± 1.79)E-02	0/1[b]	(2.34±1.64)E-04	(4.74 ± 0.56)E-03
Savannah, GA	(1.19 ± 1.65)E-02	0/1	(1.12±7.25)E-04	(1.93 ± 0.23)E-02

[a] Source: Arnett (1983).

[b] The gamma analysis package currently used by Environmental Monitoring does not force an activity determination if a threshold setting is not met. For these reported radionuclides, "No. of Samples" is displayed as "number with activities quantified/number of samples counted". Only the numbers quantified are used in the max, min, and mean generation.

Table 12.2-9. Soil Sample Analysis Results from Environmental Data Printouts

Date collected	Location[a]	Radionuclide	pCi g^{-1} Avg.	Reference
3/1/85	Savannah	^{90}Sr	0.06	KRM1997101325
5/18/87	PP NE	^{238}Pu	0	KRM1997101329
5/18/87	PP NW	^{238}Pu	0	“
5/18/87	PP SE	^{238}Pu	0	“
5/18/87	PP SW	^{238}Pu	0.02	“
5/8/87	Clinton	^{238}Pu	0	“
5/8/87	Savannah	^{238}Pu	0	“
5/18/87	PP NE	^{239}Pu	0.01	“
5/18/87	PP NW	^{239}Pu	0.01	“
5/18/87	PP SE	^{239}Pu	0.02	“
5/18/87	PP SW	^{239}Pu	0.01	“
5/8/87	Clinton	^{239}Pu	0.02	“
5/8/87	Savannah	^{239}Pu	0.01	“
5/8/87	Clinton	^{40}K	3.18	“
5/8/87	Savannah	^{40}K	0	“
5/8/87	Clinton	^{137}Cs	54	“
5/8/87	Savannah	^{137}Cs	33	“
5/5/88	PP NE	^{238}Pu	0	KRM1997101331
5/5/88	PP NW	^{238}Pu	0.01	“
5/5/88	PP SW	^{238}Pu	0	“
5/5/88	PP NE	^{239}Pu	0.01	“
5/5/88	PP NW	^{239}Pu	0.02	“
5/5/88	PP SW	^{239}Pu	0.02	“
5/5/88	PP NE	^{90}Sr	0.01	“
5/5/88	PP NW	^{90}Sr	0.02	“
5/5/88	PP SE	^{90}Sr	0.02	“
5/5/88	PP SW	^{90}Sr	0.02	“
5/5/88	Clinton	^{90}Sr	0.05	“
5/5/88	Savannah	^{90}Sr	0	“
5/18/89	PP NE	^{238}Pu	0.01	HRM199607108
5/18/89	PP NW	^{238}Pu	0.01	“
5/18/89	PP SE	^{238}Pu	0.01	“
5/18/89	PP SW	^{238}Pu	0.01	“
5/18/89	Clinton	^{238}Pu	0.01	“
5/12/89	Savannah	^{238}Pu	0.01	“
5/18/89	PP NE	^{239}Pu	0.01	“
5/18/89	PP NW	^{239}Pu	0.01	“
5/18/89	PP SE	^{239}Pu	0.01	“
5/18/89	PP SW	^{239}Pu	0.01	“
5/18/89	Clinton	^{239}Pu	0	“
5/12/89	Savannah	^{239}Pu	0	“
5/18/89	PP NE	^{90}Sr	0.01	“
5/18/89	PP NW	^{90}Sr	0.01	“

Date collected	Location[a]	Radionuclide	pCi g^{-1} Avg.	Reference
5/18/89	PP SE	^{90}Sr	0.02	“
5/18/89	PP SW	^{90}Sr	0.02	“
5/18/89	Clinton	^{90}Sr	0.10	“
5/25/90	PP NW	^{238}Pu	0.01	HRM199607106
5/25/90	PP SE	^{238}Pu	0	"
5/25/90	PP SE	^{239}Pu	0.01	“
5/25/90	PP NE	^{90}Sr	0.6	“
5/25/90	PP NW	^{90}Sr	0.41	“
5/25/90	PP SE	^{90}Sr	0.54	“
5/25/90	PP SW	^{90}Sr	0.9	“
5/25/90	Clinton	^{90}Sr	0.31	“

[a] PP = plant perimeter.

USEFULNESS AND LIMITATIONS OF THE SOIL MONITORING DATA FOR DOSE RECONSTRUCTION

We do not anticipate that the soil monitoring data collected at the SRS will be useful for later stages of dose reconstruction. Data we were able to locate generally included summaries of studies done previously and not original data. Soil data were not collected routinely before the 1970s, but rather as a part of special studies and then only very sporadically. Offsite soil data for these special studies were very limited. Most soil data were collected onsite, close to release points, where soil levels of some nuclides were elevated. In studies where samples at the Site perimeter and at offsite locations were collected, similar concentrations at the two sets of locations were evident. This indicates that Site contributions and fallout contributions are not discernible from one another.

Background concentrations are likely represented by samples collected at remote locations at Clinton, South Carolina; Savannah, Georgia; Fort Gordon, Georgia; and Bamberg, South Carolina. In some cases, 25-mile radius data were also collected. For all nuclides of plutonium, ^{90}Sr, and ^{137}Cs, concentrations at these background locations are similar to Site perimeter concentrations. These data show little evidence of Site contribution to offsite soil contamination.

Because of limited spatial and temporal resolution and the close agreement between Site perimeter and background concentrations, soil monitoring data will not be useful for reconstructing releases from the SRS. Even with the use of environmental transport models to simulate releases from the Site and deposition, validation of the deposition pattern with soil data will not be possible because of the difficulty of distinguishing between Site and fallout contributions.

SUMMARY

Offsite soils environmental monitoring data were not collected routinely at the SRS before the 1970s, and the intermittent information available is insufficient to construct a useful picture of offsite soil concentrations of radionuclides. Available information is summarized here, and it indicates little evidence of offsite soil contamination associated with SRS releases in the early years.

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CHAPTER 13

RADIONUCLIDES IN OFFSITE DRINKING WATER AND SURFACE WATER

ABSTRACT

This chapter discusses the extent and analytical results of the radiological monitoring program for water supplies of 14 communities surrounding the Savannah River Site (SRS), for one upstream and two downstream drinking water treatment plants supplied by the Savannah River, and for offsite locations from the Savannah River. The sampling locations, collection methods and frequencies, and the analyses performed are discussed.

Routine monitoring of the community water supplies began at the time of plant startup, but it was limited to gross alpha and beta analyses until 1960, when tritium monitoring was initiated. With the exception of tritium measurements, no routine, radionuclide-specific data have been generated for the community water supplies. The downstream water treatment plants were monitored for gross alpha, beta, tritium, and, in later years, for ^{137}Cs. While the Port Wentworth Plant provides most water for an industrial plant, it can also supply some water to the City of Savannah during water shortages. The Beaufort-Jasper Plant was designed to supply domestic water for a population of about 50,000 people. Since plant startup, the SRS has routinely sampled the Savannah River along the Site boundary and downstream of the Site to evaluate the impacts of liquid releases. Routine tritium monitoring began in 1958 to supplement the gross alpha and nonvolatile beta measurements. Beginning in the mid-1960s, other radionuclides, including radiocesium, radiostrontium, ^{51}Cr, ^{60}Co, were routinely monitored.

For dose reconstruction, the potential impacts of SRS operations on both the surrounding community water supplies and the downstream water treatment plants are of greatest concern because the safety of drinking water is of great interest to the public. The Savannah River monitoring data can provide the primary means for evaluating the impact of SRS releases of radionuclides other than tritium on the downstream drinking water supplies. They also may be useful to evaluate other exposure pathways, such as the ingestion of fish and other aquatic organisms and recreational use of the river.

INTRODUCTION

A thorough evaluation of radiological data for any media involves using the most basic types of data that are available. Details on the SRS water monitoring program were compiled through an extensive review of documents—such as weekly and monthly reports, internal memos, and special studies—identified during Phase I of the dose reconstruction. Emphasis was placed on reconstructing and compiling information for the 1950s through the early 1970s, when few details are provided in the semiannual and annual Site monitoring reports. By the mid-1970s, the annual reports had been expanded to include descriptions of the sampling and analytical methods and to provide quality assurance data, such as interlaboratory comparison results.

Until recent years, only summary data (semiannual or annual averages, minimums, and maximums) have been reported in the SRS monitoring reports. To provide the resolution needed for the types of assessments involved in a dose reconstruction, individual measurements or

averages calculated over shorter periods of time are needed. Furthermore, individual measurements (preferably raw, handwritten data) are needed to evaluate the reliability of summary data. This verification is needed to ensure the quality of the assessments in which the data are used and to build public confidence in the dose reconstruction process.

Fiche copies of handwritten, individual sample data were identified for a number of environmental media during *Radiological Assessments Corporation*'s (*RAC*'s) comprehensive review of SRS documents. These data, referred to as aperture card data, are available for the community water supplies and the Savannah River from the late 1950s through the early 1970s. Drinking water treatment plant aperture card data are available from the mid-1960s through the early 1970s.

Much of the available aperture card data for the offsite water monitoring programs have been compiled into three Microsoft Excel® workbooks: CWSDATA.xls (community water supply data), WTPDATA.xls (water treatment plant data), and SRDATA.xls (Savannah River data). The CWSDATA and WTPDATA workbooks also contain semiannual and annual averages from the Site monitoring reports for those years in which aperture card data are not available. The sources (e.g., aperture cards, annual monitoring reports, etc.) and their associated *RAC* document database numbers are indicated for each data entry in the offsite water monitoring workbooks.

Another source of individual sample data, covering later years for which no aperture card data are available, has been identified and catalogued in *RAC*'s SRS document database. This source is a series of Health Protection Environmental Monitoring Monthly Reports that are actually computer printouts of laboratory measurements for the various environmental media (e.g., Du Pont 1976). The data reports for 1976 through 1990 have been catalogued in the document database. However, because they were not available until late in Phase II, these data are not included in the Excel workbooks for drinking water or the Savannah River.

DRINKING WATER MONITORING

Routine monitoring of public drinking water by the SRS has consisted of sample collections from 14 communities surrounding the Site (Figure 13-1) and from two drinking water treatment plants located downstream of the SRS on the Savannah River, in Beaufort-Jasper, South Carolina, and Port Wentworth, Georgia (Figure 13-2).

Community Water Supplies

The sources of water used by the 14 communities that were routinely monitored by the SRS are Aiken-stream and well, Allendale-deep well, Augusta-river, Barnwell-deep well, Bath-deep well, Blackville-deep well, Clearwater-lake, Jackson-deep well, Langley-deep well, New Ellenton-deep well, N. Augusta-river, Sardis-deep well, Waynesboro-stream, and Williston-deep well (Arnett et al. 1992).

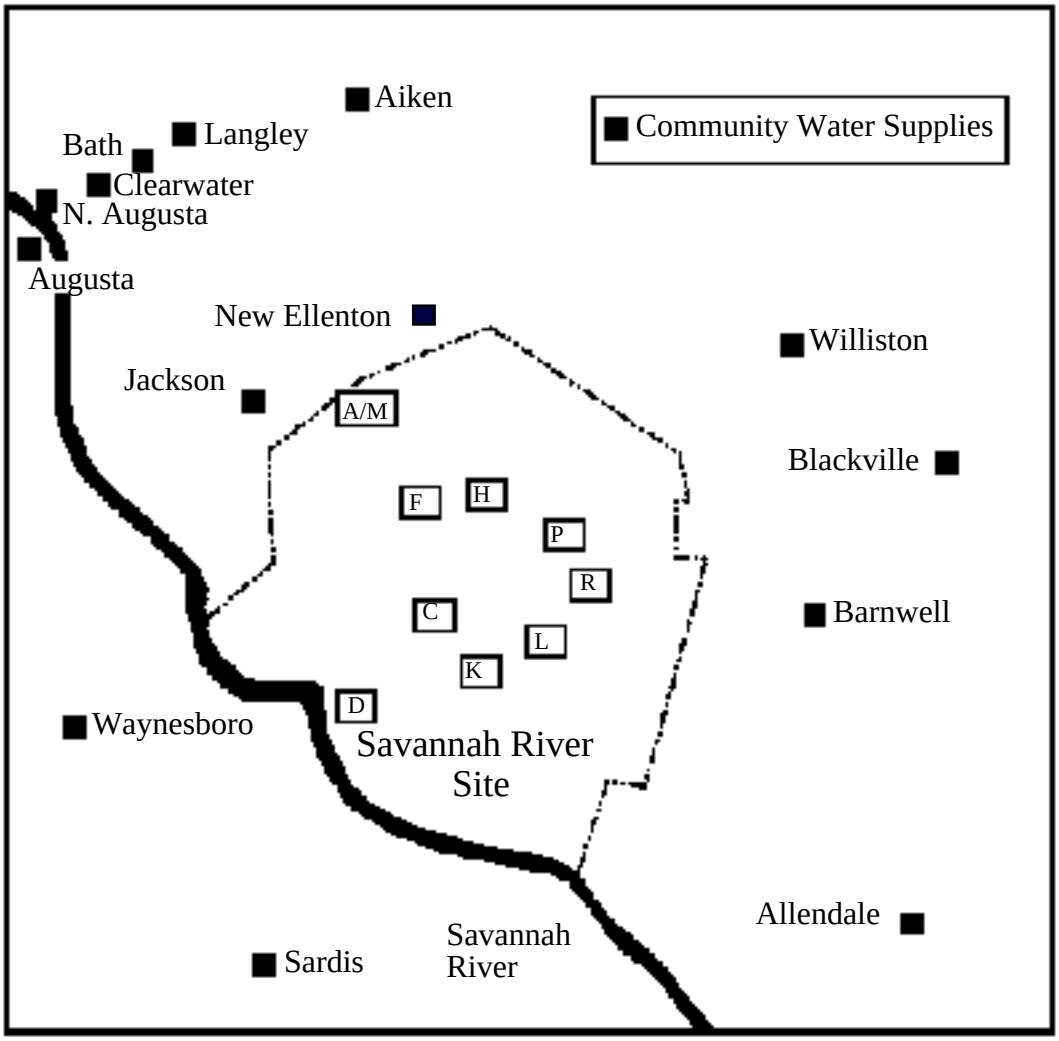


Figure 13-1. Locations of 14 communities surrounding the SRS for which routine radiological monitoring of public drinking water has been conducted.

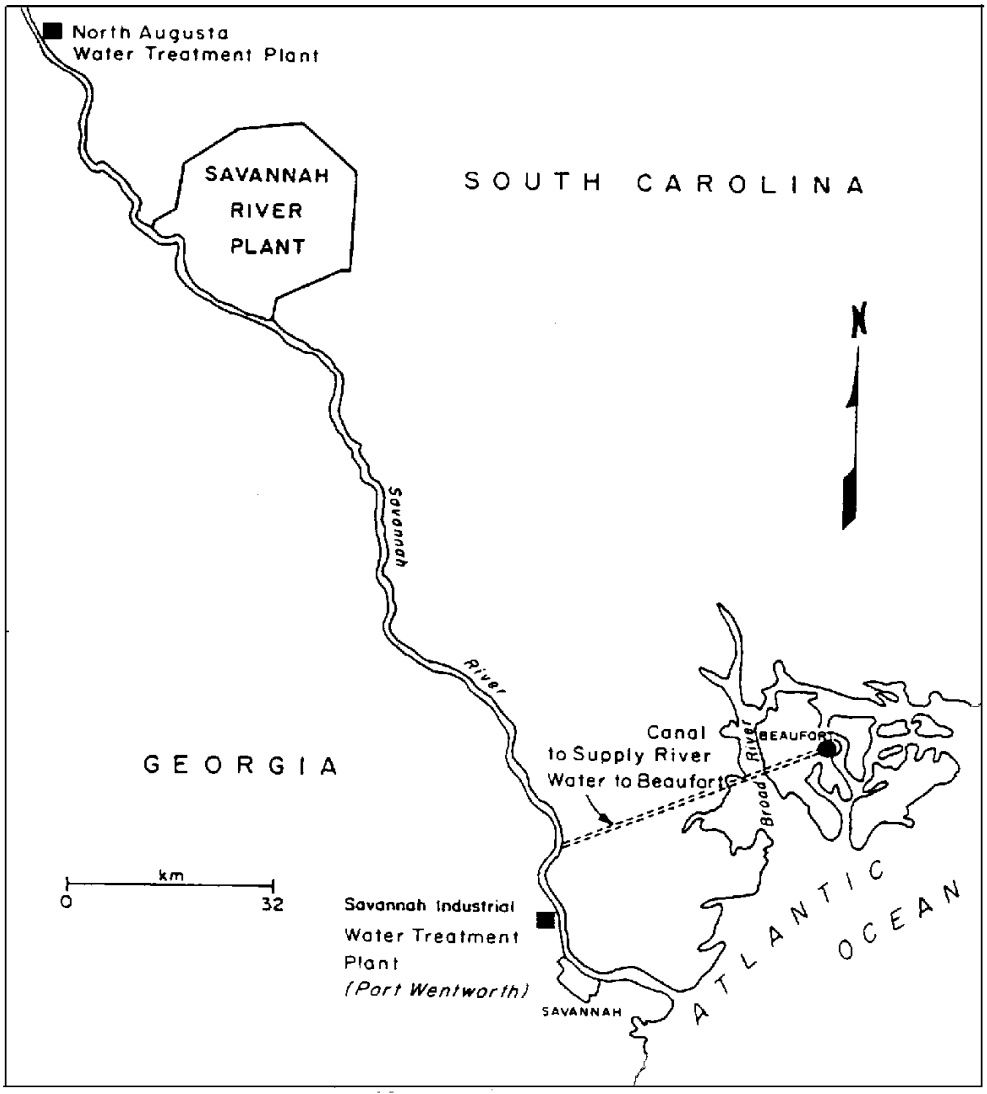


Figure 13-2. Location of drinking water treatment plants using Savannah River water in the vicinity of the SRS. The plant at Port Wentworth, which supplies an industrial complex, was in operation at the time of SRS plant startup. The Beaufort-Jasper plant, which supplies domestic water for a population of approximately 50,000, went into operation in 1965. The North Augusta plant, located upstream of the SRS, was monitored as a control location.

Monitoring History and Available Data

The routine monitoring of the community water supplies is summarized in Table 13-1. Preoperational monitoring was performed on the public water supplies from 12 of the communities shown in Figure 13-1. The New Ellenton and Jackson supplies were not operational until 1955. Samples were analyzed for gross alpha, gross beta, ^{40}K, total potassium, uranium/plutonium alpha, radium, and radon. Based on 16 samples per community, the average beta concentrations in each of the supplies ranged between 5.2 and 6.8 pCi L^{-1} (Reinig et. al 1953).

Table 13-1. History of Routine SRS Radiological Monitoring of the Community Water Supplies[a]

Year	Sampling Frequency	Radiological Analyses
1953 - 1959 [b]	Monthly	gross alpha and beta
1960 – April 1966	Monthly	gross alpha, beta, and tritium
May 1966 – 1991[c]	Semiannually	gross alpha, beta, and tritium

[a] Aiken, Allendale, Augusta, Barnwell, Bath, Blackville, Clearwater, Jackson, Langley, New Ellenton, North August, Sardis, Waynesboro, and Williston
[b] Jackson and New Ellenton not operational until 1955
[c] North Augusta sample four times in 1967 and monthly from April 1968 through 1973.

Routine monitoring of the community water supplies began at the time of plant startup, but it was limited to gross alpha and beta analyses until 1960 when tritium monitoring was initiated. The Jackson and New Ellenton water supplies began to be used in 1955 and were incorporated into the routine monitoring program at that time. Relatively high alpha concentrations were observed in the Jackson water supply (deep well) over the years, prompting periodic investigations by the SRS into the possible sources. In 1961, samples of Jackson well water were analyzed for ^{226}Ra content, and in 1963, drinking water from 11 wells in the central Savannah River area were analyzed for total radium, ^{223}Ra, ^{224}Ra, and ^{226}Ra. Total radium concentrations ranged from 0.3 to 8.9 pCi L^{-1} (Edgar 1961, 1963). In 1981, the SRS analyzed Jackson drinking water for uranium, plutonium, thorium, and gamma emitters. Only naturally occurring radionuclides, principally ^{226}Ra, ^{228}Th, and their radioactive decay products, were identified (Ashley and Zeigler 1984).

From 1953 through April 1966, samples of the community water supplies were collected monthly. After April 1966, the collection frequency was reduced to semiannually for all locations except North Augusta. Sampling of the North Augusta supply was performed four times in 1967 and monthly from April 1968 through 1972. After 1973, North Augusta was sampled semiannually. The community water supply samples have always been grab samples collected from the tap. Historically, the samples were collected at public buildings or service stations (Du Pont 1953). The 1991 SRS annual report (Arnett et al. 1992) lists the community post offices as the sampling locations.

With the exception of tritium measurements, no routine, radionuclide-specific data have been generated for the community water supplies. Furthermore, no special studies, except those related to elevated alpha levels in the Jackson supply, appear to have been performed on the community supplies.

The following individual sample data have been catalogued in *RAC*'s SRS document database:

- Gross alpha, gross beta, and tritium data for monthly samples for the years 1959 through 1966 on aperture cards (handwritten data logs)
- Gross alpha, gross beta, and tritium data for semiannual samples for 1967 through 1973 on aperture cards

- Gross alpha, gross beta, and tritium data for semiannual samples for 1976 through 1990 from the Health Protection Environmental Monitoring Monthly data reports.

No sources of individual sample data have been located for the years before 1959, and the community water supplies monitoring program was rarely mentioned in the Site's early weekly or monthly reports.

As listed above, 9 of the 14 surrounding communities are supplied from deep wells, 4 use surface water sources, and 1, Aiken, South Carolina, uses both well and surface water. Because there is no evidence that contaminated groundwater has moved beyond the Site boundary (Arnett et al. 1993), the communities that use only well water are not expected to be a focus of the dose reconstruction. Continued groundwater monitoring will provide a means of evaluating the importance of this exposure pathway into the future (see appendix J).

Of the five communities supplied by surface water, two get their water from the Savannah River (above the SRS), two from streams, and one from a lake. None of the surface water supplies come into contact with SRS liquid releases. The only pathway for SRS contaminants to enter these supplies would be via deposition following atmospheric releases. The most likely nuclide of interest from SRS releases would be tritium that could enter the surface water sources via rainwater.

All of the surface water-supplied communities are located at least 40 km from the center of the Site. Four of the five are located to the north or northwest of the SRS, and one (Waynesboro) is located to the southwest. There is no strongly prevailing wind direction at the SRS, but a slight prevalence toward the southwest is observed (Arnett et al. 1992). Tritium deposition levels have been shown to steadily decrease with distance from the Site (Murphy et al. 1991). SRS air monitoring performed in these communities during periods of relatively high tritium releases indicates that the tritium levels at these distances are low (see Chapter 8 for a detailed discussion regarding measurements made for air and rainwater samples). Therefore, the contribution of SRS-released tritium to these surface water sources is expected to be small and difficult to detect. In fact, the average tritium measurements reported for the community water supplies are frequently at or below the detection limits (Figure 13-3). For those individual measurements that are substantially above the detection limits, it would be difficult to distinguish between any SRS-contributed tritium and tritium from weapons fallout. This is true especially during the 1960s, when fallout levels were at their peak (Wyerman et al. 1970).

However, it should be noted that the SRS has occasionally reported elevated levels of tritium in these water supplies and attributed these increases to Site operations (e.g., Ashley et al. 1982).

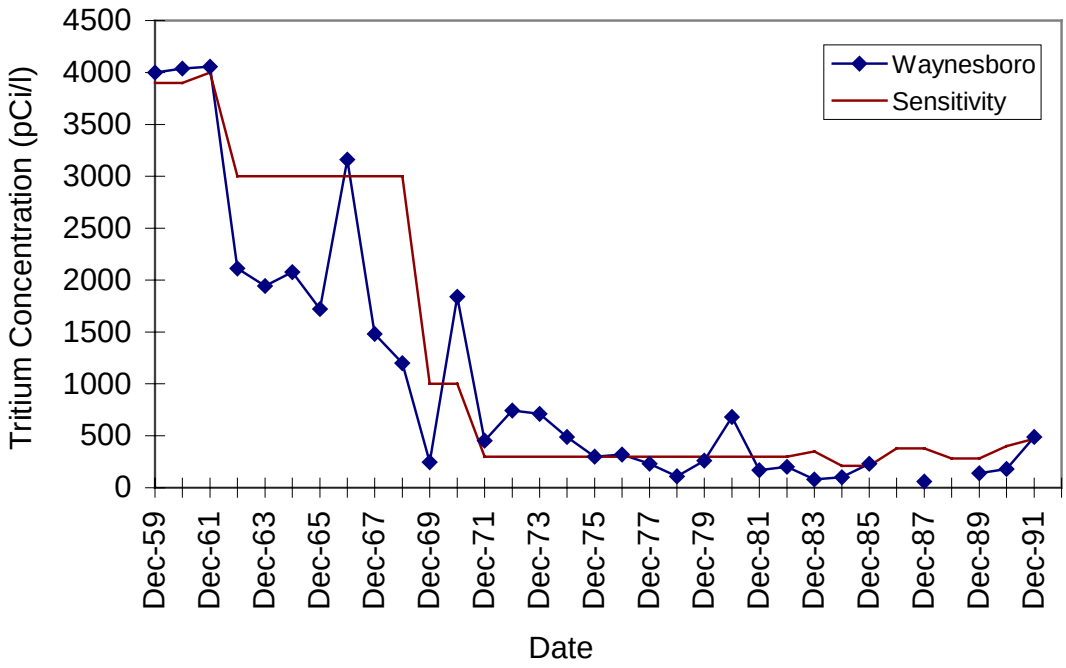


Figure 13-3. Average tritium measurements in Waynesboro, Georgia, drinking water (surface water supplied) from 1959 through 1991. Grab samples of tap water were collected monthly until 1967 when the sampling frequency was decreased to semiannually. Many of the tritium measurements were at or below the sensitivity (detection limit) of the analytical method. The general decrease over time can be attributed to decreases in the levels of fallout tritium from weapons testing and improvements in the detection sensitivities.

Community Water Supplies Data Workbook

The following data for each of the five communities supplied by surface water—taken from aperture cards and the semiannual and annual monitoring reports—have been compiled into a Microsoft Excel workbook, CWSDATA.xls:

- Gross beta and tritium data for monthly samples for 1959 through 1966
- Gross beta and tritium data for semiannual samples for 1966 through 1973
- Gross beta and tritium semiannual or annual averages for 1954 through 1992.

In addition, semiannual and annual average gross beta and tritium values for three of the well water supplied communities (Jackson, Bath, and Langley) have been compiled in the CWSDATA workbook. Annual ^{89,90}Sr data that are reported in the annual reports after 1987 have also been entered. In the electronic version, double-clicking on the following hyperlink provides access to this workbook: CWSDATA.xls.

The CWSDATA workbook contains an introductory spreadsheet that provides an overview of the workbook and individual spreadsheets for each of the eight communities for which data

have been entered (Aiken, Augusta, Bath, Clearwater, Jackson, Langley, N. Augusta, and Waynesboro). The workbook also contains a "Data Plots" spreadsheet that provides sample plots of the average and individual gross beta and tritium measurements for some of the community supplies. The "USGS" spreadsheet provides a comparison of U.S. Geological Survey (USGS) tritium measurements in southeastern streams during the early 1960s to SRS measurements in three of the surrounding communities.

Downstream Drinking Water Treatment Plants

Two drinking water treatment plants use Savannah River water downstream of the SRS: Beaufort-Jasper and Port Wentworth (also called Cherokee Water Treatment Plant) (Figure 13-2). SRS liquid effluents enter the Savannah River via the Site streams. Therefore, these downstream treatment plants are a source of direct exposure to the public. The following is a discussion of the monitoring information that is available for these two downstream plants. Until 1983, monitoring of an upstream control, the North Augusta Drinking Water Treatment Plant, was limited to the monthly or semiannual grab samples discussed above in the Community Water Supplies section. In late 1983, the SRS began collecting composite samples of raw and finished water from the North Augusta plant for comparison to data generated for the Beaufort-Jasper and Port Wentworth plants.

The treatment processes used by the Port Wentworth, Beaufort-Jasper, and North Augusta treatment plants are similar, consisting of flocculation, settling, and filtration (Figure 13A-1, Addendum 13A).

The monitoring history and available data for the drinking water treatment plants are discussed below. A summary of the data available for individual sample measurements (i.e., not summary data) is provided in Table 13-2.

Monitoring History and Available Data for Port Wentworth

The Port Wentworth Plant is named the Cherokee Hill Water Treatment Plant and is frequently referred to as Savannah or Savannah Industrial in SRS documents. It has been in operation since the time of SRS plant startup. The water plant is supplied via Abercorn Creek. In 1963, it was reported that at least 50% of the water in Abercorn Creek came from the Savannah River (Du Pont 1963a).

The Cherokee Hill Water Treatment Plant provides water for an industrial complex, and can also supply up to 10 million gallons per day to the City of Savannah in times of water shortage (Du Pont 1963a). The City of Savannah is normally supplied by well water. Approximately 15,000 workers in the industrial complex served by the Cherokee Hill plant potentially consume drinking water. Furthermore, there are indications that two soft drink bottling companies may have operated in the industrial complex for some period of time before 1990 (Hamby 1991).

Table 13-2. Available Individual Sample Data from Routine Monitoring of the Drinking Water Treatment Plants Supplied by the Savannah River[a]

Treatment Plant	Dates	Available Data	Data Source
North Augusta (located upstream of SRS, operational at time of SRS startup)	1959 – 1973	Monthly gross alpha, gross beta, and tritium	Aperture cards
	1976 – 1983	Semiannual gross alpha and beta, and tritium	Health Protection Dept. Monthly Data Reports
	1983 – 1990	Monthly gross alpha, gross beta, and tritium	Health Protection Dept. Monthly Data Reports
Port Wentworth (located downstream of SRS, also called the Savannah Industrial WTP, operational at time of SRS startup)	1/65 – 11/66	Monthly tritium	Johnson 1966 (memo)
	4/65 – 10/66	Monthly gross alpha and beta	Johnson 1966 (memo)
	1967 – 1971	Monthly tritium	Aperture cards
	1968 – 1971	Monthly gross alpha and beta	Aperture cards
	1976 – 1990	Monthly gross alpha and beta, and tritium	Health Protection Dept. Monthly Data Reports
Beaufort-Japer (located downstream of SRS, began operation in 1965)	4/65 – 10/66	Monthly tritium	Johnson 1966 (memo)
	1966 – 1973	Monthly tritium	Aperture cards
	1970 – 1973	Monthly gross alpha and beta	Aperture cards
	1976 – 1990	Monthly gross alpha and beta, and tritium	Health Protection Dept. Monthly Data Reports

[a]Does not include summary data reported in semiannual and annual site environmental monitoring reports

No monitoring data for the Port Wentworth treatment plant are included in the SRS semiannual and annual monitoring reports before 1971. However, references to monitoring, as well as some data, for the early to mid-1960s have been identified. No record of monitoring for years before 1963 has been identified. The monthly reports for 1963 and 1964 discuss a sampling program that involved collection and analysis of water at various stages of treatment to determine removal efficiencies for several radionuclides of interest (Du Pont 1963a, 1963b, 1964a, 1964b). A summary of the data collected from May 1963 to March 1964, as reported in Landon (1963) is provided in Table 13A-1, Addendum 13A. The average removal efficiencies observed for the treatment process (coagulation, settling, and filtration) over the 12-month period ranged from 35

to 79%. These averages are said to represent a wide range of treatment conditions because the quality of the raw water varied over the sampling period (Du Pont 1964a).

The monthly reports provide limited information about the removal efficiency study and few interim data are reported. However, based on one set of interim data provided for May 1963, it appears samples were collected daily and composited monthly for analysis (Du Pont 1963b). The removal efficiencies observed during May are reported for the following isotopes: ^{89,90}Sr, ^{144}CdPr, ^{51}Cr, ^{103}Ru, ^{137}Cs, ^{95}Zr/Nb, ^{54}Mn, ^{106}Ru, and ^{140}BaLa. A statement in the January 1964 monthly report indicates that, at least up to that point, sample volumes of 25 L were collected and counted for 10 minutes (Du Pont 1964b).

The sampling effort at the Port Wentworth plant also involved the installation of collection pots in the settling basins for continuous sampling of "river sediment." A similar collection system was installed at the North Augusta Drinking Water Treatment Plant, upstream of the SRS, for background sampling. The first month's sediment data, as reported in Du Pont (1964b), are provided in Table 13A-2, Addendum 13A. Although the data are presented as concentrations in river sediment, they actually represent measurements in the sludge generated during the water treatment process (i.e., flocculation followed by settling). The radionuclide concentrations in these sediments do not depend solely on the concentrations present in the raw water. Other factors that potentially affect the levels include:

- Solids content of the raw water
- Treatment process (especially the type and amount of flocculating chemicals added)
- Frequency and method by which sediments are removed from the settling basins
- Water content of the sediments (Walker 1978).

Therefore, the value of these data to the dose reconstruction would be primarily qualitative. However, in conjunction with river monitoring data, these data could provide useful information. For example, based on the expected removals during water treatment (Table 13A-1, Addendum 13A) and the measured concentrations in the sludge (Table 13A-2, Addendum 13A), it can be estimated that approximately 30 times more ^{51}Cr than ^{137}Cs was present in the raw water treated by the Port Wentworth plant during the time this sludge was generated. This estimate is in good agreement with a statement in a 1963 monthly report that ". . .the Plant has released approximately 30 times as much ^{51}Cr as ^{137}Cs. . . (Du Pont 1963c)." Despite the apparent close agreement, it should be emphasized that the estimate obtained using the sludge data represents only a rough approximation. Several important factors are not considered in this assessment, including

- Differences in the fallout levels of ^{51}Cr and ^{137}Cs present in Savannah River water
- Differences in the losses of ^{51}Cr and ^{137}Cs, primarily to sediments, during transport from the point of release to the drinking water treatment plants

The only other data from the treatment plant sediment sampling program that have been identified to date are gross alpha and beta measurements for samples from the North Augusta and Port Wentworth plants from January 1965 through November 1966 (Johnson 1966).

Water monitoring data for the Port Wentworth plant are reported in the Site semiannual and annual monitoring reports beginning in 1971. The data reported are gross alpha, gross beta, and tritium (averages, maximums, and minimums reported after 1973). Cesium-137 data are reported for the years 1983 through 1985. In addition to these summary data, the following sources of

individual water monitoring data have been identified for the Port Wentworth plant and catalogued in *RAC*'s SRS document database:

- Average ^{137}Cs concentration measured in finished water from the plant (0.29 pCi L^{-1}) during a 1-week period in December 1965 from Hayes and Boni (1983)
- Tritium concentrations in monthly grabs samples of water from April 1965 through October 1966 from Johnson (1966)
- Monthly gross beta concentrations in raw and finished water from February 1966 through November 1966 from Johnson (1966)
- Gross alpha and beta data for monthly samples of raw and finished water for 1968 through 1971 from aperture cards
- Tritium data for monthly samples of raw and finished water for 1967 through 1971 from aperture cards
- Gross alpha, gross beta, and tritium data for monthly samples for 1976 through 1990 from the Health Protection Department Monthly data reports.

Special survey data for the Port Wentworth plant (also called the Cherokee Water Treatment Plant) are included in several of the annual reports after 1980. For example, during 1986 the SRS performed a comprehensive survey of Abercorn Creek and the Port Wentworth plant to assess the impact of the restart of L-Reactor in 1985. The study consisted of three quarterly surveys during which water sediment, vegetation, and aquatic specimens were collected (Zeigler et at. 1987). In 1991, another comprehensive survey was performed in support of the proposed K-Reactor restart (Arnett et al. 1993).

Monitoring History and Available Data for Beaufort-Jasper

A second downstream drinking water treatment plant began operation in January 1965. The Beaufort-Jasper plant was constructed to supply domestic water, at a capacity of 8.8 million gallons per day, for a population of approximately 50,000 people represented by the following six major customers: Beaufort, South Carolina; Port Royal, South Carolina; Parris Island Marine Base; Laurel Bay, South Carolina; U.S. Navy Hospital; and Marine Air Base (Marter 1965).

The Beaufort-Jasper plant was designed to receive water via a 21-mi canal from the Savannah River, approximately 90 mi downstream from the SRS, to Mayer's Lake. In addition to Savannah River water, other surface water and groundwater enters the canal and Mayer's Lake. In early 1965, the treatment plant manager reportedly stated that he believed very little of the water they treated originated in the Savannah River. This belief was based on low mineral solids levels and high dissolved organics, which he believed originated in swamps that drain into the supply lake (Marter 1965). In 1985, modifications were made to the Beaufort-Jasper water pumping and transport system that increased the percentage of water supplied by the Savannah River (Hayes and Marter 1991). Before these modifications, the tritium levels reported for Beaufort-Jasper water are consistently lower than those observed in water from the Port Wentworth plant. After the changes were made, the tritium levels in the two downstream plants were essentially the same (Figure 13-4).

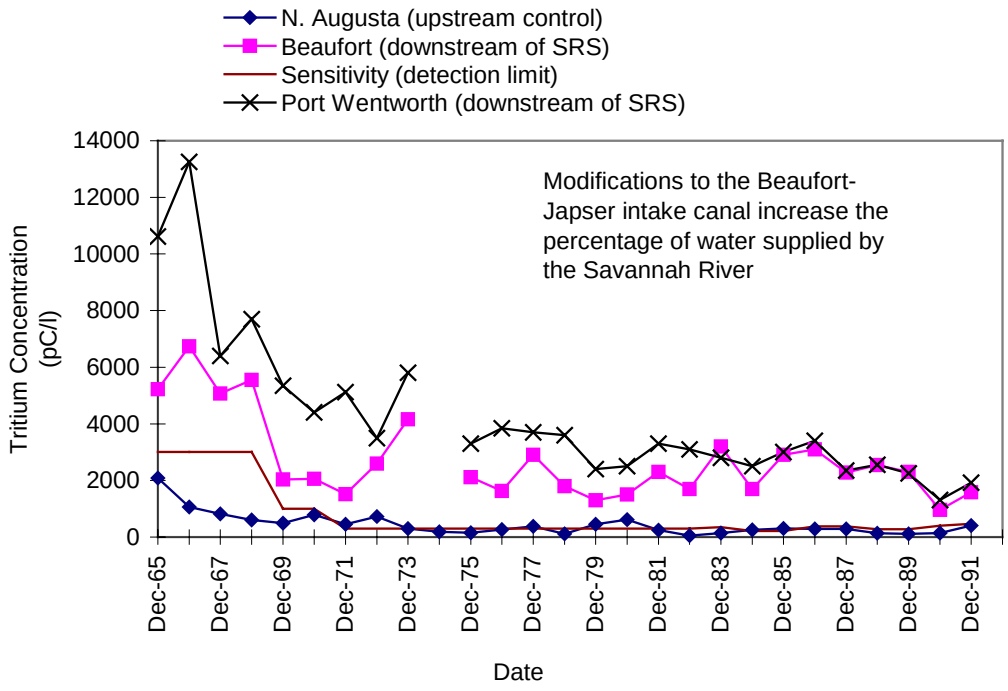


Figure 13-4. Average tritium measurements in treated water from three drinking water treatment plants supplied by the Savannah River: N. Augusta located upstream of the SRS and Beaufort-Jasper and Port Wentworth located downstream of the SRS. Until the mid-1980s when the Beaufort-Jasper supply canal was modified, the tritium levels in Beaufort-Jasper water are lower than those in the Port Wentworth plant because of dilution within the canal resulting from infiltration of surface water and groundwater.

The SRS has monitored water from Beaufort-Jasper for tritium since the treatment plant went online. Monthly data for early 1965 through late 1966 are reported in Johnson (1966). Beginning in 1967, semiannual tritium averages for the Beaufort-Jasper plant are provided in the SRS environmental monitoring report series, *Effect of the Savannah River Plant on Environmental Radioactivity* (identified by report numbers DPST-YY-30-1 and 2). Gross alpha and beta data averages are not included in the Site monitoring reports until 1971.

The following sources of individual sample data for the Beaufort-Jasper plant have been catalogued in the *RAC* document database:

- Average ^{137}Cs concentration measured in finished water from the plant (0.04 pCi L^{-1}) during a 1-week period in December 1965 from Hayes and Boni (1983)
- Tritium data for monthly samples for April 1965 through October 1966 from Johnson (1966)
- Tritium data for monthly samples (specified as raw water after 1968) for January 1966 through 1973 from aperture cards
- Gross alpha and beta data for monthly samples (raw water) for 1970 through 1973 from aperture cards

- Monthly gross alpha, gross beta, and tritium measurements for 1976 through 1990 from the Health Protection Department Monthly data reports (raw water only before 1982, raw and finished water beginning in 1982).

In addition to monitoring water from the Beaufort-Jasper plant for tritium, the SRS arranged (through the South Carolina Board of Health) to obtain urine samples from individuals who consumed the water for tritium analysis. This special sampling was part of an evaluation of the overall impact of SRS-released tritium on the radiation exposure of the surrounding population. The first sampling consisted of 21 samples collected from 14 individuals over a 1-month period. Samples from eight individuals in North Augusta, upstream of the SRS, were analyzed as controls. The average tritium concentration in the urine of the Beaufort-Jasper population was 2.51 ± 0.91 pCi mL^{-1}, and the average in the control population was 1.24 ± 0.21 pCi mL^{-1}. The samples were analyzed by a "new" chromatographic method developed at Savannah River Laboratory (SRL), referenced as DP 65-1-12. From 1967 to 1968, 140 urine samples were analyzed. The results for these samples are not provided. However, they are reported to correspond to an annual radiation dose of 0.4 mrem to the Beaufort-Jasper population as compared to 0.3 mrem for individuals using water from North Augusta (Du Pont 1966, 1968, 1969). No original data or reports relating to this study were located in the SRS records.

Primarily in support of reactor restarts, several extensive surveys of the Beaufort-Jasper plant and canal have been performed by the SRS during the 1980s and 1990s. For example, surveys that included collection of sediment cores, vegetation, water, fish, and turtles and analysis for a variety of specific radionuclides were conducted in 1982, 1986, and 1991 (Ashley et al. 1984; Zeigler et al. 1987; Arnett et al. 1993). Because, virtually no radionuclide-specific data, other than tritium, are available for the downstream drinking water treatment plants, these relatively recent surveys may provide useful data for the dose reconstruction.

Water Treatment Plants Data Workbook

All of the available aperture card and semiannual and annual report data described above for the Port Wentworth (Savannah) and Beaufort-Jasper water treatment plants have been compiled into a Microsoft Excel workbook, WTPDATA.xls. In the electronic version, double-clicking on the following hyperlink provides access to this workbook: WTPDATA.xls.

The tritium data reported in Johnson (1966) also are included. For the North Augusta Drinking Water Treatment Plant (the upstream control location), the community water supply monitoring data for 1965 through 1983 are provided along with the composite sampling data generated for this plant from late 1983 to the present. The WTPDATA workbook contains the following spreadsheets:

- Introduction
- Data plots
- Individual spreadsheets for Beaufort-Jasper, Savannah (Port Wentworth), and N. Augusta.

Water Treatment Plant-related Data

Although radionuclide-specific analyses, other than tritium, were not routinely performed on drinking water from either of the downstream treatment plants, radionuclide-specific data are available for weekly composite samples of Savannah River water from locations upstream and downstream of the SRS beginning in 1963. The downstream monitoring point is located approximately 80 and 90 mi upstream of the Beaufort-Jasper and Port Wentworth intakes, respectively.

If the Savannah River monitoring data are used to evaluate doses from consumption of water from the downstream drinking water treatment plants, additional information will be needed, including

- Transit time from points of discharge, or monitoring locations along the river, to the treatment plant intakes
- Amount of dilution within the Savannah River and within the treatment plant supply systems (canal for Beaufort-Jasper and Abercorn Creek for Port Wentworth) by incoming surface water and groundwater
- Fractions of various radionuclides that would be lost to sediments during transport within the river and within the water intake systems (i.e., in the Beaufort-Jasper supply canal and Abercorn Creek, which supplies the Port Wentworth plant)
- Removal efficiencies for radionuclides of interest during water treatment.

The transit time for contaminants discharged to the Savannah River from the SRS to reach the downstream drinking water treatment plants was estimated from a dye study conducted by the SRL Environmental Technology Section. The dye study was performed in conjunction with a water quality study performed by individuals from the U.S. Environmental Protection Agency (EPA), the South Carolina Department of Health and Environmental Control (SCDHEC), and the Georgia Department of Natural Resources (GDNR) (Hayes 1991). Hayes reports that a more comprehensive study of the river was to be performed in August 1991. Results of this further study have not been located. The dye study was used to determine travel times from or near the mouths of each of the Site effluent streams to the water treatment plants. A plot showing the time of arrival of the leading edge and peak at key locations along the river, including the treatment plant intakes, is provided (reproduced in Figure 13A-1, Addendum 13A). Table 13-3 contains values interpreted from this graph. These data show an approximate travel time from Steel Creek to the Beaufort-Jasper water treatment plant pump station of 76 hours. No information is reported about flow conditions during the time of the dye study, and the dates of the study are not provided.

Additional information on the transport and dilution of SRS-released contaminants in the Savannah River is reported in Hayes and Marter (1991). The information provided includes historical flow rates for the Savannah River at the downstream gauging station at Highway 301, which were compiled for use in retrospective dose calculations. In addition, flow rate data for the Beaufort-Jasper and Port Wentworth water treatment plants are derived by adjusting Highway 301 flow rates. The flow rates are adjusted for dilution from adding water to the river between Highway 301 and the points where the water treatment plants obtain water from the river and "additional dilution in the pumping and transport systems between the river and the water treatment plants." The annual dilution factors reported in Hayes and Marter (1991) for both

downstream drinking water plants are provided in Table 13A-3, Addendum 13A. The methodology used to calculate the dilution factors is described in Hayes (1983, cited in Hayes and Marter 1991).

Table 13-3. Results of the Savannah River Laboratory Time of Travel Dye Study on the Savannah River in 1991[a]

Location (approximate river miles above mouth)	Approximate time (hours)
From Steel Creek (142) to Highway 301 (120)	14
From Highway 301 (120) to Clyo, Georgia (63)	37
From Clyo, Georgia, (63) to Beaufort-Jasper pump station (38)	25
From Beaufort-Jasper pump station (38) to Savannah pump station (30)	5

[a] Source: Hayes (1991).

Further insight into transport and dilution of contaminants within the river can be obtained from data generated during special, more extensive, SRS monitoring activities performed following nonroutine releases—such as the December 1991 K-Reactor tritium release. This estimated release of approximately 5700 Ci of tritium, contained in about 150 gal of water from a K-Reactor heat exchanger, occurred over a 3-day period from December 22 through 25. Elevated tritium levels were first detected at the Beaufort-Jasper and Port Wentworth treatment plant intakes on the morning and afternoon of December 28, respectively. Peak concentrations of 52,000 to 56,000 pCi L^{-1} were observed at these locations approximately 30 hours after arrival of the leading edge of the plume (Arnett et al. 1992).

Dilution is not the only means by which radionuclide concentrations are reduced during transport within the river. For some radionuclides, losses to sediments and aquatic vegetation can be significant. A special study conducted in 1965 looked at reductions in discharged ^{137}Cs concentrations from these removal mechanisms. Data from the 1965 study are reported in Hayes and Boni (1983) as part of their evaluation of the potential impacts from the restart of L-Reactor. The data are used to evaluate the expected ^{137}Cs concentrations in finished drinking water from SRS releases. Information is provided on dilution and removals within the river and within the supply systems for the treatment plants. Measurements were made in the Savannah River above the SRS at Augusta, below the SRS at Highway 301, and below the water treatment plant intakes at Highway 17. Port Wentworth is approximately 7 mi upstream and Beaufort-Jasper is approximately 17 mi upstream of the Highway 17 sampling point. The study found a 48% reduction in the ^{137}Cs levels within the river between Highway 301 and Highway 17. Based on the increase in Savannah River flow between these two locations, an approximately 20% reduction from dilution would be expected. Thus, the additional 30% is attributed to the process

of deposition/sorption/reequilibration within the river channel and freshwater tidal marshes. The fresh water marshes extend upriver beyond Highway 17 to within the vicinity of Abercorn Creek.

The total reductions observed from Highway 301 to the finished water were 97.5% for Beaufort-Jasper and 79.3% for Port Wentworth. Because the treatment processes are similar, the primary reasons for the difference is attributed to dilution and losses within the Beaufort-Jasper supply canal.

The 1965 study did not include sampling of raw water from the treatment plants. Therefore, the removal efficiencies across the plants cannot be calculated. For their evaluation of potential impacts from L-Reactor releases, the authors assumed that the concentration measured in the river at Highway 17 is representative of the concentration entering the Port Wentworth treatment process (i.e., no reduction within Abercorn Creek). The validity of this assumption—the basis for which is not provided—should be further evaluated. However, if this assumption is used, a 62% removal of ^{137}Cs during water treatment at Port Wentworth is estimated. This estimate is close to the average removal of 60% observed for this radionuclide at Port Wentworth during 1963 to 1964 (Table 13A-1, Addendum 13A).

The removal of plutonium during water treatment at the North Augusta, Beaufort-Jasper, and Port Wentworth plants was investigated by the SRL in the mid-1970s. Removal factors for influent concentrations in the femtocurie range (0.1 to 3.5 fCi L^{-1}) were determined via collection of 50 to 10,000-L samples over a 9-month period. The 10,000-L samples were concentrated using online ion exchange columns at the water treatment plants. Smaller samples were concentrated in the laboratory under more controlled conditions. Following chemical separation samples were counted using an ultra low-level alpha spectrometry system. The findings are reported as removal factors (raw water concentration/treated water concentration). The reported mean removal factors ranged from 5.3 ± 3.4 in the North Augusta plant to 21.0 ± 10.2 in the Port Wentworth plant. The overall mean removal factor was 14 ± 10 (Corey and Boni 1975).

SAVANNAH RIVER MONITORING

The Savannah River forms the southwestern border of the Site and has received SRS effluents via Upper Three Runs, Four Mile Creek, Pen Branch, Steel Creek, Lower Three Runs, Beaver Dam Creek, and up through 1957, the TNX effluent ditch. Members of the public are potentially exposed to these effluents from drinking water supplied by the downstream treatment plants, ingestion of fish and other aquatic organisms taken from the river and estuary, and recreational use of the river.

Monitoring History and Available Data

Preoperational monitoring of the Savannah River was conducted in 1951 and 1952 and is reported in Reinig et al. (1953). Samples from about 15 locations along the river were analyzed for gross alpha, gross beta, uranium/plutonium alpha, radon, radium, and ^{40}K content. The preoperational monitoring report provides plots of the average concentrations for each location. The gross alpha and beta averages represent approximately 20 samples per location. Along with the plotted averages by location, an average value for all locations combined is reported. These values are provided in Table 13A-4, Addendum 13A.

Since the time of plant startup, the SRS has routinely sampled the Savannah River along the Site boundary and downstream of the Site to evaluate the impacts of liquid releases. Three locations have been consistently sampled throughout the Site's operation: (1) #8–below the mouth of Steel Creek, (2) #9–below the mouth of Lower Three Runs, and (3) #10–approximately 10 mi downstream of Lower Three Runs at the Highway 301 bridge (Figure 13-5). Six other river monitoring locations have been used: (1) #3–below the mouth of Upper Three Runs, (2) #4–above the mouth of Four Mile Creek, (3) #5– below the mouth of Four Mile Creek, (4) #6–above the mouth of Steel Creek, (5) #7–above the mouth of Steel Creek, and (6) #11–approximately 60 mi downstream of the SRS, near Clyo, Georgia (Figure 13-5).

The SRS has also consistently collected samples upstream of the Site to assess the levels of fallout and background radionuclides present in the Savannah River. A location near Augusta, Georgia, was used as the upstream control from 1954 through 1960 (#1). In 1958, a second upstream monitoring point (#1A) was added above Upper Three Runs near the Site boundary (Figure 13-5). In 1961, the designation for location #1A was changed to #2, and monitoring at Augusta was discontinued. Since that time, #2 has been consistently used as the upstream control location. The SRS also monitored the Salkehatchie River as a control location for tritium up through mid-1964. In the 1973, the SRS began routine radiological monitoring of the Edisto River for comparison to measurements in the Savannah River.

Since the early 1960s, the SRS has used the results for monitoring points #2 and #10 to evaluate the plant's contribution to radionuclide levels in the river. A summary of the monitoring performed at these locations from 1954 through 1991 is provided in Table 13-4. Location #10 was sampled continuously, using a continuous drip sampler, as early as 1958. All other river locations were grab sampled up through 1960 to 1961. Between April 1960 and August 1961, paddle wheel samplers were installed at #2 and #10 as well as five other locations along the river (Marter and Boulogne 1961). River samples have consistently been collected weekly.

At the time of plant startup, analysis of river water samples was limited primarily to gross alpha and nonvolatile beta measurements. However, some of the monthly and semiannual reports for the early years of operation do provide data from decay studies that were periodically performed on river water samples.

Routine tritium monitoring of the river began as early as 1958, and as early as 1960, samples from locations #2 (#1A) and #10 were routinely analyzed for radioiodine, radiostrontium, and radiocesium. By late 1960, ion exchange media were used to concentrate radiostrontium and radiocesium in river samples resulting in a roughly four-fold decrease in the detection limits, from approximately 4 pCi L^{-1} to 0.8 pCi L^{-1} (Du Pont 1960). Analysis for ^{90}Sr in the upstream and downstream river samples began in September 1959 (Van Wyck 1960). Routine analysis of samples from locations #2 and #10 for specific radionuclides by gamma spectroscopy did not begin until 1963.

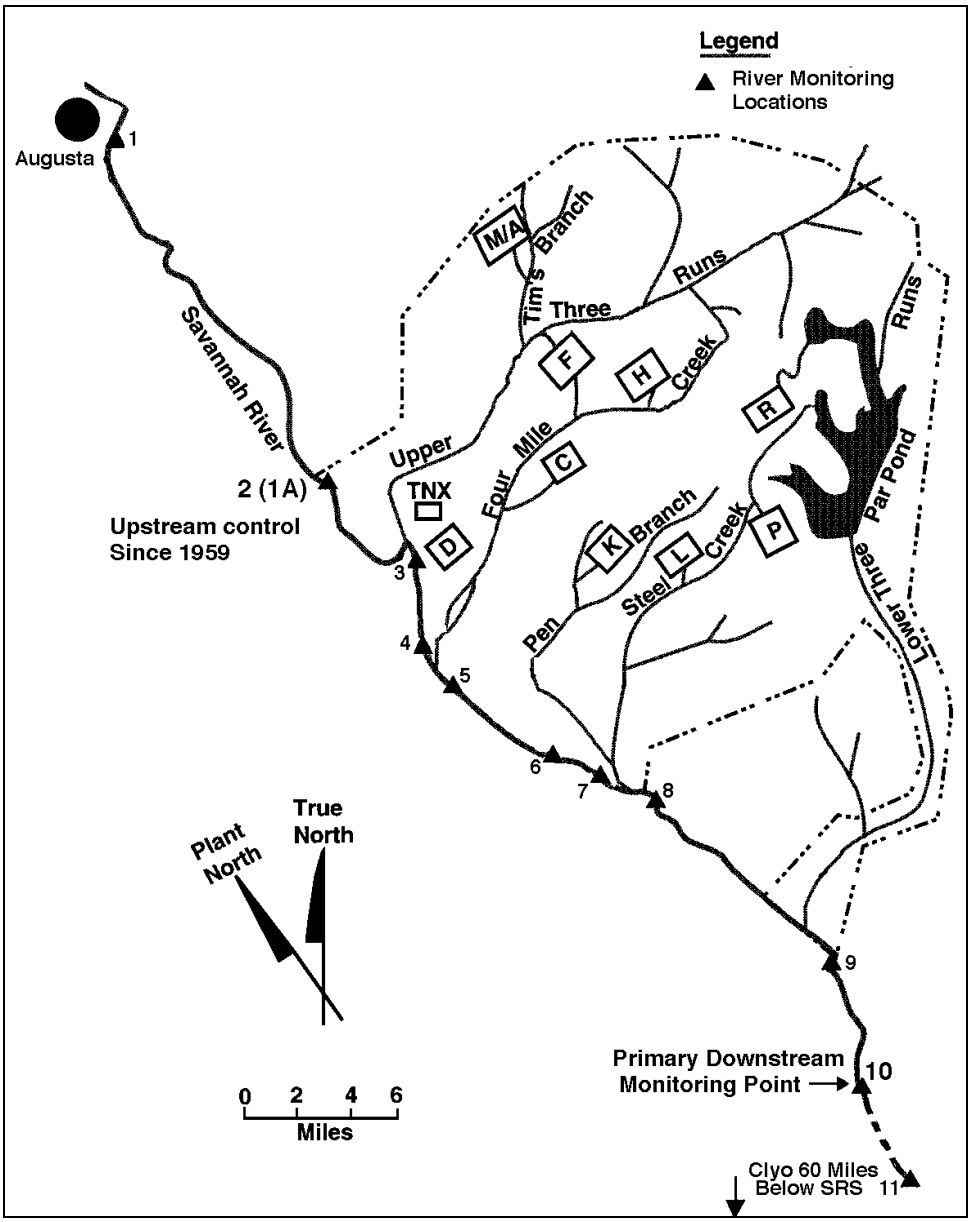


Figure 13-5. Map showing locations of SRS river monitoring points. Three locations on the Savannah River have been monitored by the SRS since the time of plant startup: #8 below the mouth of Steel Creek, #9 below the mouth of Lower Three Runs, and #10 approximately 10 mi downstream of Lower Three Runs at the Highway 301 bridge. Location #1 near Augusta, Georgia, was used as the upstream control from 1954 through 1960. In 1958, a second upstream monitoring point (#1A) was added above Upper Three Runs near the Site boundary. In 1961, the designation for location #1A was changed to #2, and monitoring at Augusta was discontinued. Since that time, #2 has been used as the upstream control location by the SRS.

Table 13-4. History of Monitoring at Upstream (#2) and Downstream (#10) Sampling Locations on the Savannah River

Sampling Point	Dates	Collection Frequency and Sample Type	Radiological Analyses
#2[a] (upstream)	1954 – 1957[a]	weekly, grab	gross alpha and beta
	1958	weekly, grab	gross alpha and beta, and tritium
	1959	weekly, grab	gross alpha and beta, tritium, and Sr-90
	1960 – 1962	weekly, continuous	gross alpha and beta, tritium, Sr-90, radiocesium, radioiodine, and radiostronium
	1963 - 1991	weekly, continuous	gross alpha and beta, tritium, Sr-90, and gamma spectroscopy
#10 (down-stream)	1954 – 1957	weekly, grab	gross alpha and beta
	1958	weekly, continuous	gross alpha and beta, and tritium
	1959	weekly, continuous	gross alpha and beta, tritium, and Sr-90
	1960 – 1962	weekly, continuous	gross alpha and beta, tritium, Sr-90, radiocesium, radioiodine, and radiostronium
	1963 - 1991	weekly, continuous	gross alpha and beta, tritium, Sr-90, and gamma spectroscopy

[a]From 1954 through 1958 the upstream was #1. A new location, #1A, located above Upper Three Runs near the site boundary was added in 1958. The designation for this location was changed to #2 in 1961.

In the spring of 1964, a new type of sampler was installed at locations #2 and #10. This sampler used an ion column to collect radioactive materials from a large volume of river water and was designed to operate continuously without electrical power. (See Appendix A for full description.) A water wheel drove two pistons pumps. One of these pumps passed water through a prefilter and the ion column. The other pump, which was geared much slower, delivered water directly into a 2-gal container. Because each pump had a fixed gear ratio, the volume of water in the container could be used to determine the volume of water that had passed through the ion column. For the first few months of operation (April through October), the volumes of water sampled ranged from 40 to 96 Ls depending on the river flow rate. A comparison of the results obtained at location #10 during this period, using both the ion exchange sampler and the standard paddle wheel sampler (Table 13A-5, Addendum 13A), found that the average concentrations for all radionuclides, except ^{131}I, were consistently higher for the ion exchange sampler. This difference was attributed to possible settling of radionuclides associated with suspended solids within the container of the paddle wheel sampler over the sampling period. Because the volume of water sampled with the ion exchange sampler is two to four times greater than the volume

collected by the paddle wheel sampler, the number of positive analyses was also consistently higher for this new sampling technique (Du Pont 1964c).

In June 1964, just after the ion-column samplers were installed at #2 and #10, data for a river sampling location #10B first appear on the river monitoring aperture cards. It is assumed that #10B was initially the collocated, standard paddle wheel sampler used for comparison to the ion exchange sampler. Based on telephone conversations with SRS environmental monitoring program personnel (Manly Grove on November 25, 1997, and Pete Fledderman on December 4, 1997), samplers #10A and #10B were "back-to-back" paddle wheel samplers in more recent years. These co-located samplers were used at the Highway 301 monitoring location up through the early 1990s when ISCO samplers were put into service for river monitoring. At this time, location #10 was moved approximately 0.25 mi downstream so that it could be physically attached to the Highway 301 bridge, and the use of co-located samplers was discontinued. While in operation, the purpose of sampler #10B was solely to provide a backup sample in the event that the #10A sampler did not operate properly. The SRS did not make comparisons of the results for #10A and #10B. Data for sampler #10B were consistently reported from 1964 through 1992. Available data for this sampler include tritium, strontium, gross alpha, and gross beta. These data could provide insight into the sampling uncertainties associated with the river monitoring data.

It is not clear from the available information how long the ion-column equipped samplers, tested in 1964, were used by the SRS. For the first few months of 1965, aperture card data for #2 and #10 are reported as prefilter, ion column, and total, indicating that these samplers were put into routine use. However, by the early 1970s when sampling methods were first described in the annual, offsite monitoring reports, ion-exchange were no longer used as part of the sampling collection process. The 1972 annual report describes continuous sampling of Savannah River water as follows:

> . . . a sampler consisting of a 'Plexiglass' water wheel suspended on two pontoons. As the water wheel is turned by flowing water, a small cup (or cups) on one paddle picks up a sample of water and deposits it into a trough. The sampled water flows by gravity from the trough through connecting tubing into a large polyethylene jug which trails the sampler. The sampled water (up to six gallons) is collected weekly at river locations above and below SRP. Increased analytical sensitivity for water samples (containing insufficient radioactivity for direct processing) is achieved through concentration of radionuclides by ion exchange. The ion exchange column is counted directly for gamma emitting radionuclides (Du Pont 1973).

The sampling equipment used by the SRS does not appear to have been modified from that time up through the early 1990s when the ISCO samplers were installed.

Savannah River Data Workbook

The Savannah River data reported in the semiannual and annual monitoring reports are limited to 6-month or annual averages, maximums, and minimums. Two primary sources of individual sample data for the Savannah River have been identified and catalogued in the *RAC*

database: aperture cards (handwritten data for roughly the years 1959 through 1973) and the Health Protection Environmental Monitoring Monthly data reports.

The aperture card data for eight radioactive materials measured at locations #2 and #10 have been compiled into a Microsoft Excel workbook, SRDATA.xls. Data were compiled for those radionuclides that were identified during the Phase I screening as contributing greater than 1% of the dose and for which specific monitoring had been performed (Meyer et al. 1995). These radioactive materials are ^{137}Cs (available data include nonspecific radiocesium and ^{137}Cs), ^{151}Cr, ^{60}Co, ^{131}I (available data include nonspecific radioiodine and ^{131}I), radiostrontium (nonspecific, ^{89,90}Sr, and ^{90}Sr), tritium, ^{65}Zn, and ^{95}Zr/Nb.

In the electronic version, double-clicking on the following hyperlink provides access to this workbook: SRDATA.xls.

The SRDATA.xls workbook contains the following individual spreadsheets:

- Introduction
- Data plots
- Individual spreadsheets forcesium, chromium, cobalt, iodine, tritium, ^{65}Zn, and ^{95}Zr/Nb

The specific data included in SRDATA.xls are as follows:

- Cesium— cesium (nonspecific)-1962 through May 1963 for location #10 only; cesium (ion column, nonspecific)-1962 through May 1963; ^{137}Cs-1963 through 1972 (excluding 1971); ^{137}Cs (low level)-1968 through 1972 (excluding 1971) for #10 only
- Chromium—^{51}Cr-1963 through 1972 (excluding 1971); ^{51}Cr (low level)-1968 through 1970 for #10 only
- Cobalt—^{60}Co-1963 through 1972 (excluding 1971)
- Iodine—radioiodine-1962 through 1967 for #10 only; ^{131}I-1963 through 1972 (excluding 1971); ^{131}I (low level)-1968 through 1970 for #10 only
- Strontium—strontium-1962 through 1964 for #10 only; strontium (ion column)-1962 through 1972 (excluding 1965 and 1971); $^{89/90}$Sr-1963 through 1972 (excluding 1971); ^{90}Sr-1962 through 1972 (excluding 1971)
- Tritium—^{3}H-1963 through 1972 (excluding 1965 and 1971), ^{3}H (sensitive method on monthly composites)-1962, 1966 through 1972 (excluding 1971)
- Zinc—^{65}Zn-1963 through 1972 (excluding 1971); ^{65}Zn (low level)-1968 through 1970 for #10 only
- Zirconium/Niobium—^{95}Zr/Nb-1963 through 1972 (excluding 1971); ^{95}Zr/Nb (low level)-1968 through 1970 for #10 only.

Other Savannah River monitoring aperture card data that are available through the *RAC* SRS Phase II document database but that have not been entered into the SRDATA.xls workbook include:

- 1965 data for tritium and strontium (nonspecific) and ^{90}Sr (not located until late in Phase II)
- 1971 data (original *RAC* copies not legible, new copies obtained late in Phase II)
- 1973 data (data for location #2 not located until late in Phase II)
- 1960 and 1961 ^{90}Sr data for #2 (#1A) and #10
- Tritium data for location #11 (1959 through June 1964 when monitoring at this location was discontinued)

- Cesium (nonspecific) data for #8, #9, #10, and #11 for 1959 through 1964
- 1963 through 1972 data for ^{54}Mn, ^{103}Ru, ^{106}Ru, ^{134}Cs, ^{140}BaLa, ^{144}Ce, and ^{239}Np for #2 and #10
- 1959 through 1973 gross alpha and gross beta data for all river monitoring locations
- 1959 through 1964 tritium data for the Salkahatchie River.

Savannah River Monitoring-related Data

The following provides a brief overview of additional information that may be useful in the evaluation of SRS releases to the Savannah River.

Flow Monitoring on the Savannah River

Weekly flow measurements for the Savannah River, as reported on the data aperture cards, for the year 1963 through 1972 (excluding 1971) are included in the SRDATA.xls workbook.

Flow rate data for four USGS gauging stations (Augusta; near Jackson [SRS Boat Dock]; Highway 301 [Millhaven, Georgia]; and Clyo, Georgia) are summarized in Hayes and Marter (1991). Monthly data (minimum, maximum, and mean) are also provided for these stations. USGS data have been generated for the Augusta and Clyo stations since 1950. Flow monitoring at the Highway 301 station also began in 1950, but it was discontinued from 1971 through 1981. Flows at Highway 301 for this 10-year period are estimated based on the correlation observed between flows at this station and Augusta for other years.

Recreational Use of the River

From December 1980 through December 1982, the GDNR collected data on the recreational use of the Savannah River. The SRL divided the GDNR data into three separate categories (boaters, swimmers, and shoreline activities) for statistical expansion. The data were used to estimate recreational use for the river above, adjacent to, and below the SRS. Estimates of time spent in recreational activity on the Savannah River are reported as person-hours per year. Estimates for boaters, swimmers, and shoreline activities adjacent to the SRS are 35,831 ± 8640, 688 ± 500, and 13283 ± 3386, respectively (Hutto and Turcotte 1983).

River Mile Designations

The river mile designations for locations along the Savannah River have changed over the years of SRS operations as the U.S. Army Corp. of Engineers has updated its navigational charts. A sketch showing mileage numbers for the river below Augusta is provided in Johnson (1968). The memo states that since plant startup, “river bends have been cut out and mileage numbers changed to the extent that Augusta is now about 20 miles closer to Savannah via the River.”

USEFULNESS AND LIMITATIONS OF OFFSITE WATER MONITORING DATA FOR DOSE RECONSTRUCTION

There are a number of factors that impact how the offsite water monitoring data may be used during subsequent phases of the dose reconstruction project. These factors include the availability of sufficient original monitoring data sets to verify reported summary data and to evaluate temporal trends, and the ability to distinguish between Site releases of contaminants and other sources of the same contaminants in the environment (i.e., to establish appropriate background concentrations).

Drinking Water Data

Because the safety of drinking water is of considerable interest to the public, the potential impacts of SRS operations on both the surrounding community water supplies and the downstream water treatment plants should be addressed as part of the dose reconstruction process. The influence of the Site is clearly seen in the data for the downstream drinking water treatment plants; however, this is not the case for the community water supplies.

Tritium is the only radionuclide for which specific monitoring has been performed in the community water supplies. The usefulness of these data for assessing contributions from SRS atmospheric releases is questionable. Tritium monitoring began at the end of 1959, but by the early 1960s, fallout tritium levels in surface waters of the U.S. were at their peak (Wyerman et al. 1970). Thus, for these years, it would be difficult to distinguish between the, presumably, low levels of tritium contributed by the SRS and that introduced from weapons fallout. The absence of a prevailing wind direction and the lack of a control location for the monitoring program would further complicate this task. By the later years, when fallout tritium levels are lower, the community water supply monitoring program is limited to analysis of semiannual grab samples. These infrequent sample collections would reduce the likelihood of identifying increases in tritium concentrations that might occur following a large, short-term release of tritium. In general, the tritium levels observed in the community supplies are low (Figure 13-3) and decreased over time following the downward trend of fallout levels and detection sensitivities.

In the case of the downstream drinking water treatment plants, a considerable amount of useful data—including original handwritten records—exists for quantifying the impacts from SRS releases of tritium to the Savannah River. It should be noted, however, that routine tritium monitoring of the Port Wentworth plant did not begin until the mid-1960s although this treatment plant had been online since before SRS operations began. Therefore, SRS tritium measurements in the Savannah River, which began in 1958, and tritium data from independent sources will be needed to estimate tritium concentrations in Port Wentworth water plant before 1965. Tritium measurements were made in the Savannah River near the downstream treatment plant intakes by the USGS and the U.S. Public Health Service beginning in the early 1960s (Wyerman et al. 1970; Chestnutt et al. 1966).

Although quantitatively distinguishing between background levels and site contribution are beyond the scope of this phase of the dose reconstruction, an effort has been made to assess the potential usefulness of upstream measurements for this purpose. In general, the upstream measurements, both in the drinking water treatment plants and the Savannah River, appear to be reliable indicators of background and fallout contributions. However, before 1983, tritium

monitoring in the upstream control for the drinking water treatment plants, North Augusta, was limited to the sampling performed under the community water supply monitoring program. Furthermore, the fact that the North Augusta plant was monitored under the community water supply program may raise concerns that this control location was potentially affected by SRS releases. Therefore, data sets from independent sources, such as the USGS and the U.S. Public Health Service, will be useful as cross-checks in the evaluation of weapons fallout tritium levels in the Savannah River. Because tritium would not be removed by the water treatment process, surface water monitoring data, as well as drinking water data, are of potential value.

The USGS began routine monitoring of surface waters across the U.S. in the early 1960s. Tritium data for 20 streams collected from 1961 to 1968 are reported in Wyerman et al. (1970). Factors affecting fallout tritium levels in streams include local precipitation conditions and the amount of groundwater, relative to runoff, that enters the stream. Because fallout tritium levels in precipitation vary with location, the most appropriate data sets for comparison to SRS monitoring information would be for streams located in the same general area as the Site.

In addition to sampling of the Savannah River near Port Wentworth, the USGS monitored three other rivers located in the Southeast: the Kissimee River near Okeechobee, Florida; the Apalachicola River at Chattahoochee, Florida; and the Neuse River near Vanceboro, North Carolina. The USGS data for the Kissimee and Neuse Rivers for the early 1960s are included in the CWSDATA workbook. A comparison of the data for 1962 through 1964 found the USGS tritium measurements in the Neuse River (Vanceboro, North Carolina) comparable to those made by the SRS in the Augusta and North Augusta Drinking Water Treatment Plants (Figure 13-6). The Apalachicola data were not included in this comparison because the USGS did not begin tritium monitoring in this river until the end of 1964.

Tritium monitoring of surface waters was also initiated by the U.S. Public Health Service in 1964. Reporting of these data began in July 1966 in Chestnutt et al. (1966). In 1969, the U.S. Public Health Service also performed tritium analysis on grab samples of drinking water from communities in at least 14 states across the U.S. Unfortunately, all of the supplies monitored in the southeastern U.S. were groundwater sources (PHS 1970).

With the exception of tritium, few radionuclide-specific data are available for the drinking water treatment plants. For the earlier years, the effect of Site releases is evident in the gross beta concentrations observed in the downstream drinking water treatment plants (Figure 13-7). Therefore, these gross beta data may be of qualitative use to the dose reconstruction.

For quantitative assessments, Savannah River monitoring data will provide the primary means for evaluating the impact of SRS releases of radionuclides other than tritium, such as ^{137}Cs and strontium, on the downstream drinking water supplies. In addition, these data are needed to evaluate the extent to which the public has been exposed to SRS-released contaminants through the ingestion of fish and other aquatic organisms and during recreational use of the river.

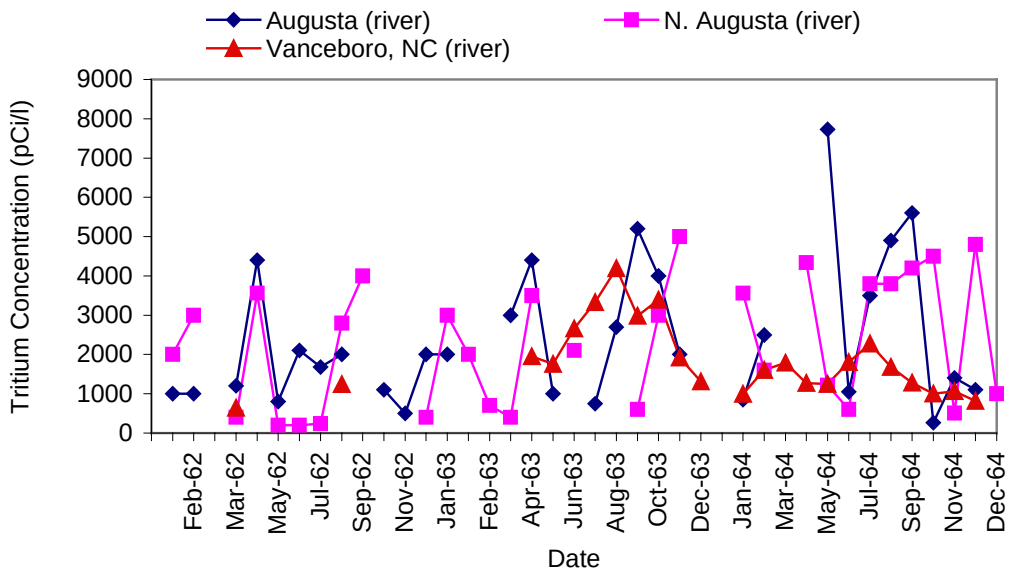
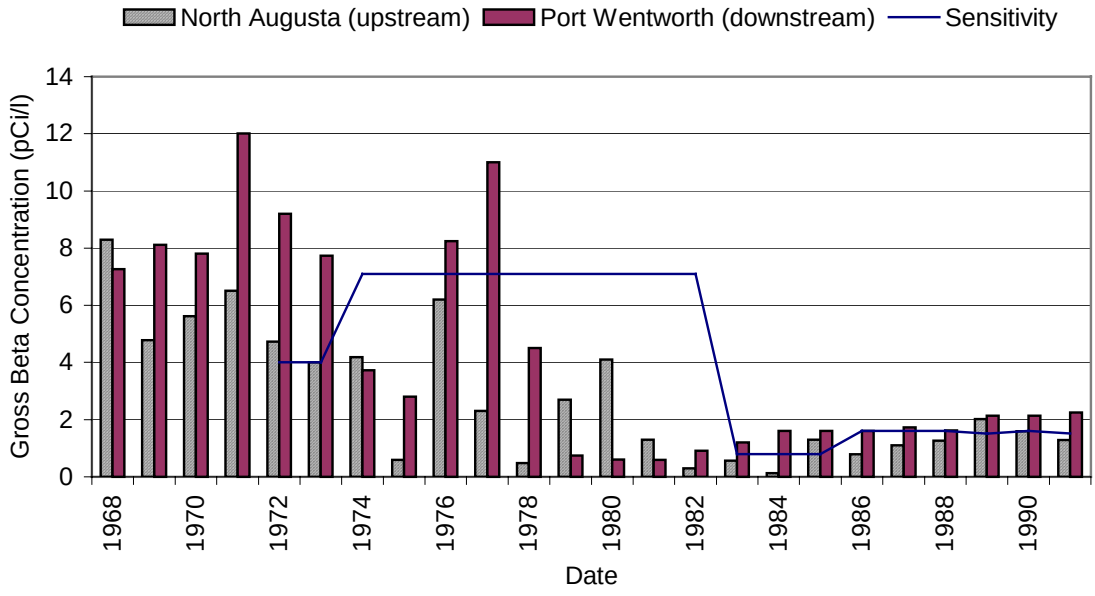


Figure 13-6. A comparison of SRS tritium measurements in drinking water from Augusta and N. Augusta (upstream controls supplied by the Savannah River) to USGS tritium data for the Neuse River in Vanceboro, North Carolina, for 1962 through 1964. Tritium is not removed by the water treatment process; therefore, the treatment plant data are reflective of Savannah River concentrations.



Figures 13-7. Average annual gross beta concentrations measured in finished (treated) water from the North Augusta Drinking Water Treatment Plant, located upstream of the SRS on the Savannah River, and the Port Wentworth plant located downstream of the Site. The influence of Site releases is evident in the gross beta concentrations up through the late 1970s. These data may be of qualitative use to the dose reconstruction.

The SRS began radionuclide-specific monitoring of the Savannah River, upstream and downstream of the Site in the late 1950s and early 1960s and has developed and extensive data set that will be of considerable use to the dose reconstruction process. As seen in a plot of ^{137}Cs concentrations measured in the river in 1963 and 1964, there have been times when the Site's contribution was masked by fallout (Figure 13-8). However, in general the upstream data provide a reliable indicator of fallout levels while the downstream measurements clearly show the influence of Site discharges. In the case of tritium monitoring, even in the early 1960s when fallout levels were at their highest in U.S. surface waters, the downstream tritium measurements are consistently higher than the levels observed upstream (Figure 13-9). Overall, the radionuclide-specific data will be useful for assessment of the Site's contribution of radionuclides to the Savannah River. (i.e., the validating source term estimates) and for directly assessing exposures of individuals who consumed drinking water form the treatment plants downstream of SRS.

The most notable gap in the offsite water monitoring data available for the SRS dose reconstruction is the lack of individual sample measurements for the mid-1950s. Based on the extensive records search performed by *RAC* during Phase I, it appears that no records of these early data have been retained by the Site. The available river monitoring data from the time of plant startup in 1954 through about 1958 are limited to sporadic reports in the weekly and monthly reports and the summary data provided in the semiannual monitoring reports.

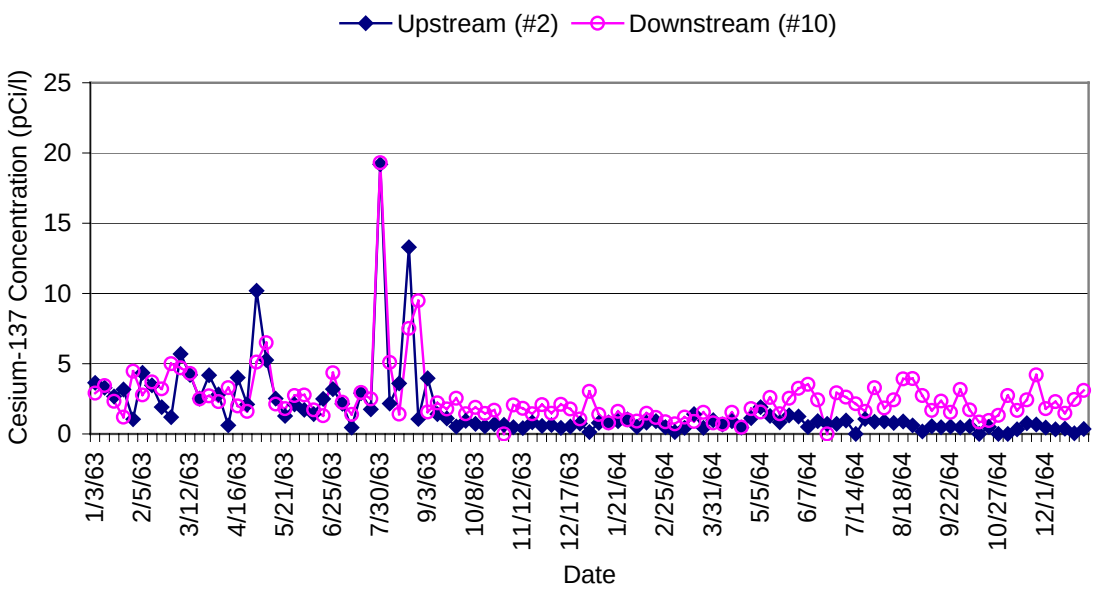


Figure 13-8. Cesium-137 concentrations measured in the Savannah River upstream and downstream of the SRS. The upstream measurements provide a means of quantifying the fallout levels of ^{137}Cs levels present in the Savannah River. There are periods of time—such as the first three quarters of 1963— when the SRS contribution is masked by fallout levels (i.e., the upstream measurements are higher than, or indistinguishable from, the downstream measurements). However, by late 1963, the downstream concentrations are consistently higher than those measured in the upstream control.

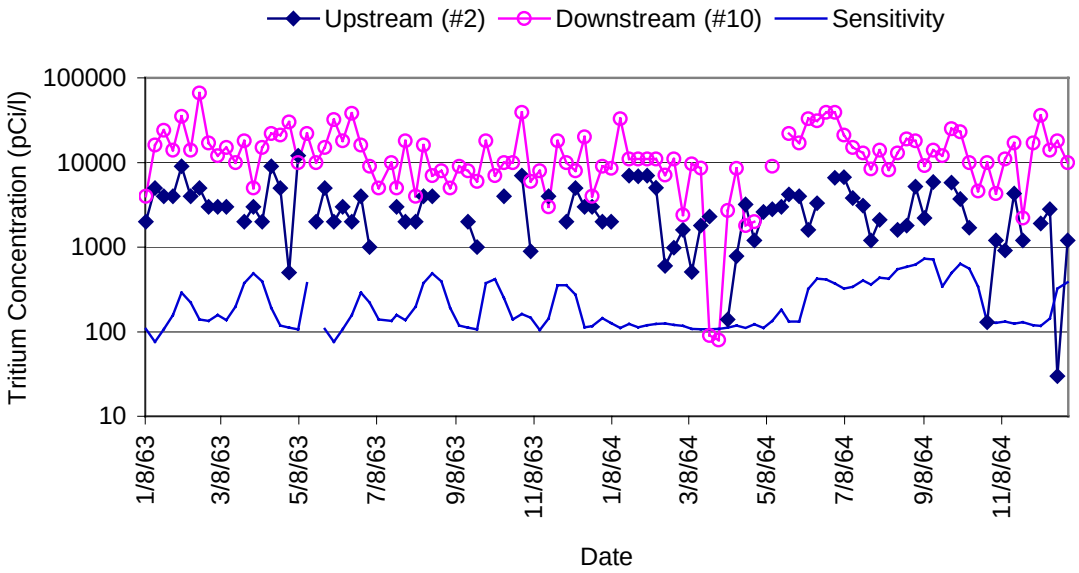


Figure 13-9. Tritium concentrations measured in the Savannah River upstream and downstream of the SRS in 1963 and 1964. The upstream measurements provide a means of quantifying the fallout levels present in the Savannah River. Even in the early 1960s when fallout levels of tritium are at a peak in U.S. surface waters, the influence of SRS releases can be seen in the downstream Savannah River measurements

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ADDENDUM 13A

ADDITIONAL WATER MONITORING DATA

ADDENDUM 13A—TABLES

Table 13A-1. Radionuclide Removal Data from Cherokee Hill Water Treatment Plant Downstream of SRS[a]

Radionuclide	SRP releases (Ci)	Influent (Ci)	Effluent (Ci)	Removal efficiency (%)	Average effluent conc. (pCi L^{-1})
Tritium	80,000	419	459		11,600
^{144}Ce	50.5	0.100	0.05	50	1.2
^{51}Cr	936	0.520	0.25	52	6.3
^{106}Ru	5.5	0.210	0.09	57	2.3
^{137}Cs	53	0.05	0.02	60	0.6
^{89}Sr	70	0.11	0.07	36	0.9
^{90}Sr	5.7	0.046	0.03	35	0.8
$^{95}Zr/Nb$	11.6	0.14	0.03	79	0.8

[a] Excerpted from SRP (Du Pont 1964a).

Table 13A-2. Comparison of Radionuclide Measurements in Settling Basin Sediment from the North Augusta Drinking Water Treatment Plant (upstream of SRS) and the Port Wentworth Water Treatment Plant (downstream of SRS)[a]

Radionuclide	North Augusta (pCi g^{-1})	Port Wentworth (pCi g^{-1})
^{144}Cd	32	49
^{51}Cr	<51	650
^{106}Ru	110	60
^{137}Cs	9	26
$^{95}ZrNb$	26	24
^{54}Mn	10	7

[a] Excerpted from SRP (Du Pont 1964b).

Table 13A-3. Estimated Annual Dilution of Contaminants in Savannah River Between Highway 301 and the Downstream Drinking Water Plants[a] Based on Tritium Measurements[b]

Year	Percent dilution below Highway 301	
	Beaufort-Jasper plant	Port Wentworth plant
1966	64	
1967	109	
1968	73	
1969	101	
1970	232	
1971	312	
1972	144	
1973	61	15
1974	104	19
1975	101	23
1976	153	20
1977	70	31
1978	131	9
1979	164	32
1980	131	30

[a] Values includes dilution between river and water treatment intakes (i.e., in canal for Beaufort-Jasper and Abercorn Creek for Port Wentworth).

[b] Source: Hayes and Marter (1991).

Table 13A-4. Results of Preoperational Monitoring of the Savannah River Performed by SRS in 1951 and 1952[a]

Analysis	Concentration[b]	Number of samples
Gross alpha	9.3 ± 2.9 ($\times 10^{-3}$ d/m mL^{-1})	20
Gross beta	0.6 ± 0.2 (pCi L^{-1})	20
Uranium/plutonium alpha	0.5 ± 0.3 ($\times 10^{-3}$ d/m mL^{-1})	3
Radon[c]	15.0 ± 6.0 (pCi L^{-1})	3
Radium	0.4 ± 0.3 ($\times 10^{-3}$ d/m mL^{-1})	3
^{40}K	2.2 ± 0.4 (pCi L^{-1})	4

[a] Source: Reinig et al. (1953).

[b] Average concentration for approximately 15 locations ± one standard deviation.

[c] Analyzed by precipitation of radon progeny for beta counting.

Table 13A-5. Comparison of Analytical Results of Savannah River Water by Two Sampling Systems-4/14/64–10/27/64[a]

	Average concentration ($pCi\ L^{-1}$)		Total number of positive analyses	
Radionuclide	Ion exchange	Standard	Ion exchange	Standard
^{144}Ce	6.9	5.8	13	5
^{51}Cr	30.4	17.4	16	12
^{131}I	1.4	1.7	4	3
^{106}Ru	5.9	4.6	11	2
^{137}C	3.7	2.3	26	24
$^{95}Zr/Nb$	0.9	0.6	16	3
Nonvolatile beta	18.5	16.4		

[a] Excerpted SRP (Du Pont 1964c).

ADDENDUM 13A—FIGURES

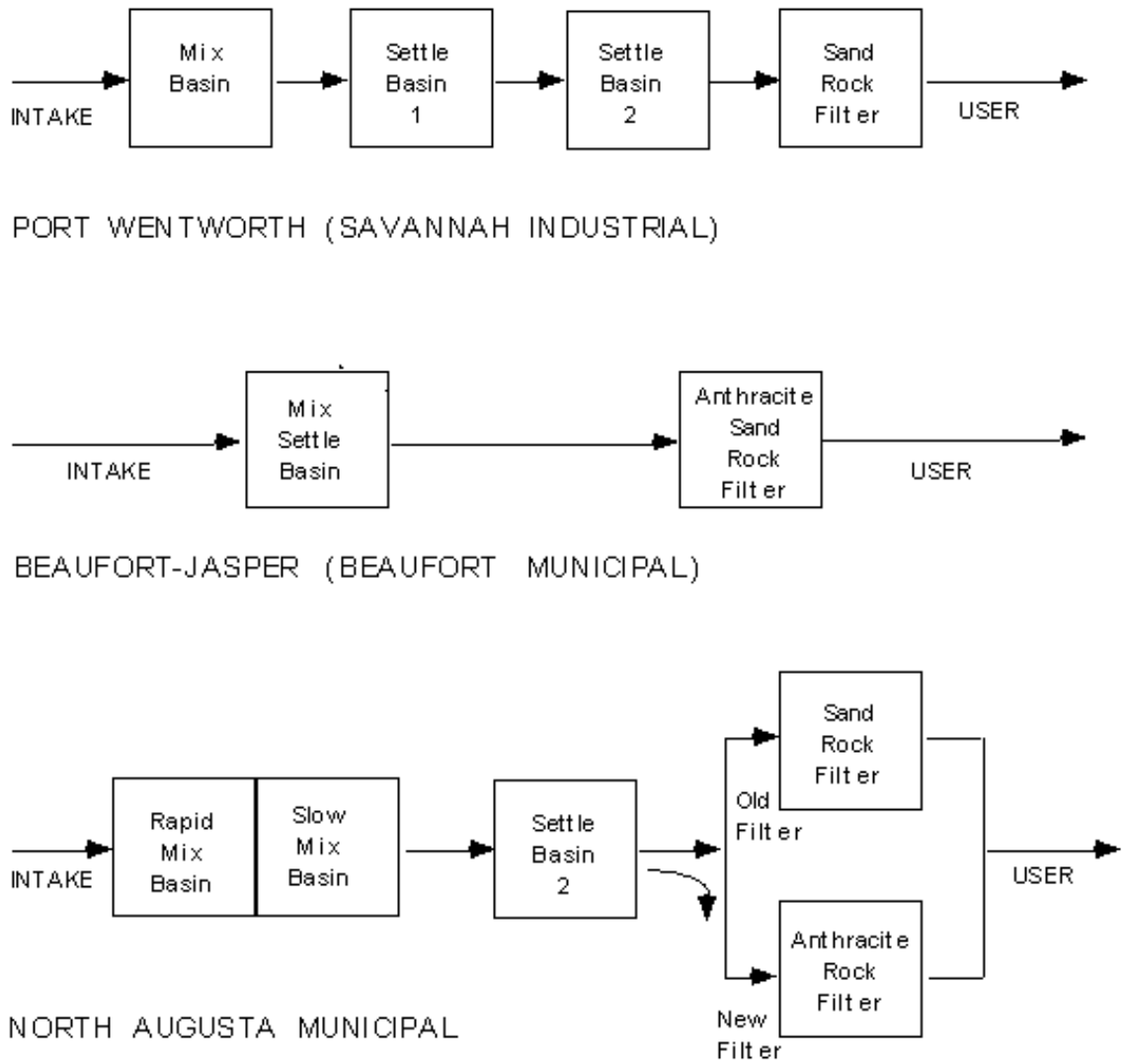

Figure 13A-1. Diagram of treatment processes used in three drinking water treatment plants that use Savannah River water. The Port Wentworth and Beaufort-Jasper water treatment plants are located downstream of SRS. The North Augusta Drinking Water Treatment Plant is upstream of the Site and is used as a monitoring control (reproduced from Figure 2 of Corey and Boni 1975).

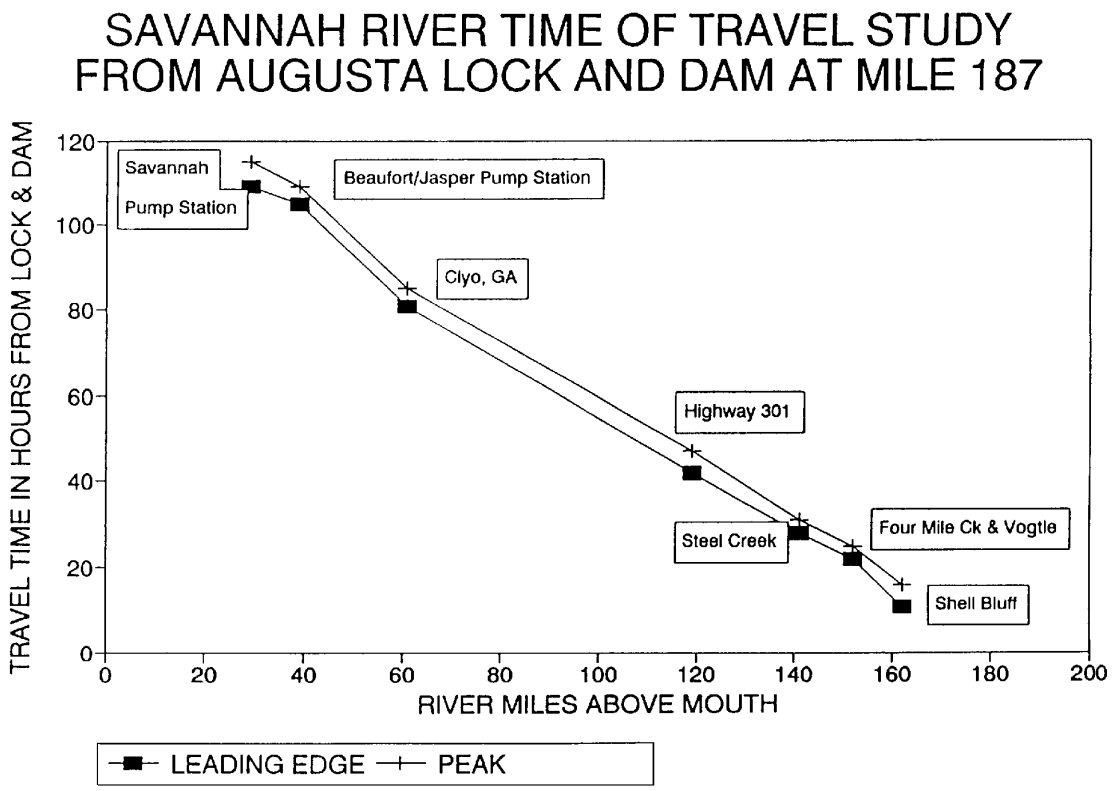

Figure 13A-2. Data obtained during a dye study conducted to determine the time of travel for contaminants discharged to the Savannah River by the SRS (reproduced from Hayes 1991). These data show an approximate travel time from Steel Creek to the Beaufort-Jasper water treatment plant pump station of 76 hours. No information is reported about flow conditions during the time of the dye study, and the dates of the study are not provided.

CHAPTER 14

RADIONUCLIDES IN FISH

ABSTRACT

This chapter discusses radionuclide concentrations measured in fish collected from onsite streams at the Savannah River Site (SRS) and offsite locations in the vicinity of the SRS, including the Savannah River. We evaluated these data with regard to their potential usefulness for dose reconstruction. We also attempted to validate the data by comparing concentrations reported in as many sources as possible. These data included original, hand-written compilations for many years. We have found these original data to consistently correspond to the data reported in monthly, semi-annual, and annual summary reports.

We compiled and examined fish data and other environmental information to determine their usefulness for source term verification, model validation, and direct exposure assessment. In general, the fish data are most valuable for direct exposure assessment, but they may also be useful for source term verification and model parameter development. Appendix K further discusses potential uses for environmental monitoring data.

INTRODUCTION

The SRS buffer area (about 300 mi^2) includes five major streams that flow into the Savannah River. This buffer area was designed to allow for direct discharge of reactor effluent to these streams with sufficient dilution occurring before the streams reached their confluence with the Savannah River, thereby minimizing offsite distribution of radionuclides. However, it was evident by the mid-1950s that fish populations in the Savannah River, particularly near the mouths of onsite streams, were being impacted by SRS activities.

Fish from the Savannah River represent a potentially important exposure pathway to people who may have relied on them for a significant portion of their diet. Although locations within the plant boundary have not been legally accessible to the general public, it is possible that some people poached fish from onsite locations. To estimate potential exposure to people ingesting fish from either the Savannah River or onsite locations during a later phase of this project, we must compile and evaluate radionuclide concentrations that have been measured in fish collected from these areas.

This section summarizes reported information regarding radionuclide concentrations in fish collected from locations on or in the vicinity of the SRS. We examined several sets of routine semiannual and annual environmental monitoring reports, prepared by the SRS contractor and spanning the years 1953 through 1992. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series.

Several additional documents have been reviewed for information pertaining to fish monitoring at the SRS. These include monthly monitoring reports from 1962 through 1965, weekly monitoring reports from 1959 through 1962, and aperture card printouts (handwritten data entry sheets) from 1970 through 1981. In general, the data reported in these documents is consistent with data provided in the monitoring reports. The radionuclides for which data are available for fish include gross alpha/beta and nonvolatile beta activity, ^{137}Cs, $^{89,90}Sr$, ^{65}Zn, ^{32}P,

and ^{3}H. Appendix A details analytical and counting procedures for fish. Mercury concentrations have also routinely been reported for fish and are discussed in Chapter 20.

Routine collection of fish began in July 1957 in response to increased releases of reactor effluent to Four Mile Creek, Steel Creek, Lower Three Runs Creek, and ultimately the Savannah River, resulting from a series of fuel element failures. Before July 1957, small numbers of fish were randomly sampled from onsite streams and the Savannah River.

Fish were collected by angling and trapping, with no mention of the specific type of trap(s). Before 1962, the frequencies of collection and analysis are unclear. From 1962 through July 1968, fish were collected and flesh and bone samples were composited for weekly and monthly analysis, respectively. A composite is a single sample comprising a number of individual fish. From August 1968 through 1970, fish were collected weekly, and flesh and bone samples were composited for monthly analysis at each location. Beginning in 1971, individual whole fish were analyzed for ^{137}Cs, except at Par Pond and Pond B, where samples were composited for monthly analysis, and bone tissue was composited for monthly $^{89,90}Sr$ analysis at all locations. The Site reports suggest that Par Pond and Pond B fish samples were composited for analysis through 1983, after which individual whole fish were analyzed. Sample collection frequencies are unclear since 1971, but they appear to have varied depending on the collection location, based on data provided in aperture card printouts from 1971 through 1983.

Little classification of fish species was made before 1971. The types of fish collected were reported to be predominantly bream including shellcracker, bluegill, and redbreast of the genus *Lempomis*. The bluegill (*Lempomis macrochirus*), a surface and bottom feeding omnivore, was reportedly selected in 1962 by the SRS as indicative of maximum uptake of radionuclides by edible species of fish (Harvey and Rabon 1965). This selection was based on the results of a nine month study conducted on bluegill, catfish, and largemouth bass collected from Par Pond in 1962 (Harvey 1963). The study indicated $^{134,137}Cs$ and $^{89,90}Sr$ to be distributed similarly in the three species of fish, but ^{65}Zn accumulated to higher levels in bluegill than in catfish or largemouth bass.

Beginning in 1971, the types of fish collected were further differentiated; the list included bream, catfish (predominantly yellow cat of the genus *Ictalurus*), and bass (predominantly large mouth of the genus *Micropterus*). Beginning in 1981, numerous other fish species were routinely reported, including carp, eel, mullet, perch, crappie, war mouth, bowfin, shad, flounder, gar, trout, suckers, chain pickerel, croaker, jack, whiting, spot, menhadden, and bluefish. Table 14-1 shows the percentages of bream, bass, catfish, and other fish collected at several locations since 1971, when classification of separate species began. The years during which these data were reported are also included.

BASIS FOR ANALYSIS

While concentrations for several different species have been reported, a detailed examination of concentrations for individual species would be tedious and difficult to interpret. In addition, no species distinctions were made before 1971 when concentrations were significantly higher than in later years. For the purposes of both brevity and clarity, and more importantly, for comparison to concentrations measured before 1971, we examined average concentrations across all species. Based on the reported concentrations for various species of fish, there is some evidence to suggest that this method of evaluation may result in data bias. Ratios of ^{137}Cs concentrations reported for

bream and catfish compared to those reported for bass from 1971 through 1976 for all onsite stream and Savannah River locations (Table 14-2) reveal some potential differences in concentration as a function of species. These years were used because species distinctions were first made in 1971, and concentrations were still significantly above the detection limit, particularly for onsite streams. Based on median ratios, concentrations in bass appear slightly higher than those reported for bream and catfish.

Table 14-1. Percentage of Bream, Bass, Catfish, and Other Fish and the Total Number of Fish Collected for Radionuclide Analysis[a] at Locations on or in the Vicinity of the SRS

		Percentage				
Location	Years	Bream	Bass	Catfish	Other	Total number
Clark's Hill/Thurmond Lake	1981–90	51.6	17.8	22.8	7.8	320
Savannah River						
Above SRS (R-2)	1971–91	37.7	4.2	46.1	12.0	1155
Adjacent to SRS (R-8)	1971–91	43.2	5.7	40.9	10.2	1083
Below SRS (R-10)	1971–91	49.9	3.5	36.5	10.2	1389
River Total		*44.0*	*4.4*	*40.9*	*10.8*	*3627*
Steel Creek						
At Road A	1971–79, 1984–91	42.8	26.2	21.6	9.5	1015
2 mi downstream from Rd. A	1971–72	65.8	14.2	20.0	0	295
Near mouth	1971–79	55.9	9.4	34.6	0	381
Four Mile Creek						
At Road 3	1971–79, 1986–91	80.4	11.2	5.2	3.2	561
Cassel's Pond	1971–79, 1984–86	65.5	17.7	14.9	2.0	666
Lower Three Runs						
At Patterson's Mill	1971–79, 1984–91	52.8	14.9	27.7	4.6	591
Reservoirs						
Par Pond	1971–78, 1984–91	33.8	50.2	1.3	14.7	920
Pond B	1984–91	43.3	47.0	7.5	2.2	268
Onsite Total		*52.5*	*15.6*	*25.6*	*6.3*	*4697*
Grand Total		48.8%	16.3%	26.6%	8.2%	8324

[a] Data are from annual environmental monitoring reports, 1971–1991.

Trophic level increases have been observed for ^{137}Cs concentrations in fish (Pendleton et al. 1965; Whicker et al. 1972). These effects result from similar assimilation of cesium and potassium but greater retention of cesium (Whicker and Schultz 1982). Whicker et al. (1990) reported ^{137}Cs concentrations for fish collected from Pond B at the SRS, and a catfish to bass concentration ratio of 0.7 and a bluegill to bass concentration ratio of 0.6 can be calculated based on these data. Similarly, a catfish to bass ratio of 0.8 and a bluegill to bass ratio of 0.4 can be calculated based on data reported by Mohler et al. (1997). Based on data provided by Harvey (1963), a catfish to bass ratio of 1.1 and a bluegill to bass ratio of 0.8 can be calculated. Such fish species are at or near the top of the food chain, would be expected to have higher concentrations than smaller baitfish, and likely represent concentrations for most commercial and sport fish. However, based on data from these studies, ^{137}Cs concentrations in bass may be as much as a factor of 2.5 greater than those measured in bluegill (bream) and a factor of 1.5 greater than those measured in catfish. During subsequent phases of the dose reconstruction process (e.g., direct

exposure assessment from ingestion of fish), it may be necessary to account for the potential differences in ^{137}Cs or other radionuclide concentrations that may exist for different species of fish.

Arithmetic mean concentrations have been historically reported by the SRS, and many of the trends and comparisons made in this section are based on these reported concentrations. Radiocesium concentrations, however, commonly fail to show a normal frequency distribution in environmental samples (Pinder and Smith 1975). This was evident with concentrations of ^{137}Cs measured in deer tissue (see Chapter 11). Medians, percentiles, and ranges are often more appropriate descriptors of these types of data. Examining concentrations for individual fish collected from Steel Creek and Four Mile Creek in 1972 and 1974 suggests a lognormal distribution, particularly for lower concentrations such as those measured in the Savannah River and in the lower parts of onsite streams. If this is the case, then arithmetic mean concentrations likely represent a conservative estimate of the central tendency for radiocesium concentrations in fish flesh.

Table 14-2. Sample Sizes and Average Concentrations of ^{137}Cs Reported for Bream and Catfish Relative to Average Concentrations Reported for Bass from 1971 through 1976[a]

	Bass	Bream			Catfish		
		Ratio of ^{137}Cs concentration in bream to that in bass[b]			Ratio of ^{137}Cs concentration in catfish to that in bass[b]		
Year	Sample size	Mean	Median	Sample size	Mean	Median	Sample size
Jan–June 1971	75	0.8	0.7	379	0.7	0.6	174
July—Dec 1971	54	0.7	0.7	329	0.9	0.9	168
Jan—June 1972	119	1.7	1.2	313	1.3	1.3	143
July—Dec 1972	76	1.0	0.6	297	2.2	1.0	192
July—Dec 1973	150	1.1	0.5	649	0.9	0.9	328
1974	163	0.5	0.5	411	0.7	0.6	278
1975	117	0.7	0.5	203	1.0	1.0	218
1976	12	1.6	1.0	29	1.1	1.0	33
Average		*1.0*	*0.7*		*1.1*	*0.9*	

[a] Data are from semi-annual and annual environmental monitoring reports, 1971–1976.

[b] Ratios were calculated based on the average concentrations reported for each location, and median and mean values were calculated based on these ratio values.

RADIONUCLIDE CONCENTRATIONS IN FISH FROM THE SAVANNAH RIVER AND ONSITE STREAMS

Assessing potential exposure to the general public from fish consumption is difficult because fish are able to move freely in Four Mile Creek, Steel Creek, and Lower Three Runs Creek, and they can potentially migrate to the Savannah River. The SRS has, however, maintained an extensive fish sampling program that, between 1962 and 1991, collected and reported concentrations for more than 13,300 fish (more than 5800 from the Savannah River and more than 7500 from onsite streams). This total is based on the collection numbers that have been reported in the monitoring reports (the number of onsite samples was not provided in 1962). This

total also does not include fish sampled from Upper Three Runs Creek from 1987 through 1991. These data were not compiled because the ^{137}Cs concentrations in fish from Upper Three Runs Creek were not significantly elevated relative to concentrations in fish from the Savannah River, which were at or near the detection limits during this time period. Before 1962, more than 4200 fish were collected and analyzed from the Savannah River and onsite locations. Figure 14-1 shows the number of fish sampled from the Savannah River and onsite locations from 1963 through 1991.

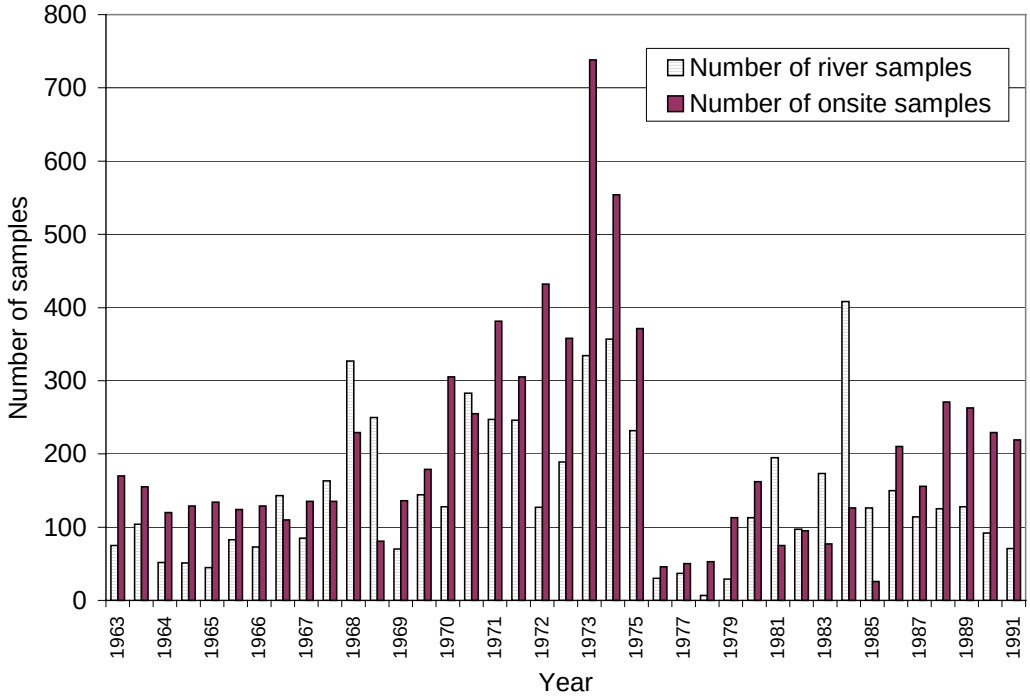

Figure 14-1. Number of fish collected from the Savannah River and onsite locations by SRS personnel from 1963 through 1991. Link to tabulated figure data.

Onsite stream sampling locations (Figure 14-2) were such that average and maximum concentrations in fish at and downstream from the most contaminated areas could be determined. Offsite locations were positioned along the Savannah River and indicate the average and maximum concentrations that have been measured in fish that were potentially caught by the general public. Concentrations in fish from two onsite reactor cooling reservoirs, Par Pond and Pond B, have also been routinely measured. However, fish in these reservoirs are isolated from the Savannah River and, therefore, are inaccessible for legal consumption by the general public. The SRS has collected fish from the Savannah River above the plant boundary, at Clark Hill/Thurmond Lake (approximately 60 mi. upriver from the SRS), and at Stoke's Bluff (approximately 60 mi. downriver from Lower Three Runs) to serve as control locations. While concentrations have not been reported continuously since 1957 for all locations, concentrations have been reported continuously since 1957 for three Savannah River sampling locations: R-2 (above SRS), R-8 (adjacent to SRS), and R-10 (below SRS). Table 14-3 provides a general summary of the routine sampling and analyses that have been completed for fish since the inception of the monitoring program.

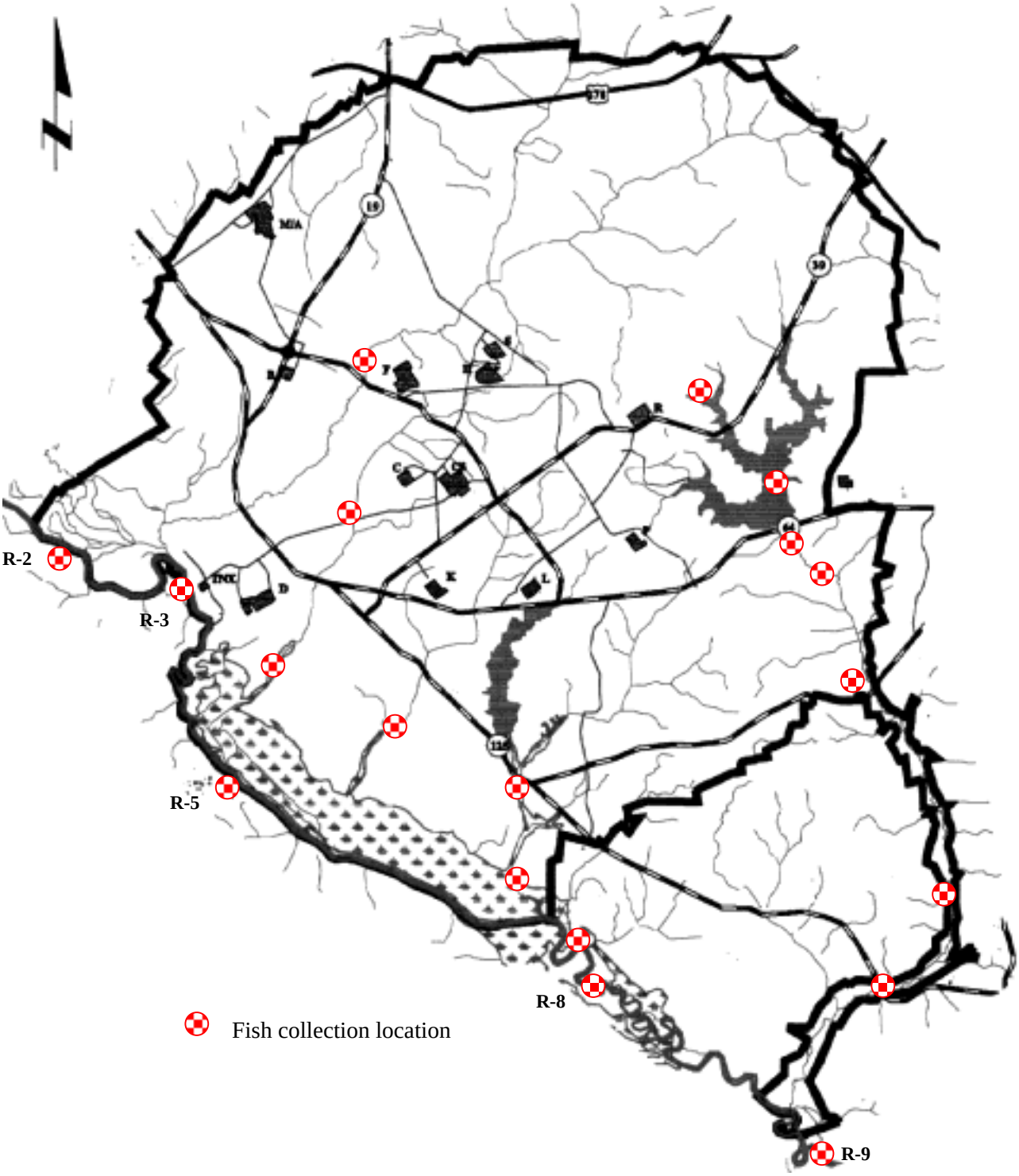

Figure 14-2. Onsite and Savannah River fish collection locations. The Clark Hill Reservoir (control) and R-10 locations are not shown. Clark Hill Reservoir is approximately 60 mi upriver from the SRS, and the R-10 location is approximately 10 mi below the SRS at the Highway 301 bridge.

Table 14-3. Summary of Routine SRS Radiological Monitoring for Fish[a]

	Sampling locations									Radiological analyses				
	Savannah River				Onsite									
Year	R-2, R-8, and R-10	R-3, R-5, and R-9	Clark Hill Reservoir	Stoke's Bluff	Par Pond (Pond B[b])	Upper Three Runs	Steel Creek	Four Mile Creek	Lower Three Runs	Gross nonvolatile beta	Cesium-137	Strontium-89,90	Zinc-65	Tritium[c]
July 1957	x						x	x	x	x				
1958	x	x		x	x		x	x	x	x				
1959	x	x		x	x	x	x	x	x	x				
1960	x	x		x	x	x	x	x	x	x				
June 1961	x	x		x	x	x	x	x	x	x				
July 1961	x			x	x	x	x	x	x	x	x	x	x	
1962	x				x		x	x	x	x	x	x	x	
1963	x				x		x	x	x	x	x	x	x	
1964	x				x		x	x	x	x	x	x	x	
1965	x				x		x	x	x	x	x	x	x	
1966	x				x		x	x	x	x	x	x	x	
1967	x				x		x	x	x	x	x	x	x	
1968	x				x		x	x	x	x	x	x	x	
1969	x				x		x	x	x	x	x	x	x	
1970	x				x		x	x	x		x	x	x	x
1971	x				x		x	x	x		x	x		x
1972	x		x		x		x	x	x		x	x		x
1973	x		x		x		x	x	x		x	x		x
1974	x		x		x		x	x	x		x	x		x
1975	x		x		x		x	x	x		x	x		x
1976	x		x		x		x	x	x		x	x		x
1977	x		x		x		x	x	x		x	x		x
1978	x		x		x		x	x	x		x	x		x
1979	x		x		x		x	x	x		x	x		x
1980	x		x		x		x	x	x		x	x		x
1981	x		x		x		x	x	x		x			x
1982	x		x		x		x	x	x		x			
1983	x		x		x		x	x	x		x			
1984	x		x		x		x	x	x		x			
1985	x		x		x		x	x	x		x			
1986	x		x		x		x	x	x		x			
1987-1991	x		x		x	x	x	x	x		x			

[a] This table is intended to provide the reader with general time periods only for collection and analysis of samples at routine locations and is not an exact representation of all collected data
[b] Reporting for Pond B began in 1974
[c] Tritium results were reported for R-2, R-8, and R-10 Savannah River locations only

Average Nonvolatile Beta, Cesium, and Strontium Concentrations

A summary for average ^{137}Cs concentrations measured in all fish at most sampling locations for 1975 and 1979 through 1991 was provided in Arnett et al. (1992). However, it was not clear how these averages were determined. In some cases the averages appeared to be calculated based on mean concentrations for each species without regard for the relative number of each species. In other cases the relative number of each species appeared to be taken into account. To maintain consistent methodology, we recalculated the weighted arithmetic average concentrations across all species based on the tabulated mean concentrations for each species considering the relative number of each species. In some cases insufficient data were available (e.g., mean for a given species was not provided), and we used the maximum measured concentration for that particular species for calculating the average across all species. In general, the recalculated averages were consistent with those reported in the 1991 report, with some slight differences.

Before 1971, concentrations for individual species were not given, so the actual tabulated data were used. Between 1980 and 1983, only concentrations measured at locations along the Savannah River were provided (concentrations measured at the control location, Clark Hill, were also provided from 1981 through 1983), so we used the average concentrations reported for onsite locations in the 1992 report (discussed above). In addition, concentrations measured in Pond B were not reported until 1974.

The radionuclide of most interest in terms of dose reconstruction is ^{137}Cs because it behaves like its nutrient analog, potassium, and tends to accumulate in muscle tissue, the portion of fish typically consumed. Strontium-89,90, ^{65}Zn, and ^{32}P may not present as important an exposure pathway since they tend to accumulate primarily in bone tissue, which is not typically consumed. However, we discuss radionuclide burdens in bone because it is possible that some people may have made fishcakes from Savannah River fish, which include some portion of bone. Also, small fish can be pan-fried without removing the skin and scales, which have been shown to contain higher concentrations of bone-seeking radionuclides than the muscle tissue (Rope and Whicker 1985).

For dose reconstruction, it is more realistic to use average measured concentrations for estimating potential exposure to individuals who may have relied upon Savannah River fish for a significant portion of their diet (e.g., 19 kg y^{-1} [Hamby 1991]). For this reason, the majority of the discussion regarding radionuclide concentrations in fish focuses on average concentrations. However, maximum concentrations are briefly discussed to provide an understanding of the maximum concentrations to which smaller numbers of individuals may have been exposed. Detailed information regarding use factors and ingestion rates for the Savannah River can be found in Hamby (1991), Turcotte (1983), and Hutto and Turcotte (1983). This type of information will be important in a future phase of this project when doses and risks are estimated.

Savannah River

The Health Physics Section at the SRS conducted Site surveys before plant startup to determine the quantities of radionuclides in several environmental media. Two reports were available for review (Reinig et al. 1953; Reinig 1952). Reinig et al. (1953) provided data for approximately 80 fish that were collected from the environs of the Savannah River Plant between

June 1951 and January 1953, before the first liquid effluent discharge to Lower Three Runs Creek from R Reactor in June 1954. The entire fish was analyzed in most cases, but specific tissues were analyzed for some fish. These fish were analyzed for nonvolatile beta concentrations, and most fish averaged less than 15 pCi g^{-1} (the apparent detection limit) with a maximum of 38 pCi g-1 detected in the flesh of one fish. Reinig (1952) provided data for approximately 46 fish collected before August 1952. The average nonvolatile beta concentration detected in the flesh and bones of these fish was 5.0 and 11.7 pCi g^{-1}, respectively.

Because only nonvolatile beta concentrations were reported before 1962, it is instructive to examine the years during which both nonvolatile beta and ^{137}Cs concentrations were reported. Table 14-4 shows the concentrations of nonvolatile beta, ^{137}Cs, and $^{89,90}Sr$ measured in fish collected from Savannah River locations from July 1958 through 1969. It is assumed that these reports provided information for the same fish, but slightly different sample numbers were reported for several years.

Table 14-4. Nonvolatile Beta[a], ^{137}Cs[b], and $^{89,90}Sr$[b] Concentrations (pCi g^{-1}) Measured in Fish Flesh and Bone Collected from the Savannah River Above, Adjacent to, and Below the SRS

Year	Month	Above SRS (R-2)				Adjacent to SRS (R-8)				Below SRS (R-10)			
		Flesh		Bone		Flesh		Bone		Flesh		Bone	
		NB[c]	Cs	NB	Sr	NB	Cs	NB	Sr	NB	Cs	NB	Sr
1958	July—Dec	4		10		4		15		4			
1959	Jan—June	4		10		5		15		4		15	
	July—Dec	4		12		6		28		4		22	
1960	Jan—June	4		14		5		35		4		15	
	July—Dec	4		10		6		21		4		17	
1961	Jan—June	4		13		5		23		4		11	
	July—Dec	4		8		4		13		5		13	
1962	Jan—June	4	nd[d]	13	9	5	1	19	15	4	nd	18	10
	July—Dec	4	nd	21	8	8	2	72	43	6	1	41	14
1963	Jan—June	4	nd	15	3	4	1	22	25	5	1	23	11
	July—Dec	3	1	18	6	4	2	43	19	3	nd	19	14
1964	Jan—June	4	nd	20	8	5	1	22	8	4	1	18	8
	July—Dec	3	nd	19	10	5	nd	57	15	5	nd	29	12
1965	Jan—June	3	1	22	17	9	8	40	12	8	12	28	19
	July—Dec	4	1	27	8	6	2	58	14	9	4	51	32
1966	Jan—June	3	<1	18	8	4	12	23	25	4	2	21	9
	July—Dec	4	<1	18	6	6	4	46	6	4	nd	25	12
1967	Jan—June	5	<1	25	9	7	22	56	19	6	4	28	17
	July—Dec		1		16		12		15		3		12
1968	Jan—June	4	1	21	9	5	11	21	14	6	2	17	13
	July—Dec	5	<1	14	8	8	3	20	10	5	3	20	10
1969	Jan—June	5	1	17	14	10	3	40	17	7	3	24	12
	July—Dec		1		9		5		14		5		15

[a] Data are from Du Pont (1959, 1960a, 1960b, 1961, 1962a, 1962c, 1962d, 1963a, 1963b, 1964a, 1965a, 1965b, 1966a, 1966b, 1967a, 1967b, 1968a, 1968b, 1969a, 1969b)
[b] Ashley (1965, 1966, 1967, 1968, 1969, 1970)
[c] NB = nonvolatile beta.
[d] nd = not detected.

Nonvolatile beta concentrations reported for fish flesh and bone before 1962 are near or below the concentrations reported from 1962 through 1969 and near or below the concentrations reported for fish collected before plant startup. Between June 1954 (date of first liquid effluent discharge) and July 1958, concentrations for Savannah River fish were not explicitly provided, but the Health Physics Regional Monitoring semi-annual reports stated that nonvolatile beta concentrations in bone and flesh from the few sampled Savannah River fish were negligible or insignificant. It appears, though, that these conclusions were based on very few fish. Muscle tissue from 10 clams collected from Lower Three Runs Creek upstream from R-Area effluent in 1957 had average and maximum nonvolatile beta concentrations of 5 and 12 pCi g^{-1}, respectively (Mealing and Horton 1957).

The nonvolatile beta data in Table 14-4 are graphically depicted in Figures 14-3 and 14-4. Based on both flesh and bone concentrations, it is clear that Savannah River fish collected from locations adjacent to and below the SRS were impacted by SRS activities. Based on fish flesh concentrations, the above SRS location does not appear to have been significantly affected by SRS activities. Bone concentrations for the above SRS location are also lower than the other two locations, but they are elevated between 1962 and 1969 compared to between 1958 and 1961. It is difficult to know whether this is a result of SRS activities or atmospheric fallout. Atmospheric deposition was greatest in 1963, with a considerable drop off during the last half of the 1960s (see Chapter 6). A similar trend is evident for fish tissue concentrations although measured concentrations near the detection limit preclude examining this data in detail.

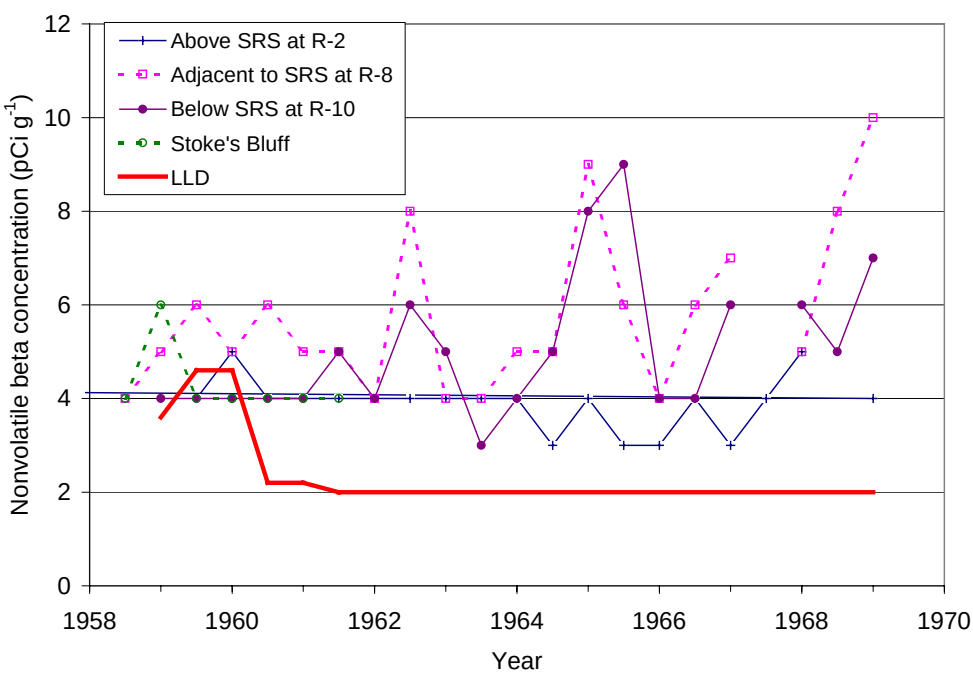

Figure 14-3. Average nonvolatile beta concentrations measured in fish flesh collected at four Savannah River locations from July 1958 through 1969 shown with the reported lower limit of detection (LLD). Link to tabulated figure data.

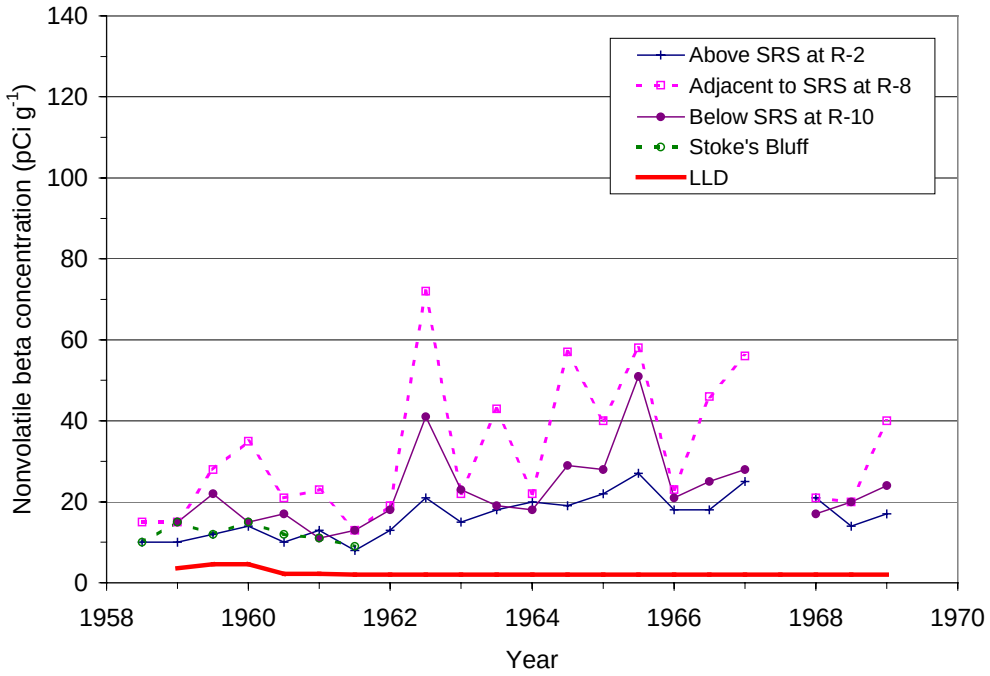

Figure 14-4. Average nonvolatile beta concentrations measured in fish bone collected at four Savannah River locations from July 1958 through 1969 shown with the reported LLD. Link to tabulated figure data.

Between 1958 and 1961, fish concentrations at the above SRS location are very similar to those at the Stoke's Bluff location, approximately 60 mi downriver from the mouth of Lower Three Runs Creek. This indicates that the above SRS river location may have been an adequate control location (i.e., one that would not have been impacted by SRS activities). Nonvolatile beta concentrations in fish from onsite streams were only reported from July 1957 through 1961 and are not shown, but they were typically about an order of magnitude greater than concentrations at Savannah River locations. Onsite concentrations in both bone and flesh were also generally highest at Lower Three Runs Creek locations during this time period.

Average ^{137}Cs and ^{89,90}Sr concentrations reported for fish from Savannah River locations are shown in Figures 14-5 and 14-6, respectively. Cesium-137 concentrations (Figure 14-5) appear elevated at locations adjacent to SRS (R-8) and below SRS (R-10) through about 1971, after which concentrations at all locations are near the lower limit of detection (LLD)[1]. Concentrations above SRS (R-2) have been near the LLD for all reported years, except during the last half of 1970 and the first half of 1971. Cesium-137 concentrations measured in fish from the control location (Clark Hill) are indistinguishable from concentrations measured in fish from river locations since 1986. From 1981 through 1984, the control location concentrations appear slightly lower than R-8 and R-10 locations but similar to R-2 locations.

[1] The lower limit of detection (LLD) values are provided in various figures throughout this chapter whenever they were reported in the environmental monitoring reports. In many cases, however, the reports state "...the sensitivity of analysis varied due to differing sample size..." and no LLD values were provided.

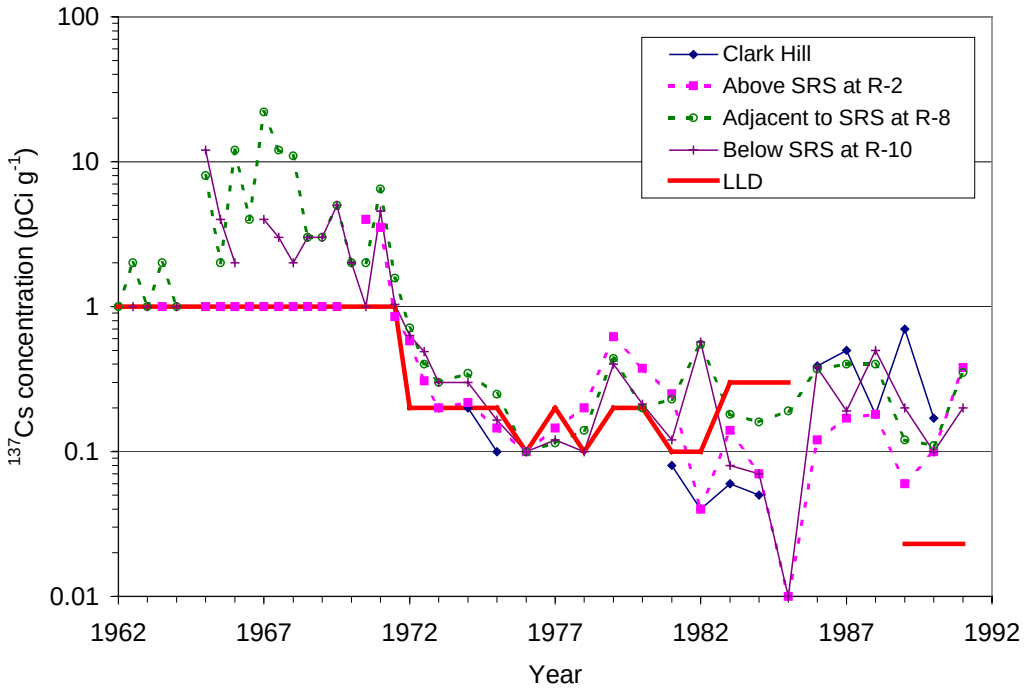

Figure 14-5. Cesium-137 concentrations in fish flesh collected at Savannah River locations above, adjacent to, and below the SRS and at Clark Hill shown with the reported LLD. Link to tabulated figure data.

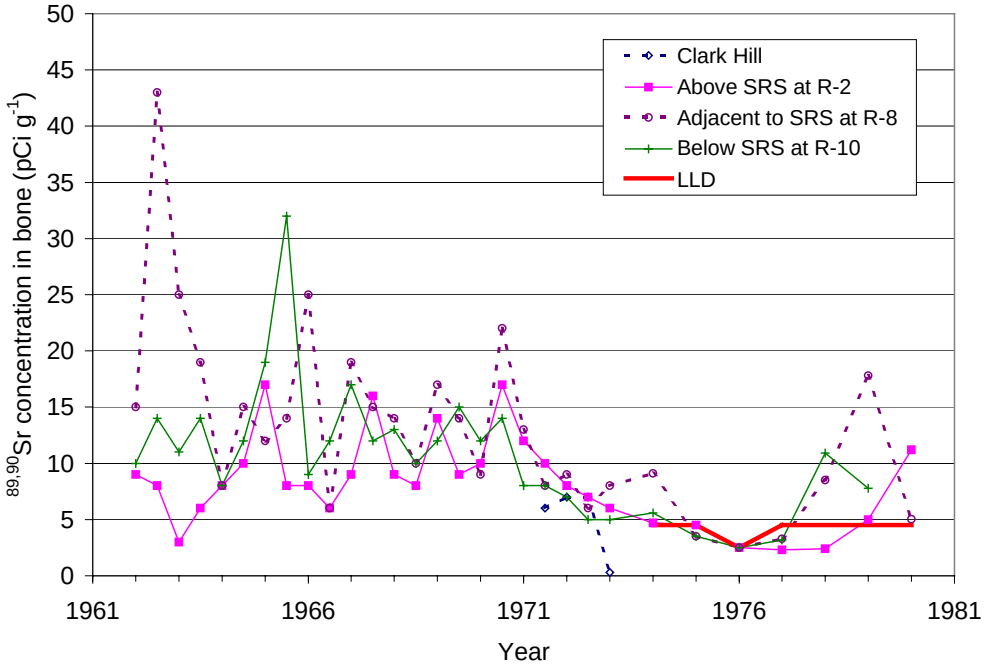

Figure 14-6. Strontium-89,90 concentrations in fish bone collected at Savannah River locations above, adjacent to, and below the SRS and at Clark Hill shown with the reported LLD. Link to tabulated figure data.

The same general trends are evident for ^{89,90}Sr in bone (Figure 14-6). River locations R-8 and R-10 appear elevated relative to the upriver location R-2 for most years through about 1972. From 1972 through 1977, concentrations are near the LLD for all locations, and from 1978 through 1980, concentrations appear to increase at all locations. Reporting of ^{89,90}Sr concentrations was discontinued after 1980. Concentrations at the control location (Clark Hill) were near the LLD for all years they were reported.

There is some question as to how appropriate the R-2 (above SRS) location was for a control location. However, based on nonvolatile beta concentrations in flesh and bone and on ^{137}Cs and ^{89,90}Sr concentrations in flesh and bone, respectively, the R-2 river location appears to have been an adequate control location. Nonvolatile beta flesh concentrations in 1952 and July 1958 through June 1962 and ^{137}Cs flesh concentrations from July 1962 through 1991 are shown for "control" and R-2 locations in Figure 14-7. Control locations are represented by Stoke's Bluff from July 1958 through June 1961 and by Clark Hill from 1972 through 1991. The nonvolatile beta concentration measured in fish before plant start-up is shown for 1952 and is similar to control and river locations from July 1958 through June 1962. Concentrations of radionuclides in fish at Stoke's Bluff and Clark Hill, both of which would not be expected to have been impacted by SRS activities, have been very similar to concentrations in fish from the R-2 Savannah River location. Concentrations measured in fish collected during May 1960 at Union Creek near Port Wentworth, Georgia, were also consistent with concentrations in fish collected from the R-2 river location. Based on these data, the R-2 river location appears to represent background concentrations resulting from atmospheric weapons testing. However, no offriver control data were available from July 1961 through 1972 or from 1976 through 1980. The R-2 river location concentrations during those time periods were at or near the LLD, except during the last half of 1970 and the first half of 1971. An explanation for elevated concentrations during these time periods is not evident, but similar increases were evident for R-8 and R-10 Savannah River locations (see Figure 14-5).

Steel Creek

To assess the potential impact of SRS activities on the environment, it is important to examine concentrations of radionuclides in fish that have been reported for various locations in onsite streams as well as in the Savannah River. Average measured ^{137}Cs and ^{89,90}Sr concentrations are shown for fish caught in Steel Creek at Road A, 2 mi below Road A, at the mouth, and in the Savannah River just downriver from the mouth (R-8) in Figures 14-8 and 14-9, respectively. Cesium-137 concentrations in fish caught 2 mi downstream from Road A are significantly lower than those reported for the Road A location. Concentrations measured at the mouth appear elevated relative to those measured at the location just downriver from the mouth of Steel Creek, which has been near the LLD since about 1971. Figure 14-9 shows the same general trends for ^{89,90}Sr, concentrations of which appear to have been near the LLD since 1975.

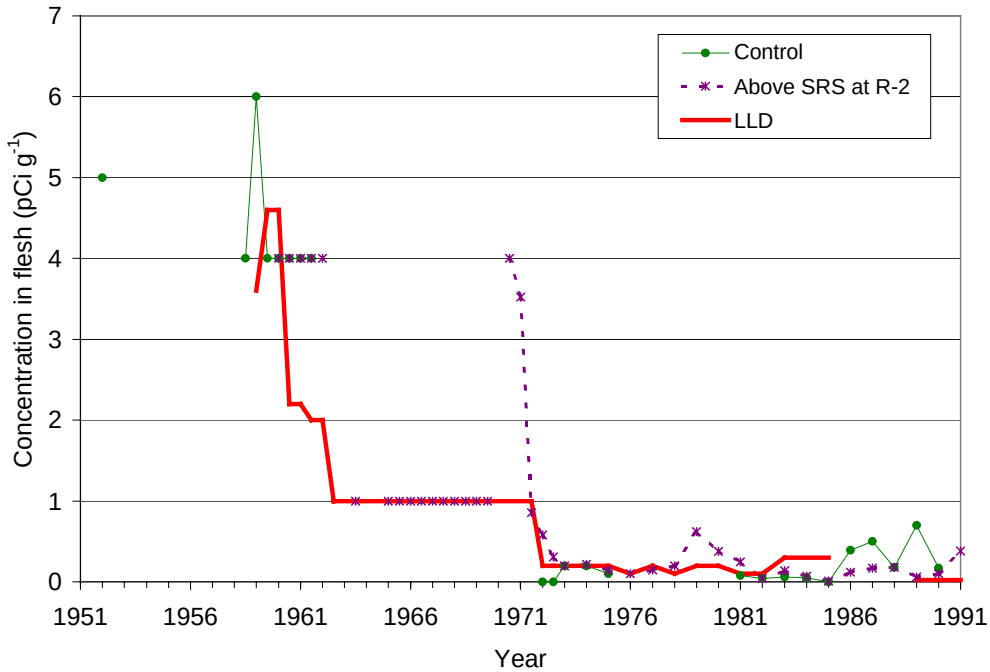

Figure 14-7. Nonvolatile beta (1952 and July 1958 through June 1962) and ^{137}Cs (July 1962 through 1991) concentrations for control (Stoke's Bluff from July 1958 through June 1961 and Clark Hill from 1972 through 1991) and R-2 river locations. Link to tabulated figure data.

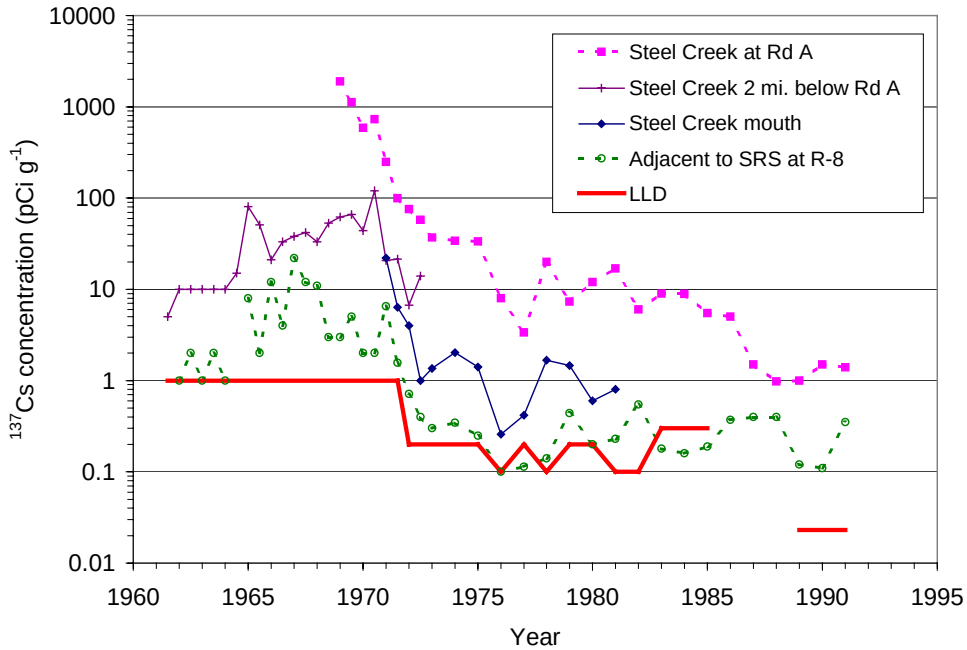

Figure 14-8. Average ^{137}Cs concentrations in fish collected at three locations along Steel Creek and at the R-8 Savannah River location shown with the reported LLD. Link to tabulated figure data.

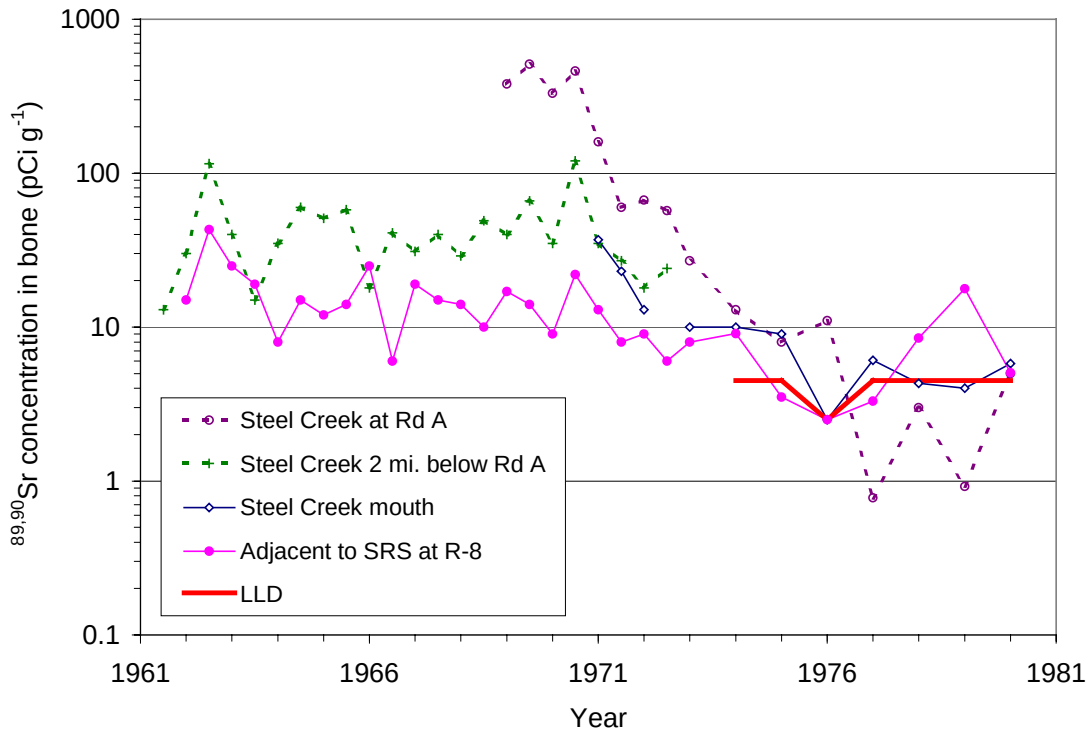

Figure 14-9. Average ^{89,90}Sr concentrations in fish collected at three locations along Steel Creek and at the R-8 Savannah River location shown with the reported LLD. Link to tabulated figure data.

Brisbin et al. (1989), however, failed to demonstrate any location differences in ^{137}Cs concentrations measured in biota collected from Steel Creek in 1981. The authors attributed this to the movement and redeposition of contaminated sediments along the stream channel. Brisbin et al. (1974) also found no differences in ^{137}Cs concentrations measured in sediment collected from upstream and swamp-delta locations in Steel Creek. These studies appear inconsistent with the significant decreases noted for fish concentrations as a function of distance downstream from reactor effluent input.

Four Mile Creek

Nonvolatile beta concentrations were reported for fish from the Four Mile Creek through 1961, but Four Mile Creek sampling was apparently discontinued in 1962 and resumed again in 1970. Figures 14-10 and 14-11 show average measured concentrations of ^{137}Cs and ^{89,90}Sr, respectively, for fish from Four Mile Creek at Road A and at Cassel's Pond (3 mi below Road A). In addition, the reported concentrations in fish from a location several miles downriver from the mouth of Four Mile Creek (R-8) are shown. Again, as with Steel Creek, the concentrations a few miles downstream from Road A are significantly lower than those reported for the Road A location, which are higher than the R-8 river location.

Strontium-89,90 concentrations in fish at Road A decreased significantly in 1976 and 1977, followed by an increase in 1978 and 1979 back to concentrations similar to those measured

before 1976 (Figure 14-11). This same trend was also evident at the location 3 mi below Road A. Concentrations in fish from the Savannah River location (R-8) several miles downriver from the mouth of Four Mile Creek also increased in 1978 and 1979. It appears this increase may be related to the significant increases noted for Four Mile Creek locations during that time period because similar increases were not evident in Steel Creek. A similar trend was noted for ^{137}Cs concentrations (Figure 14-10), but an explanation is not apparent. However, sample sizes in 1976 and 1977 were significantly lower than sample sizes for previous years (Figure 14-1).

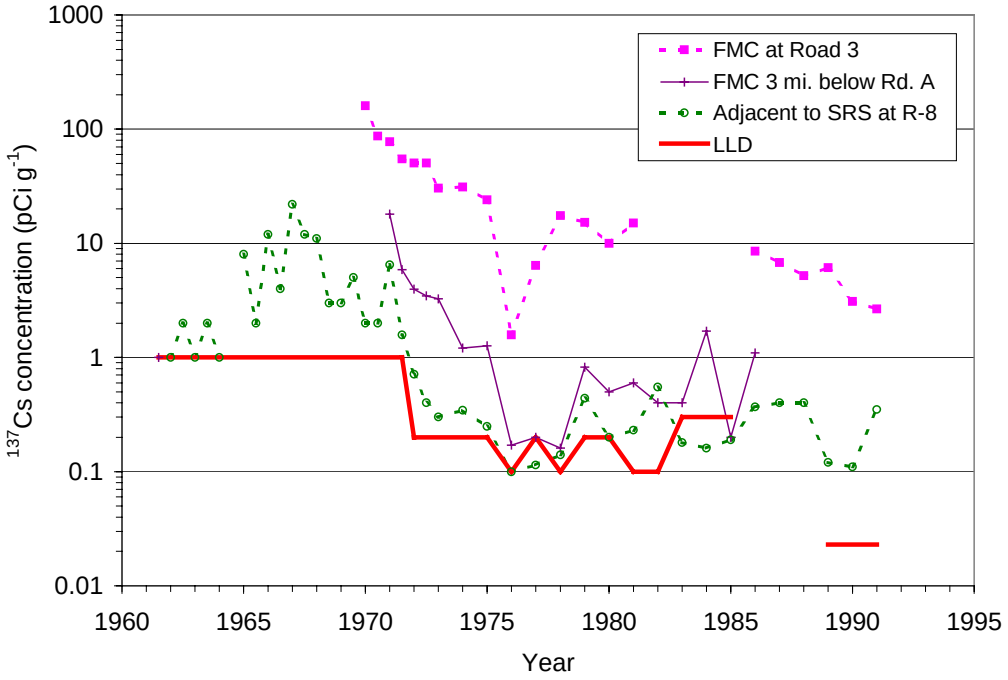

Figure 14-10. Average ^{137}Cs concentrations measured in fish flesh collected at two locations along Four Mile Creek (FMC) and the R-8 Savannah River location shown with the reported LLD. Link to tabulated figure data.

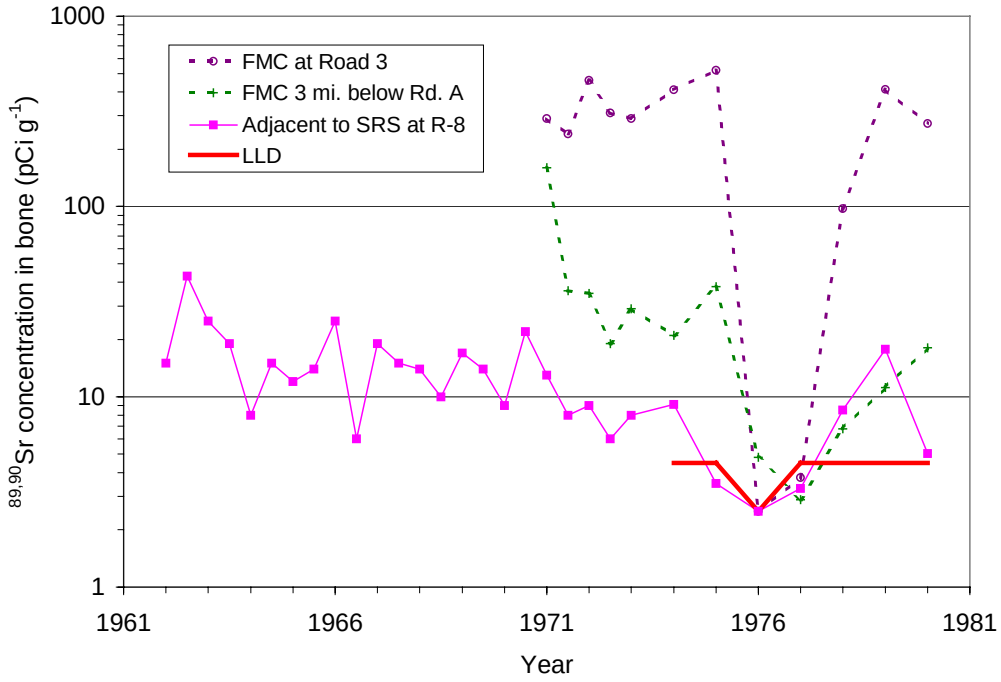

Figure 14-11. Average ^{89,90}Sr concentrations measured in fish bone collected at two locations along Four Mile Creek and the R-8 Savannah River location shown with the reported LLD. Link to tabulated figure data.

Lower Three Runs Creek, Par Pond, and Pond B

Figures 14-12 and 14-13 show the average measured ^{137}Cs and ^{89,90}Sr concentrations in fish, respectively, for Pond B (^{137}Cs only), Par Pond, Lower Three Runs Creek at Patterson's Mill, and for a location 8 mi downriver from the mouth of Lower Three Runs (R-10). Pond B received effluent via R Canal from R Reactor. The effluent stream continued to Par Pond, and water from Par Pond was released directly to Lower Three Runs Creek. Interestingly, ^{137}Cs concentrations in Par Pond appear very similar to concentrations in Lower Three Runs Creek at Patterson's Mill, which are elevated relative to the R-10 river location. However, ^{89,90}Sr concentrations in Par Pond are elevated above concentrations in Lower Three Runs Creek at Patterson's Mill, which are very similar to the R-10 river location. A significant increase in ^{89,90}Sr concentrations is seen in Lower Three Runs Creek at Patterson's Mill during the first and second half of 1970. The cause of this increase is not known, but similar increases were not seen in Par Pond or the R-10 river location. Cesium-137 concentrations in Pond B are roughly an order of magnitude higher than those measured in Par Pond.

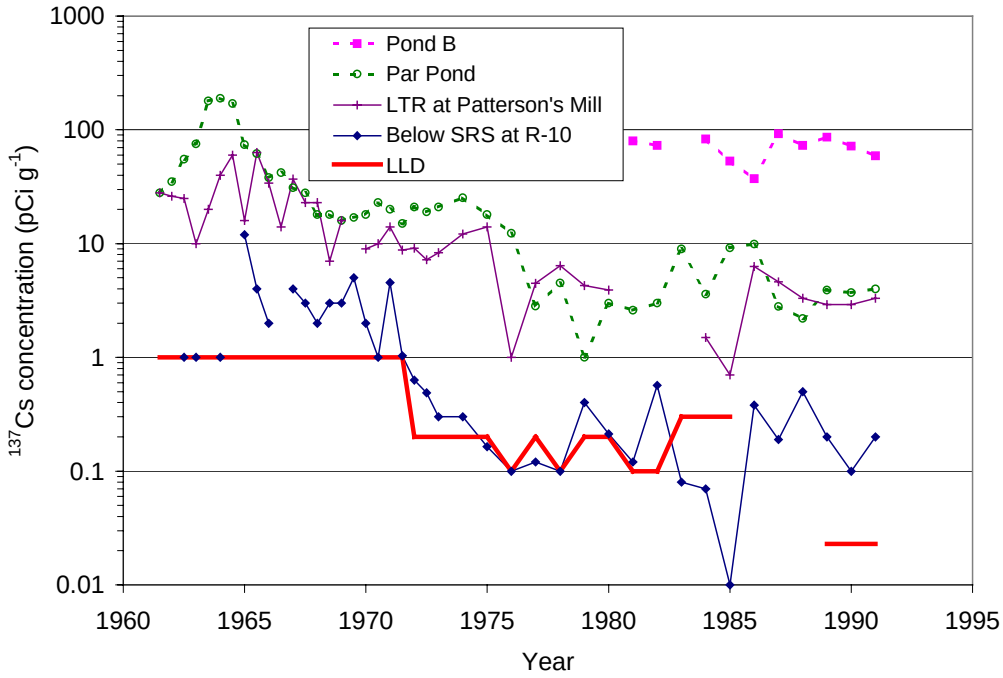

Figure 14-12. Average ^{137}Cs concentrations measured in fish flesh collected at Pond B, Par Pond, Patterson's Mill along Lower Three Runs (LTR) Creek, and the R-10 Savannah River location shown with the reported LLD. Link to tabulated figure data.

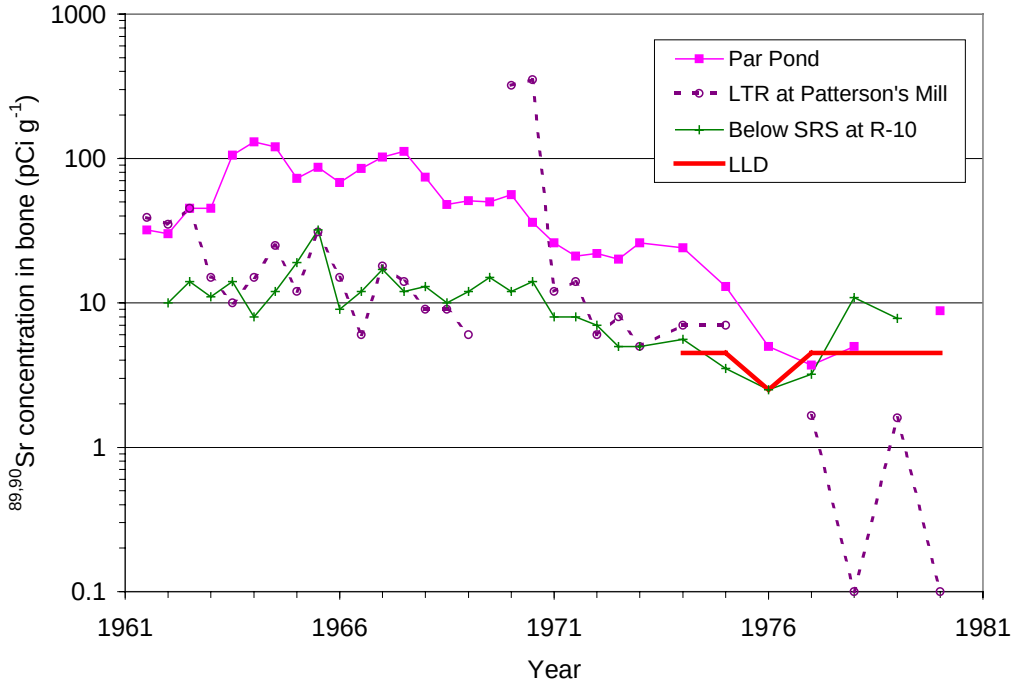

Figure 14-13. Average ^{89,90}Sr concentrations measured in fish flesh collected at Par Pond, Patterson's Mill along Lower Three Runs Creek (LTR), and the R-10 Savannah River location shown with the reported LLD. Link to tabulated figure data.

Since construction of the Par Pond dam in 1958, some flow into Lower Three Runs Creek has originated from Par Pond dam releases. Figures 14-14 and 14-15 show nonvolatile beta concentrations in flesh and bone, respectively, for various locations along Lower Three Runs Creek, Par Pond, and the Savannah River R-9 location (near the mouth of Lower Three Runs Creek) from July 1957 through June 1961. Par Pond was not constructed until July 1958, so distances relative to the dam for that time period were derived from distances relative to R Reactor. It is clear that the R-9 location concentrations are significantly lower than Lower Three Runs Creek concentrations. However, concentrations measured in fish collected from Lower Three Runs at various distances from Par Pond are generally indistinguishable and quite similar to those measured in Par Pond after construction of the dam. Before July 1958, there was a discernable decrease in concentration with increasing distance from the dam. This suggests that flow into Lower Three Runs Creek since July 1958 originated largely from Par Pond dam releases, and dilution with stream water may not have been as significant as for other onsite streams. A similar trend is evident for ^{137}Cs concentrations measured in Par Pond and Lower Three Runs Creek at Patterson's Mill from 1961 through 1991.

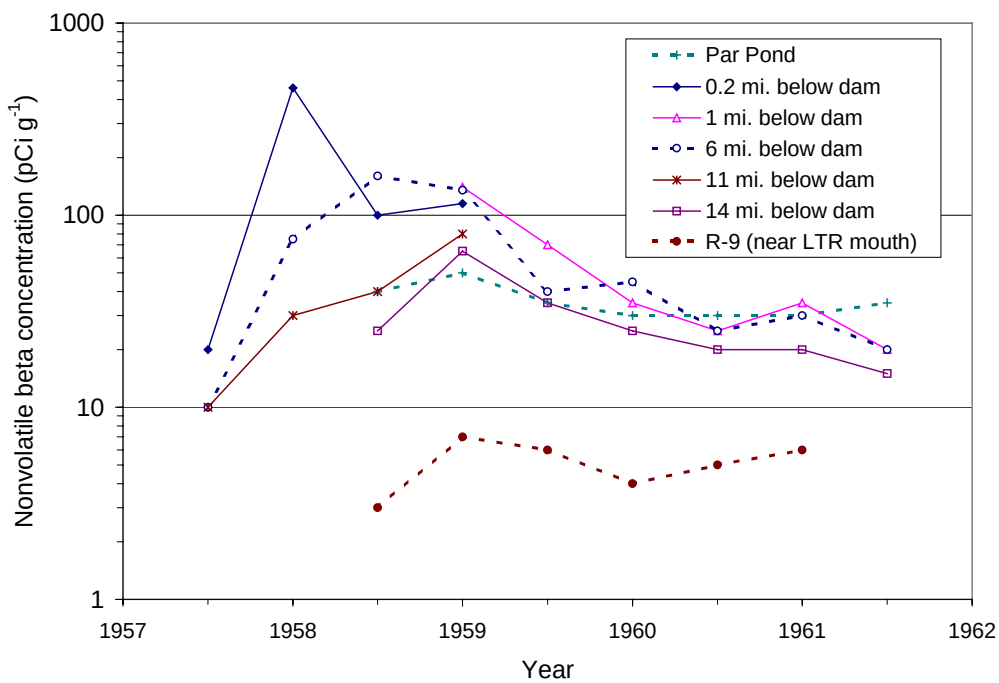

Figure 14-14. Average nonvolatile beta concentrations measured in fish flesh collected from Lower Three Runs Creek at various distances below the Par Pond dam. Link to tabulated figure data.

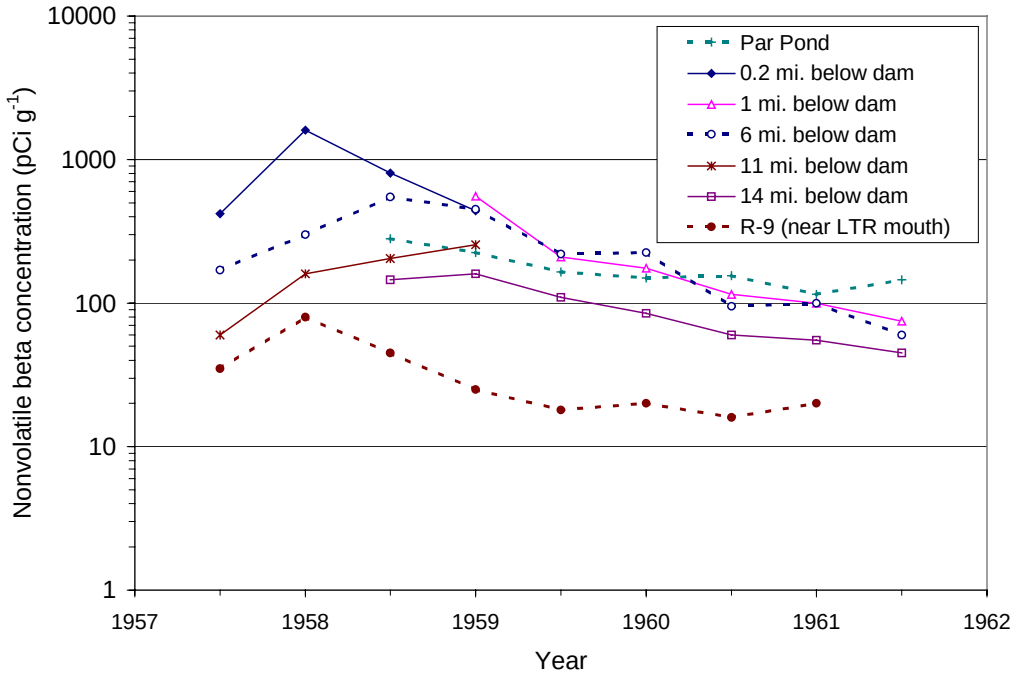

Figure 14-15. Average nonvolatile beta concentrations measured in fish bone collected from Lower Three Runs Creek at various distances below the Par Pond dam. Link to tabulated figure data.

Upper Three Runs Creek

Concentrations have not been reported consistently for Upper Three Runs Creek, which has not received reactor effluent. However, nonvolatile beta concentrations were reported from July 1959 through June 1961, and ^{137}Cs concentrations were reported from 1987 through 1991. During these time periods, concentrations were generally indistinguishable from Savannah River concentrations, indicating little radionuclide contamination in Upper Three Runs Creek. This is helpful for establishing the adequacy of the R-2 river location as representative of background concentrations.

Additional Savannah River Locations

Nonvolatile beta concentrations in flesh were reported for fish collected at six Savannah River locations from July 1958 through June 1961 (Figure 14-16). The R-2, R-8, and R-10 locations were described previously. The R-3 location was near the mouth of Upper Three Runs Creek, the R-5 location was near the mouth of Four Mile Creek, and the R-9 location was near the mouth of Lower Three Runs Creek. Based on these data, concentrations at the R-8 and R-9 locations appear similarly elevated relative to other locations, indicating the influence of Steel Creek and Lower Three Runs Creek on activity levels in fish. During this time period and through 1968, concentrations measured in fish from Steel Creek and Lower Three Runs Creek locations were similar, though concentrations were generally highest in Steel Creek. Concentrations in fish

from Four Mile Creek were not significantly elevated during this time period, and the R-5 location concentrations reflect this.

Since 1959, concentrations of radionuclides in fish from onsite streams have generally been highest in Steel Creek, and the R-8 river location has likely been representative of the highest routinely measured Savannah River location fish concentrations. However, concentrations for fish collected at the mouth of Steel Creek, which would be accessible to Savannah River fishermen, were consistently elevated relative to the R-8 river location from 1971 through 1981. Four Mile Creek concentrations were elevated relative to river concentrations since 1970 (concentrations for Four Mile Creek were not reported between 1962 and 1969), so R-5 concentrations may have been impacted. However, Four Mile Creek concentrations have been lower than Steel Creek concentrations, so R-5 location concentrations likely have been lower than R-8 location concentrations. Although Par Pond dam releases may have inhibited dilution with the stream water, concentrations in fish from the R-9 location have also likely been lower than R-8 location concentrations because Lower Three Runs Creek concentrations have been significantly lower than Steel Creek concentrations since 1959. Lower Three Runs Creek fish concentrations were, however, higher than Steel Creek fish concentrations through 1959 so it is possible that Savannah River fish concentrations were highest at the R-9 location from 1954 through 1959.

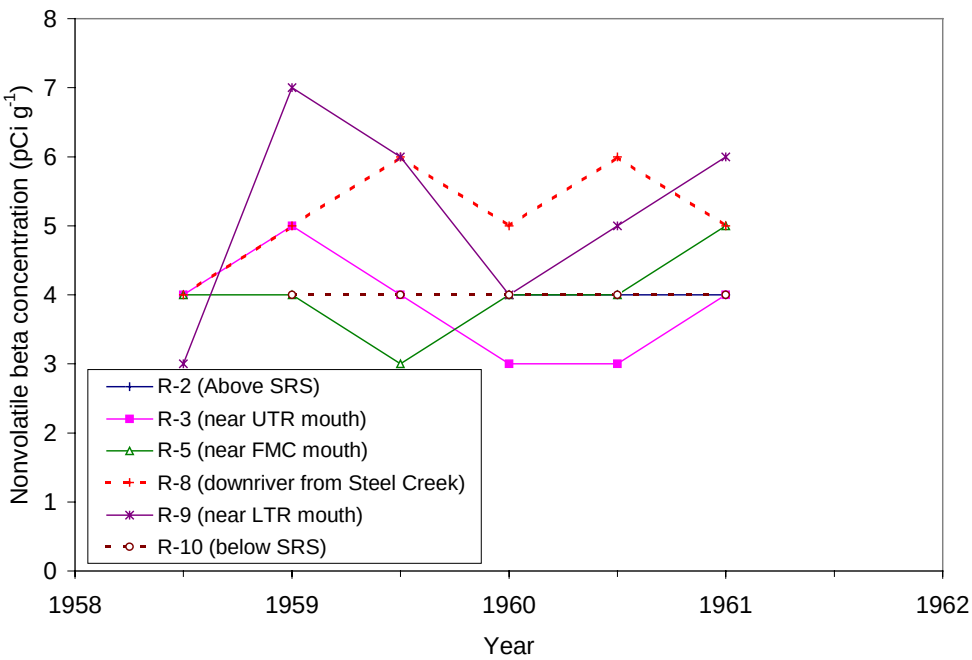

Figure 14-16. Average nonvolatile beta concentrations in fish flesh measured at six Savannah River locations from July 1958 through June 1961. Link to tabulated figure data.

Radionuclide concentrations in fish from onsite streams and ponds have consistently been significantly elevated relative to offsite control locations. There is little question that SRS activities have discharged reactor effluent to onsite seepage basins, streams/canals, and ponds, which has resulted in elevated radionuclide burdens in fish. It is also quite clear that although the SRS buffer area was sufficient to significantly dilute effluent radionuclide concentrations,

Savannah River fish have also been impacted by SRS activities. However, average concentrations of radionuclides in fish do not appear to have been impacted at the R-2 river location, and average concentrations at all river locations have been generally indistinguishable and near or below the LLD since 1972.

The measurement data suggest that fish from R-8 and R-10 Savannah River locations were most impacted by Site releases from 1962 through 1972. The R-8 river location concentrations in fish may also represent the highest concentrations of radionuclides for routinely sampled Savannah River locations because Steel Creek has consistently shown the highest concentrations for any of the onsite streams. However, concentrations measured in fish collected at the mouth of Steel Creek from 1971 through 1981 indicate that the R-8 river location may not represent the highest concentrations in fish to which Savannah River fishermen may have been exposed. Concentrations in fish at the R-5 and R-9 river locations may also have been elevated relative to concentrations at the R-2 river location, but fish were not sampled from these locations after June 1961. However, concentrations in fish from Steel Creek have been higher than concentrations in fish from other onsite streams since 1960, and fish concentrations at the R-5 and R-9 river locations were not likely higher than concentrations at the R-8 river location. Concentrations measured in LTR were, however, generally higher than concentrations measured in Steel Creek before 1960.

Maximum Nonvolatile Beta, Cesium, and Strontium Concentrations

To estimate the maximum concentrations of radionuclides in fish to which members of the general public may have been exposed, it is necessary to examine the maximum yearly reported radionuclide concentrations in fish for the Savannah River. Table 14-5 shows maximum reported radionuclide concentrations since inception of the routine monitoring program in July 1957. Figures 14-17 and 14-18 show maximum radionuclide concentrations in flesh and bone, respectively, reported for Savannah River locations.

Table 14-5. Maximum Measured Radionuclide Concentrations for Savannah River, Steel Creek Mouth, and Onsite Locations.

	Radionuclide concentration in pCi g^{-1} (location, year)[a]			
	Nonvolatile beta activity		^{137}Cs	$^{89,90}Sr$
Location	(bone)	(flesh)	(flesh)	(bone)
Savannah River	640 (R-8, 1957)	40 (R-9, 1961)	78 (R-8, 1967)	151 (R-8, 1979)
Steel Creek mouth	nr[b]	nr	280 (1971)	140 (1971)
Onsite	4500 (LTR, 1958)	985 (LTR, 1959)	3500 (SC, 1969)	960 (SC, 1970)

[a] LTR = Lower Three Runs Creek, SC = Steel Creek.

[b] nr = not reported. Link to tabulated data.

The highest **average** reported Savannah River ^{137}Cs concentration (22 pCi g^{-1}) occurred during the first half of 1967 at a location just downriver from the mouth of Steel Creek (R-8). For dose reconstruction, using average measured concentrations is more realistic for estimating potential exposure to individuals who may have relied upon Savannah River fish for a significant portion of their diet (e.g., 19 kg y^{-1}).

On the other hand, the highest **average** reported onsite concentration (1900 pCi g^{-1}) occurred during 1969 and is about two orders of magnitude greater than the highest Savannah River **average** concentration. Although onsite locations have not been legally accessed by the general public, there is the possibility that some individuals (poacher exposure scenario) have illegally collected and consumed fish from onsite locations.

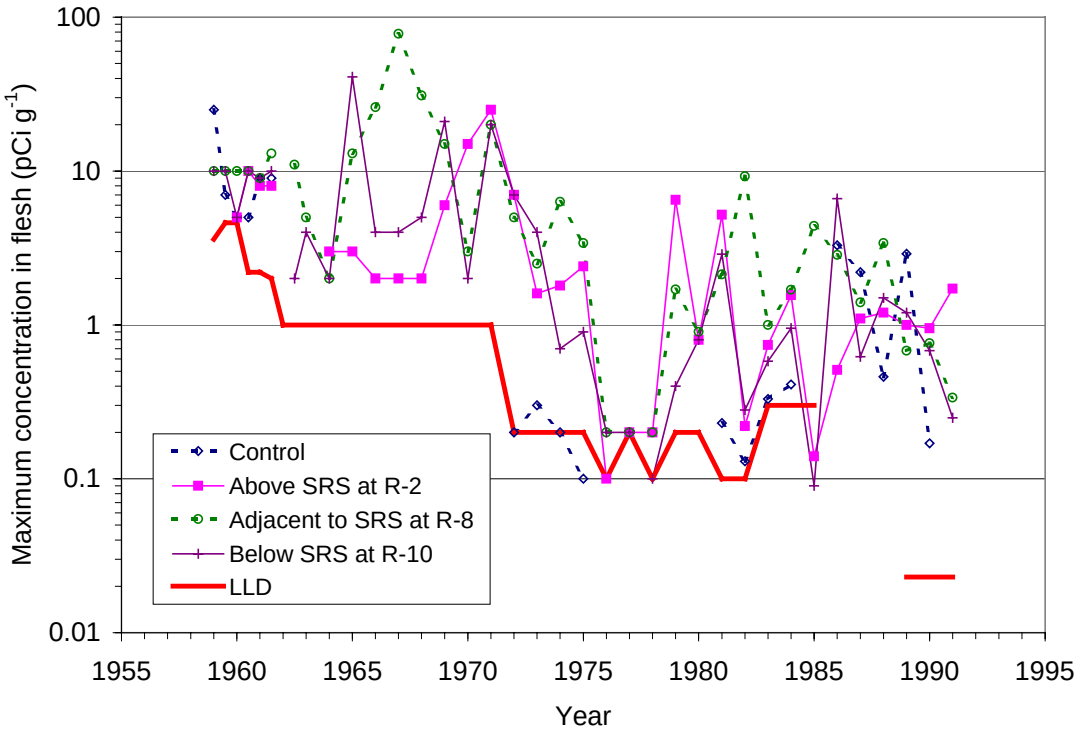

Figure 14-17. Maximum reported radionuclide concentrations in fish flesh collected from the Savannah River. Nonvolatile beta concentrations are shown before 1962 and ^{137}Cs concentrations are shown from 1962 through 1991. Link to tabulated figure data.

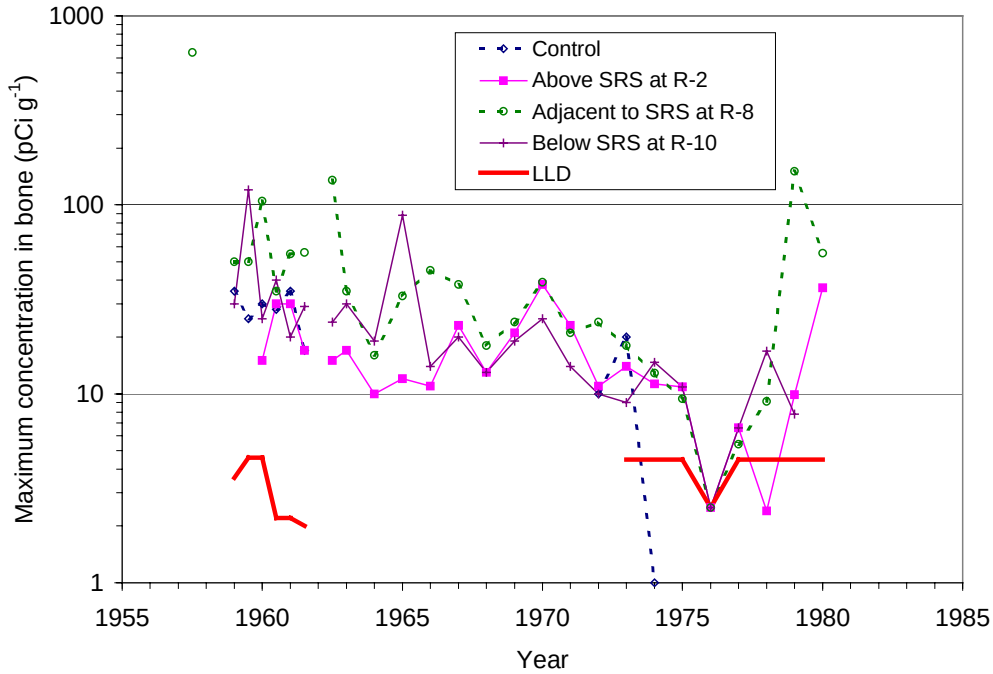

Figure 14-18. Maximum reported radionuclide concentrations in fish bone collected from the Savannah River. Nonvolatile beta concentrations are shown before 1962 and ^{89,90}Sr concentrations are shown from 1962 through 1980. Link to tabulated figure data.

Tritium

Mean and maximum tritium concentrations were reported for fish collected at locations above, adjacent-to, and below the Savannah River Site from 1970 through 1981 (Table 14-6). This beta-emitting radionuclide is incorporated into the water molecule; consequently it is present in all tissues. In general, average concentrations at adjacent-to and below-plant locations were higher than average above-plant concentrations. The maximum tritium concentration (54 pCi mL^{-1}) was measured in 1974 in a fish collected from the river location adjacent to the plant.

The reports have stated that the concentrations measured in fish reflect concentrations measured in Savannah River water. Examining annual average tritium concentrations measured in water collected at the above and below-plant Savannah River locations for the same years reveals higher concentrations at the R-10 river location. The median concentrations for fish and water are similar for the below-plant location, but median concentrations for fish and water are significantly different for the above-plant location. These results do not suggest that fish and water concentrations are not correlated, but it does not appear that they are linearly correlated with a slope near unity (at least at these relatively low concentrations). Tritium is generally not concentrated in biological tissues to levels greater than those found in water, but slightly higher levels in biological tissue may result from some degree of molecular organic binding (Whicker and Schultz 1982). This may account for the higher concentrations measured in fish at the R-2 river location.

Table 14-6. Mean and Maximum Tritium Concentrations (pCi mL^{-1}) Detected in Free Water Collected from Savannah River Fish from 1970 through 1981[a]

	Above plant (R-2)		Adjacent to plant (R-8)		Below plant (R-10)	
Year	Maximum	Mean	Maximum	Mean	Maximum	Mean
1970	6	4	8	5	11	5
1971	7	3	15	8	11	7
1972	9	4	16	7	17	8
1973	5	2	16	6	12	6
1974	8	4	54	12	12	8
1975	33	5	6	3	12	6
1976	9	5	10	5	16	8
1977	26	8	24	11	20	13
1978	1	1	4	4	7	7
1979	3	1	16	5	19	6
1980	7	3	17	5	8	4
1981	4	1	12	5	4	2
Median		3.5		5		6.5
Median[b]		0.47				5.3

[a] Data from Ashley and Zeigler (1984).
[b] Median value for river water, 1970–1981 (concentrations were not reported for the R-8 river location during this time period).

Other Radionuclides

Zinc-65 and ^{32}P were detected less frequently than ^{137}Cs (analysis frequencies were not specified), and bone tissue typically had higher concentrations than flesh. Zinc-65 is an activation product with a 245-day half-life and it is moderately assimilated in all body tissues (Whicker and Schultz 1982). Zinc-65 concentrations were reported from July 1961 through June 1970 in the Health Physics Regional Monitoring and Environmental Monitoring at the Savannah River Plant report series. The maximum reported concentrations in Savannah River fish flesh and bone, respectively, were 28 and 105 pCi g^{-1} during the second half of 1962 at the R-8 river location. In general, bone concentrations were higher than flesh concentrations. Since July 1963, concentrations in flesh and bone at all river locations have generally been less than the detection limit (1 pCi g^{-1}).

Phosphorus-32 is an activation product with a relatively short half-life of 14 days. It is rapidly assimilated primarily in bone tissue (Whicker and Schultz 1982). Maximum ^{32}P concentrations were reported from 1966 through 1968 in the Effect of the Savannah River Plant on Environmental Radioactivity report series. The maximum reported concentration in Savannah River fish flesh was 30 pCi g^{-1} during the first half of 1966 and in fish bone was 719 pCi g^{-1} during the first half of 1967.

Other gamma-emitting radionuclides, such as ^{103,106}Ru, ^{141,144}Ce, ^{60}Co, and ^{59}Fe, were rarely reported as detected in aquatic samples. However, mollusk (clam) and crustacean (crayfish and shrimp) tissues were occasionally analyzed for these radionuclides and were reported in the Health Physics Regional Monitoring report series in 1960 and 1961. Radiostrontium comprised the only reported activity in the shells of these animals, radiocesium comprised the only reported

nonvolatile beta activity reported for crayfish and shrimp flesh, and radioisotopes of cesium, ruthenium, cerium, cobalt, and iron were all reported as detected in clam flesh.

Other Sources of Data

Data regarding radionuclide concentrations measured in fish were obtained from the Georgia Department of Natural Resources (GDNR) and the South Carolina Department of Health and Environmental Control (SCDHEC) (GDNR 1997; SCDHEC 1997). These data can potentially verify concentrations reported by the SRS. However, comparisons made between these data sets are of somewhat limited use because of differing objectives of the respective analytical laboratories. Historically, the SRS analyzed a large number of fish to evaluate environmental impacts. The methods of analysis were developed to provide averages for trend analysis and throughput of a large number of samples. The GDNR and SCDHEC programs, on the other hand, were designed to analyze smaller numbers of fish to evaluate health risks. In general, more sensitive analytical procedures were used by these agencies to better estimate absolute radionuclide concentrations. However, interlaboratory comparisons between the GDNR and the SRS have indicated that both laboratories provide accurate measurements (Hoel 1991). Additionally, different sampling locations have been maintained by the three organizations, which hinders complete data comparisons. The GDNR and SCDHEC data are available in the embedded Excel® workbook discussed later in this chapter.

Figure 14-19 shows ^{137}Cs concentrations in fish measured by the SRS, GDNR, and SCDHEC at the R-10 Savannah River location. In general, the concentrations are similar, but there is a significant amount of variability in the data. Concentrations measured by the SRS appear higher, in general, than concentrations measured by the other two agencies. This difference is likely the result of the less precise analytical procedures employed by the SRS (e.g., lower detection limits for the GDNR and SCDHEC).

Figure 14-20 shows average ^{137}Cs concentrations measured in fish by these three agencies at locations that historically were not considered to be impacted by SRS activities. These locations include the R-2 river location historically sampled by the SRS, a Silver Bluff location (above the SRS at river mile 147.4) sampled by the SCDHEC, and two locations sampled by the GDNR (one near Augusta and one 90 m downriver from the SRS). In general, concentrations measured by the three agencies are similar, although there is a large amount of variability in the data. These data provide additional support for the supposition that the R-2 Savannah River sampling location has adequately estimated background radionuclide concentrations in fish.

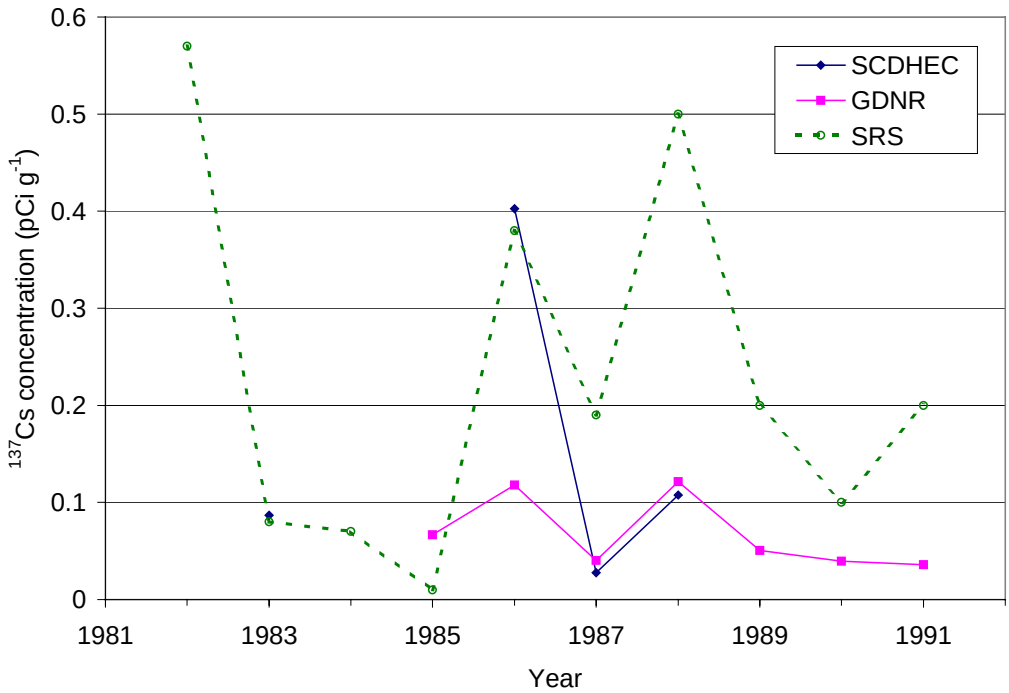

Figure 14-19. Average ^{137}Cs concentrations in fish from the R-10 Savannah River sampling location reported by the SRS, GDNR, and SCDHEC. Link to tabulated figure data.

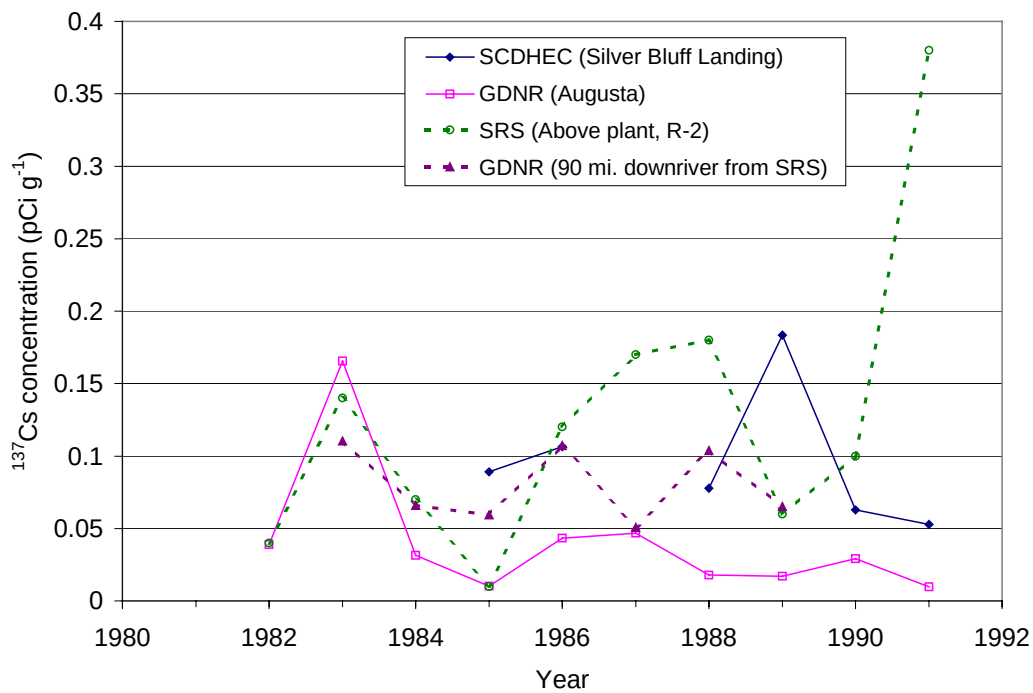

Figure 14-20. Average ^{137}Cs concentrations measured by the SRS, the GDNR, and the SCDHEC in fish collected from offsite locations not impacted by SRS activities. Link to tabulated figure data.

The GDNR has also measured tritium concentrations in free water obtained from fish tissues since 1985. At the R-10 Savannah River location, tritium concentrations reported by the GDNR from 1985 through 1986 range from 0.1 to 3.5 pCi ml^{-1} with a median of 1.7 pCi ml^{-1}. This is generally lower than concentrations reported by SRS from 1970 through 1981 (Table 14-6). A slight decrease in average concentration, however, is consistent with the decreases in release estimates of tritium to surface water after 1980 (Chapter 5).

Relative Contribution of Specific Radionuclides in Fish Tissue

To calculate a dose for a subsistence fisherman exposure pathway before 1962 (when only nonvolatile beta concentrations were reported), it may be necessary to estimate the fractional abundance of specific radionuclides in fish tissue. The median, maximum, and minimum ratios calculated as the concentration of cesium and strontium relative to the nonvolatile beta concentration for 1962 through 1969 are shown in Table 14-7 (based on data from Table 14-4). Ratios for flesh represent the relative concentration of cesium and ratios for bone represent the relative ratios for strontium. It appears that half or less of the radioactivity for both tissues can be attributed to cesium or strontium, based on median values for these years. It is clear that other radioisotopes were present in both flesh and bone tissue, particularly during the 1960s. It also appears that for a few years, measured ^{137}Cs concentrations were actually higher than measured nonvolatile beta concentrations (see Table 14-4).

Table 14-7. Ratios of Cesium and Strontium Concentrations Relative to Nonvolatile Beta Concentrations for 1962 through 1969 for Fish Collected from Three Savannah River Locations

	Above SRS		Adjacent to SRS		Below SRS	
Ratios	Flesh (Cs)	Bone (Sr)	Flesh (Cs)	Bone (Sr)	Flesh (Cs)	Bone (Sr)
Median	0.25	0.41	0.38	0.43	0.44	0.50
Maximum	0.33	0.82	3.14	1.14	1.50	0.76
Minimum	0.20	0.20	0.20	0.26	0.17	0.34

The Health Physics Regional Monitoring report series (1957 through 1962) stated that ^{137}Cs and ^{65}Zn comprised the majority of radioactivity in fish flesh and $^{89,90}Sr$ comprised the majority of nonvolatile beta activity in fish bone. The authors indicated that the dominance and magnitude of specific radionuclides varied significantly between different locations. The Environmental Monitoring at the Savannah River Plant report series (1965 through 1971) also reported that ^{137}Cs and ^{65}Zn comprised the majority of radioactivity in fish flesh and $^{89,90}Sr$ comprised the majority of nonvolatile beta activity in fish bone. Zinc-65 concentrations in fish were highest in Steel Creek and decreased through the 1960s. Zinc-65 was present in measurable concentrations in bone only by 1970, after which radiozinc concentrations were no longer reported. The Effect of the Savannah River Plant on Environmental Radioactivity report series (1962 through 1969) also reported that ^{137}Cs and ^{65}Zn comprised the majority of radioactivity in fish flesh, and $^{89,90}Sr$ comprised the majority of nonvolatile beta activity in fish bone. In 1962, radioactivity in Savannah River fish was attributed primarily to ^{40}K (Du Pont 1962a). Zinc-65 and ^{32}P were also detected in fish tissues but less frequently. The median ratio values reported in Table 14-7 indicate that radiostrontium and radiocesium comprise less than half of the nonvolatile beta activity for bone and flesh, respectively. There also appears to be significant variability in these

data. It is likely that other radioisotopes, such as ^{65}Zn, ^{32}P, and naturally occurring ^{40}K, also accounted for some portion of the nonvolatile beta radioactivity measured in both flesh and bone. Data from the SCDHEC (1997) indicate an average ^{40}K concentration in Savannah River fish of 2.3 pCi g^{-1}. This is consistent with ^{40}K concentrations of 2.6 pCi g^{-1} measured in Par Pond and L Lake in 1992 (Hinton 1997).

Figure 14-21 shows the ratio of radiocesium to radiozinc activity measured in the flesh of fish collected from Par Pond and Steel Creek. Zinc-65 has a significantly shorter half-life than does ^{137}Cs (245 days compared to 30 years) and is not as persistent in the environment. It appears that ^{65}Zn comprised a significant portion of the gamma activity detected in fish flesh through the mid-1960s. Concentrations of specific isotopes were not reported before July 1961, but ^{65}Zn likely comprised a significant portion of the radioactivity in fish flesh during the 1950s.

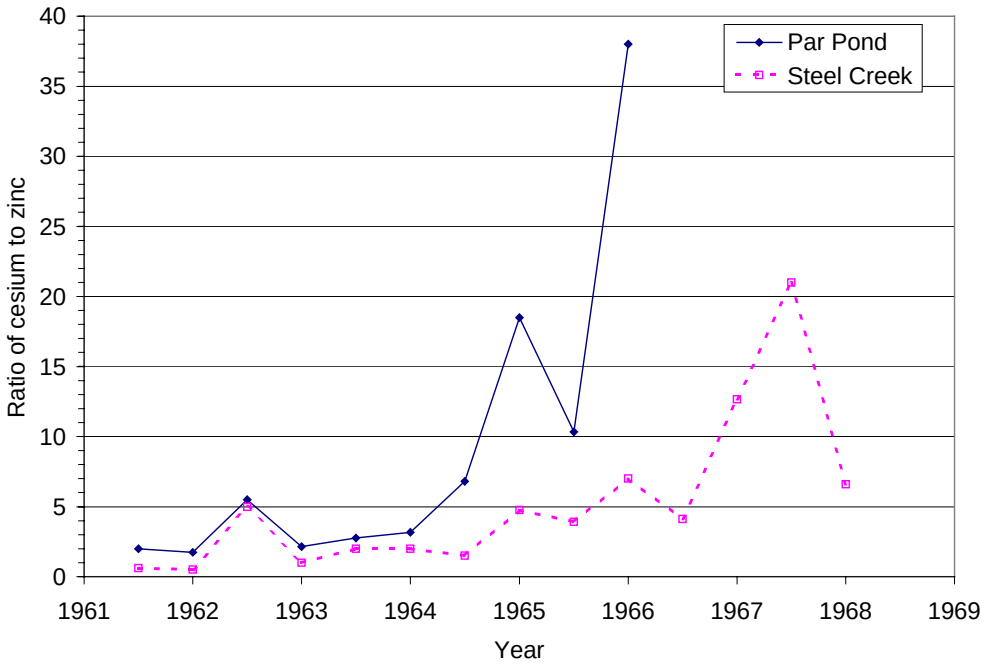

Figure 14-21. Ratio of ^{137}Cs activity relative to ^{65}Zn activity measured in the flesh of fish collected from Par Pond and Steel Creek. Link to tabulated figure data.

Harvey (1963) reported relative concentrations of various radionuclides in fish collected from Par Pond during 1962. Only $^{134,137}Cs$, ^{65}Zn, and $^{89,90}Sr$ were selectively concentrated in fish from an environment that also contained detectable concentrations of $^{141,144}Ce$, ^{51}Cr, $^{103,106}Ru$, $^{95}Zr/Nb$, ^{54}Mn, $^{59}Fe/^{60}Co$, and $^{140}Ba/La$ (^{32}P and ^{40}K concentrations were not reported in this study). Relative concentrations of $^{134,137}Cs$, ^{65}Zn, and $^{89,90}Sr$, in bluegill, catfish, and bass measured in this study are shown in Table 14-8. Radiozinc and radiocesium comprised all of the activity in flesh tissue, but radiozinc dominated in bluegill while radiocesium dominated in both catfish and bass. All three radioisotopes were present in bone tissue, with radiozinc dominating in all three species. It appears likely that the relative dominance and magnitude of specific radionuclides may vary significantly between locations as well as between species.

Table 14-8. Relative Concentrations of $^{134,137}Cs$, ^{65}Zn, and $^{89,90}Sr$ in Bluegill, Catfish, and Bass Collected from Par Pond in 1962[a]

	Bluegill		Catfish		Bass	
Radionuclide	Flesh	Bone	Flesh	Bone	Flesh	Bone
$^{134,137}Cs$	0.40	0.07	0.68	0.16	0.73	0.17
^{65}Zn	0.60	0.77	0.32	0.54	0.27	0.49
$^{89,90}Sr$		0.16		0.30		0.34

[a] Data from Harvey (1963)

During a June 19, 1991 meeting, the SRS and GDNR discussed elevated ^{90}Sr concentrations in fish flesh collected from the Savannah River (Hoel 1991). In particular, concentrations measured in bass and suckers collected from two locations in the river were evaluated. Both the GDNR and SRS laboratories analyzed composite samples. Although the SRS measured values were about a factor of 2 lower than the GDNR measured values, both laboratories measured concentrations of ^{90}Sr in flesh that were somewhat higher than would be expected for muscle tissue. Examining data supplied by the GDNR (GDNR 1997) reveals an average ratio of ^{90}Sr in edible tissue to that in nonedible tissue of 0.37. This number is based on concentrations measured in five composite samples collected on October 30 and 31, 1990, from three different Savannah River locations and includes the bass and sucker composites discussed at the GDNR and SRS meeting. If this ratio is an accurate measure of the relative concentrations of ^{90}Sr in edible (muscle) and nonedible (bones, scales, viscera) tissues, it calls into question the assumption that ^{137}Cs is the dominant radionuclide in the edible portion of fish (i.e., muscle tissue).

Evaluating data provided by the GDNR for 20 composite samples collected in 1995 and 1996 reveals an average ratio of 0.04. This ratio is approximately an order of magnitude less than the ratio calculated for the 1990 data, and is likely an overestimate of the true ratio because many ^{90}Sr concentrations reported for edible tissue were less-than values. Harvey (1963) reported $^{89,90}Sr$ concentrations for bone and flesh from bluegill, bass, and catfish collected from Par Pond in 1962. The ratio of $^{89,90}Sr$ in flesh to that in bone was less than 0.03. Whicker et al. (1990) reported ^{90}Sr levels in soft tissue to be 30 to 100 times lower than in bone tissue. Because strontium is a chemical analog of calcium, the relative distributions between bone and soft tissue for calcium are similar to those measured for strontium (Whicker et al. 1990). These data suggest that the elevated radiostrontium concentrations measured in fish flesh by the GDNR in 1990 were atypical, and it seems unlikely that radiostrontium would normally be present in significant concentrations in fish flesh. It is possible that the method of sample preparation by the GDNR in 1990 may not have adequately separated edible and nonedible portions of fish tissue.

Effect of Sample Preparation on Measured ^{137}Cs Concentration

Beginning in 1971, individual whole fish rather than dissected flesh tissue were analyzed for ^{137}Cs content. It is likely that this resulted in the reporting of concentrations that underestimated the true concentrations in flesh. Radiocesium concentrations are typically highest in muscle tissues, and analysis of additional tissues with lower ^{137}Cs concentrations (e.g., bone) would result in lower measured concentrations than analysis of muscle tissue only.

The GDNR calculated ratios of ^{137}Cs concentrations measured in edible tissue to those measured in whole fish (Hoel 1991). Ratios ranged from 1.20 to 1.49 with an average ratio of 1.4.

Concentration factors reported for whole body and muscle tissue by Gladden (1982) suggest a similar ratio of 1.5. It may be necessary to modify ^{137}Cs concentrations measured since 1971 by a factor (e.g., 1.5) to account for a reduction in measured concentrations because of a change in sample preparation procedures.

Estimating Radionuclide Concentrations in Fish at the Mouths of Onsite Streams

It is likely that the routine adjacent to SRS Savannah River fish sampling location (R-8) did not represent the maximum radionuclide concentrations in fish to which members of the public may have had legal access. Cesium-137 concentrations measured in fish from 1971 through 1981 at the mouth of Steel Creek were consistently higher than concentrations measured in fish at the R-8 river location just downstream from the mouth (Figure 14-22). The median ratio of concentrations measured in fish at the Steel Creek mouth (collected by SRS from 1971 through 1981 and by GDNR from 1982 through 1991) relative to concentrations measured in fish at the R-8 river location from 1971 through 1991 is 4. Comparing data compiled by the GDNR from 1982 through 1991 at the mouths of Steel Creek, Lower Three Runs Creek and, to a slightly lesser extent, Four Mile Creek also suggest elevated concentrations relative to those measured by the SRS at the R-8 river location (Figure 14-23).

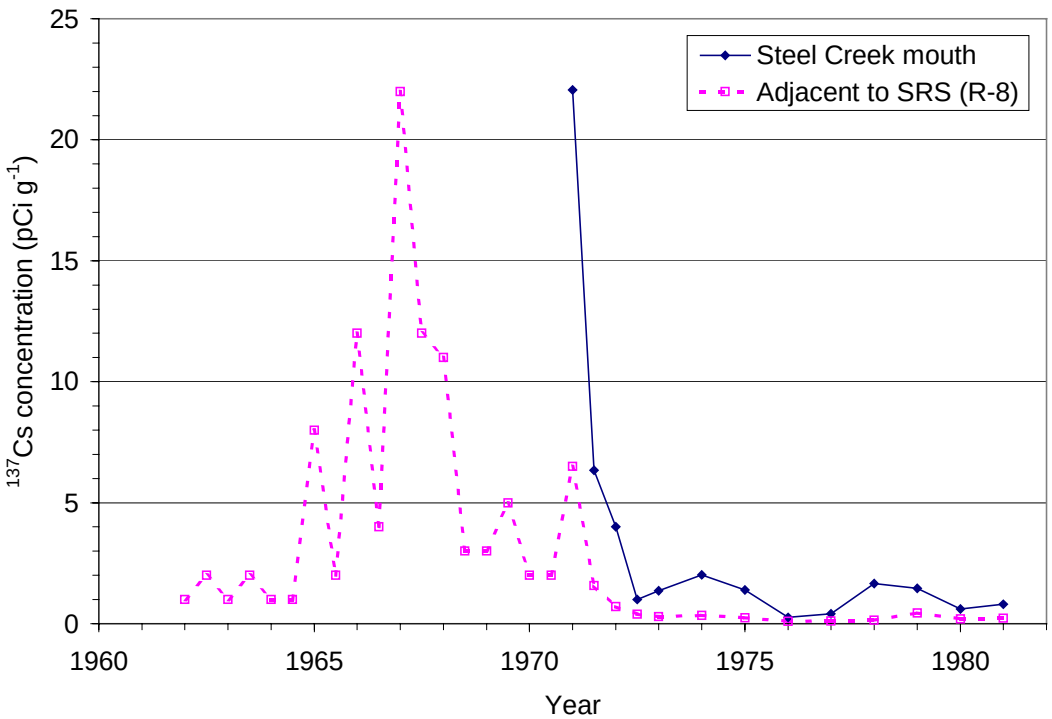

Figure 14-22. Comparison of average ^{137}Cs concentrations measured in fish collected by the SRS at the mouth of Steel Creek and at the R-8 Savannah River location. Link to tabulated figure data.

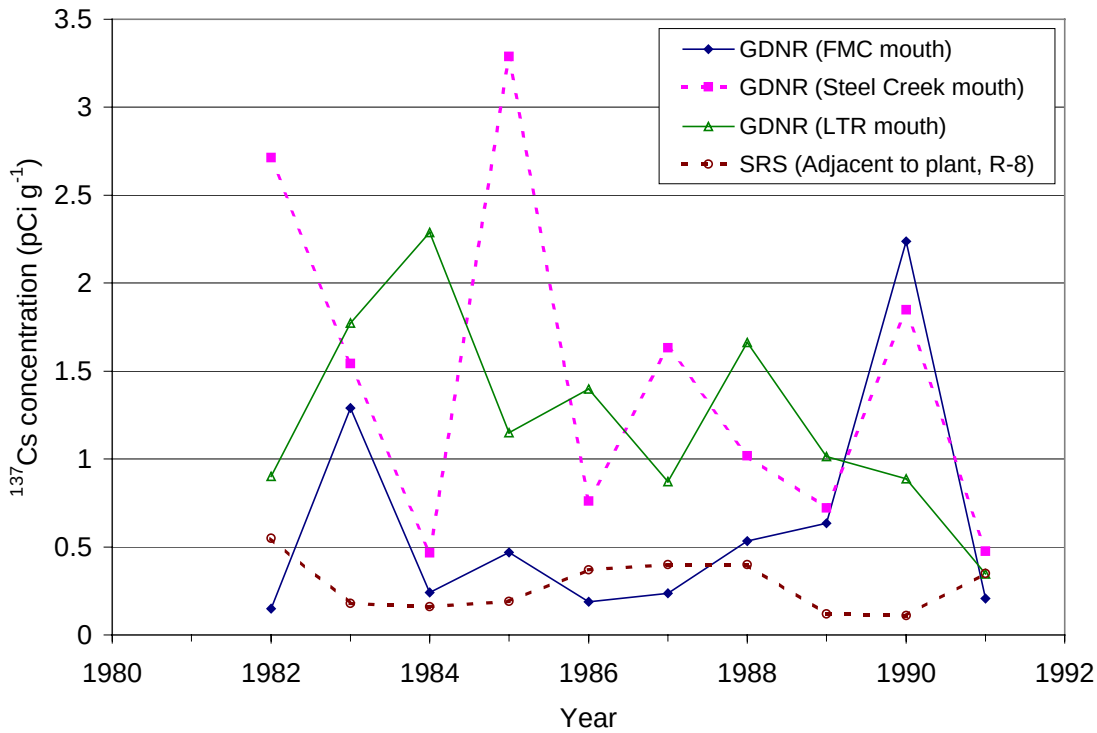

Figure 14-23. Average ^{137}Cs concentrations measured in fish collected by the GDNR at the mouths of FMC, Steel Creek, and LTR shown with concentrations measured in fish collected by the SRS at the R-8 Savannah River location. Link to tabulated figure data.

The ratio of concentrations measured in fish at the mouth of Steel Creek to concentrations measured at the R-8 river location (median ration = 4) can be used to estimate ^{137}Cs concentrations for time periods during which no concentrations were reported (i.e., before 1971). Figure 14-24 illustrates estimated concentrations at the mouth of Steel Creek from July 1958 through 1970 and measured concentrations at the same location from 1971 through 1991 that may have resulted from SRS activities. Average concentrations measured in fish at the R-8 Savannah River location between July 1958 and 1970 were used for the following calculations.

Estimated ^{137}Cs concentrations in fish for periods before 1962 were calculated from the measured nonvolatile beta concentrations in fish reported for that time and the median ratio of ^{137}Cs activity relative to nonvolatile beta activity for the R-8 river location described in Table 14-7 (median ratio = 0.38). Concentrations reported since 1971 were adjusted by an edible tissue to whole fish ratio of 1.5. These concentrations were then multiplied by a factor of four to estimate concentrations that might have been measured in fish at the mouth of Steel Creek. Finally, background concentrations (those measured at the R-2 Savannah River location) were subtracted from the estimated and measured Steel Creek mouth concentrations.

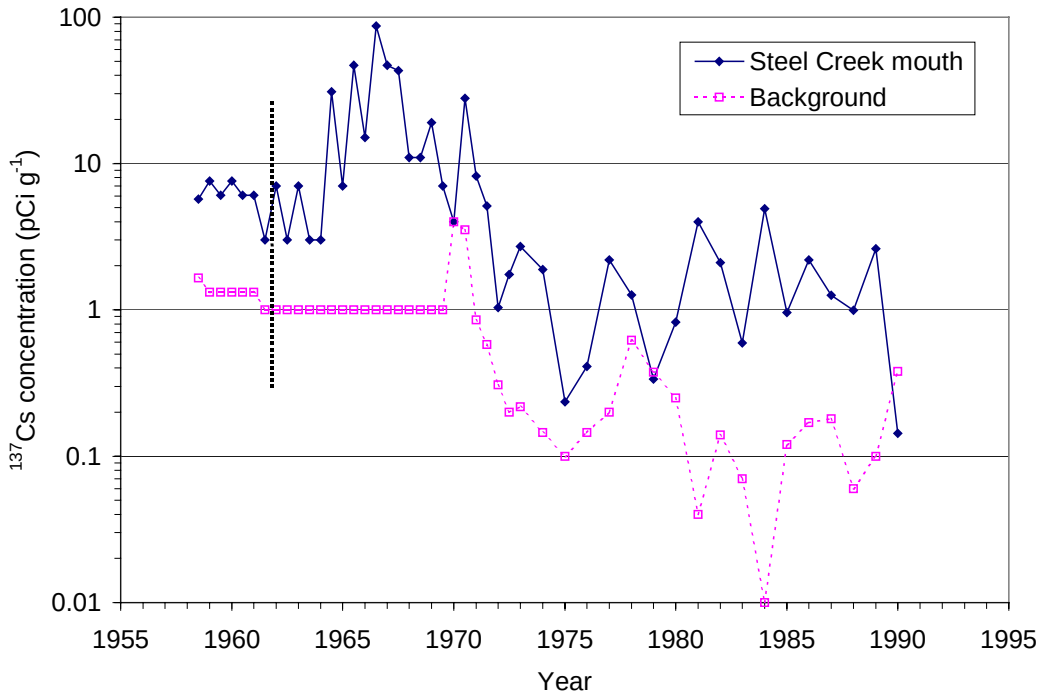

Figure 14-24. Estimated excess ^{137}Cs burdens in fish at the mouth of Steel Creek because of SRS operations shown with background concentrations measured at the R-2 Savannah River location. Values shown before the dashed vertical line were derived from reported nonvolatile beta concentrations and the median ratio of ^{137}Cs/nonvolatile beta activity for the R-8 location. Link to tabulated figure data.

For this example, average concentrations across all species have been used, but it may be necessary to account for potential differences in concentrations that may occur for different species of fish. These differences (previously discussed) and the relative percentage of bass, bream, and catfish collected at the R-8 Savannah River location (Table 14-1) may be used to estimate the concentrations that might have been measured in bass only, the species of fish that likely represents the maximum potential ^{137}Cs concentrations.

The data depicted in Figure 14-24 serve as a potential method for estimating maximum annual average radionuclide concentrations to which members of the public may have been exposed. It may eventually be necessary to evaluate concentrations at the mouths of other onsite streams using a similar approach.

It may also be possible to estimate radionuclide concentrations in fish at locations not included in the SRS routine monitoring program based on radionuclide concentrations measured in water. Figure 14-25 shows ^{137}Cs concentrations measured in water and fish collected from Steel Creek at Road A and from Lower Three Runs Creek at Patterson Mill Road. Fish and water concentrations appear well correlated.

Concentration ratios (or factors) are calculated here as the radionuclide concentration measured in fish (pCi g^{-1} wet weight) divided by the concentration measured in water (pCi mL^{-1}). The data depicted in Figure 14-19 can be used to calculate such a ratio. Table 14-9 shows the concentration ratios calculated for these data as well as concentration ratios reported by other

researchers (Harvey 1963; Gladden 1982). Concentration ratios appear to be significantly lower in Lower Three Runs Creek and Par Pond than in Steel Creek. This may be related to different water chemistry for the two locations, as different concentrations of nutrients such as potassium can markedly affect concentration ratios (Whicker et al. 1990), and concentrations may also vary as a function of water concentration. Whicker et al. (1990) reported concentration ratios for Pond B that were more than two times greater than those shown for Steel Creek in Table 14-8.

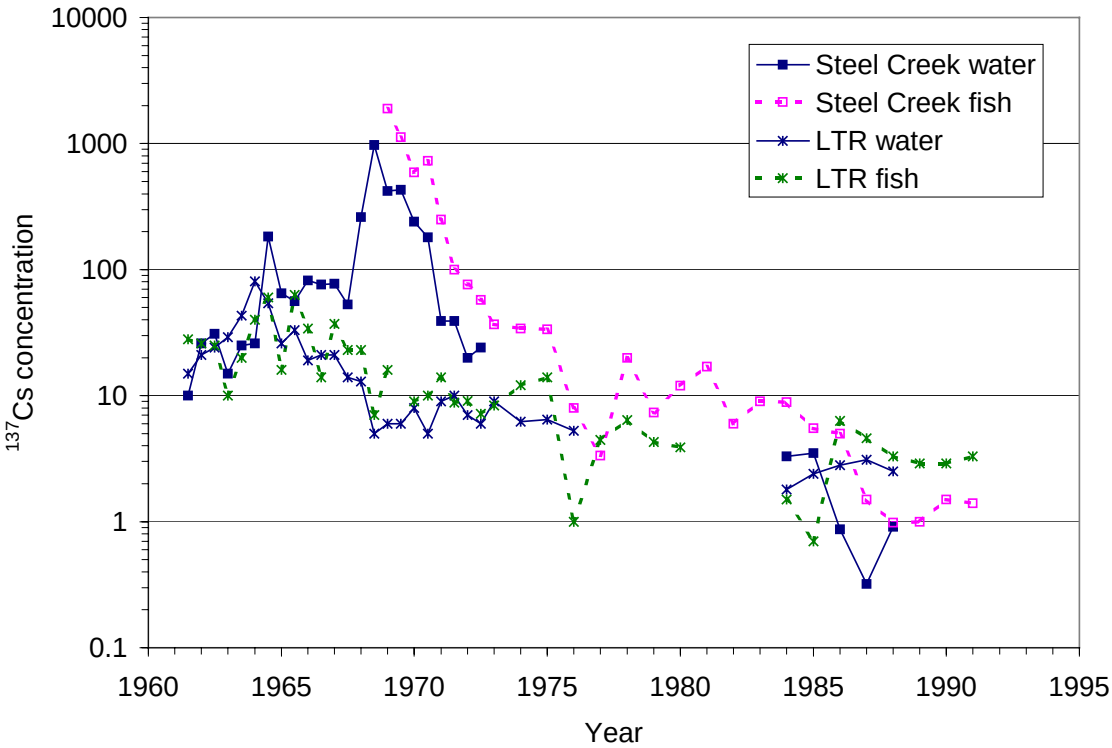

Figure 14-25. Annual average ^{137}Cs concentrations measured in fish (pCi g^{-1}) and water (pCi mL^{-1}) collected from Steel Creek at Road A and from LTR at Patterson Mill Road. Link to tabulated figure data.

Table 14-9. Mean Concentration Ratios Calculated for ^{137}Cs in Fish from Par Pond, Lower Three Runs Creek, and Steel Creek

Location	Concentration ratio (fish/water)
Lower Three Runs[a]	1258
Par Pond[b]	1100
Steel Creek[a]	3426
Steel Creek[c]	3029

[a] Ratio calculated based on data depicted in Figure 14-25.
[b] Ratio reported by Harvey (1963).
[c] Ratio reported by Gladden (1982).

Different species of fish may also exhibit significantly different concentration ratios. Gladden (1982) reported a mean concentration ratio (3029) for 527 fish collected from Steel Creek. Concentration ratios ranged from a maximum of 5688 for largemouth bass to a minimum of 963 for Savannah darters. Table 14-10 shows concentration ratios for ^{137}Cs, ^{65}Zn, and ^{90}Sr reported for Par Pond and Pond B for bass, catfish, and bluegill. It is clear that concentration ratios vary not only as a function of location, but also as a function of species and tissue type. It may be possible to estimate radionuclide concentrations in fish based on concentrations in water. However, site- and species-specific concentration ratios are clearly necessary because these ratios can vary substantially with species and, in particular, with location.

Table 14-10. Concentration Ratios for Fish Collected from Par Pond[a] and Pond B[b]

		^{137}Cs		^{90}Sr		^{65}Zn
Species	Tissue	Par Pond	Pond B	Par Pond	Pond B	Par Pond
Bass	Bone	500	nm[c]	1700	20,160	1400
	Flesh	1200	8600	<48	1088	1600
Catfish	Bone	800	nm	2100	18,240	3000
	Flesh	1200	6400	<48	20	600
Bluegill	Bone	600	nm	2400	nm	8200
	Flesh	900	5500	<48	nm	800

[a] Data from Harvey (1963).
[b] Data from Whicker et al. (1990).
[c] nm = not measured.

ELECTRONICALLY COMPILED FISH DATA

The various data summarized in this chapter are electronically compiled in two Microsoft Excel® workbooks. One workbook (Ch14-Figure_data.xls) contains the figures depicted in this chapter as well as the tabulated data that were used to produce the figures. In this workbook, there is a separate worksheet for each figure and one worksheet that contains the tabulated data for all of the figures. The second workbook (Ch14-All_data.xls) contains the data that have been tabulated from various environmental monitoring reports and aperture card printouts. The workbook contains several named worksheets that include brief summary of the compiled data.

Table 14-11 summarizes the data that have been electronically compiled for fish collected as part of the routine environmental monitoring program maintained by the SRS. Additionally, the names of the individual spreadsheets in which these data are compiled (including a brief description of the data) are provided.

Table 14-11. Description of Data Electronically Compiled for Fish

Workbook name	Worksheet name	Brief description of data
Ch14-Figure data.xls	Figures 14-1 through 14-25	Each worksheet contains a separate figure depicted in this chapter
	Data for figures	This worksheet contains the tabulated data for each of the figures
Ch14-All data.xls	Nonvolatile beta	Nonvolatile beta concentrations
	Zn-65	Zinc-65 concentrations
	Sr-89,90	Sr-89,90 concentrations
	Species %	Number and type of fish sampled, 1971–1991
	Cs-137	Cesium-137 concentrations, July 1961–1980
	Cs-137(2)	Cesium-137 concentrations, 1980–1991
	SCDHEC-Fish	Data provided by the South Carolina Department of Health and Environmental Control
	Locations	Description of sampling locations used by SCDHEC
	GDNR-Fish	Data provided by the Georgia Department of Natural Resources

USEFULNESS AND LIMITATIONS OF FISH DATA FOR DOSE RECONSTRUCTION

There are a number of factors that impact how the fish data may be used during subsequent phases of the dose reconstruction project. These factors include the availability of sufficient original monitoring data sets to verify reported summary data and evaluate spatial and temporal trends, as well as the ability to distinguish between Site releases of contaminants and other sources of the same contaminants in the environment (i.e., establish appropriate background concentrations).

In general, the aperture card printouts provided consistent verification of the summary values reported in the routine environmental monitoring reports. Information obtained from the GDNR and SCDHEC also provided general verification of the concentrations reported by SRS for fish collected from the Savannah River from 1982 through 1991.

Arithmetic mean concentrations have been historically reported by the SRS, and many of the trends and comparisons made in this chapter are based on these reported concentrations. Radiocesium concentrations, however, often fail to show a normal frequency distribution in environmental samples. This was evident for concentrations of ^{137}Cs measured in both fish and deer tissue (see Chapter 11). Medians, percentiles, and ranges are often more appropriate descriptors of these types of data. However, arithmetic mean concentrations likely err on the conservative side (i.e., provide an overestimate) of the central tendency for cesium concentrations in fish because the distributions are generally skewed to the right (e.g., log-normal).

The R-8 river location has been historically used by the Savannah River Site to represent the highest measured concentrations for fish collected from the routinely sampled Savannah River locations. However, concentrations measured at the mouth of Steel Creek from 1971 through

1981 were consistently higher than concentrations measured at the R-8 river location, just downstream from the mouth of Steel Creek. In addition, an examination of GDNR data indicates that ^{137}Cs concentrations in fish at the mouth of Four Mile Creek, Steel Creek, and Lower Three Runs Creek were elevated relative to the R-8 river location used by the SRS. Because Savannah River fishermen can potentially access fish at the mouth of Steel Creek, it may be necessary to consider concentrations measured at the R-8 river location as an underestimate of maximum concentrations to which Savannah River fishermen may have been exposed.

Concentrations for the Clark Hill and Stoke's Bluff control locations were sporadically reported. However, the concentrations reported for the Savannah River location 1 mi above the mouth of Four Mile Creek (R-2) are similar to concentrations reported for Clark Hill and Stoke's Bluff. Additionally, concentrations reported for this location are within the range of concentrations measured by the GDNR and SCDHEC at locations that would not be expected to have been impacted by SRS operations. This location appears to be adequate for establishing appropriate background concentrations.

Information is somewhat limited for the 1950s and early 1960s because of the lack of radionuclide-specific data. Therefore, it may be necessary to estimate ^{137}Cs and other radionuclide concentrations based on nonvolatile beta concentrations reported before 1962. Additionally, very little information at all is available before July 1958.

The section in this chapter entitled "Estimating Radionuclide Concentrations in Fish at the Mouths of Onsite Streams" provides a possible methodology for estimating maximum excess ^{137}Cs concentrations resulting from SRS operations in fish potentially accessible to members of the public. This methodology takes into account the fact that the R-8 location may not have represented the highest Savannah River fish concentrations, considers possible background concentrations, and estimates ^{137}Cs concentrations based on measured nonvolatile beta concentrations during time periods when ^{137}Cs concentrations were not measured.

Fish flesh was analyzed before 1971, and whole fish have been analyzed since then. It is possible that this would result in lower measured concentrations because tissues other than muscle (e.g., bone) typically do not accumulate as much cesium. However, a significant decrease in concentrations at this time is not apparent at any location, and Savannah River location concentrations have generally been near the LLD since 1971. However, for dose reconstruction, it may be necessary to modify concentrations reported since 1971 by an edible tissue to whole fish adjustment factor. Fish were also not analyzed by species until 1971, but this does not appear to be a significant limitation.

Variations in radionuclide uptake in fish can also vary as a function of location as demonstrated with the concentration ratios shown in Tables 14-8 and 14-9. This should be considered when comparing concentrations measured in fish collected from different locations. It is clear that site-specific factors (e.g., potassium and calcium concentrations) may influence the degree to which various radionuclides are assimilated in fish tissues.

It is difficult to determine the range of movement of fish living in onsite streams. However, based on the reported concentrations, the range of movement appears limited and has not resulted in significant numbers of highly contaminated fish entering the Savannah River. There is little question that some fish living in onsite streams have accumulated significant ^{137}Cs burdens, and that reactor effluent has also contributed to slightly elevated concentrations measured in fish collected from the Savannah River adjacent to and below the SRS. Significantly elevated ^{137}Cs concentrations have also been reported for fish collected from Par Pond and Pond B, but fish

living in these reservoirs have been isolated from onsite streams that drain into the Savannah River.

In summary, although mean concentrations may not be the best descriptors of the data, the available data are potentially quite useful for estimating exposure to the general public resulting from regular consumption of fish. Concentrations were not reported in the documents that have been reviewed for some onsite locations from 1977 through 1983, but concentrations have consistently been reported for Savannah River locations, which are most important for determining exposure to the general public. Very few fish were collected at any location from 1976 through 1978, but ^{137}Cs concentrations for those fish that were collected from the Savannah River were near the LLD at this time. Average ^{137}Cs concentrations in Savannah River fish appear to have decreased by more than an order of magnitude between 1963 and 1991, and nonvolatile beta concentrations before 1963 appear similar to those recorded in 1951 and 1952, before plant startup.

The fish data may also be useful to some extent for source term verification and model validation. These efforts will likely be undertaken during the next phase of the dose reconstruction and will require appropriate dispersion models and associated site-specific parameters.

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CHAPTER 15

FACILITIES AND OPERATIONS RELEVANT TO THE USE AND RELEASE OF CHEMICALS

ABSTRACT

This chapter provides information about the major facilities and operations responsible for releases of chemicals to the air and surface water at the Savannah River Site (SRS). The most important facilities and operations were the powerhouses, the separations processes, and raw materials operations. This chapter also describes key sources of information about releases of chemicals, site operations, waste disposal, water treatment, explosions, fires, and spills.

RELEASE POINTS AND PROCESSES

The SRS has several thousand process exhaust points and "literally tens of thousands of administrative events" according to the Part 70 Operating Permit Application submitted to the South Carolina Department of Health and Environmental Control (SCDHEC). Inquiries made to the Air Emissions Inventory database in October 1997 indicate that there were 527 emission points in D-Area; 3023 emission points in A-Area, which includes the powerplant, Savannah River Laboratory (SRL), and Savannah River Ecology Laboratory; 540 emission points in M-Area; 2347 emission points in F-Area, including the Naval Fuels Facility; 2123 emission points in H-Area, including the tritium facilities; 396 in T-Area or TNX; 535 in G-Area or CMX; and from 600 to 650 in each reactor area (Faugl 1996).

Six onsite process stacks emitted most of the nonradioactive materials released: three 313-M stacks and one 321-M stack in M-Area and two, 200-ft-high stacks (291/292-H and 291/292-F) in the separations areas. Because of the sensitive nature of some of the information about process equipment, design, and location, exhaust points were combined into one location for each area. Combining the exhaust points is justified because most of the toxic emissions came from the large stacks, and for air transport modeling that might be an option in later phases of the study, one exhaust point for each area would be adequate because of the size of the SRS.

This assessment focuses on major releases from processes. Activities not covered in this assessment include the operation of cars and trucks, painting, landscaping, and other maintenance work. Reinig et al. (1973) estimated that the Site had 675 vehicles that operated about 6,800,000 mi y^{-1} and consumed about 624,800 gal of gasoline per year. There are few records available to estimate releases from maintenance, construction projects, and central shop activities, which were not monitored before the late 1980s. Operations such as welding, painting, sawing, and cutting would have released nitrogen dioxides, sulfur dioxide, metals, and other pollutants. Operation of diesel generators, landscaping maintenance equipment, painting equipment, and other nonprocess-related equipment also released pollutants. Many of these sources have been included in the Air Information Reporting System (AIRS) database since about 1990.

KEY SOURCES OF INFORMATION

The monthly reports, area histories, annual reports, waste site characterization reports, environmental information documents, systems and safety analysis reports, technical progress reports, letters, memos, logbooks, and many other records were reviewed for information on chemical use and release data and how processes and pollution control measures may have changed over time. Examples of some of the process changes that would have affected releases are (a) conversion from the PUREX to the HM process in H-Area in May 1959, (b) initiation of recovery of ^{237}Np in 1963, and (c) installation of an electrolytic dissolver in H-Canyon, which could dissolve stainless and zirconium-alloy fuels, in 1969 (Westinghouse 1990).

Source term estimates were determined using inventory or usage estimates, knowledge of processes, information about release estimates as currently required by regulatory agencies, and monitoring data. There is very little monitoring data for chemical releases from the SRS. A limited amount of data are available from ambient air monitoring, water quality monitoring, and special studies, which are described in Chapter 19. Available monitoring data from the 1980s and 1990s were reviewed and compiled for selected chemicals. Chemicals detected at significant levels in the environment were added to the list of chemicals subjected to the ranking discussed in Chapter 16. For some chemicals, extrapolating back in time from 1980s monitoring data was the best way to estimate a source term, assuming the amounts and types of materials used and the processes did not change very much over the years.

The Emergency Planning and Community Right-to-Know Act of 1986 required the tracking of chemicals. Before this regulation, little or no information was available to describe or quantify the amounts of chemicals used, except in the raw materials area and tritium and separations facilities which kept inventories of process chemicals (Kvartek et al. 1994). Because chemical releases were not monitored and chemical use and disposal were not recorded before the late 1970s, much of the information on chemical use and release in the earlier years was learned from former and current Site employees. The following former and current Site personnel were interviewed about records of the amounts and uses for chemicals, including purchasing records and essential materials ledgers:

Retired	Bob Rodman, Purchasing
	Herman Drummond, Essential Materials Clerk in 300-Area
	Jim King, Essential Materials for the Power Department
	Paul Katonak, Computer Services
	Hap Holbrook , Essential Materials Clerk
	W.E. (Bill) Borders, Essential Materials Clerk in Separations
Onsite	Jim Morgan, Chemical Commodities Management Center
	Bob Harris, Procurement
	DuPree Simmons, Procurement
	Sally Strain, Industrial Hygiene
	Greg Still, Industrial Hygiene

In general, procurement records were kept for about 3 years, longer if they involved equipment for nuclear safety. Some summary information was retained for some vendors, but

most of the records on commodities and purchasing that might still be available pertain to transactions that occurred after 1990 (Morgan 1996).

Chemical storage, distribution, and record keeping practices have changed often over the years. Since 1994, all chemical purchases have gone through the Chemical Commodities Management Center, facilitating entry in the Worker Right-to-Know database and compliance with U.S. Environmental Protection Agency (EPA) regulations, according to Jim Morgan of the Chemical Commodities Management Center. Previously, many chemical purchases, primarily those with low exposure limits, were routed through the Industrial Hygiene Department. John Harris, was interviewed extensively in Phase I and provided the CIIS database information used for ranking the chemicals of concern. The Industrial Hygiene Department also maintains a comprehensive set of Material Safety Data Sheets. Chemical coordinators are designated in each major process area to function as a central point of contact for all chemical use in that area.

During interviews, essential materials clerks mentioned the existence of Cost Accounting Books, called Orange Books, which were kept in the years before the Superfund Amendments and Reauthorization Act of 1986 (SARA) Title III inventory records were required. These books were not located in the Phase I document search, and retired individuals who had kept these books and current staff in the Cost Accounting Department could not locate any of the Orange Books.

Essential materials ledgers were kept in H-Area, F-Area, and M-Area and probably in other areas as well. They typically listed the date, essential material code and/or the name of the material, the purchase order number, the receipt number, the vendor, and the quantity received. Essential materials ledgers for the separations areas and M-Area were found in Phase I and in the early part of Phase II. After the mid-1970s, receiving, inspecting, and sample logs were also used in M-Area (DPSOL-315-1060) to log materials received and to note whether they met specifications (Westinghouse 1987). Another record, called a consumption log, was kept after 1987 in M-Area. This generally contained the same information as the essential materials ledgers (Gary 1996). In addition, monthly essential materials transaction reports, also called chemical accountability reports, for M-Area were available from 1969. The information contained in the chemical accountability reports is difficult to compile into annual use amounts and is hard to interpret because many of the materials were procured by a bulk purchase order. Very large amounts of materials may have been listed when only small amounts were received and used. Total balances were sometimes reported rather than the monthly usage amounts. It became clear to both *Radiological Assessment Corporation* (*RAC*) and the Centers for Disease Control and Prevention (CDC) researchers reviewing the purchase orders in Phase I that these records would not be very relevant. In early years bulk chemicals were generally obtained through a standing order or by contract rather than through routine procurement. Many of the ledgers seem to reflect cost accounting rather than actual consumption or use by the facilities.

Preparing quantitative source term estimates for chemicals involved acquiring an understanding of their historic use in the SRS processes. A review of specific facilities and processes was presented in the Phase I Task 3 Report (Meyer et al. 1995). The processes of most concern for chemical dose reconstruction were the raw materials manufacturing processes in M-Area; the canyon processes in F-Area and H-Area; and the powerhouses in A-Area, F-Area, H-Area, and especially D-Area.

The operations of heavy water production facilities in D-Area, CNX and TNX, tritium production facilities, reactor areas, separations areas, waste disposal areas, and other facilities as

they pertain to chemical releases are also addressed further in the sections on individual chemicals in Chapters 17 and 18.

Safety analysis and systems analysis reports and standard operating procedures provided some of the best process descriptions. The 1996 operating permit application included useful descriptions of some of the most important processes and the air exhaust and liquid effluent discharges associated with them (Westinghouse 1996).

Site Studies of Nonradioactive Releases

Some research on chemical releases to the environment has been done previously at the SRS. In 1970, Monier and Bebbington compiled information on quantities and manner of disposal for nonradiological wastes in a memo. Monier and Bebbington (1970) states that "by far the largest quantities of such [nonradioactive] wastes come from the treatment of process and boiler feed water and these are almost entirely common inorganic compounds. I found no evidence of discharge of highly toxic and persistent chemicals directly to the environment." The memo acknowledged that wastes were discharged into streams that flow into the Savannah River, but it said that chemical analyses of river water above and below the Plant showed no consistent differences that could be attributed to these wastes. Most of the solvents used at the Site were thought to have been discharged to trenches and burned or buried, and a larger amount was discharged to sewers (most of which discharges to seepage basins) than directly to streams. M-Area was cited as the only area that does not have a "proper" facility for disposal of process wastes. At that time, acids, bases, salts, and chlorinated solvents were being discharged to a sewer that flows directly to Tim's Branch. The quantity of coal ash was said to be about 100 times that of the next largest waste discharge, which was caustic (sodium hydroxide). The memo recognized that significant quantities of inorganic materials might leach from the coal ash.

A summary of the estimates of the amounts of waste reported to have been "discharged" (Monier and Bebbington (1970)) is listed below. Most of the discharges were to waste sites, basins, and other contained areas rather than to areas from which the chemicals may have been transported offsite. Materials discharged to streams are listed separately below.

Discharged to Site streams	lb y^{-1}
Caustic	10^6 to 10^7
Sodium nitrate	10^6 to 10^6
Nickel sulfate	10^3 to 10^4
Aluminum nitrate	10^3 to 10^4
Chromate	10^3 to 10^4
Nickel chloride	10^3 to 10^4
Lead oil lubricant	10^2 to 10^3

Discharged to seepage basins, waste disposal pits, and trenches, etc.	lb y^{-1}
Coal ash	10^8 to 10^9
Nitric acid	10^5 to 10^6
Mercury	traces

Discharged to air	lb y^{-1}
Hydrogen sulfide	10^5 to 10^6
Ammonia	10^5 to 10^6
Hydrocarbon solvents	10^4 to 10^5

Discharged to sewers and pits	lb y^{-1}
Trichloroethylene	10^4 to 10^5
Tetrachloroethylene	10^5 to 10^6

In conclusion, Monier and Bebbington (1970) recommend that the dumping of chemicals from M-Area be reduced, the discharge of chlorinated solvents be stopped, and sediments of Tim's Branch be studied for solvent deposition. They also recommended the continuous samplers used to collect samples for radioactivity also be used to institute a program of chemical analyses in streams and the river.

In 1973, a survey of effluent monitoring at the Site was documented in Reinig et al. (1973). This survey was the report of a task force named to study releases. The task force attempted to identify release points and characterize releases from manufacturing, power generation, construction, and research activities. The report acknowledged that the characterization of nonradioactive releases from Site activities was much less complete than for radioactive releases. For many emissions points, no information was available about the concentrations or quantities of nonradioactive materials discharged in air and water effluents. Where analytical data were lacking on the nonradiological pollutants discharged from specific emission points, the task force relied on consumption of essential materials used in the particular plant operations and knowledge about the formation of substances during processes. Examples include sulfur oxides from burning coal, runoff from construction activities, and pesticide applications. They concluded that the effects of dilution, streambed deposition, chemical or biological reaction during transit, or other phenomenon that would change the concentrations of contaminants between the point of discharge and the point of stream discharge into the river needed to be better understood (Reinig et al. 1973).

For dose reconstruction, we agree that much more information on the dynamics of the system is required than is currently available. This information is needed to determine how much of a chemical released to a seepage basin may have reached groundwater, outcropped, or overflowed to Site streams and been transported to the river. Indeed, many scientists at the Savannah River Ecology Laboratory have dedicated their research careers to better characterizing SRS's streams and swamp systems and mobility and transport of materials to the river. We cannot expect to adequately define this system in a limited-duration dose reconstruction project.

DOE's (1987) *Preliminary Environmental Survey Report of the Defense Production Facilities*, included a section on the SRS. The following were of concern to the survey team because of nonradioactive materials:

- M-Area settling basin
- Sediments in Steel Creek Corridor
- Airborne mercury releases
- 1,1,1- Trichloroethane from M-Area

- Nonradioactive pits and piles in K-Area, L-Area, P-Area, R-Area, CFH-Area, CS-Area, A-Area, M-Area, and D-Area and TNX.

The M-Area unit ranked high because of the high mobility of tetrachloroethylene. Much of the concern came from scenarios involving onsite exposure to groundwater used as drinking water. This is not an applicable scenario for historical offsite exposure. See Appendix J for further details.

DOE (1987) mentioned that 1,1,1-trichloroethane releases from M-Area were derived from measured release data. However, no such records, or personnel who recollect monitoring of solvents in M-Area, have been found in the course of our study. The emission rate for trichloroethane was said to be about 200 ton y^{-1}.

The report also recognized that mercury was released to the air from the H-Area separations and tritium facilities. The survey team thought that there was a potential for stack emissions of mercury to be inhaled by the surrounding population. Again, DOE (1987) states that "the source term was derived from measured release data… Since a moderate number of assumptions were made in deriving the release rate . . . ," but the sources of the monitoring data and source terms were not given. Mercury monitoring was done in 1985 and these might be the data to which the survey referred. The potential consumption of mercury in crops and livestock drove the hazard ranking determined by DOE. They noted that the air emissions of mercury from the Site had been within the Clean Air Act standard of 200 lb y^{-1} and that mercury concentrations were also within the limit (0.25 μg m^3 at the plant boundary) of the SCDHEC Air Toxic Policy.

The nonradioactive pits and piles evaluated and ranked in the DOE Survey included: the K, L, R, C, F, D and A burning/rubble pits, Road A chemical basin, CMP pits, L-Area oil and chemical basin, the L-Area acid and caustic basin, Ford Building seepage basin, old H-Area seepage basin, hydrofluoric acid spill area, Radioactive Waste Burial Ground 643-G, Silverton Road waste site, TNX burying ground, metallurgy laboratory basin, metals burning pit/miscellaneous chemicals basin, and the SRL seepage basin. The potential for groundwater contamination from these sites was of concern. Source term data were taken from Site Environmental Information Documents. Tetrachloroethylene and trichloroethylene consumption in fish and potential ingestion of surface water while swimming drove the ranking. These hypothetical, worst-case scenarios are not applicable to dose reconstruction.

For all of the units ranked, the report says that, "This ranking… would place this …. unit with those environmental problems that are characterized as generally reaching receptors at levels below those used in regulatory decisions." The TNX Burying ground and D-Area Burning/Rubble Pit would be placed with those environmental problems "that are not projected to reach receptors"(DOE 1987).

FACILITIES OF MOST INTEREST FOR CHEMICAL RELEASES

M-Area

M-Area (also referred to as the 300-Area, the Reactor Materials Area, the Raw Materials Area, and Nuclear Fuel Fabrication Area) manufactured fuel and targets to be irradiated in the reactors (DOE 1987). M-Area is relatively close to the Site boundary, about 0.3 mi. M-Area contained five process buildings where reactor fuel elements were fabricated, extruded, cleaned,

and tested; it also included the 305-M Test reactor. Processes included uranium metal element fabrication (in Building 313-M), alloy extrusion (in Building 321-M), target extrusion (in Building 320-M), and the Chemical and Metallurgical Laboratories (in Buildings 321-M and 322-M) (Colven et al. 1985). M-Area processes were described in the Phase I Task 3 report (Meyer et al. 1995). A detailed description can be found in the technical manual written by Pelfrey (1987). The main process buildings described in the Operating Permit Application were (1) the slug production facility, which used a press, welding machines, cleaning and plating baths, autoclaves and other inspection, testing and cleaning operations, (2) the target and fuel fabrication facility, which included an extrusion press, degassing ovens, magneforming machines, radiography equipment, and other equipment used to produce aluminum clad fuel elements, (3) a target fabrication facility, which used similar equipment but primarily made lithium-aluminum tubes and control rods and tubes. M-Area operations involved chemical cleaning, etching, stripping, and plating (DOE 1987). Maximum production for M-Area occurred from about 1978–1986 (Gary 1996).

The M-Area facilities processed uranium, lithium, and aluminum into fuel and target components for the nuclear reactors. Processing included aluminum alloy formation, degreasing, etching, metal extrusion, hot-die size bonding, and nickel plating (Specht et al. 1987). Many memoranda and standard operating procedures describe the use of acids, caustics, and chlorinated solvents to etch and clean products, equipment, and tools. For example, the process for target slug fabrication used in 1985 involved degreasing in a hot nitric acid solution, dipping in Aquadag or Hydrograf solution to check cleanliness (if all lubricants had been removed these solutions would then uniformly coat the caps), dipped in nitric acid again if necessary, then cold water washing before etching. The cans were degreased in 1,1,1-trichloroethane, dipped three times in boiling 1,1,1-trichloroethane, then held in vapors above the 1,1,1-trichloroethane for about 3 minutes. The cans were etched in nitric and phosphoric acid and aluminux caustic (Pelfrey 1987).

Emission sources included solvent degreasing operations, metal cleaning and etching tanks, fixed roof storage tanks, metal extrusion, metal machining, metal casting, welding, groundwater treatment and wastewater treatment.

Much of the information used to determine emissions estimates was derived from the air emissions inventory estimates. These estimates were determined by the SRS using information obtained from interviews with operating personnel, standard operating procedures, stack testing results, and engineering judgment. Measurements of emissions were available or made for the acid etch tank and at least one degreaser. Mass balance approaches were used for the air stripper and other degreasers. Engineering calculations were used to estimate releases from the fixed roof storage tanks and open top process tanks and for releases from machining, casting, and extrusion. Four stacks were described by Hardt (1970) for M-Area:

1. The 313-M North plating line stack, 100 ft high, which vents the anodic etch, post-anodic nitric acid etch, dummy nitric acid etch, and the rinse and plating tanks
2. The 313-M South cleaning line stack, 100 ft high, which vents the pre-anodic nitric acid etch and the obsolete slug cleaning using nitric acid
3. The 313-M Final etch recovery stack, 100 ft high, which is equipped with a fume scrubber and vents the recovery and final etch nitric acid tanks
4. The 321-M Stack, also 100 ft high, which vents the tubs nickel nitric acid tank, tetrachloroethylene degreaser, and the etch caustic tanks (Hardt 1970).

The emissions sources for M-Area listed in 1992 by Radian Corporation, the contractor who estimated emissions for Westinghouse Savannah River Company, included fixed roof storage tanks; metal casting operations; metal extrusion operations; metal machining (grinding, polishing, and cutting); solvent degreasing; metal cleaning and acid etching tanks; welding; wastewater treatment; and groundwater treatment (Radian 1992b). Information sources listed include air permits, process and equipment diagrams, interviews with operating personnel, material safety data sheets, standard operating procedures, industrial hygiene data, stack testing results, and experience and engineering judgment. The approaches to estimate emissions were material balance, emission factors, stack measurements, and engineering calculations and assumptions. It appears that the methods required more detailed information than we have been able to locate for the dose reconstruction. The methods also seem to be conservative. These estimates are probably less uncertain than those we could calculate based on less information.

M-Area emissions to air included nitrogen oxides and nitric acid fumes from acid etching, cleaning, and plating processes and metal fumes from extrusion operations (Du Pont 1973; DOE 1987). Chlorinated solvents also evaporated from degreasers and from liquid effluents discharged to Tim's Branch and to the seepage basins.

The buildings are relatively close together so the emissions from the building's vents and three stacks were combined, and the four buildings were considered one release point. This is an adequate assumption for modeling transport of contaminants offsite.

Large quantities of chemicals were stored and handled in M-Area. Chemicals used in the M-Area processes included aluminum fluoride, boric acid, hydrochloric acid, lead powder, lithium fluoride, nitric acid, nickel carbonate, nickel sulfate, nickel chloride, organic phthlate compounds, phosphoric acid, sodium nitrate, sulfuric acid, tetrachloroethylene, trichloroethylene and 1,1,1-trichloroethane (DOE 1987; Colven et al. 1985). Use of these chemicals resulted in wastewater that resembled electroplating waste from metal forming and finishing processes. The waste effluent contained hydroxide precipitates of uranium, nickel, lead, chromium, zinc, and other metals as well as nitric acid and nitrates (Specht et al. 1987; Colven et al. 1985; Bradley 1981). The effluents discharged from M-Area have also included millions of pounds of chlorinated organic solvents used for cleaning and metal degreasing. Moderate quantities of acids, bases, salts, other cleaning solutions, and lubricants were also discharged to the sewer (Monier and Bebbington 1970). The sewer flowed directly to Tim's Branch, which flowed through a swampy area into Upper Three Runs Creek or to a settling basin after 1958. Not until 1979 were waste solvents barreled and not released to the sewer (Christensen and Brendell 1981). Groundwater analysis has indicated that trichloroethylene, tetrachloroethylene, 1,1-dichloroethylene, lead, and nitrates have contaminated the groundwater beneath M-Area. The groundwater has not migrated offsite (DOE 1988).

The releases to air are described in Chapter 17 for each individual chemical. Surface water and air releases of solvents from M-Area were related by the fact that the process sewer and surface water releases evaporated to the air and seeped into the ground to contaminate groundwater, which has been treated using an air stripper, resulting in additional releases to the atmosphere. Because of these relationships, Chapter 17 addresses the releases of the chlorinated solvents from M-Area liquid effluents, air strippers, and processes to the air and to surface water. Chapter 17 presents an assessment of the use and release of trichloroethylene, tetrachloroethylene, and 1,1,1-trichloroethane to the air, primarily from evaporation of solvents as they were used and discharged to streams and the settling basin.

The influent to and effluent from the M-Area settling basin was sampled and analyzed weekly for 10 weeks from March through May 1985. Based on these sampling data, a removal efficiency was calculated for the basin ([the influent concentration minus the effluent concentration] divided by the influent concentration). The results indicate that the retention of metals in the basin averaged about 80%. Although trichloroethylene and tetrachloroethylene were not being used at the time of this analysis, they were detected in the influent and effluent, presumably from the process sewer line and sludge and soils of the basin. 1,1,1-Trichloroethane was found at about 150 ppb in both the influent and effluent (Colven et al. 1985; Pickett et al. 1987). Table 15-1 summarizes this basin retention data for chemicals of concern.

Table 15-1. Average Influent and Effluent Concentrations from Nine Weekly Composite Sample and Calculated Removal Efficiency for the M-Area Seepage Basin in 1985 (Colven et al. 1985)

Chemical	Average influent concentration (mg L^{-1})	Standard deviation	Average effluent concentration (mg L^{-1})	Standard deviation	Average removal efficiency (%) for settling basin
Nitrate	361	180	156	70	57
Lead	0.407	0.50	0.110	0.31	71
Nickel	5.95	5.0	0.573	0.98	90.4
Cadmium	0.004	0.002	0.003	0.001	25
Chromium	0.037	0.037	0.006	0.006	84
Mercury	0.276	0.14	0.215	0.024	22
Zinc	0.141	0.065	0.024	0.03	83
Manganese	0.034	0.20	0.008	0.009	76
Magnesium	0.210	0.25	0.0513	0.042	75
Trichloroethane	144	79	151	67	<1
Trichloroethylene	<11	15	<18	9.5	<1
Tetrachloroethylene	<10	12	<10	6.5	<1
Uranium	50.9	70	66.7	51	<1

Uranium levels (ranging from 3.5 to 185 mg L^{-1}) were considered low, but the removal of uranium by the seepage basin was very poor. In fact, the average effluent concentration was greater than the average influent concentration, although both were extremely variable (Colven et al. 1985). The authors thought this may have been due to the high pH (averaging 11) of the water, which would tend to dissolve uranium (Pickett et al. 1987).

The release of metals from M-Area to surface water was researched. Metals were not measured in Tim's Branch discharges when process wastes were being discharged directly to the stream. In 1966, the spillway of Steed's pond gave way and allowed the pond to drain, exposing the sediments. The Site Health Protection Department conducted a study of Steed's Pond sediments in 1967. They sampled for cadmium, chromium, lead, nickel, uranium and zinc and decided that uranium was the contaminant of most concern. The study's authors concluded that the amount of sediment transported to Upper Three Runs Creek was not affected by the spillway collapse (Pickett 1990).

Merz (1982) summarized two years of effluent sampling data and reported that M-Area effluent concentrations of chemicals were less than the newly proposed EPA liquid effluent regulations for the metal finishing industry. The exceptions were the concentrations of total toxic organics, which were 1.66 mg L^{-1} in sewer effluent going to the settling basin and 0.70 mg L^{-1} in sewer effluent going to Tim's Branch, and the total suspended solids, which were 34 mg L^{-1} in sewer effluent going to the settling basin and 20 mg L^{-1} in sewer effluent going to Tim's Branch. Merz (1982) estimated that sludge in the settling basin contained about 32,000 lb of heavy metals. Groundwater samples at that time did not detect elevated concentrations of metals in the groundwater beneath the basin.

In 1984, sediment samples were collected at 31 locations along Tim's Branch and at Steeds Pond. In 1985 and 1986, monthly analysis was done for metals at four locations along Tim's Branch. Based on analyses of these samples, the effluent was thought to have contained tons of chromium, nickel, aluminum, and iron and thousands of pounds of cadmium, copper, lead, and zinc (Specht 1991). Chapter 18 describes concentrations of metals in Tim's Branch sediments found in these studies. A memo from Pickett (Pickett 1990) reports that sampling data indicate that metals are substantially elevated in the sediments of Tim's Branch, downstream of the confluence with the influent discharge ditches from the 300/700 Areas (NPDES outfalls A-014, A-011, and A-008). The contamination was attributed to historical discharges from 313-M, which were not diverted to the seepage basin until 1973 (Pickett 1990). Chapter 18 individually addresses the uses and surface water releases of nitrates, nitric acid, and the metals arsenic, cadmium, chromium, lead, nickel, uranium, and zinc.

A-Area

Many of the administrative buildings are located in A-Area. The Savannah River Technology Center (SRTC) in A-Area has done research and development and analytical support for Site operations. SRTC operations involve laboratory hoods, shielded cells, small pilot-scale facilities, waste handling facilities, glass shop, and laboratory chemicals. The radioactive hoods and cells are vented through a sand filter and one main stack. The SRTC metallurgy laboratory has been used to test corrosion and cleaning of metallurgic samples. The relatively small emissions of nitric acid and other pollutants have been exhausted through two 35-ft-tall stacks. The Savannah River Ecology Laboratory facilities include laboratory hoods, environmental sample analysis, and diesel generators.

Separations Areas

Two chemical separations plants were located near the center of the site between Upper Three Runs Creek to the north and Four Mile Creek to the south, in areas designated as 200-F and 200-H. The principal facility in each area was called the 221 Building or Canyon. Each separations plant contained remotely serviced and operated canyon facilities and directly serviced and operated finishing facilities for processing irradiated materials from the production reactors. The process dissolved the irradiated fuel and target materials and produced solutions containing various products that were separated from fission products by solvent extraction and ion exchange (Fisk and Durant 1987). Special reports, monthly progress reports, and other records provide information on how much and how many of the process chemicals were used in the

process each month and how much went to the high level and low-level waste for several years in the 1980s. However, we do not know how much went out the stacks or into Site streams because this was not measured.

Emissions examined in Chapters 17 and 18 included ammonium nitrate, cadmium, chromium, hydrazine, mercury, manganese, nitric acid, and nitrogen dioxide. Emissions of volatile organic compounds from F-Area and H-Area have not been measured, but they would be expected to be minimal based on process knowledge. The extractants, kerosene and tributyl phosphate, have relatively low vapor pressures and the amounts volatilized and exiting the process vessel vents would be expected to be small (DOE 1987).

A history for the separations areas (Du Pont 1988) contains useful information on typical operating times and process rates. The report summarizes the specific leaks, spills, and other incidents that occurred in the canyon buildings. All of the spills, leaks, and overflows described in the history and in the monthly reports seem to have occurred inside the canyons and did not involve releases to the environment (Du Pont 1988). In the late 1980s, an effort was made to reduce the volume of waste discharged from the separations areas (Pickett 1996). Several processes changes have been noted, such as the mercury reduction and deletion of the manganese oxide strike in the 1980s. The years 1987–1988 were the last two of maximum throughput through H-Canyon (Pickett 1996). F-Area processes were at peak operations in 1984 and 1986 (Villa 1996).

The 211 Buildings in the separations areas, also called the Outside Facilities, included bulk chemical storage and water handling, acid recovery, evaporation, segregated solvent facilities, water treatment, and electricity and steam generation (Fisk and Durant 1987). Spill containment dikes were installed around the 211 storage areas in 1988. Before that, any potential release would have traveled to outfall ditches and surface water. Liquid chemicals used in the separations areas were received by rail or truck by the chemical storage facilities. Tributyl phosphate, n-parriffin, aluminum nitrate, and sodium hydroxide were received in tank cars, and nitric acid was delivered by tank trailers. Bulk liquids were pumped into large tanks that were mounted in concrete saddles. The nitric acid and aluminum nitrate tanks were stainless steel and most of the other tanks were carbon steel.

Chemicals were pumped through pipelines to points of use in 221-H and F, and 211-H and F. Storage tanks and makeup tanks were on concrete pads with curbs and sumps or concrete basins surrounding them. In later years, additional confinement barriers were built and used. Most of the tanks had agitators, overflows, floor drains, drain sumps, and decant sumps, each with a pump. The tank areas had a sump collection tank and a recycle sump. A recycle vent system provided a way to filter contaminated air in tanks or vessels and vent tanks while they were being filled.

Canyon production rates were compiled and are presented in Chapter 2 of this report. Data on F-Area and H-Area production were correlated to known periods of waste generation, descriptions of volumes of chemicals used per batch or per mass of material process, and compiled as a part of waste reduction or cost accountability studies. For some of the chemicals, we used the production rates to normalize release rates in order to predict releases for the years we did not have monitoring data.

The major canyon processes were dissolution, head end, first solvent extraction cycle, second uranium solvent extraction cycle, and the second neptunium/plutonium solvent extraction cycle. Waste stream preparation and treatment included the high activity waste, low activity

waste, frames waste recovery and general-purpose evaporator systems. Chemicals used in the processes were cleaned and purified for reuse by the acid recovery, solvent recovery, waste handling, and segregated solvent systems (Westinghouse 1996).

The dissolving process was performed in two steps or batches. The first step involved dissolving aluminum cladding in caustic, which emitted ammonia, and the second step involved dissolving the irradiated uranium in nitric acid, evolving nitrogen oxides. The two dissolution steps were vented separately to separate stacks (DOE 1987). The second stack was added sometime in the 1960s to prevent the ammonia emitted from the first step from reacting with nitric acid to form ammonium nitrate solid material. This material condensed on the inside of the stack (Pickett 1996) and could be dislodged, which resulted in radioactive spalls from the stacks depositing on nearby ground and sidewalks.

The canyon stacks are 200 ft high and 10 ft in diameter. The canyon exhaust system consisted of five separate systems: the dissolver off-gas system, gang valve corridor and central exhaust system, old HB-Line exhaust system, process vessel vent system, and recycle vessel vent system. The sand filters and high-efficiency particulate air (HEPA) filters between these systems and the stack were said to provide confinement for nearly 100% of the radioactive particulates, except for the dissolver off-gas system. The dissolver off-gas was vented through an iodine reactor, followed by a fiberglass filter to remove particulates, then exhausted through the stack. The central exhaust system drew air from the hot and warm canyon cells through HEPA filters using exhaust fans. The process vessels for the head end process, the first solvent extraction cycle, the second uranium cycle, the second neptunium/plutonium solvent extraction cycle, solvent recovery system for each extraction cycle, the frames waste recovery system, low activity and high activity waste process vessels, and rerun system were all vented through the process vessel vent system fiberglass filters followed by sand filters then out the stack. The acid recovery system, general purpose evaporator, segregated solvent system, third-level cold feed process, sump exhaust, waste handling facility, and uranyl nitrate storage tanks were vented to the recycle vessel vent system fiberglass filters followed by sand filters then out the stack. Chapter 17 describes mercury and nitrogen dioxide emissions from the canyon stacks.

Tritium Facilities

The 1996 operating permit application, an unclassified document submitted to the SCDHEC, described the tritium facilities operating in 1994. In 1994, Lines I and II of the 232-H tritium operations included separations, cryogenic distillation, gas stripping, Z-Bed recovery, degreasing, metallography, and cutting. The process gases from 232-H were passed through a uranium and zeolite bed to remove water vapor from the hydrogen isotopes. They were then passed through a palladium-silver diffuser to separate the isotopes from argon, helium, and nitrogen. The purified isotopes were then fed into a cryodistillation process. If the off-gas met the stripper feed limits, it was discharged through the stack stripper to the stack. If it did not meet limits, it was recycled back to the diffuser for the removal of more tritium. Tritium was separated from protium and deuterium by cryogenic distillation using a batch still. The product stream was loaded into gas containers in 232-H or sent to 233-H or 234-H for loading into reservoirs. The off-gas from 232-H and purged nitrogen collected in the nitrogen collection tank was collected in a stack tank and passed through a stripper system involving zeolite beds to remove tritium, and then was discharged to the stack. Regeneration of the zeolite beds involved the use of magnesium. The

process also involved moderate quantities of freon and degreasing solvents, such as 1,1,1-trichlorethane. Exhaust air from these processes, including the laboratory, degreasing, metallography, and cutting hoods, as well as the processes just described, was released from the 200-ft-tall, 10-ft-diameter, 295-H stack with an exhaust velocity of about 13 ft s^{-1}. Most of the processes have been in place in some form since 1954. The metallography processes were installed in 1986. The release of chlorinated solvents out the stack were said to have been negligible. Relatively small nitrogen oxide emissions were a result of nitric acid etching, performed on a small laboratory scale. The degreasing hood emissions were primarily freons (Freon TF or 1,1,2-trichloro-1,2,2-trifluoroethane) (Westinghouse 1996).

Line III of 232-H involved the extraction of tritium from target assemblies. These were loaded into crucibles and melted in a vacuum extraction furnace. The product gases were purified in Lines I and II as described above. Most of the off-gas treatment was concerned with reducing the amount of tritium released. The line III off-gas was released through a 200-ft exhaust stack, 297-H, and subject to pre-filter and HEPA filters. The extraction furnace described in 1994 has been in place since 1961 (Westinghouse 1996).

The tritium loading facilities, 234-H, include the finishing operations for reservoirs loaded in 233-H, receiving and unloading reservoirs, packaging and shipping reservoirs, inspecting and storing reservoirs, providing tritium assay of filled reservoirs, providing stripper for the off-gas treatment from 236-H and 232-H operations, proof testing new reservoirs, and sealing reservoirs for disposal. Before 1994, reservoirs received were unloaded, the gas was transferred to 232-H Lines I and II, and the reservoirs were sent to 238-H for reclamation. Emissions were released from a 200-ft-tall stack, 296-H. Emissions of nickel from the reservoir finishing operations were classified, but they would be expected to resemble other fabrication processes.

Emissions from the reservoir reclamation facility in 238-H were released from a 75-ft stack. Processes include milling and machining, lathe, and decontamination of reservoirs. The reservoir loading facility, 233-H, receives filled reservoirs, unloads them, transfers gas to Lines I and II, compresses the gas, and loads it into new or reclaimed reservoirs. Emissions were released from a 50-ft-tall stack. No chemical emissions were listed in the operating permit application, which focused on releases of tritium and control equipment to reduce these emissions (Westinghouse 1996).

The Systems Analysis for the Tritium Processing Facilities in 234-H states that chemical toxicity hazards in the building are small and that the only hazard identified by the industrial hygienists was associated with careless handling of cleaning solvent [freon] and exposure to mercury only specific chemical toxicity hazard is associated with the careless handling of cleaning solvent, [and] mercury (Haynes and Stoddard 1984).

Naval Fuel Manufacturing Facility

The Naval Fuel Manufacturing Facility (FMF) Project began in 1981. Pilot plants for the processes were operated at the SRTC in 1983. From 1986 to 1989, the facility manufactured highly enriched uranium fuel to be used by the nuclear Navy, from uranium shipped in cylinders from the gaseous diffusion plant at Portsmouth, Ohio. The FMF, housed in a single building in F-Area, 247-F, operated independently from other facilities in F-Area and maintained its own analytical laboratory (Zeigler et al. 1987; Evans 1998).

The Naval Fuels processes resulted in the release of relatively small amounts of nitric, sulfuric, and hydrochloric acids; nitrogen oxides; sulfur compounds; and other chemicals from stacks and vents (Du Pont 1994). Some of the process exhaust was released through roof top vents (DOE 1982). The facility was designed with a 7-ft diameter, 108-ft-tall stack with a predicted flow rate of 54 ft s^{-1} (Du Pont 1994). The facility's off-gas treatment system was constructed with filters, vapor coolers, a cyclone separator, a venturi scrubber, and caustic scrubbers to remove aerosols, vapors and hydrogen fluoride, sulfates, sulfuric acid, ammonia, and hydrochloric acid (DOE 1987; Du Pont 1994). Predicted emissions were described in detail in the construction permit filed with the state and in subsequent operating permit applications. Air exhausted from the facility was first treated using HEPA filters. Liquid effluent was subject to evaporation then was either put into saltstone and buried or discharged to a treatment plant and then to Upper Three Runs Creek, as a permitted discharge. The liquid wastes likely contained nitrates, chlorides, fluorides, and trace quantities of process solvents. The process wastes were neutralized and evaporated, and the evaporator condensate was mixed with concrete and encapsulated as a block in steel containers and buried in the burial ground. Organic wastes were placed in solvent storage tanks in the burial ground (DOE 1982). Although the information about the processes, capacities, and end products are not presented here because some of this information is sensitive, *RAC* researchers holding the appropriate clearances have reviewed and assessed this information. Releases were included in the source term estimates for the separations areas.

The Reactor Areas

The reactor areas and special reactor projects, such as HWCTR, which operated from March 1962 to December 1964, did not use large quantities of hazardous chemicals. The reactor building stacks were 200 ft high with a normal flow of 120,000 cfm. Building roof vent releases of trichloroethylene used for fuel element cleaning were reported (Reinig et al. 1973).

Each reactor area had a solvent degreaser, but the details of solvent use and waste solvent discharge are unknown. The solvent used in the reactor areas was procured through M-Area, so the M-Area inventories and materials ledgers include reactor area solvent use. The releases to the air are included in the release estimates calculated for M-Area (in Chapter 17), which were based on total inventory amounts.

After 1989, each reactor area submitted an inventory to be included with the Section 313 Hazardous Chemical Inventory Report as required by EPA's SARA Title III Regulations. A memo, Mundy (1989), submitted for the SARA regulations, listed moderate quantities of nitric acid, freon 113, sodium hydroxide, phosphoric acid, and trichloroethane for the reactor areas. The reactor areas released sulfur dioxide and nitrogen dioxide from coal burning and the use of diesel fuel burning generators. Small emissions from the reactor areas may have resulted from the use of solvents for degreasing and cleaning, the use of other cleaners and disinfectants, and the use of paint, solder, and halon fire protection systems (Radian 1992a).

G-Area

The G-Area or general area includes areas and facilities outside of other designated areas. G-Area operations include the Forest Station, the Railroad Classification Yard, the Central Sanitary

Waste Water Treatment Facility, and the Par Pond Radioecology Laboratory. Emissions from the G-Area facilities include criteria pollutants from the railroad locomotives and generators as well as water treatment chemicals and small amounts of laboratory chemicals.

CMX

CMX performed research and development and testing for the reactor areas. This included water treatment and testing of fuel assemblies, temperature, and other sensors (Crawford 1995).

TNX

TNX facilities, also called the 600 Area, handled support services and conducted research and development for the 200 Areas. Many chemicals were used in bulk quantities at TNX. Canyon processes were tested with pilot-scale plants. Research and development for the Defense Waste Processing Facility (DWPF) was conducted at TNX through the 1980s. The DWPF involved waste processing into immobile forms. The TNX facility had pilot equipment, models, and mockups of DWPF equipment. The facility vented air exhaust to a 50-ft stack. DOE (1987) stated that stack exhaust was tested in 1984 and average emissions were found to be 0.037 lb h^{-1} nitric acid, 0.00074 lb h^{-1} depleted uranium, and 0.022 lb h^{-1} benzene. We could not find a copy of the report about this testing referenced in the DOE Survey. We did not locate another mention of TNX nonradiological monitoring and data are not available on emissions from this facility to the air until the air emissions inventory was begun in 1989.

Powerhouses

As many as seven coal-fired powerhouses operated onsite and released sulfur dioxide, ash particulates, oxides of nitrogen, and other pollutants to the air. Chapter 17 addresses the powerhouse operations, off-gas systems, pollution control, and releases in the “Coal” section.

Defense Waste Processing Facility

The DWPF is a glassification plant for high activity waste, located in S-Area. Constructed in 1985, cold (nonradioactive) runs began in 1990 (Cummins et al. 1991).

Saltstone Facility

The Saltstone Facility or Saltcrete Facility in Z-Area was designed to stabilize a salt solution containing waste in concrete. Many of the chemicals currently listed in inventories onsite are used at the Saltstone Facility. The facility began test runs in 1989. The DWPF and the Saltstone Facility are not included in the historical dose reconstruction study because they began operating after 1989 and releases from these facilities are subject to more stringent, current air quality and air toxics regulations. These plants also have a Chemical and Industrial Waste Treatment Facility to treat liquid waste effluent.

Waste Handling Facilities and Disposal Sites

RAC reviewed documentation on chemical and solvent burning and disposal pits and other waste areas. Only those waste disposal sites with a potential for offsite releases of chemicals are discussed in the sections on source term determination for chemicals released to the air and water in Chapters 17 and 18.

The waste storage and disposal areas reviewed include:

- Bingham pump outage pits
- C-Area burning/rubble pit
- Central Shops burning/rubble pits
- CMP pits
- D-Area burning/rubble pits
- F-Area burning/rubble pit
- K-Area burning/rubble pit
- L-Area burning/rubble pits
- Miscellaneous chemicals basin/metal burning pit
- Metals burning pit
- P-Area burning/rubble pit
- R-Area burning/rubble pits
- 488-D ash basins
- 716-A Motor Shop seepage basin
- A-Area coal pile runoff basin
- D-Area oil seepage basin
- F-Area and H-Area seepage basins
- G-Area oil seepage basin
- K-Area reactor seepage basin
- L-Area oil and chemical basin
- M-Area process sewers
- M-Area seepage basin
- Old F-Area seepage basin
- Road A chemical basin
- Tank farm
- Hydrofluoric acid spill area.

Chemicals of concern from the waste areas include chemicals that might have volatilized into the air, or leaked into the soil and then become suspended in air, or migrated from soil into groundwater (which could have outcropped into surface water that flowed to the Savannah River). Chemicals of concern released from the F-Area, H-Area, and M-Area seepage basins; the ash disposal and coal runoff basins; the M-Area process sewers; and open burning pits and pans are discussed further in the chapters on the releases of chemicals to the air and water. The remaining waste disposal sites listed above and those described below do not appear to have caused releases of chemicals offsite, and release of chemicals from these sites do not comprise a complete exposure pathway for people offsite.

The seepage basins and disposal sites that released materials into the air are addressed in Chapter 17. Releases from the basins and sites that contaminated groundwater that outcropped to

surface water or that overflowed to surface water are discussed in Chapter 18. Although many of the basins or waste areas characterized below caused groundwater and soil contamination onsite and have required remediation, their contribution to releases to the Savannah River and air is considered negligible.

Seepage, Settling, and Retention Basins

Seepage basins are shallow earthen excavations used for disposal of wastewater containing chemicals and radionuclides. Some of the wastewater discharged to the basins evaporated while some seeped through the bottom of the basin into shallow groundwater, which in some cases traveled horizontally and discharged to surface streams. Before reaching the streams or the groundwater, contaminant levels were reduced by processes like radioactive decay, filtration, absorption on the soil, and ion exchange. Determining what fractions of contaminants that were discharged to the Site seepage basins, eventually reached surface streams, and were subsequently released to the Savannah River is a part of Phase II of the dose reconstruction study and is addressed in Chapters 5 and 18.

The history of the seepage basin operations was obtained from monthly progress reports and separations histories. For example, the Monthly Progress Report for 1964 reported that failed tubes in the reboiler at 211-F acid recovery unit necessitated the transfer of about 1.1 million gallons of 5–9% nitric acid, containing 85,000 lb of 100% nitric acid and 13 Ci of gamma activity to the F-Area seepage basins. Enough caustic to neutralize this amount of acid was sent to the seepage basins from March 8–11 (Du Pont 1964). Environmental Information Documents contain estimates of the amounts of liquid effluent and contaminants that may have been discharged to the basins. Site characterizations and remedial investigations, required by the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA), have also been helpful for characterizing releases to the basins.

The basins received waste oils, solvents, and chemicals resulting in groundwater contamination by metals and solvents. F-Area, H-Area, and M-Area seepage basins received the most chemicals. Before the Resource Conservation and Recovery Act (RCRA), most of the seepage basin monitoring effort was devoted to radioactivity. Under RCRA, basins had to be evaluated for hazardous waste criteria by sampling the basin influent and liquid. Three basins onsite have contained waste considered hazardous: 200-F for pH; 200-H for pH, mercury, and chromium; and 300-M for pH and organics. Some of the radioactive seepage basins also received chemical wastes, but most of the decontamination solutions and oily wastes were discharged to the reactor area's oil and chemical pits (Peralta and Lewis 1982).

Of interest for dose reconstruction are those seepage basins and waste disposal areas that had the potential to contribute to offsite releases. Seepage basins that contaminated groundwater, outcropped into surface streams, and eventually empty into the Savannah River are of most concern. The four seepage basins for which this applies are the F-Area and H-Area seepage basins that outcrop to Four Mile Creek; K-Area retention basin, which outcrops to Pen Branch and Indian Grave Branch; and the P-Area seepage basin, which outcrops to Steel Creek.

The Old TNX basin was evaluated because it has overflowed into a swamp. In addition, the settling basin at M-Area is of interest because large amounts of chemicals were released to this basin, including volatile organics that evaporated into the air. Coal and ash pile runoff basins

were also considered and are described in Chapter 18 with the discussion of releases of chemicals to surface water.

This chapter provides a brief characterization of the seepage basins in F-Area, H-Area, P-Area, and K-Area that have contaminated groundwater that outcrops to surface water. Chapter 18 provides a further analysis of F-Area, H-Area, and M-Area basins in the sections on releases of mercury, nitrates, chromium, and the chlorinated solvents.

Separations Area Basins. Four seepage basins and one lined retention basin are located in both F-Area and H-Area, for a total of 10 basins. In contrast to seepage basins that are unlined, retention basins are lined pits that allow radioactive decay, evaporation, and storage. Historically, the safety analysis reports for these areas have characterized the flow of cooling water, contaminated storm water, and process fluids through the separations processes and described the potential of radioactive contamination of wastewaters; however, they have not addressed nonradioactive constituents (Holcomb and Emslie 1984). The seepage basins were operated in a cascade arrangement, overflowing from one basin to the next. Liquid waste going to the basin system was proportionally sampled by a trebler sampler, and chemical analysis of some of the samples for nonradioactive metals began in 1982 (Holcomb and Emslie 1984). A special study on influent composition was done in 1975. This and other studies to characterize the release of metals and nitrates to the basins and their transport to groundwater or surface streams are compiled with the releases of chemicals to surface water in Chapter 18.

Overheads condensed from various evaporators (such as the nitric acid recovery unit and general-purpose evaporators) were the major contributors to the volume of water going to the separations areas seepage basins. Nitrates and mercury appear to be the chemical contaminants of most concern for surface water releases from the 200 Areas. The primary sources of effluent sent to the seepage basins were the nitric acid recovery unit overheads, the general-purpose evaporator overheads, waste tank farm evaporator overheads, and other evaporator overheads. The wastewater flowed to the basin through an underground pipeline into basin 1, which overflowed into basin 2, then 3 through underground pipelines. Wastewater was sampled and flow measurements were taken at trebler monitors upstream of basin 1 (Killian et al. 1987).

Groundwater beneath the F and H seepage basins areas flows toward Four Mile Creek, which is 2200 ft from the nearest basin in F-Area and 500 ft from the nearest basin in H-Area. The seep from the basins that eventually outcrops into Four Mile Creek is being evaluated as a part of the dose reconstruction because Four Mile Creek eventually empties in the Savannah River.

The F-Area and H-Area seepage basins were characterized by Christensen and Gordon (1983). The groundwater flow rate was reported to have been 0.5 ft d^{-1} in F-Area and 1.0 ft d^{-1} in H-Area. The travel time calculated from the basin to the creek was 9 years for F-Area and 4 years for H-Area. Low pH, increased conductivity, and tritium were noted along the seep line in the 1960s and were attributed to the F-Area and H-Area seepage basins (Du Pont 1967; Arnett 1993). An extensive study was conducted by SRTC to characterize the shallow groundwater that was seeping into Four Mile Creek and its associated seep line in 1988 and 1990 (Haselow et al. 1990; Arnett 1993). Further evaluation of the outcropping and chemical releases to Four Mile Creek is presented in the sections on nitrates, mercury, and chromium in Chapters 18 and 20. The F-Area and H-Area seepage basins were not used after 1988 when the Effluent Treatment Facility for the separations areas began operation (Arnett et al. 1992).

Old F-Area Seepage Basin. The first seepage basin (Building 904-49G) was built north of F-Area and received effluent from 221-F from November 1954 until May 1955. Three additional basins were constructed south of F-Area in 1955, and the 49-G basin was abandoned. It is often referred to as the Old F-Area Seepage Basin or the abandoned seepage basin (Odum et al. 1987). Chemical discharges to the basin were not recorded. Sediment core samples have shown that mercury and uranium concentrations were greater than background levels. Manganese, nitrates, lead, chromium, cadmium, and chlorinated solvents have been identified in groundwater under the basin (DOE 1987).

F-Area Seepage Basins. The PUREX process, which has always been in operation in F-Area, involved the dissolution of uranium fuel in nitric acid followed by solvent extraction. The process used three cycles of solvent extraction to separate actinides (uranium and plutonium) from each other and from fission products. The actinides were extracted from an aqueous stream to a solvent stream, scrubbed, then stripped back into a dilute nitric acid stream. High activity waste from the solvent extraction process was sent to waste tanks. Beginning in 1955, the F-Area seepage basins received wastewater from F-Area containing low level radioactivity and chemicals including chromium, mercury, nitric acid, and sodium hydroxide. The basins were taken out of service in 1988 and clay capped in 1991 (DOE 1987).

In F-Area, waste streams discharged to the basins included 1CI and 1EU evaporator overheads, hydrate evaporator overheads, general purpose evaporator overheads, laboratory waste evaporator overheads, acid recovery unit overheads, laundry wastewater, 211-F chemical drain systems, 211-F and A-Line Sumps, and A-Line pad drainage. Some of the overheads were run through a skimmer to remove organic solvents. The acid recovery unit was a fractional distillation unit used to recover nitric acid from acidic overheads generated from evaporation of wastes. The acid recovery unit bottoms, which were about 50% nitric acid, were recycled back through the canyon processes, and the overheads were discharged to the seepage basins. The overheads were typically 0.001 to 0.1 molar nitric acid with an average pH of 2.0. The general-purpose evaporators were used to concentrate a variety of waste solutions, including those from sumps, pads, catch tanks, and vessels that collect rainwater and process leaks and spills. The bottoms from these were sent to the waste tanks, and the overheads were sent to the seepage basins. The 221-F chemical drains included those in the cold-feed preparation area and all of the process and nonprocess sumps. Wastewater could be fed to the general-purpose evaporator if contaminated with radioactivity. Decontamination solutions were also probably fed into this system. Before 1983, the acid recovery overheads were sent to the seepage basins. In 1983, an attempt was made to recycle most of the overheads back into the canyon processes, which reduced the nitric acid and nitrate releases (Holcomb and Emslie 1984). During the early years, waste was sampled for radioactivity before being discharged to the seepage basin or evaporated depending on the activity. The concentrations of chemicals in many of the waste streams related to the PUREX process were nearly linearly related to the production rate. Evaporators, for example, should be directly related and the gallons of wastewater discharged were related to the amount of uranium processed. About 80 million gallons of waste liquid was discharged to the F-Area seepage basin each year (Killian et al. 1987).

The plant laundry, Building 723-F, served the entire Site. It contained five washers with capacities of 300 lb of dry clothes each, five water extractors, four dryers, two wastewater tanks in a concrete box underground, and a lint and dust collector. The wastewater tanks were discharged to the seepage basins. The laundry wastes could also be sent to the general-purpose

evaporator. The Environmental Information Document for the Old-F-Area Seepage Basin reported that laundry employees said that only water soluble detergents were used by the laundry and no other chemicals were present in the building.

H-Area Basins. H-Area basin operations began in July 1955. The HM process was similar to the PUREX process; however, much larger quantities of mercury were used in H-Area to dissolve aluminum. The evaporators and acid recovery operations were similar to F-Area, but quantities of mercury discharged in evaporator overheads were much larger than the quantities discharged to F-Area seepage basins.

From July 1955 to December 1982, the discharge to the H-Area seepage basins was estimated to have totaled 1.24 billion gallons. The trebler sampler was used to sample liquid effluent discharged to the seepage basins. Weekly composites from the trebler samplers were analyzed for mercury and chromium concentrations routinely after 1982 (Holcomb and Emslie 1984). In 1987, levels of nitrate and mercury were greater than the drinking water standards in groundwater below H-Area seepage basins. Nitrate levels were as high as 118 mg L^{-1} and mercury concentrations up to 0.0082 mg L^{-1} were measured (Mikol et al. 1988). Special studies have been done by the Site to characterize potential transport of mercury from the H-Area seepage basins. Most of the mercury was said to be accounted for in the basin soil. Chapter 20 discusses mercury monitoring and Chapter 18 presents estimates of the rate of mercury outcropping into Four Mile Creek.

K-Area Basins. Pen Branch received heat exchanger cooling water from K-Area and flow from Indian Grave Branch. Tritium migration from the K Reactor retention basin outcropped into Indian Grave Branch, which flowed into Pen Branch (Cummins et al. 1991). Tritium migration from the K-Area containment basin has been measured by weekly flow measurements and a continuous sampler in Indian Grave Branch (Gladden et al. 1985; Arnett et al. 1992). However, no mention has been found of chemicals in the water that might have migrated from the basin. In Looney et al. (1987), a chemical constituent selection study, no nonradiological constituents from K-Area basins were selected for consideration.

P-Area Basins. Liquid wastes from P-Area have been released to the seepage basins since 1978. Monitoring in Steel Creek above L-Lake indicated tritium migrated from the P-Area seepage basin to Steel Creek (Arnett 1993). Concentrations of radionuclides in Steel Creek decreased after the construction of L-Lake in 1985. After 1985, the releases first entered L-Lake, then Steel Creek (Cummins et al. 1991). Nitrate, lead, and trichloroethylene have been detected in groundwater below the P-Area basin, but significant amounts of these contaminants have not been reported in Steel Creek.

M-Area Settling Basin. The M-Area settling basin was used from about 1958 to 1982. In 1981, the Site estimated that M-Area processes had used an estimated 6500 tons of chlorinated degreasing solvents. From this quantity it was estimated that about 1000 tons may have been discharged to the sewer leading to the M-Area settling basin and about 750 tons to Tim's Branch through the A14 outfall (Christensen and Brendell 1981). After 1982, all waste went to the settling basin. In 1985, the settling basin was removed from service and wastes were sent through a treatment facility. Lead, manganese nitrates, and solvents reached the groundwater below the settling basin. Concentrations of organics in groundwater have exceeded 250 mg L^{-1} tetrachloroethylene and 150 mg L^{-1} trichloroethylene (DOE 1987).

Old TNX Seepage Basin. The old TNX seepage basin was operated from 1958 to 1980. The basin received process water from tests conducted at TNX in support of the separations areas and

the defense waste processing facility, including discharges of uranyl nitrate and mercury nitrate (Simmons et al. 1985). In 1981, the basin was closed, backfilled, and capped with clay (Simmons et al. 1985). Basin sediments contained chromium, uranium, nickel, mercury, and a number of radionuclides (DOE 1987). Chromium, mercury, nickel, and nitrates have been detected in groundwater below the basin. Inventory calculations estimate about 318 kg of mercury may have entered the basin (Simmons et al. 1985; DOE 1987). Another estimate for mercury discharged to the old TNX seepage basin was 292 kg (Zeigler and Lawrimore 1987).

Chemicals and radionuclides from the old TNX seepage basin have been released to a swampy area located adjacent to and below the basin, and the basin itself was referred to as a swamp in some documents. There has been a loss of forest canopy in this swamp area from the discharges. Mercury is well dispersed in the swamp and has been detected in the vegetation. The swamp water was reported to contain 50 times the drinking water standard for mercury (Zeigler and Lawrimore 1987). The swamp sediments were characterized in 1984 and chromium, mercury, and uranium were found in the top 0.6 m of sediment. Chromium and mercury were found in the swamp vegetation at levels greater than 10 ppm and 3000 ppm, respectively (Simmons et al. 1985; DOE 1987). However, the swamp water and sediments have not been transported offsite.

Material in the basins was pumped into Upper Three Runs Creek on two occasions: once in 1955 and once in 1965. In 1969 and 1970, this basin received etching solution wastes (Odum et al. 1987).

Savannah River Laboratory Seepage Basins. The Savannah River Laboratory seepage basins consist of four earthen basins. Two have received waste since July 1954, one was added in 1958, and one was added in 1960. All of the basins were inactive in October 1982. Most of the waste was laboratory waste, which was pH adjusted before release to the basins. The basins received low-level waste generated in laboratories in 735-A and 773-A. Pipes transferred wastes from building drains to underground tanks. The liquid waste was sampled for radioactivity, and if the levels were sufficiently low, it was discharged to the basin (Looney et al. 1987; Fowler et al. 1987).

Soil cores suggest that relatively small, 10–350 kg amounts of chromium, mercury, manganese, uranium, arsenic, and nitrates have been released to the basin. Christensen and Gorden (1983) suggested that chlorinated solvents were disposed of through drains leading to the basins, but later reports with more detailed history of laboratory practices suggest that significant amounts of chlorinated organics were not sent to the basins. Estimates of solvent release also included freons, which Fowler et al. (1987) said were “improperly assumed to be solvents such as trichloroethylene.” Fowler et al. believed former estimates of solvent discharged mistakenly included other materials and were overestimates. Fowler et al. came to this conclusion after investigating historical handling practices, which involved evaluating disposal records, reviewing waste handling procedures for the SRL, interviewing SRL staff, and assessing data in previous documents used in making various estimates. Most of the solvent used was trichloroethylene in the fabrication laboratory. None of the metals would have been expected to have been released to the air from the seepage basin (Fowler et al. 1992; DOE 1992).

Metals Burning Pit and Miscellaneous Chemical Basin. The metals burning pit and miscellaneous chemical basins were located 1.5 mi south of A-Area and M-Area, 3 mi east of the SRS boundary. The basins were used for liquid wastes from 1956–1974. There are no records of the materials disposed of at this site, but volatile organics have been found in the soil (Arnett

1993; DOE 1987). Some information was compiled for reports about the impact and cost of closure (Muska and Pickett 1985). The Metals Burning Pit was in service from 1960–1974. The pit was used to burn waste and scrap from A-Area and M-Area operations. The waste in the pit consisted mainly of solid lithium and aluminum waste materials generated by the metal finishing operations in M-Area (Muska and Pickett 1985). Metals, especially lithium and aluminum, and chlorinated solvents were the contaminants of concern for cleanup of these areas (Westinghouse 1992). It is thought that barrels of waste solvents, primarily tetrachloroethylene, were emptied at the basin then the empty containers were disposed of in the pit (DOE 1987). The metals and other waste deposited in the pit were burned periodically. In addition, miscellaneous chemical wastes were placed in a small 6 × 6-m basin within the metals burning pit area. The pit was backfilled and graded in 1974. No inorganic chemicals or metals have been detected in monitoring wells, but trichloroethylene has been found at relatively low levels in two of the wells (Muska and Pickett 1985). Soil gas analysis indicated that high levels of organic solvents were present and trichloroethylene was detected in the groundwater (Westinghouse 1989). Trichloroethylene and tetrachloroethylene and a breakdown product, 1,2,-dichloroethylene, have been detected in soil (DOE 1987).

716-A Motor Shop Seepage Basin. The 716-A Motor Shop seepage basin was used from 1977 to 1983 for the disposal of oils, degreasers, and miscellaneous waste. Solvents and metals are present in the basin soil (Westinghouse 1992).

D-Area Oil Seepage Basin. The D-Area oil seepage basin operated from 1952 until 1975. In 1973, when the plant stopped open burning in a number of areas around the Site, the waste going to the basin increased. Numerous 55-gal drums were believed to have been disposed of in the basin (Westinghouse 1992).

Huber et al. (1987) stated that the D-Area waste oil seepage basin accepted things that could not be burned by the powerhouse. We can assume this may have included halogenated compounds and polychlorinated biphenyl-contaminated oils during the time period 1952–1975. In 1975, the basin was backfilled (Westinghouse 1992).

F-Area and H-Area Retention Basins. The F-Area and H-Area retention basins were used from 1955 to 1973. These were open, unlined basins for emergency storage of contaminated cooling water. Only trace quantities of chemicals were thought to have been discharged to the basins (Westinghouse 1992).

G-Area Oil Seepage Basin. The G-Area oil seepage basin took liquid wastes from 1951 to sometime in the 1960s. The exact dates of operations and the nature of the liquids disposed of are unknown. Lead, chromium, chlordane, and solvents have been detected in basin soil and sediment (Westinghouse 1992).

L-Area Oil and Chemical Basin. The L-Area oil and chemical basin was used to dispose of oils and small volumes of other, miscellaneous liquid wastes from 1961 to 1979. Metals, trichloroethylene, and tetrachloroethylene have been found in the basin soils (Westinghouse 1992).

Based on groundwater and sludge and soil concentrations, Looney et al. (1987) estimated that 3 kg of cadmium, 500 kg of chromium, 50 kg of lead, 0.3 kg of mercury, 28 kg of nickel, and 1.4 kg of tetrachloroethylene had been discharged to the basin.

Road A Chemical Basin. Road A chemical basin soils contained trichloroethylene and perchloroethylene. The dates of operation and wastes disposed of were not recorded. This basin

was closed and backfilled in 1973 (Westinghouse 1992). Looney et al. (1987) estimated that about 14 kg of lead has been discharged to the basin.

Acid/Caustic Retention Basins. The acid/caustic retention basins were constructed in the early and mid-1950s in F-Area, H-Area, K-Area, L-Area, P-Area, and R-Area. They are unlined earthen pits that received dilute sulfuric acid and sodium hydroxide solutions used to regenerate ion-exchange units used in the water purification processes for the reactors and separations areas. In addition, steam condensate and runoff from spill containment areas were discharged to these basins. The basins in R-Area and L-Area were closed in the 1960s. Basins in the other areas were used into the 1980s. The basins were taken out of service in 1982 and replaced with neutralization systems. Groundwater contamination under these basins suggests that metals and halogens were also discharged to the basins (DOE 1987).

New TNX Seepage Basin. The new TNX seepage basin has been used to dispose of waste solutions containing chromium, lead, and a large amount of nitrate (Looney et al. 1987).
Ford Building Seepage Basin. The Ford Building seepage basin was constructed in 1964. It received wastewater containing surfactants, oils, and grease from many different sources. Levels of chromium, nickel, and solvents below the drinking water standards were found in groundwater near the basin (DOE 1987).

Metallurgical Laboratory Seepage Basin. The Metallurgical Laboratory seepage basin received wastewater effluent from the Metallurgical Laboratory from 1956 to 1985. Small quantities, usually less than 10 gal d^{-1}, and noncontact cooling water, about 900 gal d^{-1}, were also discharged. The basin was closed and filled in 1992 (Arnett 1993). Based on soil core, groundwater, and process data, Looney et al. (1987) estimated that 4 kg of chromium, 4 kg of lead, 0.05 kg of mercury, and 250 kg of trichloroethane may have been disposed of in this basin over the years.

Other Disposal Pits and Waste Sites

Chemical, Metal, and Pesticide Pits. The Chemical, Metal and Pesticide (CMP) pits were seven trenches, 8 to 10 ft deep, used from 1971–1979 or 1980 (Anonymous 1981; Beckwith 1983). The pits were located on a hill near L-Area, about 7 mi from the nearest plant boundary and about 1200 ft from the nearest surface water outcrop into Pen Branch Creek. Four pits were dedicated for disposal of chemicals, (18G, 18.1G, 18.2G, and 18.3G); two for pesticides (17G and 17.1G); and one for metals (19G). Waste disposal records were kept from 1977 to 1979. No record system was used before 1977, and records of what was disposed of after 1977 were said to be very incomplete (Beckwith 1983). Trichloroethylene, tetrachloroethylene, lithium nitrate, and hydrazine were listed in some of the records (Anonymous 1981). Waste was dumped into the pits by truck. Some of the documentation for remediation (Anonymous 1981) said that, “many containers of chemical wastes were shot by patrol after disposition in pits to absorb chemicals into the soil.” Solvent drums were stacked by dropping them in place and some were said to have been leaking. Pickett et al. (1987) found that little qualitative or quantitative data on the waste disposed of were available. Scott et al. (1987) listed 71 contaminants of potential concern in the pits and concluded that Silvex and toxaphene were the contaminants of greatest concern. Waste disposal records indicate that more than 9550 kg of trichloroethylene and more than 24,100 kg of tetrachloroethylene were buried in the CMP pits. Soil cores taken adjacent to the pits contain 30 ppm tetrachloroethylene and 3 ppm trichloroethylene. Shallow groundwater near the pits

indicates that these solvents have migrated through the soil. However, results of groundwater sampling in the early 1980s suggest that contamination of groundwater by pesticides stored in the pits had not occurred (Beckwith 1983). Yet another document reported that diedrin was detected in groundwater near the pits at concentrations as high as 0.22 μg L^{-1} in 1979 and at a concentration of 0.08 μg L^{-1} in 1981. Lindane and chlordane were detected at low levels in 1981. Details of how this sampling was conducted and pesticides that may have been analyzed for but were not detected are not given (Anonymous 1981). The pits were excavated in 1983 and 1984, and about 75,000 ft^3 of soil was removed, put into drums, and was said, at that time, to be awaiting incineration. The pits then were covered and bermed to protect the soil from rainwater runoff (Beckwith 1983).

DOE (1987) estimated that about 50,000 kg of solvents (most likely tetrachloroethylene, trichloroethylene, and freon) and 50,000 kg of oils were excavated and 250 to 290 kg remained after the removal. Concentrations of DDT, lindane, chlordane, endrin, toxaphene, and methoxychlor were detected in soils excavated from the CMP pits (DOE 1987). There is not enough information on the quantities or disposal practices to estimate how much of these materials could have been released into the air. There is no indication that burial of chemicals at the CMP pits resulted in releases that may have traveled offsite.

Radioactive Waste Burial Ground. Contaminants in the groundwater at the Radioactive Waste Burial Ground include mercury, lead, cadmium, and chlorinated solvents, but the groundwater has not moved offsite (Looney et al. 1987; DOE 1987). Mercury releases from the burial grounds are addressed further in the section on releases of mercury to the air.

Bingham Pump Outage Pits. The Bingham pump outage pits were unlined earthen pits that received equipment removed during the Bingham pump shutdowns in 1957 and 1958 while modifications were made to the reactor's primary and secondary cooling systems. One pit was located in K-Area, one in P-Area, two in L-Area, and three in R-Area (Westinghouse 1992).

Silverton Road Waste Site. The Silverton Road waste site received drums, tanks, and metal shavings. In 1987, estimates of disposal amounts included 14 kg of lead, 5 kg of tetrachloroethylene, and 42 kg of trichloroethylene (Looney et al. 1987). The site was closed in 1974 (DOE 1987). In 1983, the groundwater at the Silverton Road site was found to contain measurable concentrations of chromium, manganese, lead, zinc, nitrates, and chlorinated organics (Ziegler et al. 1985).

Tank 16. Tank 16 in the H-Area High Level Waste tank farm developed a below ground leak that overflowed subsurface containment structures. Liquid waste containing mercury, chromium, and lead leaked from the tank. The quantities of these metals in the soil were said to be relatively small and radioactivity was the primary concern (Westinghouse 1992).

Hydrofluoric Acid Spill Area. The hydrofluoric acid spill area is located in the southwest part of the Central Shops Area. It is uncertain whether acid was spilled or if acid-contaminated soil or acid-filled containers were buried at the site. The spill or disposal occurred sometime before 1970. Records and interviews with former and current employees regarding the history of this waste unit, conducted for the remedial investigation, were said to have been inconclusive (Westinghouse 1992). Fluoride and lead have been found in the groundwater (Huber and Bledsoe 1987; Looney et al. 1987).

M-Area West. M-Area West consists of two small areas where empty drums were found. Markings on the drums indicated they once contained chlorinated solvents and were about 30 years old (Westinghouse 1992).

TNX Burying Ground. The TNX Burying Ground was used for material contaminated or damaged during the 1953 evaporator explosion. Nitrates and uranyl nitrate were contaminants of concern for this area (Westinghouse 1992).

Burning Rubble Pits. The burning rubble pits are described in more detail in the Chapter 17 section discussing incineration and burning.

SRL Oil Test Site. The SRL oil test site disposed of paint thinner, hydraulic fluid, and waste-cutting contaminated with heavy metals (Looney et al. 1987).

Chemical Treatment of Water

Well water and Savannah River water has been treated before use for cooling water, drinking water, and other uses. The water is subject to sedimentation, filtration, and pH adjustment. In D-Area, precipitates from the sedimentation process and filter backwash was pumped to the D-Area ash basin. In the 1980s, drinking water was treated with polyphosphate and chlorinated. Of more concern for chemical releases was the chromium, biocides, and other chemicals added to very large quantities of cooling and process water. Process and cooling water for the reactor areas was pumped from the river into the 186-Basin reservoirs. Water to be used for processes and cooling was chlorinated, and water used for the boilers in the powerhouses was demineralized. The demineralized regenerants were neutralized and discharged to the ash basin. All domestic well water was chlorinated and in some areas it was also degassed and pH adjusted. M-Area used a granular activated carbon filter to adsorb chlorinated organics from the groundwater before its use (DOE 1987).

Corrosion inhibiting chemicals, water treatment chemicals, and biocides were added to maintain chemistry control of plant water and to prevent corrosion and proliferation of algae and bacteria in cooling systems. Most of the water flowing down the creeks was pumped from the Savannah River and treated with alum and lime as flocculants, chlorine as a biocide, sodium sulfite, and phosphates; used for cooling or process water; and then discharged. An essential materials listing for water treatment chemicals estimated the monthly consumption from 1951 to September 1954, characterized by area. The specifications for purchasing were also given in this document (Du Pont 1954). Alum, hydrate lime, slaked lime, sulfuric acid, 50% caustic, clay, sodium sulfite, trisodium phosphate, chlorine, tannin, and sodium silicate were listed. For example, reported values for chlorine are summarized in Table 15-2. This suggests that a total of about 120 ton mo^{-1} of chlorine was used in 1954, more than 80% of this for water clarification in the reactor areas (Du Pont 1954).

The only water treatment chemical to come out of the ranking described in Chapter 16 was chromium, which is carcinogenic. The air emissions inventory included cooling tower drift emission estimates for chromium for the 185-K towers for 1985–1990. These estimates were made using groundwater quality data and EPA's AP-42 emission factor for drift loss as a function of cooling water circulation rate. Adequate records on cooling water flows were not available, so cooling water requirements of similar systems were reportedly used.

Table 15-2. Chlorine Use for Water Treatment in 1954

Area	ppm in water	Ton mo^{-1} range	Ton mo^{-1}average	Use
A	20,000		2.7	Cooling tower
A	500	0.09–0.18	0.13	Domestic
D	60,000	5.4–10.8	8.1	River water
D	18,500		2.5	Cooling tower
D	500	0.09–0.18	0.13	Domestic
RPLKC	80,000	43–144	100	Clarification
RPLKC	27,500		3.7	Cooling tower
RPLKC	500	0.09–0.18	0.13	Domestic
FH	3,500	0.31–0.63	.47	Clarification
FH	15,000		2.03	Cooling tower
FH	500	0.09–0.18	0.13	Domestic

Accidental Releases of Chemicals

RAC reviewed documentation on spills, explosions, and fires. The accidental releases involving the chemicals of concern are summarized in the section for each chemical in Chapters 17 and 18.

Explosions and Fires

The presence of an organic material with nitric acid at high temperature can result in a rapid exothermic nitration of the organic material, which is a reaction called a red-oil explosion. Two such explosions have occurred at the SRS. In 1975, a chemical explosion and fire occurred in the A-Line of F-Area, a result of the contamination of uranyl nitrate solution with process solvent (tri butylphosphate). Reports indicate that no contamination was spread outside the fenced A-Line facility (Bebbington 1990; McKibben 1976). On January 12, 1953, an explosive chemical reaction occurred in evaporator building 678-G in the CMX Area. Uranium dust was a worker safety concern (Du Pont 1971). These accidents were described in detail in Durant (1983).

The 200-F and H-Area have sustained about 500 fires, many of which are described in safety analysis reports or incident reports. Solvent fires and red-oil explosions caused by the nitration of extraction solvent were two of the industrial hazards addressed in the separations area safety analysis reports (Fisk and Durant 1987).

Chemical Spills

In addition to routine releases during processing, chemicals have been released to the environment because of inadvertent leaks, transfer errors, spills, overflows, and uncontrolled reactions. We reviewed records of spills involving chemicals of concern and the amounts released were included in the source term estimates. Since the mid-1980s, any spills of toxic materials must be reported to the EPA and the State of South Carolina. In the 1970s, many of the spills were reported in the fault tree databanks. A number of minor spills involving relatively small quantities are compiled in annual reports, monthly reports, and fault tree databank records. For

example, there have been 23 transfer errors reported in the H-Area outside facilities since 1962. Most of the spills reported in the 1970s and 1980s involved fuel oil, sodium hydroxide, and acids used in bulk quantities, usually involving loss to seepage basins when valves were accidentally left open. The Fault Tree Data Storage and Retrieval System lists incidents, occurrences, failures, maintenance problems, etc. (McCulloch 1980). It appears that most of the leaks, spills, and accidents reports resulted in contamination in buildings rather than release to Site streams. There was mention of leaks to sidewalks and ground, but they all involved radionuclides. Reporting of chemical spills before 1980 was uncommon. Inadvertent additions of materials to seepage basins were usually reported in monthly reports for the separations areas. When a leak in a solvent tank or a spill during transfer of a chemical occurred, often the name of the chemical was not reported and the amount released was not mentioned. Only the amount of radioactivity released was reported, which is an example of the concern over radionuclides and lack of concern about reporting chemical releases.

In the 1980s and 1990s, spill incidents and their containment and cleanup had to be reported to the EPA and State. We reviewed the documentation for these reports, and Chapter 18 summarizes information on spills of chromium-treated water, mercury, sulfuric acid and hydrogen sulfide, and chlorinated solvents. Most of the spills influenced surface water. Some of the more notable spills and leaks are described in the section on chemical releases to surface water. The end of Chapter 18 summarizes spills to Beaver Dam Creek, Site-wide oil spills, and fish kills.

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CHAPTER 16

RANKING AND SELECTION OF CHEMICALS OF CONCERN

ABSTRACT

This chapter describes the ranking method used to determine which chemicals were to be evaluated during Phase II of the Savannah River Site (SRS) dose reconstruction project. The method involved comparing amounts of chemicals that could have been released to the environment in a worse-case situation to those in air or water that would not be expected to cause adverse health effects. The comparison resulted in a ratio that indicated whether there was a sufficient quantity of a chemical onsite to have presented a hazard. From these ratios, we concluded that is was important to derive a source term that described how much, when, where, and in what form the following chemicals were released from the Site: arsenic, benzene, cadmium, chromium, coal and coal ash, hydrogen sulfide, hydrazine, lead, manganese, mercury, nickel, nitric acid, nitrogen dioxide, sulfur dioxide, and zinc. Chapters 17 and 18 describe the estimates of the amount of chemicals released to the air and surface water and the information used to develop them. Chapters 19 and 20 describe additional environmental and effluent monitoring information used to develop or support chemical release estimates.

PURPOSE

Thousands of chemicals have been used at the SRS over the years., We used a ranking method to determine which chemicals were important to evaluate for Phase II of the dose reconstruction study because they may have been discharged to the air or water in quantities that could have posed an offsite hazard.

We found documents and records that describe how chemicals were used, stored, purchased, and disposed of as a part of specific operations. We used essential materials lists, purchasing records, and inventories to try to determine what chemicals were onsite, when, and in what quantities. From this information, we developed a list of potential chemicals of concern and subjected the chemicals to a ranking exercise.

We determined a ratio for each chemical that compared an approximation of the amount of chemical that may have been in the environment to the concentration in water and air that was not expected to cause adverse health effects. To develop the ratios, we derived the amounts of chemicals that may have been in the environment from inventory amounts using conservative release fractions and air dispersion and water dilution factors. We used the term toxic levels of concern or toxicity values to refer to the threshold concentrations in water and air that were not expected to cause adverse health effects. The ratio indicates whether there was a sufficient quantity of a chemical onsite to have presented a hazard. The more toxic or carcinogenic a substance, the smaller the amount dispersed or released needed to exceed toxic levels of concern. These ratios involved several very conservative assumptions and represent a worse-case or upper-bound situation.

RANKING METHOD

Inventory Information

A list of potential chemicals of concern was compiled primarily from three sources: (1) a list of 34 essential materials in a *Survey of Effluent and Environmental Monitoring at the SRP*, from 1973 (Reinig et al. 1973), (2) the *Company Chemical Inventory (CCI) Report* compiled by Du Pont in the 1970s (Du Pont 1979), and (3) the 1994 *Chemical Information and Inventory System Database* of materials onsite (Information Systems Engineering 1994). The latter two inventories were developed to comply with U.S. Environmental Protection Agency (EPA) reporting and listing requirements and are both described in more detail in the Phase I Task 3 Report (Meyer et al. 1995) and below. In addition, we used lists of materials in safety analysis documents and other documents that described processes and operations. Materials listed in the SRS Ground Water Monitoring Program's quarterly report (Westinghouse 1994) as being found above levels of concern in groundwater were also included.

The May 1959 Monthly Progress Report from the Works Technical Department reported that in response to a request by the Atomic Energy Commission Division of Biology and Medicine, a survey of potentially hazardous materials used in bulk quantities at SRS was made. The survey was said to identify a total of 48 potentially hazardous materials on the plant. The monthly report stated that these survey results were issued as a report (Du Pont 1959). Tom Cavenaugh and the SRS Central Records staff conducted a search of the central records and the PINT database for the 1959 report during Phase I in 1994 when mention of the survey was first found in a monthly report. They were unable to find a report of the survey

Radiological Assessments Corporation (RAC) team members interviewed many former and current Site workers with knowledge of chemical usage, receipts, and records in Phases I and II. They agreed that purchasing records would not be useful and purchase order and related records would not have been retained far enough back in time to be useful. Some essential materials ledgers were found and were used to develop the list of chemicals of potential concern. Inventory amounts were all converted from pounds, tons, or grams per week or month to kilograms per year using 2000 lb ton^{-1}, 0.4536 lb kg^{-1}, 1000 g kg^{-1}, 12 mo y^{-1}, and 52 wk y^{-1} conversions.

The list of materials in the 1973 *Survey of Effluent and Environmental Monitoring at the SRP* included estimates of the amount of material used each month (Reinig et al. 1973). These estimates provided the largest amounts listed in any of the inventories found, for several materials.

Du Pont developed an index of chemicals at the SRS, called the *CCI Report* (Du Pont 1979), in the mid-1970s to comply with the Toxic Substances Control Act. The index was a list of about 400 chemicals that were present onsite. A printout of the index can be found in Appendix A of the Task 3 Report for Phase I (Meyer et al. 1995). The inventory cataloged each chemical by name; registration number; jurisdiction values (whether the material is in a published inventory, reportable, user exempt, etc.); quantity; and date the material was added and/or deleted from the inventory. Most of the chemicals were identified as a chemical synthesized at the Site, imported onsite, a process chemical, a support substance that does not end up in the commercial product, an intermediate chemical, a laboratory chemical, a nonwaste impurity, a waste by-product, or a research chemical. Quantities were given by letter designations corresponding to ranges of less than 454 kg; 454 to 4540 kg; 4540 to 45,400 kg; 45,400 to 454,000 kg; 454,000 to 4.54 million

kg; 4.54 to 22.7 million kg; 22.7 to 45.4 million kg; 45.4 to 227 million kg; 227 to 454 million kg; over 454 million kg; and quantity not reported or reported under another synonym. For the ranking, the upper bound of the corresponding range was used as the inventory amounts for materials. 'Quantity not reported' was the most common entry. The Du Pont inventory contained quantity entries for only 65 chemicals, which accounted for about 15% of the chemicals listed. Of these, seven chemicals (copper, dioctyl phthalate, gadolinium nitrate, lead, mercury, nickel and nickel sulfate, and uranium) were listed as being present in quantities less than the quantities reported in the Chemical Information and Inventory System (CIIS) database. Amounts for five chemicals (cadmium, ceric ammonium nitrate, ferrous sulfamate, phosphoric acid, and potassium permanganate) in the Du Pont inventory were consistent with the CIIS database. Quantities of nine chemicals (hydrazine, hydrofluoric acid, hydrogen sulfide, hydroxylamine nitrate, hydroxylamine sulfate, manganese nitrate, napthalene, oxalic acid, and sodium hydroxide) were listed in the Du Pont database with quantities greater than CIIS database amounts. The 44 chemicals presented below were listed with quantities in the Du Pont database, but they were not included on the list to be ranked because, for various reasons, they are very unlikely to present an offsite hazard.

Acetylene	Aluminum	Aluminum nitrate
Aluminum sulfate	Ammonium sulfamate	Aquadag
Ascorbic acid	Calcium	Calcium fluoride
Several paints, thinners and primers	Calgon	Carbon
Calcium oxide	Dodecane	Ethylene glycol
Formic acid	Gelatin	Glucose
Helium	Iron	Methane
Nitrogen	Oxygen	Polyethyleneimine
Propane	Sodium aluminum silicate	Sodium carbonate
Sodium hypochloride	Sodium nitrate	Sodium nitrite
Sodium salicylate	Sodium sulfite	Steel
Sucrose	Urea	

The CIIS database, maintained by Westinghouse SRS, is a comprehensive listing of all hazardous materials used or stored in the workplace. The database was designed to fulfill the requirements of Worker-Right-To-Know legislation and to help organize inventory data for EPA reporting requirements. It is an ORACLE relational database that runs on the VAX computer. Information in the database is derived from material safety data sheets (MSDSs) that are also stored in hard copy form in notebooks. The database is designed to be updated each year and to provide information for annual reports. The SRS CIIS database is not historical but contains materials currently inventoried onsite. It does not provide specific information on toxicity, use, monitoring information, or time periods of use.

Two products were sought from the CIIS. Cheryl Hardy and John Harris, Westinghouse SRS Environmental Protection Department personnel, selectively extracted this information from the database. One product was a list of all of the chemicals in the database with a Chemical Abstract Service (CAS) number. The material name, CAS number, average amount, and number of hits (number of times the material is reported onsite) were printed for 1994. A list of about 4000

chemicals resulted. This list is printed in Phase I Task 3 report (Meyer et. al. 1995). The CIIS list appeared to be the most complete listing of chemicals. It seemed prudent to start with a comprehensive list and eliminate materials not of concern for dose reconstruction. Although some of the chemicals used onsite at one time may not have been stored onsite in 1994, many chemicals used historically were included in this large database. The second product was the result of several data extractions. The resulting database of chemicals was transferred to *Radiological Assessments Corporation* (*RAC)* on disk, converted to a FoxPro format, and incorporated into the *RAC* SRSCHEM database developed for Phase I. This database included information on the chemical name, manufacturer synonyms, symbols, quantities reported, and uses onsite.

The following fields were entered into FoxPro for about 30,000 records:

MSDS number
CAS number
Material name
Manufacturer
Form (liquid, gas, solid, or mixture)
Maximum amount (onsite in 1994)
Average amount (onsite in 1994)
Synonyms
Formula
EPA storage code

A	Above ground tank	H	Silo
B	Below ground tank	I	Fiberdrum
C	Tank in a building	J	Bag
D	Steel drum	K	Box
E	Plastic or nonmetallic drum	L	Cylinder
F	Can	M	Glass bottle
G	Carboy	N	Plastic bottle

Usage code

1	Import
2	Byproduct
3	Impurity
4	Reactant
5	Repackaged
6	Laboratory
7	Manufacturing aid
8	Processing aid
9	Other use.

The department, area, building, and room where the chemical was currently used or stored could have been extracted from the CIIS. However, because many of the release point locations were classified and the ‘area’ provided sufficient information about the location of the release, this information was not included to expedite the security review.

Several subsequent extractions of this database were conducted. One extraction targeted all chemicals on EPA lists and other lists including, Superfund Amendments and Reauthorization Act (SARA) Title III; Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA); Resource Conservation and Recovery Act (RCRA), Toxic Substances Control Act (TSCA); and a list of reproductive hazards. The extractions excluded chemicals with no hazard rating (no acute toxicity, chronic toxicity, flammability or reproductive hazard). We made a second extraction of all chemicals in quantities greater than 50 lb. This, we felt, captured all of the chemicals that might be of concern based on toxicity and quantity. We then conducted a line-by-line review of the database to delete materials that were unlikely to present an offsite hazard, such as concrete, plastic materials, and solid building materials. Records that involved the following materials and/or synonyms for them were deleted:

Joy dishwashing	Markers	Crayons
Glass cleaner	Copier	Typewriter
Glue	Analytical standard	Deep Woods Off
Paint	Correction fluid	Ajax
Welding rod	Battery	Comet
Toner	Air freshener	Carpet
Saddle Soap	Ink	Crayons, pens
Mounting solutions	GC column packing	Mortar
Weatherstrip	Cutting oils	Enamels
Adhesives	Polyurethane	Shellacs
Resins	Welding wire	Salt solutions
Cements	Sealants	Buffer solutions
Lubricants	Oil absorbants	Gaskets
Insect repellents	Spill absorbents	Office supplies
Small volume cleaners	Abrasives	Laboratory supplies
Steel	Pump oils	Grease
Cast iron	Art supplies	Tape
Deodorizers	Acetylsalicylic acid	Solder and flux
Inert solids	Sand and aggregate	Scintillation fluids
Salts	Insulation	Lotions

We then reviewed the list of deleted items line-by-line and added back in items that could not be identified by name (for example ‘fuel additive number 13,’ or ‘accelerator SFA342’). A total of 168 materials were listed in the database by trade name and with insufficient manufacturer or synonym information to allow identification. A list of these was sent to Dick Reynolds, Manager of the Chemical Commodity Group at SRS, for further identification. The task of looking at these materials was assigned to Mark Lloyd, who provided synonym, carcinogenicity, or reproductive hazard (yes or no) information and the hazardous constituents of each material if known. Based on this information, trade name materials were deleted, retained as discrete materials, or combined with and listed under their most hazardous component.

The following groups of materials were removed from the list of chemicals of potential concern. The following materials contain noncarcinogenic components and relatively nontoxic components or ingredients:

Mod U Formula	Crimstar 30
CP 220 ChilWet	Solucom Clear
Cronwear Eagle	Electrostatic solution
Apiezon Q Compound	Mojave
Inhibitor 526C	Snapback
Magicfloc 985N	Corrosion Inhibitor GC Formula
Sediperse All	Drewguard 315
Polysperse Plus	Monosodium titinate slurry
Bycothane 300	Formed Molecular Sieve
Durapox Primer	CI Mastic
Industrial Grade PVC solvent	Paraffin Wax

The following are relatively nontoxic and are used as food additives, medicines, anesthetics:

Sodium nitrate	Sodium nitrite
Potassium nitrate	Polyethylene glycols
Propylene glycol	Phospholene
Sodium sulfite	Fumaric acid
Hexane	Boric acid
Trisodium phosphate	Sodium sulfite
Sodium phosphate	Nitrous oxide
Ascorbic acid	

The following materials are used in office equipment, primarily copiers. Some of these materials contain usually less than 1% of formaldehyde, which is a carcinogen:

Cronar reducer	Itek activator
Cronolith liquid blender	

The following materials contain components that are not chronic hazards, have relatively low acute toxicity, and are not carcinogenic:

Doubleteam
Alconox cleaner (sulfonate)
Cryosan (bleach)
Chloroethane
Calcium
Barium
Graphite
Paraffin
Chloride
Phosphate
Silver
Calcium hydroxide
DPD free Cl reagent
(disodium phosphate)
Synthetic sludge
Exotherm (calcium chloride)
Sulfamic acid
Biphenyl
Cerium
Hydrogen peroxide
Oils and lubricants
Aluminum
Iron
Selenium
Silver nitrate
Butyl stearate

The following materials contain components that are not carcinogenic but are sensitizers or irritants:

Rynothane activator
(diisocyanate)
Fortify (acrylic polymers 11–25%)
HitCZO Part A (diglycidyl resorcinal ether)
R process gum (<1% formaldehyde)
diethyl hexyl phosphoric acid
Duraskid
Phosphates
Lanthanum
Terphenyls
Polyethyleneamine
LTC accelerator
ChemTreat
Concresive liquid hardener
(polyaminoamide)
Virchem 931 hardener
Scotchcast (vinylcyclohexanedioxide)
Diphenyl methane diisocyante
Cloro-m-cresol
Epon
Neodynium oxide
Polyphosphates
Erbium oxide
Saframine
CreteLease

The database contained many paints, paint thinners, and epoxies that may be hazardous in a liquid form but once dried would not be expected to pose an offsite hazard. These materials may have been stored in large amounts, but they were likely to have been used in relatively small amounts. Although resins and enamels may contain toxic components, they are generally used for coating and painting structures and equipment and would be found in a polymerized state or stored as active ingredients, separated in small quantities. Epoxy components known to be potent sensitizers in occupational settings were listed above as sensitizers.

In another data extraction, substances with maximum annual inventory amounts of less than 0.25 lb were deleted. This extraction eliminated the thousands of hazardous materials that were used in very small volumes, such as laboratory reagents, analytical standards, or samples. These materials would have been present and used in such small amounts that they would not pose an offsite hazard. The cut off of 0.25 lb was chosen from the results of an assessment of a hypothetical release of TCDD, the most potent carcinogen and developmental toxin ever evaluated by the EPA. The proposed cancer potency factor for TCDD yields a daily intake level

corresponding to a 10^{-6} cancer risk level of 0.01 picograms per kilogram per day (pg/kg/day). In this assessment, we attempted to back calculate an amount that if released over a very short time, would be low enough not to be harmful. If 100 g (0.22 lb) of a chemical was released at one time and a conservative dispersion factor (χ/Q) of 10^{-5} sec m^{-3} was used, the resulting exposure would correspond to a maximum daily intake of about of 0.00065 pg/kg/day. This value is about 15 times less than the daily intake level for a 10^{-6} cancer risk. This justifies 0.25 lb as a very conservative cut off amount. For similar exercises, others have chosen 1 or 0.5 lb, which are also justifiable limits. Using 0.25 lb seemed to accomplish our goal of eliminating most of the chemicals used in very small amounts.

Again, the list of deleted materials was reviewed line-by-line and exceptionally potent carcinogens (such as some of the polyaromatic hydrocarbons and pesticides that were listed with inventory amounts of less than zero but were likely to have been used onsite in the past) were added back to the list. Many of the materials remaining on the list with inventory amounts of less than 0.25 lb were not derived from the CIIS database. They were taken from essential material lists or the list of groundwater contaminants of concern, so they were not eliminated as a result of this data extraction.

Inventory amounts were not available for all materials. Many materials in the CIIS database had a zero inventory amount. The staff at Westinghouse SRS who maintained the database in 1994 believed that the zero amounts on our ranking list were a result of less than 0.001 lb being reported and rounded to zero. Benzo(a)anthracene, benzo(a)pyrene, benzofluoranthene, benzoperylene, and chrysene were listed in the database with inventory quantities of zero or less than 0.001. These were probably used as analytical standards and were evaluated using a quantity of 0.001.

Gasoline and diesel fuels were added back to the list and were included in the ranking because they may have leaked from tanks into soil and groundwater. A large amount of developer was currently and has been historically used at the Site. Photographic developer was used in the Site's photo labs to develop photos and x-rays of welds. Developer and toner were also used in all of the copy machines and in other equipment onsite and it may have been ordered in large quantities by some departments. It is unlikely that developer would have caused an offsite hazard, and entries totaling approximately 11,043 lb were deleted from the list. We also deleted many materials that contain small amounts of carcinogens or are carcinogens but were used in small quantities onsite. For example, we deleted Joy dishwashing liquid (which contains ethanol), upholstery cleaners containing chlorinated solvents, nickel-containing lubricants, roofing tars, chromium-containing corrosion preventatives used in small amounts (less than 2 lb), and formaldehyde-containing solutions used in small quantities. We also deleted EDTA and similar chelators from the list.

Although they present little or no chronic health hazard, several caustics (such as nitric acid, sulfuric acid, sodium hydroxide, and hydrochloric acid) were used and stored in very large quantities; therefore, we retained them on the list for the ranking. Other materials that generally present no chronic health hazard unless large amounts are ingested or they were used in quantities or ways that releases or transport offsite seems unlikely include waxes, Freons, antifreezes, diatomaceous earth, carbon monoxide, calcium carbonate, carbon, propane, and other inert and asphixiant gases. Fly ash, a large volume of which exists onsite, may have been stored outdoors and may have been subject to atmospheric dispersion or runoff into water; therefore, it was

retained on the list. Because polyaromatic hydrocarbons and toxic metals may have been leached or contained in rainwater runoff from coal piles, coal inventories were also retained.

Although we used the initial data extractions for the CIIS from Phase I and the FoxPro database derived from the CIIS database was the starting point for the ranking, subsequent treatment of the list of chemicals differed. Therefore, the list of potential chemicals of concern in the Phase I Task 3 report (Meyer et al. 1995) is not the same as the list developed, with more care, in this phase of the study.

Toxicity Values

Toxicity values were obtained or derived from the EPA's 1995 HEAST Tables; 1995 National Ambient Air Quality Standards (NAAQS) in the Code of Federal Regulations, Part 50; the 1995 Integrated Risk and Information Systems (IRIS) Database; the Agency for Toxic Substances and Disease Registry's (ATSDR) Toxicity Profiles; and Workplace standards published by the American Conference of Governmental Industrial Hygienists (ACGIH) in 1994 and by the Occupational Safety and Health Administration (OSHA) or National Institute of Occupational Safety and Health (NIOSH) in 1994. The documents and databases used are listed in the references for this chapter. The toxicity values are listed in Appendix C3, Tables C3-1 and C3-2. We used the shaded values to calculate the ranking ratios.

The tables include a qualitative developmental effects designation (D) and reproductive effects designation (R). The number of Ds and Rs corresponds to potency or certainty of the information available. A chemical with a DDD or RRR designation is more potent or more positive human data were available for the chemical than for chemicals with a DD, RR, D, or R designations. The designations were determined from information obtained from a search of the Reprotext® Database, information found in ATSDR Toxicity Profiles, and data reported on developmental and reproductive hazards in the workplace in ACGIH (1994), NIOSH (1994), or Lewis (1993).

A carcinogenicity designation was listed for all of the chemicals that might be carcinogenic. In an effort to provide consistency, published EPA designations were used when available, followed by International Agency for Research on Cancer (IARC) designations, ACGIH categories, and any new information from the National Toxicology Program (NTP).

The EPA has evaluated many of the chemicals in Tables C3-1 and C3-2 for carcinogenicity and has assigned the chemical a designation based on the weight of evidence. There are five designations or groups:

Group A- Known human carcinogen. There is sufficient evidence from human epidemiological studies to support a causal association between the substance and cancer.

Group B - Probable human carcinogen. There is limited evidence of human carcinogenicity based on epidemiological studies but sufficient evidence of carcinogenicity based on animal studies.

Subgroups

B1 - Limited epidemiological but sufficient animal data

B2 - With sufficient animal data but inadequate or no data from human epidemiological studies.

Group C - Possible human carcinogen. There is limited evidence in animals and no human data.
Group D - Not classifiable, inadequate data or no data.
Group E - No evidence for carcinogenicity. Negative test results, usually in at least two animal species, or adequate negative epidemiological data.

Many of these chemicals have not been evaluated by the EPA and are classified as Group D because their carcinogenicity has not been adequately determined or are currently under evaluation. Some of the chemicals have not been tested for carcinogenicity.

The IARC has a similar ranking scheme. The scheme defines *sufficient evidence* as when a causal relationship has been demonstrated in humans; *limited evidence* as when there is causal relationship but chance, bias, or confounding factors cannot be discounted; *inadequate evidence* as when available studies cannot determine the carcinogenicity; and *evidence suggesting a lack of* when there are adequate studies that are negative. IARC groups chemicals into four groups:

Group 1 - Compounds are carcinogenic to humans.
Group 2 - Compounds cause cancer in animals.
Subgroups
2A - Probable carcinogen in humans. There is limited evidence of carcinogenicity in humans but sufficient evidence in animals and causal relationship is clear in multiple species or strains in independent experiments.
2B - Possible carcinogens in humans. There is insufficient evidence in humans or animals.
Group 3 - Compounds are not classifiable. There is inadequate evidence and neither the presence nor absence of a carcinogenic effect can be demonstrated.
Group 4 - Compounds are probably not carcinogenic. There are adequate studies involving at least two species to suggest that the substance is not carcinogenic.

The ACGIH also has carcinogenicity designations for chemicals used in the workplace:

A1 - Confirmed human carcinogen
A2 - Suspected human carcinogen
A3 - Animal carcinogen
A4 - Not classifiable as a human carcinogen
A5 - Not suspected as a human carcinogen.

We also consulted reports published by the NTP of cancer bioassays that report ‘clear’ evidence, some evidence, equivocal evidence, no evidence, and inadequate evidence of carcinogenicity. If available, an EPA designation was listed. If an EPA designation was not available or was Group D, the IARC or ACGIH designation was entered. We listed several chemicals that are not yet designated by the EPA, IARC, ACGIH, or examined by the NTP but are mutagens or structurally resemble other carcinogens with the carcinogens in Table C3-2.

For noncarcinogens, we used a reference dose (RfD) for ingestion or a reference concentration (RfC) for inhalation. The RfC or RfD is defined by the EPA as a provisional estimate (with uncertainty spanning perhaps an order of magnitude) of the daily exposure to the

human population (including sensitive subgroups) that is likely to be without appreciable risk of deleterious effects during a portion of a lifetime in the case of subchronic RfD or RfC or during a lifetime in the case of a chronic RfD or RfC (EPA 1995). When available, we used chronic RfCs in units of milligrams per cubic meter air for continuous exposure and chronic RfD values in units of milligrams per kilogram per day. The oral RfD was converted into a corresponding water concentration in milligrams per liter by the equation:

$$\text{mg / L in water} = \frac{\text{oral RfD in mg / kg / day} \times 70\text{ kg}}{2\text{ L / day}} \tag{16-1}$$

which assumes a average human body weight of 70 kg and average water consumption of 2 L day^{-1}. The RfC, in milligrams per cubic meter or micrograms per cubic meter, corresponds to an ambient air concentration for continuous, 24 hr day^{-1} exposure.

For carcinogens, cancer potency factors, called slope factors, have been determined by the EPA for many environmental carcinogens. The slope factor is an upper-bound estimate. It is estimated using mathematical extrapolation models, most commonly the linearized multistage model that estimates the largest possible linear slope, within the 95% confidence limit. The EPA believes true cancer risk to humans is not likely to exceed this upper limit and is likely to be lower. The slope factor is expressed as risk per unit dose in units of risk per milligram/kilogram/day (EPA 1995).

Another useful value is the unit risk value, which is the risk per unit concentration. The unit risk for inhalation is the risk per concentration unit in air expressed as risk per micrograms per cubic meter. The unit risk for oral exposure is the risk per concentration unit in water expressed as risk per micrograms per liter. The unit risk is calculated by dividing the slope factor by the body weight of 70 kg and multiplying by an average breathing rate of 20 m^3 day^{-1} for air or 2 L day^{-1} average consumption for water (EPA 1995).

The risk-specific air or water concentrations can be estimated using the unit risk value at a given risk level. We choose to use a risk level of 1 in 100,000 or 10^{-5}. The concentration in air in micrograms per cubic meter corresponding to

$$\text{a lifetime cancer risk of } 10^{-5} = \frac{10^{-5}}{\text{unit risk per } \mu\text{g / m}^3} \ . \tag{16-2}$$

Risk-specific concentrations in drinking water can be estimated from the oral slope factor. The water concentration corresponding to an upper-bound increased lifetime cancer risk of 1×10^{-5} is calculated as

$$\text{the risk per mg/L in water} = \frac{10^{-5} \times 70\text{ kg}}{\text{slope factor (mg/kg/day)} \times 2\text{ L/day}} \ . \tag{16-3}$$

These values are published in units of micrograms per cubic meter, milligrams per cubic meter, micrograms per liter, and milligrams per liter. There are 10^6 µg g^{-1}, 10^9 µg kg^{-1}, 10^3 mg g^{-1}, 10^6 mg kg^{-1}, and 10^3 µg mg^{-1}. Appropriate conversion factors were applied so that

concentrations corresponding to a 10^{-5} risk level, drinking water standards, workplace standards, and other toxicity values could be compared in Tables C3-1 and C3-2.

The NAAQS are primary standards designed to protect public health. Six have been established to date. The NAAQS include an extensive database that has been rigorously reviewed. The primary NAAQS and the inhalation RfC have essentially the same function, and the EPA has stated that, except for lead, the NAAQS with annual averaging times can be used instead of the RfC.

The time weighted average (TWA) threshold limit value (TLV) established by the ACGIH or the permissible exposure limit (PEL) established by OSHA (in milligrams per cubic meter or fiber per cubic centimeter) were used when EPA toxicity values were not available. Because the TLVs are designed to protect healthy workers, these were divided by 10 to help account for the fact that some members of the general public (especially young children, elderly people, or people with preexisting health conditions) are more sensitive to the toxic effects of chemicals.

Drinking water standards (specifically the EPA maximum contaminant levels [MCLs]) were used if the RfD, oral slope factor or drinking water unit risk value was not available. We preferred to use the toxicity values in the order presented in Tables 16-1 and 16-2.

Table 16-1. Order of Preference for Carcinogens

Air	Water
10^{-5} unit risk	10^{-5} unit risk
RfC	RfD converted to a water concentration
NAAQS	MCL
TLV ÷ 10 or PEL ÷ 10	

Table 16-2. Order of Preference for Noncarcinogens

Air	Water
RfC	RfD converted to water concentration
NAAQS	MCL
TLV ÷ 10 or PEL ÷ 10	

Because they are more relevant to environmental contaminants, we first chose to use the EPA's RfD or concentration, followed by unit risk values, NAAQS values, MCLs, then TLVs. In general, the concentrations corresponding to a risk level of 10^{-5} are the lowest concentrations of concern. However, in a case where the MCL, TLV/10, RfD, or RfC was lower, we used the lower, more stringent level to calculate the ranking ratios.

In some cases, a single toxicity value and combined inventory quantity was given to a group or class of compounds that contain a common element or component. Although different valence states of metals may vary tremendously in their toxicity, for the purposes of screening, the toxicity value we used was based on the toxicity or carcinogenicity of the most toxic or elemental form depending on the data available. For some materials, like manganese, sodium, and potassium compounds, more toxicity information is available on specific compounds, so we used it. We combined compounds of arsenic, cadmium, chromium, mercury, nickel, and tin for screening. For example, the chromium compounds include chromium metal; chromium oxide;

dichromic acid; lead chromate; potassium chromate; potassium dichromate; sodium chromate; sodium dichromate; chromium azo dye; chromic acids and salts (acetate, chloride, nitrates, sulfates); and ^{51}Cr. We used the toxicity values for carcinogen hexavalent chromium.

If both the anion and the cation of a compound are responsible for the toxicity, then we considered both. For example, zinc cyanide was included in both the zinc and cyanide inventory amounts, and we included both compounds in the ranking. Some metal salts (like mercuric nitrate) were used in large quantities, so we considered this compound separately from elemental mercury.

Many radionuclides, such as ^{51}Cr, are hazardous because of their radioactivity and chemical toxicity. Uranium, for example, is toxic to the kidney. Although uranium will be evaluated with the radionuclides of concern, for the purposes of this ranking we treated uranium as a toxic metal and assessed it using the RfD for kidney toxicity.

Isomers of tetrachloroethane, dichlorobenzene, dichloroethane, dichloroethylene, dimethylphenol, dinitrotoluene, dichlorophenol, chlorophenol, nitrophenol, trichlorobenzene, trichloroethane, tetrachloroethane, and trichlorophenol were summed, and we used the toxicity value for the most toxic or carcinogenic isomer. All of the inventories for various arochlors and polychlorinated biphenyls (PCBs) were summed and the toxicity values for arochlor 1260 were used for the PCBs.

We included chemicals used at the Naval Fuel Facility in the ranking. The names of these materials are classified; however, the materials of concern were not unique to the Naval Fuels Facility and were already included on one or more of the Site inventories.

Ranking Ratio Calculation

Using inventory amounts and toxicity values described above, we calculated ranking ratios for 170 chemicals in Tables C3-1 and C3-2. Initially, we did not calculate ranking ratios for 12 chemicals, primarily pesticides, for which there was no inventory amount or for which there was an inventory amount that seemed far too small. An effort was made in April 1996 to actively solicit information on inventory amounts from former and current Site employees who might have knowledge of chemical usage, purchase, storage, disposal, or other relevant operations onsite. A letter was sent to 118 individuals by post or electronic mail, and 18 individuals responded. Several respondents agreed that some of the pesticides listed may have been used onsite in the 1960s and 1970s; however, we did not obtain new or different inventories for any of the chemicals.

No published toxicity values could be obtained for 11 chemicals. We looked at the MSDSs for these chemicals in the SRS files and sought toxicity testing data and structure activity information for these compounds to determine air and water concentrations of potential concern. Because better toxicity values have not been determined, we used LD50 divided by 100,000 (Layton et al. 1987) as a toxicity value.

Some chemicals (for example, dichlorodifluoromethane and sulfur oxides) are only a concern if released to the air and inhaled. We did not calculated ratios for water for these types of chemicals. We did not calculate air ratios for chemicals, such as dichlorobenzidine and chlorophenol, because toxicity values have only been determined for consumption of water.

We calculated the ranking ratio for materials released to air using the following equation:

$$\text{air ratio} = \frac{Q_a\,(\text{kg / s}) \times \chi / Q\,(\text{s / m}^3) \times 10^9\ (\mu\text{g / kg})}{\text{Tox}_a\ (\mu\text{g / m}^3)} \qquad (16\text{-}4)$$

where

Tox_a = air concentration that corresponds to a level of concern. The preferred toxicity values used are described on page 14.

χ/Q = atmospheric dispersion factor. The average χ/Q, based on tritium monitoring done in 1990, for onsite locations of 5.4×10^{-8} s m^{-3} (Cummins et al. 1991), was used

Q_a = the release rate

$$Q_a\,(\text{kg / s}) = \frac{I\,(\text{kg}) \times RF_a}{3.16 \times 10^7\ \text{s / year}}$$

where I = annual inventory amount in kg and Rf_a = the release fraction.

Release fractions account for the fact that some materials may be released in total (like volatile solvents or gases that are put out the stacks) and others (like solids) will not all be released. We chose to conservatively assume, for this first screening, that 100% of the inventory was used. Although more material is usually stored than is used, it is reasonable to assume that it all was used and the entire annual inventory was subject to release each year.

We assigned release fractions according to the volatility of the compound (that is, how readily it evaporates). For materials released to the air, we chose a release fraction (RF_a) of 1.0 for volatile liquids and 0.005 for nonvolatile liquids and solids. It is likely that workers would have made some effort to conserve reagents, keep lids on solvent baths, and prevent spills and leaks of volatile substances. However, for the purposes of this ranking, we assumed that all of the volatile material inventoried was eventually released to the atmosphere and we used the conservative release fraction of 1 (signifying 100% of the inventory quality was released).

The ranking ratio for materials released to water was calculated using the following equation:

$$\text{ratio}_w = \frac{Q_w\ (\text{kg/s}) \times 10^6\ \text{mg/kg}}{DF\,(\text{L/s}) \times \text{Tox}_w\ (\text{mg/L})} \qquad (16\text{-}5)$$

where

Tox_w = water concentration that corresponds to a level of concern. The preferred toxicity values used are described on page 14.

DF = the dilution due to flow of the river. We assumed that the Savannah River is the surface water of concern and that it was used for drinking water (untreated). Other activities, such as fishing, boating, swimming, etc., will be considered in later phases of this work. The 1990 *SRS Environmental Report* includes a plot of the flow rate of the Savannah River for 1980–1990. The median value corresponds to a DF of 2.55×10^5 L s^{-1} (Cummins et al. 1991).

Qw = the release rate.

$$Q_w\ (kg/s) = \frac{I\ (kg) \times RF_w}{3.16 \times 10^7\ s/year}$$

We used the release fractions (RF_w) 0.02 for volatile liquids, which would tend to evaporate out of the water, and 0.1 for nonvolatile liquids and solids.

To compare among values, we converted all of the concentrations corresponding to 10^{-5} risk levels, MCLs, TLVs, and NAAQS to units of milligrams per cubic meter or milligrams per liter in the tables. In summary,

$$\text{air ratio} = \frac{I\ (kg) \times RF \times 5.4 \times 10^{-8}\ s/m^3 \times 10^6\ mg/kg \div \text{toxicity value}\ mg/m^3}{3.16 \times 10^7}. \qquad (16\text{-}6)$$

If all the toxicity values are in units of milligrams, then the two ranking ratio equations can be summarized as

$$\text{ratio air} = \frac{\text{inventory amount in kg} \times \text{release fraction} \times 1.7 \times 10^{-9}}{Tox_a} \qquad (16\text{-}7)$$

$$\text{ratio water} = \frac{\text{inventory amount in kg} \times \text{release fraction} \times 1.24 \times 10^{-7}}{Tox_w}. \qquad (16\text{-}8)$$

Ranking Results

Tables C3-1 and C3-2 in Appendix C3 list the resulting ranking ratios for 166 chemicals. Ratios could not be determined for methyl bromide and methyl chloride (two chemicals detected in groundwater but not used onsite and for which there was no inventory) and ozone, an air pollutant produced as a byproduct for which there is no inventory. Materials with the highest ratios are listed in Tables 16-3 through 16-6.

Table 16-3. Chemicals with Ranking Ratios > 1.0

Chemical	Ranking ratio	Ratio
Coal	254	Water
Hydrazine	36.7	Air
Uranium	1.87	Water
Gasoline	1.77	Water
Hydrazine mononitrate	1.26	Air

Table 16-4. Chemicals with Ranking Ratios < 1.0 but > 0.1

Chemical	Ranking ratio	Ratio
Tetrachloroethylene	2.29×10^{-1}	Water
Hydroxylamine sulfate	1.10×10^{-1}	Water

Table 16-5. Chemicals with Ranking Ratios < 0.1 but > 0.01

Chemical	Ranking ratio	Ratio
Benzene	1.04×10^{-2}	Air
Nickel compounds	1.20×10^{-2}	Water
Mercury	1.21×10^{-2}	Water
Sulfuric acid	1.32×10^{-2}	Air
Trichloroethane	1.54×10^{-2}	Air
Chromium compounds	2.32×10^{-2}	Water
Lead compounds	3.25×10^{-2}	Water
Mercuric nitrate	3.53×10^{-2}	Water
Asbestos	9.40×10^{-2}	Air

Table 16-6. Chemicals with Ranking Ratios < 0.01 but > 0.001

Chemical	Ranking ratio	Ratio
Zinc compounds	1.03×10^{-3}	Water
Manganous nitrate	1.28×10^{-3}	Air
Copper compounds	1.35×10^{-3}	Water
Nitric acid	1.48×10^{-3}	Air
Aldrin	2.48×10^{-3}	Water
Cadmium	2.52×10^{-3}	Water
Hexanol	3.44×10^{-3}	Air
Chlorine	3.95×10^{-3}	Air
Gadolinium nitrate	4.27×10^{-3}	Air
Coal tar	4.45×10^{-3}	Water

In addition, we found no inventory amount or the inventory amount seemed far too small, for 19 chemicals:

Aldrin	Carbon tetrachloride	Chloroethane
Chloromethane	DDT	Dieldrin
Endosulfan	Endrin	Chlordane
Heptachlor	Hydroxyquinoline	Lindane
Toxaphene	Trichlorophenol (2,4,5-T)	Velpar-L
Chloroethane	DDT	Chloromethane
PCBs		

Because many hazardous, persistent pesticides (like DDT) have been banned for some time, we might expect little or no inventory amounts although these materials may have been used in the past. We found little mention of these materials in the periodic reports. We compiled recent monitoring study results to see if they could help us quantify some of the pesticides of concern. Although we will could not determine an inventory amount for many of these pesticides and many could not be adequately evaluated in the ranking, it seemed inappropriate to simply dismiss these pesticides at this early stage of the ranking simply because of a lack of inventory.

Limitations of the Ranking

This approach to ranking chemicals of concern has several limitations that are important to recognize. The amount of material listed on an inventory is not necessarily related to the amount that was discharged out a stack as a result of a process. For example, nitrogen dioxide stored in tanks or cylinders onsite is in no way related to the amount of nitrogen dioxide produced by a process and discharged to the environment. A careful examination of processes and materials that may have been produced and released at certain points of an operation will be another important part of this work.

Another problem is that the CIIS database contains the inventory for 1994. The amounts onsite now may be more or less than when facilities at the plant were in full operation. We compared the amounts in the 1994 CIIS database and the Du Pont Index from the 1970s, and noted differences. We used the largest amounts (usually those in the listing from 1973 or the upper value from the range in the Du Pont database). It is unfortunate that we could not locate additional inventories from the 1950s, 1960s, and early 1970s.

One of the most significant problems with this approach is a lack of information. Information on inventory amounts and toxicity is missing for many of the materials. Often, toxicity values are available for inhalation or ingestion but not both pathways. Information on reproductive and developmental effects is not available for most of the materials. Eleven materials have no published toxicity values, so we used a very conservative and uncertain value of the lowest LD50 value reported (usually an oral LD50 in rodents or rabbits) divided by 100,000 (Layton et al. 1987). For several materials on which no toxicity testing has been done, we used values derived for similar compounds.

Second Stage of the Ranking

After the ranking ratios were calculated, it was obvious that the ratios alone could not adequately be used to prioritize chemicals of concern. The conservative assumptions used created unrealistic scenarios for many of the chemicals that had ranking ratios greater than 10^{-3}.

The next step was to further evaluate the chemicals identified in the ranking by considering environmental fate and transport characteristics, information on chemical use at the plant; and release potential. We also assessed the chemicals based on the physical and chemical properties relevant to their behavior in the environment, such as water solubility, volatility, susceptibility to biodegradation and chemical breakdown, and mobility. These properties can be very important to environmental behavior. For example, highly water-soluble compounds are generally less likely to adsorb to soils and sediments, more likely to remain in the water, more biodegradable, and less likely to volatilize from water. The octanol-water partition coefficient, K_{ow}, is a measure of the degree to which an organic material will preferentially dissolve in octanol compared to water. The greater the K_{ow}, the greater the tendency for the material to partition from water to a more organic phase.

Some chemicals, for example titanium tetrachloride, break down so rapidly that environmental exposure for people living offsite from this chemical is unlikely. Other chemicals, for example asbestos, were used in building materials and were not subject to storage in large amounts, leakage, spillage, or routine release.

Chemicals with ranking ratios greater than 10^{-3} are evaluated further in the following paragraphs.

Alcohols

Large amounts of ethanol and hexanol and smaller amounts of propanol, ethoxyethanol, and other alcohols were used for cleaning and as part of processes at the SRS. The ranking ratios for these compounds were very low. The highest ratio was for hexanol, with an air ratio of 3.4×10^{-3}. Because toxicity information on hexyl alcohol was limited, it was ranked using the very conservative LD50/100,000 value. Hexanol is a volatile skin and eye irritant that appears to cause liver toxicity and other toxic effects similar to those caused by other alcohols. The availability of documentation specific for this alcohol limited our ability to develop a source term. It is likely subject to biodegradation and has fate and transport characteristics similar to other alcohols. Emission estimates in the 1985, 1987, and 1990 Air Emissions Inventory are low, and significant offsite concentrations would not be expected. Therefore, we did not develop a source term for this chemical.

Aldrin

Aldrin is a particularly toxic chemical for which we have no reasonable SRS inventory and usage information. In general, in the U.S., it was used as an agricultural pesticide until 1970 and for termites until 1987. Aldrin and its toxic breakdown product, dieldrin, are very persistent, not soluble in water, and tend to bioaccumulate. Further evaluation of this chemical was not possible because documentation on its use was not found.

Asbestos

Asbestos is a known human carcinogen. It causes lung cancer and lung disease. Asbestos insulation was used extensively in the 1950s, when many facilities at the SRS were initially constructed. Asbestos materials can be found in roofing, pipe, and vessel insulation; building insulation; gaskets; packing; siding; and other building materials. The materials have been removed as they deteriorate or as renovations, maintenance, and repairs have been done. Worker protection has necessitated that asbestos controls be in place during all removal operations (DOE 1987).

More than 80,000 linear feet of asbestos-containing materials were reported to have been removed in 1984. Nonradioactive asbestos has been disposed of in a separate trench in the sanitary landfill. Radioactive asbestos was buried in the Solid Waste Disposal Facility (formerly referred to as the Radioactive Waste Burial Grounds). An asbestos disposal site in C-Area was closed and capped in 1984 (DOE 1987).

Although a concern for workers onsite, asbestos has not been considered an environmental contaminant and has not been a concern for offsite exposure. In 1967, asbestos concentrations in air at several plant locations were measured. The maximum concentration was “1 million particles per cubic foot,” which was said to have been one-fifth of the TLV for workers at that time (Du Pont 1967). No facility has listed asbestos as an emission for the Air Emissions Inventory (Faugl 1996). We did not evaluated asbestos further because it is primarily found in

building materials that are resistant to environmental transport unless seriously degraded or the building is demolished. If a building is imploded, exploded, or demolished all at once, the asbestos would be released to the air and would be subject to resuspension with time unless precautions were taken to contain it. It is likely that most of the buildings containing asbestos materials have been remediated or destroyed in the last decade, when awareness of asbestos hazardous was high and precautions were taken to protect workers and limit dispersion of asbestos fibers. For the purposes of the conservative stage one ranking, if all of the asbestos reported onsite was released to the air, concentrations about 100 times the level of concern could have been reached. This is a very unreasonable scenario, however, because most of the asbestos onsite is in insulation and building materials. Asbestos has not been subject to storage in large amounts, leakage, spillage, or routine release to the environment; therefore, we did not estimate a source term for asbestos.

Benzene

Benzene is a very volatile, slightly water-soluble chemical used in the past as a solvent. It is a contaminant of several other materials used at the SRS, including gasoline. Chemical degradation, primarily reaction with hydroxyl radicals, limits the persistence of benzene in air to a few days or even a few hours. Benzene released to soil and water is subject to biodegradation, photooxidation, and volatilization. The half-life in surface water has been estimated to be about 17 days for photolysis. Biodegradation half-lives are estimated to be about 8 to 16 days in surface water and about 30 days in groundwater. Although a large volume of benzene is released to the environment from a variety of sources, environmental levels are generally low because of rapid removal and degradation. The Log K_{ow} is 2.15. Benzene is considered highly mobile in soil and water, but it will adsorb to organic matter in soil. Benzene does not bioconcentrate, and it is a known human carcinogen. Because large amounts of petroleum products were used at the SRS, we investigated environmental monitoring information and releases of benzene further.

Chlorine

Chlorine is a severely irritating gas that is also more of a hazard in the workplace than an environmental contaminant of concern. Chlorine is reactive and would not be expected to persist in the environment as an irritating gas. It is not a carcinogen or chronic toxicant and would not be likely to have caused health effects offsite unless released in very large quantities at one time. Therefore, chlorine was not included in the chemicals to be considered further.

Coal

Coal is primarily carbon containing varying amounts of toxic or carcinogenic metals, sulfur, and other contaminants. Coal is regulated in the workplace as a nuisance dust (Lewis 1993). Although the ranking ratio is high, coal should not be the first priority in evaluating source terms for chemicals because of the ultraconservative ranking assumptions applied. For the purposes of consistency in the ranking exercise, it was assumed that all the coal piles were discharged to the Savannah River and that the toxicity value for the entire pile was that of benzo(a) pyrene, with an MCL of 0.002 mg L^{-1}. This led to a ranking ratio of 254. If the entire store was evaluated using

the MCL of 0.005 mg L^{-1} for benzene, another carcinogenic component, the ranking ratio would be 101. It is unreasonable to treat the entire coal inventory as if it were carcinogenic aromatic hydrocarbon or benzene. We could conservatively estimate that coal tar and coal tar creosote material could contain as much as 3% aromatic hydrocarbons (Gosselin et al. 1984) and as much as 10% benzene (ATSDR 1995). If the inventory amount is reduced by these percentages, the resulting ranking ratios are 7.6 for benzo(a)pyrene and 10.1 for benzene. It is also unreasonable to assume that all of the coal inventory was discharged into the river. We know the piles remain onsite, but we do not have an estimate of the amount of material that left the pile in runoff or leachate. A preliminary evaluation of the runoff history, means taken to prevent runoff, and the results of environmental sampling around the coal piles was worthwhile for this phase of the study. We also evaluated coal ash.

Coal Tar

Coal tar and coal pitch are volatiles formed during the distillation of coal, and they contain carcinogenic polyaromatic hydrocarbons like pyrene, benzo(a)pyrene and chrysene, benzene, and phenol. If stores of coal tar were not contained, they could be subject to rainwater runoff and leaching, and they could contaminate surrounding soil, surface waters, and underlying groundwater. We investigated this material as a part of the qualitative evaluation of coal stores.

Freons

Chlorinated, fluorinated hydrocarbons, also called Freon (a registered trademark of Du Pont), are clear, colorless, noncombustible liquids. Perhaps best known for their use as refrigerants, they were also used at the Site for cleaning, degreasing, and as decontamination solutions. Freons were used in solvent degreasers, especially after 1988. Freon was also used as a coolant during operation of saws and lathes in M-Area. About 530 gal y^{-1} was used during the peak production times of 1985–1988. All of this was probably released to the atmosphere (Radian 1992). Freon was also released from heating, ventilation, and air-conditioning equipment.

Freon vapors are four to five times heavier than air and tend to accumulate in tanks, pits, sumps, and other low places; therefore, inhalation of concentrated vapors can be an occupational hazard (Haynes and Stoddard 1984). In a 1975 industrial hygiene summary report, mention is made of 800 gal of Genetron 11 being removed from a refrigeration unit and disposed of. The Du Pont Freon Products division memo informed SRS staff that the company had “no method or requirement for chemically changing these materials into other substances that would be more acceptable for release to the environment the recommendation (and practice) is to remove the material to a remote area and allow it to evaporate” (Harper and Croley 1976). Although release of these compounds to the environment is recognized to contribute to ozone depletion, it is very unlikely these materials would have presented a health hazard offsite; therefore, we did not determine releases of Freon.

Gadolinium Nitrate

Because toxicity information on this chemical is limited, we ranked gadolinium nitrate using the very conservative LD50/100,000 value reported in the MSDS. Gadolinium nitrate is described

as a severe irritant and no chronic health effects are known. It is a relatively stable material, but environmental fate and transport of this material has not been studied. It is likely that precautions were taken to conserve this material and large releases were probably uncommon. The unavailability of documentation specific for this material would have seriously limited our ability to develop a source term; therefore, we did not evaluate gadolinium nitrate further.

Gasoline and Other Fuels

Gasoline is a complex mixture of hydrocarbons containing small amounts of benzene, toluene, xylene, 1,3-butadiene, and sometimes lead. Gasoline may have been released to the air or water and it may have seeped into groundwater from leaking storage tanks, pipelines, or as a result of spills. For the purposes of the ranking, we assumed all gasoline stores spilled into the river or discharged into the air, and the entire amount was ranked using the drinking water standard for a carcinogenic component, benzene.

The composition of gasoline varies tremendously. Compositional data studies sponsored by the American Petroleum Institute found the benzene content of gasoline mixtures were as high as 2% (ATSDR 1994). The IARC estimated that gasoline typically contains 0.5 to 2.5% by volume benzene (ATSDR 1994) or as high as 3.2% by weight. Benzene, ethylbenzene, and other aromatics related to benzene (like toluene and xylene) may make up as much as 30.5% by weight of a gasoline mixture (ATSDR 1994). Measurements of the amount of benzene versus total hydrocarbons gasoline released to the atmosphere vary greatly when tanks are vented, gasoline is pumped, or storage tanks are excavated. However, measurements do suggest benzene vapors could account for nearly 10% of the gasoline vapors under certain conditions of discharge. Studies have shown that benzene concentrations in air are highest during refueling operations. Studies on service station attendants suggest that the gasoline in the air they breathed averaged about 0.25% benzene. Taken together, these studies suggest an upper-bound estimate of 30% for the percentage of benzene in gasoline or gasoline vapors. Using the toxicity values for benzene and 30%, the inventory amount results in a ranking ratio of 0.531 for gasoline.

Gasoline is very volatile and does not dissolve readily in water. Most of the hazardous components of gasoline are broken down in a number of hours to weeks after their release. The Log K_{ow} for gasoline ranges from 2.13–4.87. Most chemicals in gasoline do not bioaccumulate. After it is released to the environment, gasoline is not transported as a mixture. The components of the mixture selectively partition into different environmental media according to their individual chemical and physical properties. The compounds of greatest health concern, like benzene, are water-soluble and are transported in groundwater, surface water, and through soils. These compounds are also subject to photochemical oxidation in air and have half-lives on the order of one or several days. They are also subject to biodegradation in water and volatilization, photooxidation, and biodegradation in soil. Although gasoline has caused liver and kidney tumors in animal studies, there is no evidence that gasoline causes cancer in humans. We did not find documentation, interview notes, or other evidence of leaks, spills, and large releases of gasoline that may have traveled offsite; therefore, we did not determine a source term estimate for gasoline released to surface water.

Hydrazine

Hydrazine is a reactive, flammable liquid used as a reducing agent for nuclear fuel reprocessing. It has also been used as an intermediate in the production of agricultural and industrial chemicals, as rocket fuel, and as a medication for sickle cell disease and cancer. Hydrazine has a relatively low vapor pressure and is soluble in water. Hydrazine could have been released to air, water, or soil. This chemical rapidly degrades in most environmental media by oxidation and biodegradation. Hydrazine in air is quickly destroyed by chemical reaction within minutes or hours, depending on the concentration of ozone and hydroxyl radicals in the air. Most hydrazine in air would be expected to have degraded within several hours of its release. Hydrazine released to water and soil can become dissolved in water or bind to soil. Hydrazine can sorb onto clay soils. The Log K_{ow} is –3.08. Hydrazine is subject to oxidation and biodegradation, and most of the hydrazine in water and soil would be expected to be gone within a few weeks. Hydrazine does not tend to biomagnify up the food chain. The potential for hydrazine to have been transported offsite is limited by its rapid degradation. Hydrazine causes several types of tumors in animals and is classified as a probable human carcinogen by the EPA (ATSDR 1994). We reviewed the use, storage, release history, and environmental monitoring results for hydrazine and hydrazine mononitrate.

Hydroxylamine Sulfate

Hydroxylamine sulfate is a white crystalline material that is very corrosive and can cause severe burns, ulceration, and sensitization reactions. There is a lack of toxicity information for this material, which is listed as being used in relatively large amounts in the 1970s Du Pont Index. The ranking ratio for hydroxylamine sulfate is comparatively large because we used the upper value of the range reported in the Du Pont inventory and the high LD50/100,000 value to calculate it. The LD50 was derived from data on mice given hydroxylamine sulfate i.p. and reflects the corrosive, irritant effects rather than any chronic effects. Although specific environmental fate data on this chemical are also lacking, it is a reactive chemical and would not be expected to persist in the environment. Based on this and the extreme conservatism of the toxicity values, we did not develop a source term for hydroxylamine sulfate.

Manganese

Manganese was evaluated using the EPA's RfC value, which is quite conservative, with an uncertainty factor of 900 to account for a lack of toxicity data. Available monitoring data and source term information were examined, but the lack of documentation specific to this material limited our ability to develop a source term. We reviewed and evaluated monitoring and usage data on manganese compounds to the extent possible.

Mercury

The physical and chemical characteristics of metals that influence their behavior in the environment include solubility, oxidation state, and tendency toward sorption on materials in soils and sediments. Mercury exists in the elemental form (a volatile liquid), in the +1 and +2

oxidation states, and as organic mercury. Mercury sorbs strongly to organic material and oxides and tends to accumulate in sediments. Microorganisms in sediments can convert mercury to methymercury, which bioaccumulates. Mercury and methylmercury are very potent neurological toxins. The inventory amounts used for the ranking may not include mercury in pumps that was disposed of by burial through the years. We further evaluated discharges of mercury to the air and water and evaluated mercuric nitrate along with elemental mercury.

Other Metals

Chromium, lead, and nickel had ranking ratios for water that were greater than 10^{-2}. Zinc, copper, and cadmium compounds had ranking ratios for water that were greater than 10^{-3}. These metals are toxic, and chromium, nickel, and cadmium are carcinogenic. Lead is a potent developmental neurological toxin.

We used combined inventories of small metal stores all over the Site and numerous metal-containing compounds like rust and corrosion inhibitors that were used and stored in relatively small amounts to obtain the ratio. Many of these materials were primarily found in solid form, like sheeting, bricks, pellets and pipes. Especially large amounts of lead shielding, and lead pellets were used at the SRS. These materials were relatively resistant to environmental transport and were not likely to have been subject to resuspension in air or leaching into surface or groundwater. For the purposes of ranking, we assumed that all stores of these materials were placed in the Savannah River. This is especially unreasonable because these materials were not stored in large piles or tanks but were used all over the Site in many different facilities, in small and large amounts, and in many forms.

The mobility of all of the metals depends on soil and water chemistry and pH. For example, cadmium usually occurs as the Cd^{+2} ion at pH levels less than 8 or quite commonly as cadmium sulfate. Cadmium will adsorb to soils and sediments by cation exchange. Chromium occurs in the +3 or +6 oxidation state in water. Cr(III) is insoluble and readily absorbs to metal oxides in soils. Cr(VI) is soluble and is more mobile in the environment.

We compiled environmental monitoring data and information useful for source term determination for arsenic, cadmium, chromium, lead, manganese, mercury, nickel, and zinc. These metals were released into surface water from their use as cooling water treatment chemicals, during surface water runoff from coal and ash piles, and as a component of process waste in M-Area, F-Area, and H-Area.

Nitric Acid

Nitric acid is a caustic, severely irritating compound that is more of a workplace hazard than an environmental contaminant of concern. It is subject to rapid degradation. It is not a carcinogen or chronic toxicant and would not likely have caused health effects offsite unless released in very large quantities at one time. We compiled releases of nitric acid fumes and further evaluated nitrates and nitrogen dioxide, which are chemicals associated with the use of nitric acid.

Sulfuric Acid

Large amounts of sulfuric acid were used onsite, and several spills of sulfuric acid to Site streams occurred. Sulfuric acid is a caustic, corrosive material that would not be expected to persist in the environment. We noted documentation of sulfuric acid releases, but it did not seem necessary to develop a source term for this material because transport of hazardous amounts offsite would have been limited by its rapid degradation or conversion into other materials.

Tetrachloroethylene

Tetrachloroethylene is a volatile liquid used as a solvent, cleaner, and vapor degreaser. Tetrachloroethylene is slightly water-soluble, and tetrachloroethylene released to surface water would be expected to rapidly evaporate into the air. Tetrachloroethylene also evaporates from soil, but it can easily travel through soil into groundwater. The Log K_{ow} is 3.40. Tetrachloroethylene in air is photochemically degraded. The half-life in air has been estimated to be 3 to 4 months, while degradation in water is much slower. Tetrachloroethylene is subject to biodegradation, but it can persist in soils and groundwater for decades. Tetrachloroethylene has a very low tendency to bioaccumulate. Tetrachloroethylene has caused cancer in animal studies, but it has not been shown to cause cancer in humans. The ranking ratio for tetrachloroethylene in water was quite high because the drinking water standard for this chemical is quite protective. It is likely that most of the tetrachloroethylene used onsite was released into the atmosphere. We evaluated the discharge of this solvent and others to M-Area sewers, into groundwater, and into the air; however, the priority assigned to tetrachloroethylene was not as high as the water ranking ratio might suggest.

Trichloroethylene

Like tetrachloroethylene, trichloroethylene was used as a solvent and cleaner. Trichloroethylene released to surface water would be expected to rapidly evaporate into the air. Trichloroethylene also evaporates from soil, but it can easily travel through soil into groundwater. Large amounts of trichloroethylene were used in M-Area, and we further evaluated discharges to M-Area sewers, into groundwater, and into the air.

Trichloroethane

Trichloroethane is a volatile cleaner and solvent that was released to the air and to M-Area process sewers. It dissolves slightly in water and would be expected to evaporate rapidly from soil and water. Once in the air, it is estimated to persist for about 6 years. Trichloroethane is thought to be important in reducing the stratospheric ozone layer. Trichloroethane in soil and water is also subject to biodegradation. An estimated half-life for degradation in groundwater is about 10 months. Trichloroethane does not bioaccumulate. 1,1,1-Trichloroethane is not carcinogenic, and the likelihood that environmental exposure would cause significant health effects is low. 1,1,2-Trichloroethane has caused cancer in some animal studies, but it has not been shown to cause cancer in humans. For the purposes of the screening, we combined different isomers of trichloroethane and calculated a ranking ratio using the more conservative toxicity

values for the carcinogenic isomer. We further evaluated discharge of this solvent and others to M-Area sewers and into groundwater and the air.

Uranium

Uranium is a radioactive metal that can cause cancer and kidney disease. In general, uranium releases were reported in curies rather than kilograms or pounds of material. Uranium is one of the radioactive materials being evaluated and we developed a source term for it. Understanding the kidney toxicity of uranium may be an important step in later phases of the project when health risks will be determined.

Other Chemicals of Potential Interest

There are several chemicals that, as a result of the screening, were not evaluated further but are of public interest. A discussion of these chemicals follows.

Anthracene. Anthracene is a polycyclic aromatic hydrocarbon. Unlike similar aromatic hydrocarbons, anthracene is not carcinogenic. Benzanthracene and other similar carcinogenic compounds have been listed in essential materials ledgers, but how they were used was unclear. Benzo(a) anthracene, benzo(a)pyrene, benzofluoranthene, benzoperylene, and chrysene were listed in the 1994 SRS CIIS database with inventory quantities of zero or less than 0.001 lb, which is consistent with use as an analytical standard or laboratory reagent. A September 1953 monthly progress report for the Works Technical Department mentioned that assistance had been given to the instrument department concerning the use of anthracene in relatively large quantities to coat tubes in building 773-A. The Works Technical Department recommended that this be done in a hood, using protective clothing and a respirator, and special attention be given to personal clean up after each job (Du Pont 1953).

Carbon Tetrachloride. An inventory amount for carbon tetrachloride was not given in the CIIS database. The inventory amount in the CIIS database would not have been particularly useful for estimating quantities of carbon tetrachloride used in the past because it is likely that use of this chemical as a solvent was phased out. As at other U.S. Department of Energy facilities, it is likely carbon tetrachloride was replaced with less toxic solvents, such as tetrachloroethylene, trichloroethylene, and trichloroethane. Very large amounts of carbon tetrachloride were used at the Rocky Flats Plants in Colorado. However, no documentation has been found to support the use of large amounts of carbon tetrachloride at the SRS. It seems that tetrachloroethylene, trichloroethylene, and trichloroethane were the solvents used in M-Area to clean materials such as targets and cans. Recent RCRA and CERCLA monitoring data have detected carbon tetrachloride in groundwater but not in quantities suggesting a significant use in the past. Carbon tetrachloride was not reported in the 1974 inventory. Based on an apparent lack of inventory, we did not evaluate carbon tetrachloride further.

Fluoride. Hydrogen fluoride was released from the JB-Line stack in F-Area (Reinig et al. 1973). Fluoride was also a component of liquid waste from the separations area processes. Potassium fluoride was used in the frames process to isolate ^{238}Pu. A 1988 summary of separations activities reported that 59 lb of fluoride was discharged to the seepage basins without evaporating because of corrosion problems with evaporators. This amount was said to represent

the maximum annual amount that would have been discharged during 1 year from one separations plant (Du Pont 1988).

Fluorides are ubiquitous in food and water. Human exposure to toxic amounts of fluorine and hydrogen fluoride is unlikely outside of an occupational setting. Fluoride is highly electronegative and reacts vigorously with other compounds. Fluoride, fluorine, and hydrofluoric acid released into soil or surface waters would rapidly react to form fluoride salts. Fluorides discharged to the seepage basins have been retained by the soil.

Hydrofluoric acid was spilled on the grounds west of central shops sometime before 1970. Monitoring wells have been installed in the area. Contaminated groundwater has not moved offsite (Christensen and Gordon 1983), and it is not likely to move offsite in the near future because the central shops are centrally located. A hydrofluoric acid solution has been used in the 773-A glass shop, which makes glassware used at the Savannah River Technology Center. The 1996 Operating Permit Application described the releases as being exhausted from two small stacks. The application considered the releases to air to be very small (Westinghouse 1996). A 1987 annual environmental report describing ambient air quality monitoring onsite and offsite said that gaseous fluorides were not monitored because the potential release was insignificant compared to the standard (Mikol et al. 1988). We did not evaluate fluorides further.

Scintillation Fluids. Scintillation fluids are photofluoric compounds and surfactants in organic solvents, such as xylene, toluene, dioxane, napthalene, or trimethylbenzene solutions. An annual waste generation rate for scintillation fluids used to analyze samples for radioactivity was estimated to be about 200 gal y^{-1} (Smithwick 1984). Liquid scintillation solutions have been buried at the Radioactive Waste Burial Grounds since 1965. As of 1984, the amount sent to the burial grounds was estimated to total 10,000 gal, most of which had been used for tritium analysis. The solutions were buried in plastic or glass vials in containers with absorbent material. After 1987, all liquid scintillation solutions were supposed to be incinerated (DOE 1987). There is no evidence that these fluids traveled offsite; therefore, we did not evaluate them further.

CONCLUSIONS

Based on the ranking results and the discussion above, we concluded at the beginning of Phase II of the project that a source term should be developed for the following chemicals:

Benzene	Mercury and mercuric nitrate
Coal	Nitric acid
Coal Ash	Trichloroethylene
Hydrazine	Tetrachloroethylene
Gasoline	Trichloroethane

We concluded the following metals should also be evaluated:

Arsenic	Cadmium
Chromium	Lead
Manganese	Uranium
Nickel	Zinc

A source term was estimated for chromium, cadmium, and lead releases to air and nickel releases to Site streams. We did not find enough information to estimate a source term with reasonable certainty for arsenic in coal and ash pile runoff; chromium releases to surface water; or nickel, zinc, and arsenic releases to air. We compiled monitoring data for these metals and described releases using all available information. Information on gasoline, coal, and ash storage, use, transport, and disposal was reviewed and summarized. We made release estimates for benzene and toxic components of metals in coal and ash and releases of toxic metals and other pollutants from coal burning. In addition, we developed source terms for nitrogen dioxide, sulfur dioxide, ash particulates, and hydrogen sulfide. The SRS operations had the potential to release large amounts of these pollutants into the air.

The following chemicals may have been released, but additional analyses were impossible because of a lack of available inventory or toxicity information:

Aldrin/dieldrin	Chloroethane
Chloromethane	DDT
Endosulfan	Endrin
Heptachlor	Hydroxyquinoline
Lindane	Toxaphene
Trichlorophenol (2,4,5-T)	PCBs (polychlorinated biphenyls, arochlors)

An inventory for these chemicals was not reported in 1974 or 1994. Chloroethane and chloromethane have been detected in onsite groundwater. There is no amount listed in the inventory for PCBs, but PCBs have undoubtedly been used in electrical equipment onsite, and some of this equipment was likely to have been buried in onsite waste areas. The *RAC* researchers reviewing monthly reports and other documents for Phase II of the dose reconstruction study were given a list of these compounds and asked to flag any information regarding their use, release, monitoring. or disposal. All of this documentation was reviewed and used to develop a source term estimate for as many of these chemicals as possible. The lack of documentation for some of the chemicals seriously limited our ability to develop a source term for them. However, where a release estimate could not be calculated, a qualitative evaluation of the use and potential release of the compound was conducted and the resulting characterization is provided in Chapters 17 and 18. Environmental monitoring for chemicals is addressed in Chapters 19 and 20.

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CHAPTER 17

RELEASES OF CHEMICALS TO AIR

ABSTRACT

This chapter discusses releases of chemicals to the atmosphere from Savannah River Site (SRS) facilities. Releases were determined using emissions estimates from the air emissions inventory, Operating Permit Application, Toxic Release Inventory; environmental and effluent monitoring data, process records and/or information obtained by interviewing SRS personnel. Releases from manufacturing and processing facilities, power plants, generators, open burning, construction and maintenance work were included in the release estimates. Large amounts of sulfur dioxide, nitrogen dioxide and particulates (ash) were released from seven coal-fired power plants. Thousands of tons of chlorinated solvents were released from the raw materials area (M-Area). Releases of mercury, lead, manganese, nickel, nitric acid, chromium, cadmium and hydrogen sulfide from the SRS were estimated. Release estimates are summarized in the tables and conclusions at the end of this chapter.

INTRODUCTION

We determined releases of chemicals to air at the SRS using several types of information, including the air emissions inventory; estimates in the SRS's Air Information Reporting System (AIRS) database; estimates made in support of the Title V Operating Permit Application submitted to the State of South Carolina in 1996; the Toxic Release Inventory (TRI); special stack monitoring or ambient air monitoring studies; and worker knowledge of processes, operations, and releases. Information obtained from interviews with current and former workers has been particularly important for understanding how chemicals were used, disposed of, and may have been released in the 1950s and 1960s. A limited amount of attention was given to chemical spills and releases to Site streams in reports from the 1970s. Most of the information and records we have found on chemical use and release pertains to operations after 1985. If we assume that the amounts and types of materials used and the processes did not change significantly over the years, then we can extrapolate this data back in time from the mid- and late-1980s.

Most of the release estimates are based on very limited information. The emissions estimates in the Air Emissions Inventory, the TRI and the Operating Permit Application are reported in units of tons per year. Throughout this chapter, amounts are given in tons, pounds, and kilograms (1 ton = 2000 lb and 1 kg = 2.2 lb), whichever units best facilitate comparisons.

Key Records and Resources Used to Reconstruct the Use and Release of Chemicals

There are many regulatory requirements for air pollution characterization and reduction, such as the Clean Air Act; the State of South Carolina's Air Toxics Rule; the Superfund Amendments and Reauthorization Act of 1986 (SARA), which require emergency planning and annual reporting of potentially hazardous chemicals; the 1970 National Emission Standards for

Hazardous Air Pollutants (NESHAPs) Program; and the Clean Air Act Amendments of 1990. Because of these requirements, the Site must submit to the State and the State must submit to the U.S. Environmental Protection Agency (EPA), emission inventories, operating permits, annual reports of toxic chemical releases to the environment, and chemical inventories—all of which have been useful to this study. The reports have only been required since the mid- to late-1980s, and data on the use, storage, and release of chemicals before this time were not common.

Several sources of information on air emissions were used to determine source terms. This section describes data found in the following recent reports on air emissions: (a) the air emissions inventory and the AIRS database maintained by the SRS to help prepare reports needed to comply with EPA and South Carolina Department of Health and Environmental Control (SCDHEC) regulations, (b) the Standard 8 and Standard 2 Air Pollutant Reports sent to SCDHEC to comply with operating and permitting regulations, (c) the Title V, Part 70 Permit Application submitted to the SCDHEC in 1996, (d) available calculational notebooks, worksheets, and other records used to derive the emissions in the AIRS database and the Part 70 Permit Application, and (e) the Toxic Release Inventory (TRI) reported to the EPA. Information in these reports and database may be the only information available on the potential release of some of the chemicals of concern. Although they pertain to operations after 1985, and in some cases, after 1990, the information may be applied to earlier time periods and has been useful for determining hypothetical maximum emissions estimates.

Air Emissions Inventory and AIRS Database

Following promulgation of the South Carolina Air Pollution Control Regulation for Air Toxic Pollutants (Standard 8) in 1991, the SRS began to conduct an air emission inventory for all facilities onsite. The inventory was intended to

- Help demonstrate compliance with the requirements of Standard 8
- Fulfill the requirements of the Air Toxics Rule promulgated by the SCDHEC
- Provide information, required by the State and EPA, on release points for air pollutants
- Account for sources of pollution
- Better characterize releases from Site processes.

The inventory includes information on each source, such as stack height, diameter, location, pollutant emission rate, production, and operating rate. Data collection forms (referred to by site personnel as the Air Emissions Inventory Worksheets) were used in this effort. Information from the forms was entered into the AIRS database. The initial air emissions inventory was finished in 1993, and it estimated air emissions for the years 1985 through 1991. Air emissions estimates for 1990 were used, along with air dispersion modeling, to demonstrate compliance with Standard 8. The Air emissions inventory has been updated annually since 1993. The air emissions inventory data for 1994 were used for the 1996 Part 70 Operating Permit Application (Westinghouse 1996a).

In 1985 and 1986, the EPA and the State environmental agencies conducted a comprehensive audit of the Site. They asked that all stacks, vents, and other discharge points for atmospheric emissions be identified for all areas. The AIRS database also fulfilled this requirement. The AIRS information goes back to 1985, a year when the last four reactors in use were still running and both canyons were operating. With time, the information in the database

has become more complete and more effort has been put into calculating the estimates, resulting in improved estimation methods. The estimates for recent years are considered to be more defensible (Faugl 1996a).

SARA Section 313 requires annual reporting of releases to all media of toxic chemicals with usage exceeding 10,000 lb y^{-1}. The emissions inventory and AIRS database was put in place at the SRS to

- Respond to SCDHEC requests for a air toxic emissions inventory
- Provide a basis for operating permits
- Estimate air emissions for SARA regulations and reporting
- Provide baseline estimates to evaluate emissions reductions.

The Site has put all of the emissions estimates used for the various reports just described in an ORACLE database called the AIRS database. The emission estimates for each year are compiled in the database. Several reports use the information in the database, but the estimates are not available in a hardcopy report. Estimates must be extracted from the database. We requested several database excerpts.

The database has space for four types of emission estimates:

- Uncontrolled actual emissions
- Uncontrolled maximum design capacity emissions
- Controlled actual emissions
- Controlled maximum design capacity emissions.

Uncontrolled emissions are theoretical emissions that might occur if pollution control equipment was not in place or not operating. Controlled emissions account for the pollution control equipment in place and working. The actual emissions are those predicted under normal operating capacities and times. Maximum design capacity emissions are theoretical for 24 hours a day, 365 days a year, 8760 hours a year operations. Of most interest for this study for determining a best estimate of a source term are the controlled actual and controlled maximum design capacity emissions. These reflect likely actual emissions under real or typical operating conditions and maximum theoretical emissions at full production capacity after reduction by any pollution abatement equipment. The uncontrolled maximum design emissions represent an unrealistic, extreme upper bound scenario. All of these emissions were calculated, rather than measured; however, some measurements for sulfur dioxide and nitrogen dioxide (opacity) have been used to corroborate the estimates. The database includes a code to reference how the estimates were calculated. The codes for the estimation methods were

01 EPA AP-42
02 EPA speciate
03 Engineering calculations
04 Material balance
05 Stack test data
06 Monitor—sampler data
07 EPA CHEMDAT7-LAND7
08 EPA SIMS Model
09 MSDS Data Sheet
10 Other.

Almost all of the estimates were calculated with method 03, engineering calculations.

After reviewing the air emission inventory forms and worksheets that show the fields of data potentially available from the AIRS database, *RAC* requested an initial search for process releases from any facility for the chemicals of concern. We asked for a search of the emissions for 1985, 1987, 1990, and 1992 and a printout of the following fields for benzene, cadmium, chromium, hydrogen sulfide, lead, manganese, mercury, nickel, nitric acid, nitrogen dioxide, sulfur dioxide, tetrachloroethylene, trichloroethane, and trichloroethylene: chemical (pollutant); source ID; building number; building name; source type; source description; form (solid, gas, liquid); controlled maximum; and controlled actual emissions in tons per year. Some of the information on the location of the source was sensitive, so the request was abbreviated to eliminate this information to expedite security review. For some chemicals, known emissions from a source, for example mercury from the 291-H stack, were not in the database, so a search for 1994 and 1995 emissions was done for these missing estimates. Although more emissions estimates have been calculated each year, not all of the emissions of interest for the historical dose reconstruction study have been estimated.

After examining these data, another request for database information was made for key emission points and sources. We asked for emission type (point, volume or fugitive); height aboveground; height above building, diameter (of vent, stack etc.); exit velocity; exit temperature; release height; continuous detection (monitoring in place); estimation method (key 1-10); raw materials (used in process); process rate; product; pollution abatement equipment data; and control device (key). Not all of this information was available for all of the emission points. The results of the AIRS database runs for each chemical (except sulfur compounds, which were grouped together) were given *RAC* document numbers. This report references database runs as Faugl 1996a, Faugl 1996b, Faugl 1996c, Faugl 1996d, etc. because Timothy Faugl, with the Environmental Protection Department at the SRS, conducted the database searches or arranged for them to be done by a subcontractor.

The AIRS database includes emissions from maintenance and fabrication shops, controlled burns, welding operations, batteries, combustion engines, compressors, and generators. The uncertainties associated with the total release estimates are large enough to justify subtracting from our analysis all the emission points that are not process related (such as minor maintenance operations, small emergency generators, portable grinders, or welders) or those that contribute little to the total emissions. Many sources of chemical releases are very minor and localized and these were not considered by *RAC*, for example, electric oven exhausts, paper shredders, painting stations, brush cleaners, paint rollers, small diesel pumps, and mobile air compressors.

A typical diesel engine will release lead, oxides of nitrogen, oxides of sulfur, benzene, arsenic, cadmium, chromium, manganese, mercury, nickel, and organic pollutants. Emissions estimates for some individual diesel engines were also made in the Title V Operating Permit Application. Worksheets from the M-Area Air Emissions Inventory indicate that emissions of sulfur dioxide, nitrogen oxides, arsenic, beryllium, cadmium, chromium, mercury, manganese, nickel, and benzene were calculated for generators using AP-42 emission factors (Radian 1992a).

In the case of nitrogen oxides and benzene, the emissions estimates from generators and other engines are some of the highest at the SRS. The most numerous sources of cadmium, chromium, mercury, and nickel emissions to the air were generators. The emergency generators are used in emergency power failure situations and most are usually started and run once or twice

a year and tested. Some are tested monthly or quarterly. Many are tested for about 1 hour each week to keep batteries charged. Generally, all emergency generators are run less than 100 hours per year. A few generators are the main power source for equipment, and some run continuously at a maximum of 8760 hours per year assumed for the estimates in the AIRS database. Emissions from generators are addressed in a section of this chapter called, "Diesel-powered Generators."

Most of the emissions points for metals were grinding, cutting, torch cutting, belt sanders, lathes, drilling, and other similar maintenance facility and machine shop operations. Estimates for releases of cadmium, manganese, mercury, and nickel were estimated for the H-Area and K-Area coal pile and transfer operations, and chromium and lead were included in estimates for the K-Area coal pile. Manganese releases were included for the D-Area coal pile. It appears that the contractor or Westinghouse Savannah River Company department responsible for the K-Area coal pile estimates provided the most comprehensive estimates. The K-Area coal pile releases included all of the metals, while the H-Area coal pile estimates included four metals, and the other coal piles included none or no emissions estimates were reported. Why other metal releases were not included for each coal pile is unknown.

Estimates of total emissions generally increased with time after 1985 because more processes were included in the inventory. For example, nickel emissions for 232-H included six processes in 1985 and eight in 1987 and 1990. All eight processes operated in 1985, but emissions were not estimated for metallography and cutting operations before 1987. In 1992, the number of emissions points reported more than doubled because release points were added from the vitrification process and there was an increased reporting of tanks, generators, and other sources.

Because different contractors may have been responsible for calculating emission estimates from different facilities or areas, similar processes may have different estimates and some emissions may be reported for one facility but not another. The emission estimates for F-Area and H-Area were developed by two different contractors. The pollution control equipment, maximum design rates, tank capacities, etc. should have been the same, but emission estimates were different. For example, the highest benzene emissions for 1985 were for the two paraffin tanks in the F-Canyon building. H-Area did not report such tanks. The highest emission for oxides of nitrogen in 1985 was the second uranium cycle discharge to the 291-F stack (an emission of 49.3 ton y^{-1}). No emission for oxides of nitrogen was listed for H-Canyon stack in 1985 or any H-Canyon processes except the diesel generators for the canyon exhaust fan house, two generators used in the HB-Line and the metal manufacturing processes.

Some of the processes and the locations of some of the emissions points are sensitive and they may not be identified specifically in this analysis; however, all of their emissions are included in the source term estimates for individual compounds. In the AIRS database, all of the emissions points at the SRS are given an identification number. For example, F-Canyon emission points are numbered 001–0059, for a total of 59 emission points for that one building, 221-F. The emission points include major points, such as the stack, vents and hood exhausts, and minor points, such as welding booths, grinding areas, and various tank and vessel vents. The pollution control equipment is described, for example scrubbers, high-efficiency particulate air (HEPA) filters, adsorbers, sand filters, and dust filters designed to remove particulates and oxides of nitrogen (Faugl 1996a). Almost all of the radionuclides and chemicals of concern were discharged from the stacks. For the purposes of this study and to avoid identifying emission sources that are sensitive, all of the emissions from the canyon building were combined. The SRS

emission point identification numbers and all of the sources going into each emission point were made available to *RAC*, but this information was not compiled here because of the sensitive nature of some of the information. For the purposes of this study and transport modeling and dose calculation in subsequent phases of the dose reconstruction, it should be sufficient to combine the emissions for each area to one or several points with appropriate release heights.

Data from the AIRS database runs were reviewed and useful information was compiled for each chemical of concern. We compiled a description of the types of emissions reported, the controlled maximum design emissions estimates, and the controlled actual emission estimates in tons per year for major sources from the AIRS database. This information is presented in tables for each chemical in this chapter.

Part 70 Operating Permit Application

The Title V Clean Air Act amendments promulgated in 1990 required that estimates of possible releases at maximum capacity be developed and the SRS was required to submit a permit application, under R61-62.70 Title V Operating Permit Program, Clean Air Amendments for Title III, Part 70 Regulations. The SCDHEC Air Pollution Control Regulation, Title V Operating Permit Program requires that all of sources of air pollution subject to the regulations have a permit to operate that assures compliance with all applicable regulations. To obtain a permit, the regulation requires that each source must submit a Part 70 Permit Application that identifies all sources of air emissions at the facility and identifies all state and federal regulations and requirements pertaining to the sources. Many of the regulations are quite general and have been applied to the Site as a whole rather than to individual facilities or sources (Westinghouse 1996a).

The State of South Carolina requires new SRS construction permits to include emissions information and modeling analysis for air pollutants. The emission rates are calculated based on process knowledge and EPA Air Pollution Emission Factors (AP-42) or other models available through the EPA. The maximum potential emission rates used for the dispersion modeling are based on maximum estimates of operating capability based on process design capabilities while operating within permit restrictions. Some of the modeling for metal emissions has been done using actual operation times and emission rates rather than potential maximum values.

The permit currently on file for the SRS was prepared in March 1996 in 23 volumes with three additional binders of amendments from 1997 (Westinghouse 1996a, 1996c, 1997c). This document is kept by the Environmental Protection Department in 742-A. We reviewed these binders and photocopied sections relevant for Phase II of the project.

The SRS developed an emission source identification system for the air emissions inventory. Point sources, volume sources, and area sources were identified. Exhaust points, sources, and pollution control devices were numbered. Fortunately, the same codes were used in the air emissions inventory and the Part 70 Operating Permit Application. The identification code and procedure for implementing the identification system was included in Chapter 5 of the permit, which *RAC* photocopied. Some of the information about process throughputs and concentrations of chemicals is currently sensitive with respect to national security and was not photocopied as a part of the application or worksheets supporting it. *RAC* researchers with the appropriate clearance did review this and other classified information relevant to the dose reconstruction project.

Title V of the Clean Air Act amendments require operating permits that detail the emission rates of regulated pollutants, describe fuels and materials use, emission points, control equipment, monitoring equipment, stack or release point parameters, area sources, and fugitive sources. The Criteria Pollutants (nitrogen oxides, sulfur dioxide, ozone, lead, and particulates) are subject to the National Ambient Air Quality Standards (NAAQS) and were addressed in the Standard 2 report. Hazardous air pollutants regulated under the 1970 NESHAPs Program, such as asbestos, beryllium, and mercury were addressed in the Standard 8 Report (Westinghouse 1993a). These reports are described in more detail in the next section of this chapter.

The Part 70 Operating Permit Application included large sections of air emissions inventory-derived emissions calculation sheets that list the approach, assumptions involved, input data, and emissions for processes (such as abrasive cleaning, bulk material handling, cutting, grinding and metal fabrication, diesel combustion, open burning, tanks and vessels, and welding). Calculations associated with many of these processes are important for establishing emission estimates for the chemicals of concern for the dose reconstruction. Those calculations used to establish estimates for emissions of the chemicals of concern are discussed in the section for each chemical.

The permit application may also have information on the design capacity of process equipment; the major raw material and quantity used; and the product for individual process source, operating schedule and production rate, stack height, diameter, distance to the plant boundary, control equipment, and maximum uncontrolled and controlled emissions estimates for NAAQS pollutants and Hazardous Air Pollutants or Air Toxics. Monitoring devices and regulatory requirements and compliance status are also given if applicable.

The assumptions and values used to calculate the emission estimates were described in the Title V Operating Permit Application and accompanying material. The calculations were based on a review of process flowsheets, safety analysis reports, engineering notes, and interviews conducted in 1984, 1985 and 1986. For example, the F-Canyon Purex process dissolver and head end were evaluated for the permit based on interviews with F-Area engineers, historical process data, abatement devices and their efficiencies, stoichiometric calculations, and characteristics of the fuel dissolved. In most cases, the information used for the estimates appeared to be more detailed than any *RAC* could collect in 1997; therefore, we consider these data to be the best available for estimating releases. The assumptions about operations, throughputs, volumes, and usage seem conservative. The actual estimates are likely to be upper bound rather than central or best estimates.

Included in the permit, for most sources, were air pollution control equipment forms that include efficiency estimates, gas flow rate, pressure drops, gas temperature, and description of the unit. These data were used by the Site and contractors to estimate the controlled emissions. The information does not indicate when the equipment was first put into place.

After inquiring about where to find supporting documentation and calculations for the AIRS estimates, we were advised that similar, if not identical, methods were used by the Facilities and the Environmental Protection Departments to calculate emissions for the Title V Part 70 Air Permit Application.

Generally, these records were called Air Emissions Inventory Data Sheets, Worksheets, or Supporting Information. Most of these records have been kept in the Environmental Protection Department at the SRS. Calculations that the facilities may have used to make the estimates may have been submitted as worksheets and supplemental information to the information submitted for the AIRS database and the Operating Permit Application. The worksheets were sometimes

submitted with the estimates to the Environmental Protection Department, but often, supplemental information and worksheets were kept at the facility. Environmental Protection Department personnel thought that once their contractor had entered the annual AIRS information into the database, the supplemental data were usually sent to be microfiched and then to central records. Most the information used for this study was found in the Environmental Protection Department records. A few pertinent files were found in central records, and records that were kept at the facilities were also photocopied for this study.

The permit application also included useful descriptions of some of the most important processes and the air exhaust and liquid effluent discharges associated with them. Chapter 15 summarizes the useful process or facility descriptions. Many of the storage tanks addressed in the permit application were waste tanks and process chemical tanks associated with new facilities, such as the consolidated incineration facility, Defense Waste Processing Facility (DWPF), in-tank precipitation, saltcrete, and others. These processes are subject to air quality regulations designed to protect the offsite public.

Standard Two and Standard Eight Reports

SCDHEC Bureau of Air Quality Control promulgated Operating and Construction Permits Regulations in June 1991. This health-based regulation established maximum allowable ambient air concentrations at the facility boundary in micrograms per cubic meter for 256 identified pollutants. To comply with this rule, the SRS submits air emission source information and modeling input/output data and maintains the air emissions inventory and the AIRS database for all potential criteria and toxic air pollutant emission sources. The AIRS identified 138 Standard 8 air toxics with the potential to be emitted. Emissions calculations were based on process knowledge, EPA emissions factors (AP-42), and other EPA or vendor-developed models. Bowman's Industrial Source Complex-Short Term II (ISC-ST II) dispersion model was used to demonstrate whether any of the identified air toxics have compromised air quality standards. The first report submitted to comply with the regulation, (which was also the most recent report as of 1997), was sent to SCDHEC in 1993 and was based on the 1990 air emissions inventory data and 1991 meteorological data. The maximum emissions were based on maximum design capabilities or maximum potential while operating with permit restrictions in place. None of the air toxics concentrations modeled exceeded the standards. The modeling results showed that the SCDHEC standard boundary concentration would not be exceeded by maximum potential emissions from the SRS. However, the potential emissions for two of the criteria pollutants could be in excess of the air quality standards. Using maximum values, the modeled concentrations of sulfur dioxide and nitrogen dioxide exceeded the standards at the Site boundary near D-Area and TNX. The largest contributing source of emissions was predicted to be the D-Area powerhouse boilers. Minor exceedances were also predicted for the A-Area boundary for the 3-hour sulfur dioxide standard (Dukes 1993; Westinghouse 1993a).

The Environmental Protection Department staff submitting the report to SCDHEC believed that the exceedances were due to extremely conservative approaches being used in the modeling analysis. They planned to remodel for the standards using more accurate operating data and system configurations. They also said they planned to develop a control strategy plan for the D-Area powerhouses (Dukes 1993).

Table 17-1 lists the air pollution standard, estimated concentration in air at the Site boundary, and the percentage of the standard this concentration represented for the chemicals of concern for this study.

Table 17-1. Air Pollution Standards and Concentrations Predicted by Air Dispersion Modeling at the SRS boundary, in 1993, for Air Toxics and Criteria Pollutants[a]

Chemical	Maximum allowable concentration set by SCDHEC ($\mu g\ m^{-3}$)	Concentration predicted at the Site boundary ($\mu g\ m^{-3}$)	Percent of the standard
Air toxics			
Chlorine	75.0	7.63	10.17
Nitric acid	125.0	50.97	40.77
Trichloroethylene	6750.0	6.43	0.10
Hydrogen sulfide	140.0	0.20	0.14
Tetrachloroethylene	3350.0	2.0	0.06
Aldicarb	6.0	.007	0.12
Benzene	150.0	31.71	21.14
Cadmium	0.25	0.00028	0.11
Hydrazine	0.50	0.0018	0.36
Manganese	25.0	0.82	3.29
Mercury	0.25	0.01	5.57
Nickel	0.50	0.27	54.21
1,1,1-trichloroethane	9550.00	80.83	0.85
Criteria pollutants (standard)			
Sulfur dioxide (3 hour)	1300	2319.06	178.39
Sulfur dioxide (24 hour)	365	1039.10	284.69
Sulfur dioxide (1 year)	80	75.21	94.02
Nitrogen dioxide (annual)	100.0	125.41	125.42
Lead (quarterly mean)	1.50	0.0015	0.10

[a] Source: Dukes (1993).

The modeling results predicted that the 3 and 24-hour standards for sulfur dioxide and the annual standard for nitrogen dioxide could be exceeded. The criteria pollutant with the largest potential offsite impact was sulfur dioxide, modeled to be 289.69% of the maximum 24-hour average Site boundary concentration at maximum operations.

Plans Applicable to Chemical Releases

The Best Management Practices Plan was required by the State of South Carolina, which administers the National Pollutant Discharge Elimination System (NPDES) program in South Carolina, and regulatory requirements of the Federal Clean Water Act. The plan was required to "identify and control the discharge of hazardous and toxic substances listed in 40 CFR Part 122"

(Westinghouse 1997a). The plan has been maintained by the Environmental Protection Department in 742-A. It contained potential spill sources for each area and listed the chemical, department, tank, building, outfall, and equipment to which it is related. It also described the quantity and secondary containment that was in place (Westinghouse 1997a).

A Spill Prevention Control and Countermeasures Plan was also required for identification and pollution prevention measures for oil facilities at the Site. This document characterizes polychlorinated biphenyl (PCB) oils in transformers onsite, as well as non-PCB oils. It lists the underground and aboveground fuel and oil tanks, tank trucks, and pipelines that store oils, fuels, and solvents. It also describes liners, basins, concrete pads, and other secondary containment measures in use. Outfalls are listed and drawings of the surface water flow, basins, and facilities were included for some of the areas. The Spill Prevention Control and Countermeasures Plans for both 1987 and 1997 (Graham et al. 1987; Westinghouse 1997b) were reviewed and entered into the Phase II document database.

From these plans, the total amount of chemicals, the total number of tanks, and volume of a chemical could be compiled. The plans are also a relatively condensed source of information about which areas, processes, and facilities are associated with which NPDES outfalls. Data on the storage of toxic chemicals could also be derived from inventories, such as those taken for the SARA Title III reports, which we used to select chemicals of concern using methods described in Chapter 16.

Toxic Release Inventory

From 1987 to 1995, the TRI reported to the EPA and SCDHEC included the pounds of benzene, lead, manganese compounds, nitric acid, and 1,1,1-trichloroethane released per year from the SRS (Westinghouse 1996b). It is likely that in years with no amount reported some amount below the reporting limit was released. Release estimates reported in the TRI are summarized in the sections on each chemical that follow.

CHEMICALS RELEASED TO AIR

Ammonia

Ammonia was used onsite but not in sufficient quantities to result in a screening ratio greater than 1. Like sulfur dioxide and the oxides of nitrogen, ammonia was a product of the dissolving processes at the canyons, TNX operations, and Fuel Production Facility processes and was released from process stacks. Aluminum dissolution in sodium hydroxide can produce hydrogen gas, a serious explosion hazard, so sodium nitrate was added to the reagents producing ammonia and nitrate rather than hydrogen. About 0.167 kg of ammonia was produced per kilogram of aluminum dissolved. A large percentage of the ammonia (50–80%) could remain in solution or condense before the gas was exhausted, depending on the temperature of the off-gas system. The total ammonia released was estimated to be about 0.050 kg NH_3 kg^{-1} of aluminum dissolved (Allender 1985). Ammonia is irritating but is relatively nontoxic and would not be expected to have been a concern offsite.

Ammonium Nitrate

Ammonium nitrate flakes or spalls were released to the air from the stacks in F-Area. These were of concern because they caused radioactive contamination of sidewalks and other surfaces in F-Area. Sodium hydroxide was used in F-Area to dejacket (remove the cladding) from slugs. This reaction gave off ammonia (NH_3), which could combine with nitric acid fumes from other parts of the process and result in a precipitate of ammonium nitrate (NH_4NO_3). This precipitate adhered to the inside of the stack and was jarred out periodically to result in the 'spalls' on the sidewalk. The precipitate was water-soluble, so washing the stack with water from time to time helped remedy the problem. A 1957 monthly report says that solid particles discharged from the F- Area stack during August and September were principally ammonium nitrate and mortar, carrying ruthenium and plutonium activities and that flushing the stack with water removed essentially all of the ammonium nitrate (Du Pont 1957a). The wash water was probably transferred to a sump. Sometime after 1962, the ammonia and dissolving effluent was separated by the addition of two small stacks that run along the main stack. The ammonia was discharged out the small stacks and the rest of the dissolving/nitric acid process fumes were exhausted up the large stack (Pickett 1996). Ammonium nitrate was also periodically removed from the hot and warm canyon process vessel vent filters. A monthly report from 1976 described the removal of ammonium nitrate canyon process vessel vent filters after a 28-month operating period. A total of 4900 kg of ammonium nitrate was removed during two flushes of each filter. The authors expressed concern that ammonia in waste tanks from these flushes might reach explosive limits (Du Pont 1976). The primary concern with the ammonia nitrate flakes that came out of the stack was their radioactivity. This is addressed in Chapter 4.2, in the discussion about releases of beta emitters.

Benzene

Gasoline and volatile solvents, like xylene and toluene, contain benzene. Benzene was released from gasoline tanks, gasoline transfer equipment and tanks, and equipment for other solvents and fuels used at the SRS.

Most of the emissions for benzene in the AIRS database were for the DWPF, which is not being considered as a part of this study because it began operations in 1990. Research and development for the DWPF was conducted at TNX through the 1980s, and this testing also resulted in the release of benzene, which was included in our estimates. The Standard 8 modeling results submitted to SCDHEC in 1993 stated that, in 1991, benzene was released from sources in all of the areas at the SRS. The maximum 24-hour average Site boundary concentration was calculated to be 31.7 $\mu g\ m^{-3}$, well below the ambient air standard of 150 $\mu g\ m^{-3}$ (Westinghouse 1993a).

Emission estimates for benzene in the AIRS database included those from 10 waste storage tanks, each emitting less than 10^{-6} ton y^{-1}. Many gasoline tanks, diesel fuel storage tanks, and underground fuel tanks were listed with emissions ranging from less than 10^{-6} to 3.8×10^{-3}. Benzene emissions were also calculated for gasoline generators, diesel generators and emergency generators (Faugl 1996b). Some of the larger emissions estimates are listed in Table 17-2.

Some of the additional emissions noted in 1990 were for operations at the vitrification plant. Numerous organic liquid storage tanks were listed with small emissions. Emission estimates for

A-Area waste tanks were less than 10-6 ton y^{-1}. Waste storage tank purges were listed but were blank except for one. Waste storage tank number 48 in H-Area had a maximum emission estimate for tank purges of 2.65 lb h^{-1}. No ton per year values were given and how often the tank was purged is unknown. If we assume that the tank was continuously purged, as much as 11.6 ton y^{-1} may have been released from this tank. The highest benzene emissions for 1985 were from the two paraffin tanks in the F-Canyon building. H-Canyon did not report such tanks. The next highest emission was for the F-Area seepage basin, followed by the diesel generators in the generator house in H-Area, Building 254-05 with actual emission estimates of 9.45×10^{-2} ton y^{-1} and 9.36×10^{-2} ton y^{-1} (Faugl 1996b).

Benzene emissions were calculated for 18 waste storage tanks; 31 gasoline, diesel, and other fuel tanks; 2 paraffin tanks; 48 generators 2 diesel pumps; 5 painting and related activities; and 2 tanker truck loading stations (Faugl 1996b). If we assume that gasoline, diesel fuel, paraffin, and organic solvent use in 1987 was similar to uses in years past, we could apply the 1987 totals to the entire time period and estimate that 2.3 ton y^{-1} actual and 17.8 ton y^{-1} maximum of benzene may have been released. This provides an estimate of 87 ton actual and 676 ton maximum of benzene emissions from 1952 to 1989. We would expect that more vehicles would have been in use in 1987 than in earlier years and that using the 1987 values would tend to overpredict the fuel use and resulting emissions. It may be that use in the 1950s and 1960s was more careless and more care to prevent spills, worker inhalation, and transfer errors was taken in the 1970s and 1980s. Fuel efficiency may also have been important for consumption during the energy crisis in the 1970s. More information on fuel use can be found in the section about gasoline in this chapter.

The Part 70 Operating Permit Application contained emission estimates for almost all of the diesel and gasoline fueling stations as well as storage tanks thought to emit volatile organic compounds (VOCs). The air quality permit gave example calculations for gasoline tanks and gasoline and fuel-dispensing stations, which were generally considered exempt because of insignificant activity, based on emission levels. A maximum emission rate for a dispensing station with four pumps was reported to be 0.0166 ton y^{-1} for benzene. Maximum emissions estimates for a 5000-gal underground gasoline fuel tank was 0.0215 ton y^{-1} for benzene (Westinghouse 1996a).

Two identical aboveground gasoline tanks at 715-6A, with a capacity of 10,000 gal each, are used to store gasoline for the entire Site. These tanks have been used to fill tanks at the gasoline station and to fill Site gasoline trucks. Gasoline vapors are continuously released from the tank vents. Because of the large volume of the tanks, the maximum uncontrolled emissions estimates for the transfer pump are high and would exceed air toxics limits. However, the actual throughput was about 2000 gal of gasoline per year, which corresponds to what the air quality permit terms as “insignificant air emissions of all components.” The actual emissions estimate for benzene from both tanks totaled 4.13×10^{-2} ton y^{-1} (Westinghouse 1996a). We can multiply this by 38 years for the years 1952–1989 for a total estimate of 1.6 ton for the entire time period.

Table 17-2. AIRS Database Emissions Estimates for Benzene for 1985, 1987 and 1990[a]

Year Source Description	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
1985 Total	**1.16×10^{1}**	**1.50**
F-221 Canyon, N-Paraffin Tank 21	4.0×10^{-1}	4.0×10^{-1}
F-221 Canyon, N-Paraffin Tank 22	4.0×10^{-1}	4.0×10^{-1}
F-Area Tank Farm degreaser	2.50×10^{-3}	2.50×10^{-3}
G F-Area Seepage Basin	2.14×10^{-1}	2.14×10^{-1}
G Old Sanitary Landfill	1.21×10^{-2}	1.21×10^{-2}
S Area Organic Waste Storage Tank Building 430	3.00×10^{-1}	0
T-Area Building 672, DWPF Semiworks Building Process tank	5.0	2.70×10^{-3}
T-Area 678 Chemical Semiworks Building, Organic Storage Tank A-3	1.00×10^{-3}	1.00×10^{-3}
T-Area 678 Chemical Semiworks Building, Organic Storage Tank	1.00×10^{-3}	1.00×10^{-3}
E 643007 burial ground, 643-7E buried waste	1.0×10^{-6}	1.0×10^{-6}
T Pilot Plant building incinerator	1.86×10^{-4}	2.33×10^{-7}
1987 Total	**1.78×10^{1}**	**2.29**
F-221 Canyon, N-Paraffin Tank 21	4.0×10^{-1}	4.0×10^{-1}
F-221 Canyon, N-Paraffin Tank 22	4.0×10^{-1}	4.0×10^{-1}
F-Area Tank Farm degreaser	2.50×10^{-3}	2.50×10^{-3}
G F-Area Seepage Basin	2.14×10^{-1}	2.14×10^{-1}
G Old Sanitary Landfill	1.21×10^{-2}	1.21×10^{-2}
S Area Organic Waste Storage Tank Building 430	3.00×10^{-1}	0
T-Area Building 672, DWPF Semiworks Building Process Tank	5.0	2.70×10^{-3}
T-Area 678 Chemical Semiworks Building, Organic Storage Tank A-3	1.00×10^{-3}	1.00×10^{-3}
T-Area 678 Chemical Semiworks Building, Organic Storage Tank	1.00×10^{-3}	1.00×10^{-3}
E 643007 burial ground, 643-7E buried waste	1.0×10^{-6}	1.0×10^{-6}
T Pilot Plant building incinerator Building 667	1.86×10^{-4}	1.79×10^{-6}
Naval Fuels Glove Boxes, Building 247	1.50×10^{-1}	1.50×10^{-1}
1990 Total	**1.64×10^{1}**	**2.06**
Open Burning Fire Training Pit #1, in D-Area, fire fighting simulator	8.92×10^{-4}	1.25×10^{-4}
F-221 Canyon, N-Paraffin Tank 21	4.0×10^{-1}	9.0×10^{-2}
F-221 Canyon, N-Paraffin Tank 22	4.0×10^{-1}	9.0×10^{-2}
F-Area Tank Farm degreaser	2.50×10^{-3}	2.50×10^{-3}
G F-Area Seepage Basin	2.14×10^{-1}	4.22×10^{-3}
G Old Sanitary Landfill	1.21×10^{-2}	1.21×10^{-2}
S Area Organic Waste Storage Tank Building 430	3.00×10^{-1}	0
H-Area Seepage Basin	1.85×10^{-10}	5.51×10^{-12}
T-Area Building 672, DWPF Semiworks Building Process tank	5.0	8.20×10^{-3}
T-Area 678 Chemical Semiworks Building, Organic Storage Tank A-3	1.00×10^{-3}	1.00×10^{-3}
T-Area 678 Chemical Semiworks Building, Organic Storage Tank	1.00×10^{-3}	1.00×10^{-3}
T Pilot Plant building incinerator	1.86×10^{-4}	1.79×10^{-6}

[a] Source: Faugl (1996b).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Purges from waste tank 22, a 1,300,000-gal tank installed in 1962, resulted in the release of benzene. Although some measurements were said to have been taken, they were not included in the worksheets and no report or reference to them could be found. The maximum emission rate for the tank purge was calculated to be 0.044 ton y^{-1}. The tank was permitted to release an annual average of 0.01 lb h^{-1} of benzene. It is unclear from the air permit forms whether other waste tank purges also result in benzene releases. The total VOCs released from the tanks was compiled for the air emissions inventory, and this value may include benzene (Westinghouse 1996a; Faugl 1996b). We can estimate that at a rate of 0.044 ton y^{-1} for the 28 years from 1962–1989, the total benzene emissions from regular purges of this tank would be a maximum of 1.2 ton.

The 760-2G diesel fuel dispensing station was included in the permit application. Fugitive emissions of benzene had estimated emissions rate of 1.19×10^{-2} ton y^{-1}. Estimates for the 620-G diesel fuel station fugitive emissions were 1.44×10^{-2} ton y^{-1} for benzene. Emission estimates for xylenes and toluene were similar. The estimates involved worse case assumptions of continuous emissions from seals, valves, and flanges. Benzene was assumed to be 6% of the total VOCs in diesel fuel and 7% of the total in gasoline. Pumps were assumed to operate at maximum capacity, and lines were assumed to be filled with fuel at all times. Emissions for the gasoline refueling station were 1.90×10^{-2} ton y^{-1} for benzene for each of the gasoline dispensing stations 715-1G and 715-2G (Westinghouse 1996a).

Benzene emissions from process-related operations in 1985 and 1987 included 0.4 ton y^{-1} from the F-Canyon building, 2.50×10^{-3} ton y^{-1} from the F-Area tank farm, 0.214 ton y^{-1} from the F-Area seepage basins, and 0.012 ton y^{-1} from the sanitary landfill (Westinghouse 1996a). These are relatively small amounts compared to the amount probably released from gasoline and fuel and stations.

The actual emissions estimate for the 200-H emergency power generator, assuming 2080 hours of operation each year, was 6.46×10^{-5} ton y^{-1} for benzene (Westinghouse 1996a). Information in the K-Area Air Emission Inventory Calculation Procedures Notebook, prepared by Radian in 1992 for the Air Emissions Inventory, included a computer printout of a speciation report for VOCs emitted from the 184-K powerhouse boilers (Radian 1992b). It is not clear if releases of the compounds were measured or estimated but no concentration data were provided. The design maximum emission for benzene was calculated to have been 0.17 ton y^{-1}, and the actual emission was 0.075 ton y^{-1} in 1988 based on the amount of coal burned (Radian 1992b). The air quality permit application estimated that the two 784-A coal-fired stoker boilers burned about 8500 ton of coal per year each and released a maximum total of 2.27×10^{-2} ton y^{-1} of benzene (Westinghouse 1996a). Estimates for benzene from coal burning were not included in the AIRS database information. If the air permit calculation for A-Area boilers is applied to coal burning Site-wide, then an estimate of 0.68 ton y^{-1} benzene corresponds to burning 500,000 ton of coal per year, which was the amount reportedly burned in the early and mid- 1970s. The emissions sources reported in the AIRS database for F-Area and H-Area should be similar, but F-Area included the canyons, seepage basin, and paraffin tanks, while H-Area included generators and the seepage basin. The F-Area emissions totaled about 2.04 ton y^{-1} maximum, while H-Area emissions totaled about 6.5×10^{-5} ton y^{-1}. The emissions reported for the F-Area seepage basin were 9 orders of magnitude greater than those estimated for the H-Area seepage basins. If we assume that all of the sources in H-Area were also in F-Area, then maximum emissions of about 2.04 ton y^{-1} should apply to both areas. Fuel facilities tank emissions, including purges and waste

areas such as the sanitary landfill, totaled about 0.14 ton y^{-1}. Totaling the individual estimates for the largest sources reported by the AIRS database in 1985, 1987, or 1990 totals 4.2 ton y^{-1}.

From 1987 to 1995, the TRI that was reported to the EPA and SCDHEC included estimates of the pounds per year of benzene released from the SRS (see Table 17-3).

Table 17-3. Benzene Release from the SRS (1989–1995)

Year	Air emissions (lb y^{-1})	Ton y^{-1}
1989	1590	0.79
1990	2850	1.42
1994	5878	2.94
1995	3000	1.50

It is likely that in years with no amount reported, some amount below the reporting limit was released (Westinghouse 1996b). It is important to recognize that automobile and truck traffic, refueling, refinery operations, and many industrial operations also release benzene in similar quantities. The benzene released from the SRS historically has not been the result of a special process or large-scale use. Most has come from fueling and engine use.

In summary, the AIRS database actual emissions estimates seem to range from 1.5 to 17.8 ton y^{-1}; the TRI estimates range from 0.79 to 2.94 ton y^{-1}. Emissions from coal burning were probably less than 0.68 ton y^{-1}. Taken together, the total maximum benzene emissions for the entire Site in the late 1980s from the sources described above were likely to have ranged from 1.8 to about 18 ton y^{-1}. If we assume that benzene releases from 1952–1989 were similar to the values estimated for 1985–1990, then a total benzene release for SRS operations from 1952–1989 might be estimated between 68 and 684 ton. The highest maximum release total would correspond to 684 ton for the entire period. The central value is more likely to be between 60 and 200 ton for the period from 1952–1989.

Although there was more than one available estimate for benzene released from the Site, the range of 68 to 684 ton over the 38-year operating period spans only one order of magnitude and is probably quite reasonable given the available information. Uncertainty calculations would not offer any more detailed information on benzene, and it is difficult to measure uncertainty in estimating historical releases from current data. For these reasons, uncertainty calculations for benzene were not made.

Cadmium

Inhalation of cadmium can cause lung cancer. Cadmium was not used in the F-Area, H-Area, A-Area or M-Area processes. However, cadmium was found to be elevated at the Four Mile Creek seepline and in the sediment of the M-Area settling basin. The source is unknown. Evidence of cadmium release to the air from plant processes was not found.

Cadmium has been released from the combustion of fuel and coal. The AIRS database contained emission estimates for cadmium from 41 emergency and other diesel generators and pumps and a variable number of welding and brazing operations. Cadmium emissions from the coal storage piles in H-Area and K-Area were also calculated for the AIRS database (Faugl 1996c). Table 17-4 contains the total cadmium emissions reported in the AIRS database.

Table 17-4. Total Cadmium Emissions Reported in the AIRS Database

Year	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
1985	5.83×10^{-3}	4.97×10^{-4}
1987	5.96×10^{-3}	4.71×10^{-4}
1990	6.38×10^{-3}	3.48×10^{-4}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Emissions estimates for individual sources were provided in Table 17-5. The estimates were the same for the three years examined. The estimates for H-Area and K-Area were calculated using conservative assumptions.

Table 17-5. Cadmium Emission Rates for 1985, 1987, and 1990

Source description	Maximum (ton y^{-1})	Actual[a] tons (y^{-1})
H-Area powerhouse Building 284; coal storage pile	1.70×10^{-4}	1.0×10^{-5}
H-Area powerhouse ash sluice sump	1.45×10^{-5}	3.16×10^{-6}
K-Area coal pile	5.40×10^{-6}	5.40×10^{-6}
K-Area coal storage runoff basin	5.40×10^{-6}	5.40×10^{-6}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

The highest release estimated for cadmium in 1985, 1987, and 1990 was 1.35×10^{-4} actual ton y^{-1} from the H-Area diesel house diesel generator, followed by 2.0×10^{-5} for each of eight diesel generators in K-Area engine house 108001 and 108002 (Faugl 1996c). Emissions from the powerhouse from coal combustion were not given, and emissions from D-Area powerhouse coal and ash handling and burning were not included. We assume that emissions from D-Area coal pile and ash basin would have been higher than for H-Area or K-Area because more coal was stored and burned and more ash was created and disposed of in D-Area.

Actual emissions estimates in the operating permit application for metals from the fuel oil-fired package boilers in K-Area included cadmium with, 2.04×10^{-4} ton y^{-1} (Westinghouse 1996a). The coal crushing operation at 784-A was also evaluated for the air quality permit. No data about the cadmium content of the coal burned were found. The air quality permit application, using AP-42 values, estimated that the two coal-fired stoker boilers in 784-A burned 8500 ton y^{-1} of coal each and released a total of 6.25×10^{-3} ton y^{-1} of cadmium (Westinghouse 1996a). If 6.25×10^{-3} ton of cadmium was produced from burning 17,000 ton of coal, then burning 500,000 ton Site-wide may have produced 0.184 ton (367 lb or 167 kg) of cadmium each year. Releases from coal burning dominate the emissions.

If we assume combustion engines were used the same amount in past and recently, then the average of the 3 years AIRS database totals about 6.05×10^{-3} ton y^{-1} maximum and 4.38×10^{-3} ton y^{-1} actual. When we add the 0.184 ton y^{-1} cadmium produced by coal burning during the

maximum coal burning year (1972), we estimate a total of approximately 0.19 ton y^{-1} (380 lb or 172 kg) of cadmium were released each year. The uncertainty in this estimate is very large. We do not know how the coal burned in A-Area in 1994 relates to the coal burned throughout the Site in earlier years. We do not know how much coal was burned before 1972 or the cadmium content of the coal. We do not know how many generators and other small engines were used in the past compared to their use in 1985, 1987 and 1990. The estimates in the permit and the AIRS database, on which this estimate is based, are conservative so our estimate likely overestimates the release. The available release estimates, however, are all point values. Calculating uncertainty from these values would not be useful, and we have little other information to support ranges of releases. Therefore, uncertainty was not calculated for cadmium. Cadmium releases from the SRS to surface water from coal and ash seem to be more worthy of concern than cadmium releases to air. Releases to surface water are discussed in Chapter 18.

Chlorinated Solvents

Three solvents were used in large quantities at M-Area and in moderate amounts at other facilities at the SRS and are evaluated in this phase of the study: tetrachloroethylene, trichloroethylene, and 1,1,1-trichloroethane. Tetrachloroethylene is also called perchloroethylene, Perc, Perclene® and PCE. Trichloroethylene is also called Trike, Triclene®, and ethylene trichloride and is often abbreviated as TCE. Trichloroethane is abbreviated TCA and is also called methyl chloroform. Abbreviations for the solvents have varied over time and were different in different Site reports. Care must be taken in interpreting tables where TCE has been used to refer to both trichloroethylene and trichloroethane. To avoid any confusion in the text of this report, these solvents will not be abbreviated.

Trichloroethylene was used from startup in 1952 until 1962 in Building 313-M and until 1970 in Buildings 320-M and 321-M. Tetrachloroethylene was used from 1962 in Building 313-M and from 1970 in Buildings 320-M and 321-M. In 1979, tetrachloroethylene was no longer used and was replaced with 1,1,1-trichloroethane (Gordon 1982; Christensen and Gordon 1983).

Many of the reports characterizing groundwater contamination in M-Area and documents characterizing liquid effluents for planning the effluent treatment facility have estimated the amount of chlorinated solvents that were discharged to the process sewers, seepage basins, and Tim's Branch. Often, the same estimates seem to be perpetuated in subsequent reports, and the basis for the original estimates is not always clear.

This section describes the use and disposal of solvents in M-Area, discharges to Tim's Branch and the M-Area settling basin, evaporation of solvents from M-Area operations and surface water, the air strippers being used to remediate M-Area groundwater, and more recent air quality permit and air emissions inventory estimates of trichloroethane releases from M-Area. From all of this information, we estimated the releases of trichloroethylene, tetrachloroethylene, and trichloroethane to the air. Although releases to surface waters were notable, most of the solvents released would have evaporated into the air before reaching people offsite. When these chlorinated solvents have been released or spilled to surface waters, the primary removal process is evaporation, with half of the solvent volatilizing within minutes, days, or up to several weeks depending on the conditions (Howard 1990). Almost all of the solvents in Site streams probably evaporated or were degraded long before they reached the Savannah River.

Chlorinated Solvents in Liquid Effluents Discharged from M-Area

M-Area wastewater drained to two process sewers. One discharged to Tim's Branch, which flowed into Steed Pond (also called Steed's Pond or Forrest Oakely Pond) then into Upper Three Runs and the Savannah River. The other emptied into the settling basin, which overflowed forming a small stream (often called the seep area) that seeped into the ground adjacent to Lost Lake, described as an upland depression or Carolina Bay (Christensen and Gordon 1983; Merz 1982).

The history of liquid effluent discharges in M-Area is important for characterizing releases. From 1952 (production started in 1954) until 1958, all effluents were discharged directly to Tim's Branch. The canning process effluents discharged from Building 321-M contained uranium. Because of the uranium in the effluent, a seepage basin was constructed and put in service in 1958 (Christensen and Brendell 1981). Several documents suggest the seepage basin was first used in 1973 (Specht 1991; Pickett 1990). Colven et al. (1985) suggests that 313-M effluents first went to the seepage basin in 1960. In their discussion of the solvents released to the M-Area sewers, Christensen and Brendell (1981) indicate that solvents had been released to sewers leading to the seepage basins since 1958. We believe that 1958 is the correct year, but more effluent was diverted to the seepage basin with time and some effluents may not have gone to the seepage basin until 1973.

In 1970, a program to decrease the amount of liquid waste discharged from 300-M Area was initiated. It involved repairing the Steed Pond Dam, recovering waste tetrachloroethylene (Du Pont 1972b), neutralizing caustics, and monitoring uranium oxide releases (Du Pont 1970). In 1970, a memo (Hardt 1970) explained that the process wastes from Buildings 313-M and 320-M were discharged through a process sewer through a uranium oxide sampler to the Tim's Branch. Tim's Branch emptied into Steed's Pond, located about 1.5 mi from M-Area. The dam partially washed away in 1966, reducing the size of the pond from 10 acres to about 2 acres. The dam was repaired in December 1970, restoring the pond to its original size. The larger pond facilitated settling of particulate material (Du Pont 1970). The overflow from the pond ran into Upper Three Runs and then the Savannah River. About 5×10^6 gal of water per week (or about 2.6×10^8 gal y^{-1}) was discharged from the Raw Materials Area via this route in 1970 (Hardt 1970).

In 1972, a new sewer line was constructed to discharge 313-M process wastes into the 321-M settling basin instead of into Tim's Branch. In 1973, all discharges except laboratory drains were routed to the settling basin. In 1979, the laboratory drain discharges were diverted to settling basin. In May 1982, discharges of process effluent to the Tim's Branch, which totaled about 370,000 gal d^{-1}, or about 1.35×10^8 gal y^{-1} were stopped and diverted to the basin. In November 1982, most of the cooling water and noncontact process effluent was directed back into Tim's Branch (Gordon 1982; Colven et al. 1985). In July 1985, the M-Area Liquid Effluent Treatment Facility (LETF) was completed and all effluents were treated and discharged to Tim's Branch (Specht 1991; Zeigler et al. 1986).

The process sewer line to the basin was described as a 76-cm diameter underground vitrified clay sewer line. Each process building had leader sewer lines 15–30 cm in diameter that intersected the 76-cm line. The process sewer line to the settling basin was about 715 m long with a slope of about 0.03%. Many cracks and misalignments in the sewer pipe were discovered in 1981, when the sources of groundwater contamination were being investigated. The sewer lines were lined with a 12-in. PVC liner in 1983 (Christensen and Gordon 1983). Wastes from 321-M

and 322-M flowed through the process sewer to the setting basin, which overflowed to Lost Lake, a Carolina Bay about 2–3 acres in size. About 5×10^5 gal wk^{-1} or 2.6×10^7 gal y^{-1} of wastewater was discharged to the settling basin in 1970 (Hardt 1970).

The first comprehensive attempt to estimate the amounts of chlorinated solvents used and released from M-Area seems to have been summarized in a report by Christensen and Brendell (1981) published the same year the groundwater contamination was discovered. The total solvent use and percentage thought to have leaked or have been discharged to the process sewer was estimated for each year. Christensen and Brendell (1981) acknowledge that estimating these quantities was "difficult, since no measurements of solvent loss to the process sewers or annual use in M-Area were ever made. The solvent use and loss estimates are derived from past solvent purchase orders, past operational events and M-Area personnel experience."

Separating out the solvents discharged for each time period, based on the solvent reported to have been used at the time and calculating the amount discharged from the percentage discharged and use estimates, resulted in estimates for each solvent for each year summarized in Table 17-6.

Potential understatement of the use from 1952–1970 was acknowledged by Christensen and Brendell (1981) on whose data many subsequent Site evaluations and our estimates are largely based. Because their estimates of use and release were the best data available and are the basis of our source term estimates for the early years, the solvent releases will be addressed for three time periods: (1) from startup until 1981, (2) after 1981, and (3) after 1984 when the air emissions inventory and air permit application calculations were done.

Chlorinated Solvents Use and Release from 1952–1981

An inventory of the number of degreasers and their volumes was found in the memo written by Christensen and Brendell (1981). Building 313-M had three 200-gal degreasers; Building 320-M had one 1000-gal and one 200-gallon degreaser; and Building 321-M had two 1000-gal degreasers and one 200-gal degreaser in 1981 (Christensen and Brendell 1981).

Degreasers are large vats partitioned in halves that contained heated and cooled solvent. Reactor components were cleaned by passing them through hot solvent vapor. Some of the vapor was condensed by water coils around the top of the vat. Some of the degreasers also had refrigerated coils that improved recovery. Periodically, as the oil and grease removed from the parts accumulated in the solvent, the vats were emptied and the solvent was replaced with fresh. In the early years, the dirty solvent was drained directly to the process sewers. At some point, some of it was funneled into holding tanks until it could be distilled for reuse. Occasionally, during the 1970s the still bottoms, degreaser sludge, and dirty solvent were drummed and stored until distillation recovery. When storage became a problem or drums began to rust, their contents were often emptied into the seepage basin process sewer. Most of the solvent used in these degreasers eventually evaporated. It seems that all of the solvent that did not evaporate was discharged to the process sewer between 1952 and 1979. Beginning in 1979, waste solvent and sludges were drummed and stored in the Hazardous Waste Interim Storage Facility, Building 710-U (Christensen and Brendell 1981).

From 1952 to 1958, trichloroethylene was used as a degreasing agent in Buildings 313-M and 320-M. The degreasers each had a still and a hold tank. By 1958, the stills and hold tanks had corroded and replacements had not been installed. The degreasers had also began to leak. Operations began in Building 321-M in 1958, with three new degreasers. Process liquid effluent

from 321-M went to the new M-Area seepage basin. The basin was intended to keep uranium from reaching surface waters (DOE 1987). By 1961, the basin began to overflow. The overflow traveled along an engineered ditch to Lost Lake. In 1962, the process in 313-M changed and tetrachloroethylene was used in the place of trichloroethylene. The tetrachloroethylene waste was released into Tim's Branch. Leakage from the degreasers continued. In 1971, 320-M and 321-M processes substituted tetrachloroethylene for trichloroethylene. In 1973, about one-half of the solvent discharged to Tim's Branch from 313-M was diverted to the seepage basin. In 1976, the remainder of the 313-M solvent discharged was going to the seepage basin. The leaking degreasers in 313-M were replaced in 1974. The degreasers in 320-M and 321-M continued to leak. Spot welding, installation of stainless steel liners, and other repair efforts were said to have been unsuccessful. In 1977, one of the three original degreasers in 321-M was replaced, but the new unit was not put into use until 1979. In 1978, the 320-M degreaser was replaced with a cold solvent dip tank. In 1979, concern over the potential carcinogenicity of tetrachloroethylene was raised and an effort to replace this solvent with trichloroethane was made. Another one of the old degreasers in 321-M was removed and a new degreaser, with refrigerated cooling coils, was installed. In 1979, the sewer line from 313-M to the seepage basin became blocked and discharges were routed to Tim's Branch. The solvent residues and sludge were being barreled, but some trichloroethane, although small amounts compared to discharges in earlier decades, were still being routed to Tim's Branch. The cold solvent dip tank in 320-M was replaced with a new degreaser in June 1981 (Christensen and Brendell 1981).

Although solvent consumption was said to have been estimated from former purchase records, Christensen and Brendell (1981) states that "area triclene records were not available and use-estimates are derived from M Area personnel judgment." The M-Area cost charging ratios suggested that solvents were allocated so that 20% went to 313-M, 25% went to 320-M, and 55% went to 321-M. Each 105 building in the reactor areas obtained one or two barrels of solvent from M-Area stores. The 105 degreasers were 1000 gal each (the same size as the degreasers in 320-M and 321-M). Degreaser solvents used in 773-A were also taken from the M-Area stocks. Allocations to other areas from M-Area were estimated to be from 25,000 to 50,000 lb each year. This was taken into account in estimates of M-Area consumption.

The leakage of solvent to the process sewers from the older, corroded degreasers was thought to have been proportional to solvent use. Christensen and Brendell (1981) estimated that 20–50% of the tetrachloroethylene used was released to the process sewers, higher than the 15–20% released for trichloroethylene and 3–5% for trichloroethane. Christensen and Brendell (1981) reported that site personnel felt that by the time tetrachloroethylene was in use, in about 1971, the degreasers in 313-M and 320-M were about 19 years old and those in 321-M were 13 years old. Corrosion and leaking increased during the 1970s. The increase in the amount of solvent purchased during this time was thought to reflect the deterioration and leakage problems and resulting losses. Tetrachloroethylene is also less volatile that trichloroethylene or trichloroethane. More tetrachloroethylene would have been expected to reach the process sewers if less evaporated (Christensen and Brendell 1981).

Christensen and Brendell concluded that since 1952, M-Area has used about 13 million lb (6,000,000 kg) of chlorinated solvents as degreasers and cleaners. They believed that from 50–95% of the solvents evaporated during degreasing operations, and the rest went to the M-Area process sewer systems. They estimated that about 2 million lb (901,100 kg) of solvents had been released to sewers leading to the seepage basins since 1958 and about 1.5 million lb (682,020 kg)

of solvent had been released to sewers going to Tim's Branch since 1952. The total discharges for the time period they evaluated are given in Table 17-6.

Table 17-6. Discharge Estimates for Trichloroethylene, Tetrachloroethylene, and Trichloroethane from M-Area Based on Christensen and Brendell (1981) Solvent Use and Process Sewer Discharge Estimates for M-Area Solvents[a]

Year	Estimated trichloro-ethylene use (ton y^{-1})	Estimated tetrachloro-ethylene use (ton y^{-1})	Estimated tri-chloro-ethane use (ton y^{-1})	% to sewers from 321-M	% to sewers from 320-M	% to sewers from 313-M	Trichloro-ethylene to the settling basin (ton y^{-1})	Trichloro-ethylene to Tim's Branch (ton y^{-1})	Tetra-chloro-ethylene to the settling basin (ton y^{-1})	Tetra-chloro-ethylene to Tim's Branch (ton y^{-1})	Trichloro-ethane to the settling basin (ton y^{-1})	Trichloro-ethane to Tim's Branch (ton y^{-1})
1952	76.5				15	15		11.5				
1953	76.5				15	15		11.5				
1954	76.5				15	15		11.5				
1955	76.5				15	15		11.5				
1956	76.5				15	15		11.5				
1957	76.5				15	15		11.5				
1958	76.5			20	15	15	11.5	12.5				
1959	140			20	15	15	11.5	12.5				
1960	140			20	15	15	11.5	12.5				
1961	140			20	15	15	11.5	12.5				
1962	89	54		20	25	25	12.5	7.5		13.5		
1963	89	54		20	25	25	12.5	7.5		13.5		
1964	89	54		20	25	25	12.5	7.5		13.5		
1965	89	54		20	25	25	12.5	7.5		13.5		
1966	89	54		20	25	25	12.5	7.5		13.5		
1967	89	54		20	25	25	12.5	7.5		13.5		
1968	89	54		20	25	25	12.5	7.5		13.5		
1969	89	54		20	25	25	12.5	7.5		13.5		
1970	89	54		20	25	25	12.5	7.5		13.5		
1971		185		20	25	25			25	24.5		
1972		315		25	30	30			51.5	49.5		
1973		415		35	35	40			96	53		
1974		335		35	35	20			70.5	42.5		
1975		575		45	45	20			153.5	76		
1976		500		45	45	20			143.5	56.5		

Table 17-6. Discharge Estimates for Trichloroethylene, Tetrachloroethylene, and Trichloroethane from M-Area Based on Christensen and Brendell (1981) Solvent Use and Process Sewer Discharge Estimates for M-Area Solvents[a]

Year	Estimated trichloro-ethylene use (ton y^{-1})	Estimated tetrachloro-ethylene use (ton y^{-1})	Estimated tri-chloro-ethane use (ton y^{-1})	% to sewers from 321-M	% to sewers from 320-M	% to sewers from 313-M	Trichloro-ethylene to the settling basin (ton y^{-1})	Trichloro-ethylene to Tim's Branch (ton y^{-1})	Tetra-chloro-ethylene to the settling basin (ton y^{-1})	Tetra-chloro-ethylene to Tim's Branch (ton y^{-1})	Trichloro-ethane to the settling basin (ton y^{-1})	Trichloro-ethane to Tim's Branch (ton y^{-1})
1977		560		45	45	20			161	63		
1978		412		45	20	20			130	20.5		
1979–		412		25	20	20			73.5	20.5		
–1979			85	5	5	5					2.5	3
1980			150	5	5	5					4	3.5
1981			100	3	3	3					3	1
Totals from 1952–1981							158.5 (25,000 gal)	191.5 (30,000 gal)	900 (133,000 gal)	500 (77,000 gal)	9.5 (14,000 gal)	6 (8850 gal)

[a] Solvent releases to the process sewers in tons per year (1 ton = 2000 pounds) for trichloroethylene, tetrachloroethylene, and trichloroethane estimated by Christensen and Brendell (1981). The 1981 total includes an accidental spill of 300 gal of trichloroethane. Discharges from the three M-Area facilities (313-M, 320-M, and 321-M) were combined.

Table 17-7. Estimates of the Chlorinated Solvents Used and Released in M-Area[a]

Solvents used and released	Solvent and years of use			Totals
	Trichloroethylene 1952–1970	Tetrachloroethylene 1971–1979	1,1,1-Trichloroethane 1979–1982	
Total used	3,700,000 lb	8,700,000 lb	670,000 lb	13,070,000 lb
	1,680,000 kg	3,950,000 kg	305,000 kg	5,941,000 kg
Released to the	317,000 lb	1,800,000 lb	19,000 lb	2,136,000 lb
Settling Basin	144,100 kg	820,000 kg	8,600 kg	971,000 kg
Released to Tim's	383,000 lb	1,000,000 lb	12,000 lb	1,395,000 lb
Branch	175,000 kg	450,000 kg	5,500 kg	634,090 kg

[a] Source: Christensen and Brendell (1981).

Both the draft (Colven et al. 1985) and final (Pickett et al. 1987) versions of the Environmental Information Document relevant to the M-Area settling basin (which had a different order of authors and much revised text but the same Site document number) reported the same numbers as Christensen and Brendell (1981). Slight differences, with the exception of one obvious typographical error, seemed to be due to differences in rounding amounts or rounding kilogram values converted from pounds.

The values reported by Merz (1982) are summarized in Table 17-8 in a format similar to Table 17-7. Merz does not reference the source of his estimates, but they were likely to have been based largely on Christensen and Brendell (1981).

Table 17-8. Estimates of the Chlorinated Solvents Releases to the M-Area Process Sewers[a]

Solvents used and released	Solvent and years of use			Total solvent released 1952–1981
	Trichloroethylene 1952–1971	Tetrachloroethylene 1971–1979	1,1,1-Trichloroethane 1979–1981	
Released to the	320,000 lb	1,700,000 lb	17,000 lb	2,037,000 lb
Settling Basin	145,450 kg	772,730 kg	7,730 kg	925,910 kg
Released to Tim's	383,000 lb	1,100,000 lb	21,000 lb	1,504,000 lb
Branch	175,000 kg	500,000 kg	9,550 kg	683,640 kg
Total released	703,000 lb	2,800,000 lb	38,000 lb	3,541,000 lb
	319,550 kg	1,272,730 kg	17,300 kg	1,609,550 kg

[a] Source: Merz (1982).

Other estimates of releases seem to have been based on those of Christensen and Brendell (1981) and were consistent with their conclusions. The estimates in Christensen and Brendell (1981) average to 118,303 lb y^{-1} or 53,650 kg y^{-1} discharged to surface water from M-Area. Another report from 1970 suggested that about 90,000 lb (40,900 kg y^{-1}) of tetrachloroethylene had been discharged annually to the plant streams (Du Pont 1970).

In summary, Christensen and Brendell (1981) estimated that about 6 million kg of solvents was used in M-Area, 1 million kg was released to the settling basin, and 0.6 million kg was released to Tim's Branch from 1951 to 1981. The remaining 4.4 million kg may have evaporated. Certainly, much of the 1.6 million kg discharged in liquid effluent also evaporated, although some seeped into the groundwater and some was degraded.

Perhaps Christensen and Brendell's estimates are far better than any estimates we can make because they interviewed Site personnel in 1981. These personnel had far more institutional memory and understanding of historical practices than we would expect to find in 1996 and 1997. We attempted to verify their arithmetic and observe whether the percentage of usage amount estimated to have been discharged to the sewer correlated with process changes involving solvent use (such as replacement of old degreasing units and changes in drumming of solvents) described above. The process and equipment changes described above appear to agree with those described in the *Savannah River Plant History for the Raw Materials Area for July 1954 through December 1972* (Du Pont 1972b). This report is a likely source for some of the historical information compiled by Christensen and Brendell (1981) about process changes and installation of new degreasers. The estimates seem to correlate with the practices described for each time period. It is not clear how the percentages of solvent going to Tim's Branch and the settling basin were determined to vary from 15–40% in different years. The percentages assigned to each year appear to have changed as the degreasers aged and presumably corroded and leaked more often. It is difficult to improve on the approach and estimates of Christensen and Brendell (1981) because of a lack of knowledge and information.

An attempt to further characterize the M-Area solvent releases was published in a report, in a draft document (Colven et al. 1985) and then in a final version (Pickett et al. 1987). These documents suggested that M-Area processes began in 1954 rather than 1952. Other records (Christensen and Brendell 1981) suggest that processing in 313-M and 320-M began in 1952 and processing in 320-M, which resulted in releases of uranium to the liquid effluent from the canning process, began in 1954. Other than this, discrepancies between the 1981 analysis and later reports are not evident and no new information on solvent discharges was presented.

Chlorinated Solvent Use and Release After 1981

In May 1982, process sewer discharges to Tim's Branch were stopped and all effluents went to the settling basin. In November 1982, waste considered nonradioactive went back into Tim's Branch (Pickett et al. 1987).

The Environmental Survey (DOE 1987), conducted in the late 1980s, described vapor degreaser operations in Buildings 313-M, 320-M, and 320-M in the 1980s. Used solvent was recycled using a solvent still. Evaporation of 1,1,1-trichloroethane from the degreasers was limited by refrigerated cold traps, condensers, and degreaser tank covers. Local ventilation of the areas was required for worker safety. Vapors from the degreasers were vented to the outside air through hoods. Ventilation of the vapor degreasers was important for worker protection and appears to have been sufficient in the late 1980s; however, the survey team found that one cold trap was out of service and covers were not always in place (DOE 1987). One of the key findings of the Environmental Survey was that emissions of 1,1,1-trichloroethane to the air from the 300-Area degreasing operations were large and may have been exceeding 200 ton y^{-1} (DOE 1987) for an undetermined number of years.

Monitoring data of use to help verify release estimates for chlorinated solvents are not available. The available surface water and sediment monitoring data, most of which was collected to characterize M-Area effluent and settling basin sludge in preparation for the Effluent Treatment Facility, were reviewed and are summarized here.

In 1982, less than 5 lb d^{-1} (2.3 kg d^{-1}), or less than 829 kg y^{-1} of solvent was said to have been discharged in M-Area effluents. Samples taken during "routine operations" indicated about 0.2 mg L^{-1} 1,1,1-trichloroethane in the sewers with "occasional surges to about 10 mg L^{-1} (ppm) when cleaning tanks are being drained"(Merz 1982).

Concentrations of 1,1,1-trichloroethane measured in a 24-hour composite sample of sewer effluent sampled in December 1981 when the plant was operating were 0.47 mg L^{-1} in sewer effluent going to the settling basin and 0.09 mg L^{-1} in sewer effluent going to Tim's Branch (Merz 1982). Colven et al. (1985) implied that peak process discharges to the Tim's Branch in 1982 were 1.35×10^{8} gal y^{-1} or 5.1×10^{-8} L y^{-1}. Using volume data from Colven et al. (1985) and concentrations reported by Merz (1982), we can estimate that a total of 286 kg y^{-1} released to surface water in the 1980s. Of this, about 240 kg may have gone into the settling basin and 46 kg into Tim's Branch each year.

Chlorinated Solvents Released to Tim's Branch

In 1968, an organic compound, discovered to be tetrachloroethylene, was observed in a Tim's Branch water sample. The memo describing this discovery said that, "discussions with M-Area personnel indicated that 313-M is the only building using large quantities of tetrachloroethylene and that they do routinely dispose of this material into the sewer system" (Johnson 1968).

Hardt (1970) described releases to Tim's Branch and predicted the release of the same materials after initiation of proposed waste reduction measures, which included recovery of waste tetrachloroethylene by distillation.

Volumes and concentrations of waste discharged to the Tim's Branch, 300-Area effluent, and to the 321-M settling basin corresponding to production data from November 1968 to February 1969 were estimated in Monier (1970). Monier (1970) estimated the amount of solvents in the in 300-Area liquid effluent based on an analysis of 4 months of production data from November 1968 to February 1969. He estimated that 6000 gal (29,809 kg) of trichloroethylene and 13,000 gal (64,586 kg) of tetrachloroethylene (all were 100% solutions) had been discharged to Tim's Branch in 1 year, and that about 5000 gal (24,840 kg) of trichloroethylene were discharged to the 321-M settling basin in a year. References for the estimates and for the data from which they were determined were not included, and it was not clear how the estimates were determined (Monier 1970). If we assume a conversion of 10.93 lb gal^{-1} and 2.2 lb kg^{-1}, then 29,809 kg and 24,840 kg of trichloroethylene were discharged that year to the Tim's Branch and the settling basin and 60,816 kg of tetrachloroethylene was discharged to surface water in that year (Monier 1970).

In a study of waste reduction methods, Hardt (1970) estimated that each year about 91,000 lb (41,364 kg) of tetrachloroethylene, and 12,000 lb (5455 kg) of trichloroethylene were being released to Tim's Branch. These amounts represented 100% solutions, calculated from larger amounts of more dilute solutions actually discharged. At that time, tetrachloroethylene from Building 313 and trichloroethylene from Building 320 were discharged to the process sewer. The site was said to be planning to convert from trichloroethylene to tetrachloroethylene in 1971 and to distill waste tetrachloroethylene. The authors predicted that in the future all but 1% of it would be recovered, and the 1% in the heel could be barreled and buried (Hardt 1970).

In a handwritten note or logbook entry about a meeting with Raw Materials and Health Physics personnel, Johnson (1978) said that tetrachloroethylene waste from 321-M was collected in 560-gal tanks and released to the seepage basin every 2 weeks. The potential for low-level uranium release from this practice was of concern. Samples of the 321-M 'cleaning solution' were to be analyzed for uranium, and the possibility of installing a trebler sampler at the seepage basin was discussed.

In 1985 and 1986, a study was done to determine any adverse effects of Outfall A-014 effluent, which received M-Area effluents, on the Tim's Branch and Upper Three Runs Creek. The study was required as a part of the NPDES permit modification for the M-Area Liquid Treatment Facility. Chemicals detected in Tim's Branch at the outfall at concentrations higher than upstream locations included nitrate, zinc, uranium, and trichloroethylene (Specht et al. 1987).

In January 1985, 75 gal of trichloroethylene was spilled onto the ground in M-Area and the soil was removed and taken to the M-Settling Basin (Jewell 1990). We have no information on spills that occurred before 1980. It is likely that spills occurring before about 1985 would have been allowed to evaporate or would have been routed to the sewer or seepage basin, where most evaporated.

The solvents reaching the basin or Tim's Branch either evaporated, seeped into the ground and eventually into groundwater, accumulated in basin sludge and sediments, or were biodegraded.

Evaporation of Chlorinated Solvents from Surface Water

Evaporation of chemicals with a high vapor pressure (volatile chemicals) and low solubility from settling ponds and seepage basins can be important for evaluating releases to the air. Looney et al. (1987) used a simple mass balance model to determine the fraction of tritium and volatile organic compounds that evaporated rather than entered the soil. Equations incorporating volatility, solubility, inflow, outflow, seepage rate, and other parameters were used for each surface impoundment or seepage basin examined. The goal seemed to be to determine the fraction of chlorinated organics that entered the groundwater and identify which materials were of concern for groundwater contamination at each waste site. Appendixes in Looney et al. (1987) include data tables with an estimated discharge mass for each site, which were developed from analysis of groundwater and soil data as well as process knowledge. A fraction to convert the mobile mass (moving to groundwater) to the total disposal mass was developed for common contaminants. The disposal mass estimates were calculated from groundwater concentrations and seemed very uncertain. Soil concentrations and groundwater concentrations were compared to multiples of drinking water standards and other criteria to select contaminants of concern for each disposal site. Background concentrations of chemicals in groundwater from SRS wells were also compiled.

The data tables in Looney et al. (1987) for M-Area do not include estimates for the amount of solvents that may have evaporated, even though these were said to have been calculated and accounted for in the estimates of releases to groundwater. The total disposal mass was calculated to have been 84,000 kg of tetrachloroethylene, 15,000 kg of trichloroethylene, and 1000 kg of trichloroethane for 1971–1985. The soil core inventory estimated in 1985 was 3600 kg for tetrachloroethylene, 170 kg for trichloroethylene, and 140 kg for trichloroethane. The maximum

groundwater concentration measured was 427,000 μg L^{-1} tetrachloroethylene, 161,000 μg L^{-1} for trichloroethylene and 203 μg L^{-1} for trichloroethane from well MSB-3A (Looney et al. 1987). The total amount of chlorinated solvents in the groundwater was not estimated. These data are not sufficient to allow us to use a mass balance approach for estimating the amount of solvents that evaporated.

Trichloroethane Use in the 1980s and 1990s

Another potentially helpful handwritten ledger called *the 300-Area Essential Materials Inventory Control* covered the receipts of some of the process chemicals, including 1,1,1-trichloroethane (Westinghouse 1986b, Westinghouse 1989). The ledger for 1,1,1-trichloroethane was a handwritten log that listed the railroad cars that delivered 1,1,1-trichloroethane, the balance of the purchase order, and the amount "consumed" by building from July 1, 1979, to June 1, 1986. There are very few entries for 320-M, and some of the values under 313-M and 321-M appear to have been corrected. It is not clear whether the distribution to the facilities was a cost accounting or the actual amount delivered. There are many handwritten corrections and a few illegible entries, but most of the values are legible. However, notes on the lack of any delivery correspond to a consumption entry left blank, and some of the entries are out of sequence. A note is written on the top of the page, "20%-313M, 15%-320M and 65%-321M," which appears to be the formula by which the total amount used was partitioned between the three buildings. This is most likely a cost accounting breakdown, but it was useful in estimating values that were illegible.

From February 1981 to August 1982, and again from January 1983 to December 1983, three values were entered into the consumed column. These were added together to calculate a number that presumably represented the total consumption for all buildings. Most of the values are positive, except for several Decembers when consumption numbers are negative, presumably to allow the totals to match the year-end balance. To avoid implying a certainty that is lacking, 3-month totals were compiled rather than monthly totals. The approximately quarterly totals are shown in Table 17-9. Monthly averages and annual totals are shown in Table 17-10. The ledger contained entries of both gallons and/or pounds and stated that a conversion of 10.926 lb gal^{-1} was used to convert the gallons to pounds. The values were converted to kilograms using 10.93 lb gal^{-1} and 2.2 lb kg^{-1}.

Table 17-9. Quarterly 1,1,1-Trichloroethane Consumption for 1979-1986 from Handwritten 300-Area Essential Materials Inventory Control Ledgers

Time period	kg
July, Aug, Sept. 1979	3.5×10^4
Oct., Nov., Dec. 1979	7.8×10^4
Total for the last 6 months of 1979 (× 2)	2.3×10^5 kg y^{-1}
Monthly average	1.9×10^4 kg mo^{-1}
Jan., Feb., March 1980	3.2×10^4
April, May, June 1980	4.3×10^4
July, Aug., Sept. 1980	2.7×10^4
Oct., Nov., Dec. 1980	2.9×10^4
Total for 1980	1.3×10^5 kg y^{-1}
Monthly average	1.1×10^4 kg mo^{-1}
Jan., Feb., March 1981	4.1×10^4
April, May, June 1981	3.2×10^4
July, Aug., Sept. 1981	4.9×10^4
Oct., Nov., Dec. 1981	5.3×10^5
Total for 1981	1.8×10^5 kg y^{-1}
Monthly average	1.5×10^4 kg mo^{-1}
Jan., Feb., March 1982	5.4×10^4
April, May, June 1982	7.1×10^4
July, Aug., Sept. 1982	6.8×10^4
Oct., Nov., Dec. 1982	5.5×10^4
Total for 1982	2.5×10^5 kg y^{-1}
Monthly average	2.1×10^4 kg mo^{-1}
Jan., Feb., March 1983	3.3×10^4
April, May, June 1983	4.9×10^4
July, Aug., Sept. 1983	1.2×10^5
Oct., Nov., Dec. 1983	7.4×10^4
Total for 1983	2.8×10^5 kg y^{-1}
Monthly average	2.3×10^4 kg mo^{-1}
Jan., Feb., March 1984	3.7×10^4
April, May, June 1984	1.1×10^5
July, Aug., Sept. 1984	9.6×10^4
Oct., Nov., Dec. 1984	3.9×10^4
Total for 1984	2.8×10^5 kg y^{-1}
Monthly average	2.3×10^4 kg mo^{-1}
Jan., Feb., March 1985	6.4×10^4
April, May, June 1985	7.3×10^4
July, Aug., Sept. 1985	1.4×10^4
Oct., Nov., Dec. 1985	8.9×10^4
Total for 1985	2.4×10^5 kg y^{-1}
Monthly average	2.0×10^4 kg mo^{-1}
Jan., Feb., March 1986	5.3×10^4
April, May, June 1986	5.8×10^4
Total for the first 6 months of 1986 (× 2)	2.2×10^5 kg y^{-1}
Monthly average	1.8×10^4 kg mo^{-1}

Table 17-10. Approximate Monthly and Annual Amounts of Solvent Used Based On the Handwritten 300-Area Essential Materials Ledgers

Year	Monthly average (kg)	Total for each year (kg)	Ton y^{-1}
1979	1.9×10^4	2.3×10^5	253
1980	1.1×10^4	1.3×10^5	143
1981	1.5×10^4	1.8×10^5	198
1982	2.1×10^4	2.5×10^5	275
1983	2.3×10^4	2.8×10^5	308
1984	2.3×10^4	2.8×10^5	308
1985	2.0×10^4	2.4×10^5	264
1986	1.9×10^4	2.2×10^5	242
Total 1979–1986	1.5×10^5	1.8×10^6	1,980
Average	$1.5 \times 10^5 \div 8\ y = 1.9 \times 10^4$	$1.8 \times 10^6 \div 8\ y = 2.3 \times 10^5$	248

Source: Westinghouse 1986b, Westinghouse 1989.

Essential materials records from M-Area for 1984, 1985, and January through April 1986 were found in Phase I of the dose reconstruction study (Westinghouse 1983, 1984, 1985, 1986a, 1987a). These are computerized (as opposed to the handwritten ledgers just described) cost accounting records that included monthly inventories of the amount of process chemicals consumed in gallons. Three values for 1,1,1-trichloroethane are printed for each month, which correspond to the amount delivered to each of the three process buildings, each designated by a charge code. The monthly values do not seem to be accurate. The quantities were apparently overestimated for some months and then given negative values for the following month. It seemed best to average values over a longer time period, at least 1 year. Annual averages are presented in Table 7-11. The monthly values vary but generally range between 2,000 and 12,000 gal mo^{-1}. The values printed for October 1984 totaled 61,248 gal. What appears to be a handwritten correction of 7743 gal is written over the values. The values reported for November 1984 are unusually large, totaling 83,866 gal, then the values for December total –6786 gal. The values for November are about 10 times the monthly average, which suggests a decimal place error occurred, but the negative numbers in October and December may have been an attempt at correcting the balances. If the numbers in the printout are taken at face value, and the corrected value for October is assumed to be accurate, the total consumption for 1984 is 149,700 gal (7.5×10^5 kg) averaging 12,474 gal mo^{-1} (6.2×10^4 kg mo^{-1}). If we disregard the unusual monthly values for October, November, and December, the total for January through September 1984 is 64,875 gal or 7208 gal mo^{-1}. The consumption for 1985 totaled 29,108 gal (1.4×10^5 kg), which averages out to be 2,425 gal mo^{-1} (1.2×10^4 kg mo^{-1}). A total of 24,478 gal was recorded as consumed from January 1, 1986, to April 30, 1986; an average of 6120 gal mo^{-1}; and a projected average for the year of 73,434 gal y^{-1} (3.6×10^5 kg y^{-1}) (Westinghouse 1984, 1985, 1987a). These records suggest that the consumption of 1,1,1-trichloroethane was very inconsistent from month to month and may have ranged from about 2500 to 12,500 gal mo^{-1} during peak periods of production.

Table 17-11. Summary of the Average Amount of Trichloroethane Used in M-Area for Three Years Based on Computerized Essential Materials Ledgers

Year	Average (kg mo^{-1})	Average (kg y^{-1})	Ton y^{-1}
1984	6.2×10^4	7.5×10^5	825
1985	1.2×10^4	1.4×10^5	154
1986	3.0×10^4	3.6×10^5	396
Average	3.5×10^4	4.2×10^5	462

The annual use estimates derived from averaging the values in the essential materials ledgers differ from estimates derived from the Inventory Control ledgers for 1984–1986. Using the highest total estimated for each year from either source, we can estimate that as much as 2.4×10^6 kg or 2662 ton of trichloroethane was used from 1979–1986.

Chlorinated Solvents in M-Area Groundwater

Although most of the solvent released in liquid effluent evaporated, some entered the groundwater because of (a) seepage through the basin and from basin overflow areas through cracks in the pipelines to the basins and (b) leaks and spills in M-Area (Bradley 1981).

Groundwater under M-Area was found to be contaminated with metal degreasing solvents in June 1981 (Zeigler et al. 1985). Groundwater samples from four monitoring wells found that trichloroethylene and tetrachloroethylene were approaching the parts-per-thousand (g L^{-1}) range (Christensen and Brendall 1981). At that time, Du Pont estimated that 60,000 lb of chlorinated solvents had contaminated 330 million gal of groundwater in a plume that had traveled a distance of about 2200 ft (Zeigler et al. 1985). A 1982 fact sheet on M-Area solvent contamination and a Technical Data Summary prepared by Gordon (1982) both said 59,000 lb of solvents had entered the groundwater. Gordon (1982), Du Pont (1982b), and Bradley (1981) used a value of 60,000 lb in a preliminary scoping documents, fact sheets, and evaluations prepared for planning groundwater cleanup. Test well samples in 1982 were said to have identified a 330-million gal plume of groundwater contaminated with about 60,000 lb of chlorinated organic solvent (Merz 1982). Additional groundwater monitoring wells were drilled and the contamination was studied over the next several years. By 1985, the plume of contaminated groundwater in M-Area had been defined by more than 700 samples and an additional 200 monitoring wells. In 1987, the inventory of solvents in the groundwater was estimated to have been between 260,000 and 450,000 lb, with concentrations as high as 300 mg L^{-1}. About 70% of the solvent was thought to be trichloroethylene (Colven et al. 1987). Bebbington (1990) reported that 450,000 lb of chlorinated solvents had seeped or leaked into the groundwater.

No chlorocarbons have been detected in offsite drinking water in the vicinity of the 300/700-Area (Zeigler et al.1986). Groundwater studies led Site researchers to predict that the plume of solvent-contaminated water beneath M-Area might reach the Site boundary by the year 2005 (DOE 1988; Gordon 1982). Although the groundwater does not represent a complete exposure pathway for people offsite, the groundwater has been and is still being remediated. This has resulted in solvents being aerated into the air.

M-Area Air Strippers and Soil Vacuum Extraction Unit Emissions. Air strippers have been and are still being used to remove volatile chemicals from groundwater under M-Area. Groundwater is pumped through the stripper where the water is aerated and the solvents are released to the air. Stripped water is returned to the groundwater. The M-Area Groundwater Remedial Action Program began in 1983. Additional recovery wells and a pilot air stripper began operating in 1984. A full-scale recovery system with 11 recovery wells and a 400-gal min^{-1} air stripper was put in place in September 1985, and it removed an estimated 53,000 lb (24,100 kg) of solvent in the first year it operated. The remediation plan was to remove and treat groundwater for about 30 years (Colven et al. 1987). The goal of the groundwater treatment systems was to reduce the concentrations in groundwater from the parts per million to the parts per billion range (Bradley 1981).

Detailed descriptions of the air stripper can be found in several Site documents (Looney et al. 1991; Gordon 1982). The air stripper removed solvents from the groundwater and discharged them through a stack into the air. The groundwater was pumped to the top of the air stripper's column and allowed to trickle down over packing. Volatile organics evaporated into the air, which was blown into the bottom of the column and exited out the top (Gordon 1982). It was recognized that concentrations of solvents in the air exiting the stripper might be in excess permissible occupational exposure limits, but ground concentrations were predicted to be well within the threshold limit values (Gordon 1982).

A full-scale test of the air stripper near at 300-M Area was described by Looney et al. in 1991. During the test, from 100 to 140 lb (45 to 63 kg) of volatile organic chemicals was extracted each day or 16,000 lb (7272 kg) over 139 days. The air containing the chemicals was recovered by vacuum extraction. The concentration of the contaminated vapors extracted from the horizontal vacuum extraction well was measured approximately three times each day. Concentrations after the process first started were as high as 5000 ppm. The concentrations stabilized to about 300 ppm after several days. The vacuum extraction process removed 109 lb d^{-1} (49.5 kg d^{-1}) and the air stripping component removed an additional 20 lb (9.1 kg). Groundwater samples were taken daily (Looney et al. 1991). If this removal rate would have been sustained, up to 565 ton y^{-1} or 3.6 kg h^{-1} would have been released from the vacuum extraction and groundwater stripping operation. To address the question about potential air pollution, Site engineers also modeled the transport of solvents released to the air from the air stripper and attempted to calculate redeposition and expected concentrations in rainfall. Using an atmospheric concentration of 10 μg m^{-3}, they estimated that less than 0.002% of the trichloroethylene would be redeposited. The remainder would be diluted, transported, and photodegraded. The predicted maximum rainfall concentrations were 25 μg m^{-3} (Looney 1984). Air quality effects were also examined by Colven et al. (1987). Air emission testing was required for the air stripper's air emissions operating permit. In October 1985, stack emissions were tested for trichloroethylene, tetrachloroethylene, and 1,1,1-trichloroethane. Based on the test results, an average emission rate of 7.87 lb h^{-1} (3.6 kg h^{-1}) was predicted, just in compliance with the permit emission limit of 7.9 lb of volatile compounds per hour. An adsorbent sampling train monitoring method approved by the EPA and SCDHEC was used. The error of the method used was estimated to be as high as ±50%. Effluent water samples were also tested for solvents. The total chlorocarbon removal rate varied from about 3.5 to 10 lb h^{-1}. Water samples analyzed by the laboratory in 321-M was used to determine an average air emission rate of 6.50 lb h^{-1} (2.95 kg h^{-1}). In addition, modeling studies were conducted to predict the dispersion of organic compounds in the air and to estimate

concentrations at several locations. Dispersion calculations using EPA's Industrial Source Complex model were made and included as a part of the materials accompanying the permit application. Meteorological data for 1976, which were thought to produce the highest predicted concentrations, were used. Modeling for trichloroethylene was done and downwind concentrations for tetrachloroethylene and 1,1,1-trichloroethane were predicted using the ratio of these compounds to trichloroethylene in the stack. The maximum trichloroethylene concentrations predicted for the plant property line were predicted to be less than 0.013% of the threshold limit value (occupational exposure limit). Values for 1,1,1-trichloroethane and tetrachloroethylene were less. The authors of this report concluded that "predicted values for downwind organic concentrations are judged to be insignificant and below the sensitivity of the most sophisticated measurement techniques" (Colven et al. 1987).

Another report described the air stripper that began operating in 1985 as having a capacity of 400 gal min^{-1} and emitting about 7.9 lb h^{-1} (3.6 kg h^{-1}) of chlorinated solvents into the air, roughly equivalent to 35 ton y^{-1}. Groundwater inlet and treated water outlet analyses, said to be more accurate than exhaust air measurements, were used to estimate solvent emissions (DOE 1987).

By the second quarter of 1988, cumulative throughput of groundwater through the stripper was estimated to have been 532 million gal, with 130,300 lb (59,000 kg) of solvent removed since startup in September 1985. A total of 164,500 lb (75,000 kg) was calculated to have been removed, including all of the other experimental removal programs, up to 1988. The influent concentrations to the air stripper ranged from about 22,800 to 23,978 μg L^{-1} (DOE 1988).

The system was upgraded in 1990. In 1992, it was estimated that 285,000 lb (129,550 kg) of volatile organics had been removed from 1.5 billion gal of groundwater (Arnett et al. 1993).

For the air emissions inventory, trichloroethylene and tetrachloroethylene emissions from the air stripper used to treat groundwater (shown in Table 17-12) were estimated from the difference in inlet and outlet concentrations and information about treatment system flow rates (Radian 1992a).

Table 17-12. Estimates of the Amount of Chlorinated Solvents Removed by the 323-M Air Stripper, Reported in an Operating and Performance Summary Published in 1992 by Radian for the Building 323-M Air Stripper

Year	Total VOC removed (lb)	Operating time (h)	Emission rate (lb hr^{-1})	Trichloroethylene emissions (ton y^{-1})	Tetrachloroethylene emissions (ton y^{-1})
1985	19,523	2339	8.34	6.74	3.03
1986	48,756	8419	5.79	16.58	7.80
1987	44,346	8388	5.29	15.96	6.21
1988	36,790	8632	4.26	12.51	5.89
1989	26,024	8194	3.18	9.11	3.90
1990	28,323	8313	3.41	9.77	4.39
Total	203,762			70.67	31.22

Release of trichloroethylene is a concern for ozone depletion. In recent years, the air stripper and the vapor extraction units are connected to catalytic oxidation (Catox) equipment that

converts most of the chlorinated solvents to hydrogen chloride (hydrochloric acid) which is also a regulated air pollutant (Faugl 1996a).

The air stripper contribution to surface water releases seems to have been negligible. Trichloroethylene did not exceed the permit limits at the air stripper outfall, M-005 (Specht et al.1987).

Other Air Strippers and Soil Vacuum Extraction Units. The A-1 groundwater air stripper, installed in 1990, was described in the Part 70 Operating Permit Application. Releases for trichloroethylene, tetrachloroethylene, and hydrochloric acid from the unit were modeled for compliance with Standard 8 regulations. The limits required by the State Bureau of Air Quality Control to be in compliance with Standard 8 was 5.34 lb h^{-1} for hydrochloric acid and 6.4 lb h^{-1} for the solvents. The emission estimate for volatile compounds was 0.267 ton y^{-1}. The maximum design capacity emissions estimates were 9.55×10^{-3} ton y^{-1} for tetrachloroethylene and 2.94×10^{-3} ton y^{-1} for trichloroethylene. These were based on 1994 estimates that the unit operated for 7705 hours, removing a total of 517.3 lb of trichloroethylene and 16.8 lb of tetrachloroethylene.

In 1994, six soil vapor extraction units (SVEUs) were operating. These were somewhat like large vacuums used to remove solvents from soil. The emissions from all of these units totaled about 1/10th of the emissions estimated for the M-Area air stripper in 1994 (Faugl 1996m). The Savannah River Technology Center (SRTC) soil vacuum extraction unit, used for field demonstrations of pollutant treatment systems, was also described in the operating permit. The unit was used to extract contaminated vapors from the soil and send the contaminants through whatever technology was being demonstrated. The unit exhausted through a 15-ft stack. Using engineering calculations, operators estimated that the actual emissions of volatile compounds were 4.87×10^{-3} ton y^{-1}. Most of this, 1.01×10^{-3} ton y^{-1}, was trichloroethylene. 1,1,1-Trichloroethane emissions were estimated to be 4.80×10^{-5} ton y^{-1}, and the estimate for tetrachloroethylene was 1.01×10^{-3} ton y^{-1} (Westinghouse 1996a).

Estimates for a rate of discharge of trichloroethane from M-Area operations and also for trichloroethylene and tetrachloroethylene to the air from the air stripping operation were made as a part of the air emissions inventory (Radian 1992a).

Emissions in the AIRS database for trichloroethylene and tetrachloroethylene from air strippers, soil vacuum extraction units, basins, and tanks in 1985, 1987, and 1995 are summarized in Table 17-13.

Table 17-13. Emissions in the AIRS Database for Trichloroethylene and Tetrachloroethylene from Air Strippers, Soil Vacuum Extraction Units, Basins, and Tanks in 1985, 1987, and 1995

Emission source	Year	Actual[a] emission estimate (ton y^{-1})
Trichloroethylene		
H-Area tank farm	1985	0.0003
	1987	0.0003
M-Area air stripper M-1	1985	6.74
	1987	16.0
	1995	3.31
321-M SVEU	1995	0.054
Tetrachloroethylene		
F-Area seepage basins	1985	0.0214
	1987	0.0214
M-Area settling basin	1985	0.205
	1987	0.154
M-Area air stripper M-1	1985	3.03
	1987	6.21
	1995	2.15
5-SVEU in M-Area (totaled)	1995	0.697
321-SVEU	1995	0.425

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

About three times as many emissions points were listed for trichloroethylene in 1994 than in 1985. In 1994, more estimates for the tank farm and SVEUs in M-Area were listed. Table 17-14 presents the 1994 sources listed in the AIRS database. Table 17-15 provides the totals for trichloroethylene emissions.

The total actual trichloroethylene emissions for 1994 in the AIRS database were 5.65 ton y^{-1}. These emissions included releases from the burial ground solvent tanks; solvent tank trailer; tank farm painting; small degreasers in separations areas; tanks and vessels in H-Area; M-Area SVEUs and air strippers; and equipment used after 1990 like the ICF Tank farm, mobile experimental bioreactor, and demo waste incinerator. About one-half as many of the emission points were listed in 1992 than in 1994. The 1992 actual emissions totaled 8.66 ton y^{-1}. The actual emissions for 1990 totaled 9.78 ton y^{-1}. As expected, the air stripper in M-Area dominated the estimates, followed by the A-1 air stripper in 1992 and 1994. For all years, almost all of the trichloroethylene emissions were from the M-1 air stripper. About 95% of the total was from the M-1 air stripper in 1994. The next largest source was the A-1 air stripper, accounting for 0.05%.

In 1990, the M-1 air stripper accounted for 99.9% of the total. The highest annual emissions totaled 16 ton y^{-1} in 1987, with almost 100% coming from the M-1 air stripper. Trichloroethylene emissions from M-Area degreasers, sewers, and liquid effluent occurred long before the air emissions inventory was being done.

Table 17-14. Trichloroethylene Emission Sources for 1994 in the AIRS Database[a]

Source description	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
F-Area tank farm degreaser and miscellaneous uses	2.3×10^{-2}	3.0×10^{-4}
Air stripper G-Area A1	2.9×10^{-1}	2.6×10^{-1}
H-Area tank farm	1.84×10^{-2}	2.40×10^{-4}
Consolidated Incinerator (CIF) tank farm emissions total	6.6×10^{-2}	1.1×10^{-5}
SVEU For M-Area process sewers	1.05	0
SVEU For M-Area seepage basin	1.05	0
IOU-SVEU 782-7M	4.4×10^{-2}	2.3×10^{-2}
SVEU for A014 Outfall	1.05	0
SVEU	1.05	0
Air stripper M1	5.63	5.35
321-M SVEU	1.05	0
Groundwater air stripper	2.19×10^{-1}	blank

[a] Source: Faugl (1996l).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Table 17-15. Total Trichloroethylene Emissions from the AIRS Database[a]

Year	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
1985	11.5	2.64
1987	25.	16.0
1990	25.2	9.78
1992	34.6	8.66
1994	12.9	5.65

[a] Source: Faugl (1996l).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Tetrachloroethylene emissions in 1985 included waste handling and tanks, burial ground, laboratories, F-Area, H-Area and M-Area seepage basins, landfill and tank farm emissions, Pilot Plant equipment, painting, and the air stripper. The actual total emissions were 3.26 ton y^{-1}, with 3.03 ton y^{-1} from the M-1 air stripper. The next highest release was from the M-Area settling

basin with an estimate of 0.205 ton y^{-1}. In 1987, the total actual emissions were 6.39 ton y^{-1}, with 6.21 ton y^{-1} from the M-1 air stripper. The 1990 total was 4.4 ton, with 4.39 ton from the M-1 air stripper. The M-Area settling basin was not listed for 1990. Actual emissions in 1992 totaled 4.35 ton y^{-1}, with 99.5% from the M-1 air stripper (Faugl 1996m).

The AIRS database also contained emissions estimated for tetrachloroethylene from waste storage tanks, incinerators seepage basins, disposal and landfills, painting and printing operations, and laboratories (Faugl 1996l) (see Table 17-16). Table 17-17 provides the totals for tetrachloroethylene emissions from the AIRS database.

Table 17-16. Emissions in the AIRS Database for Tetrachloroethylene from Waste Storage Tanks, Incinerators Seepage Basins, Disposal and Landfills, Painting and Printing Operations, and Laboratories[a]

Year Source description	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
1985		
A-Area waste storage tanks (each)	3.63×10^{-7}	1.82×10^{-7}
F-Area seepage basin	2.14×10^{-2}	2.14×10^{-2}
H-Area seepage basins	1.47×10^{-10}	1.47×10^{-10}
723-A Met Lab basin	1.10×10^{-5}	1.10×10^{-5}
Landfill 740-G	2.20×10^{-3}	2.20×10^{-3}
H-Area tank farm, use for maintenance	2.40×10^{-4}	2.40×10^{-4}
M-Area settling basin	2.05×10^{-1}	2.05×10^{-1}
M-Area air stripper M1	1.13×10^{1}	3.03
M-Area use for maintenance	3.70×10^{-3}	3.70×10^{-3}
Pilot Plant incinerator	3.74×10^{-6}	4.69×10^{-9}
Burial Ground 643	1.4×10^{-8}	1.4×10^{-8}
1987 was the same as 1985 except for the addition of the		
M-Area air stripper M1	1.13×10^{1}	6.21
The same emissions estimates were given in 1990 except for		
H-Area seepage basins	1.85×10^{-10}	1.85×10^{-10}
M-Area air stripper M1	1.13×10^{1}	4.39

[a] Source: Faugl (1996l).

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Table 17-17. Total Tetrachloroethylene Emissions from the AIRS Database

Year	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
1994	13.6	3.55
1992	20.7	4.35
1990	11.3	4.40
1987	11.5	6.39
1985	11.5	3.26

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Operating Permit and Air Emissions Inventory Information about M-Area Operations

M-Area emissions estimates for the permit application included volatile solvents emitted from degreasers and operations of the cap/can and plating lines. For the solvents, a mass transfer coefficient was calculated assuming an equilibrium from liquid to vapor taking into account mixing and aeration. For calculating trichloroethane emissions from the cap/can line, the process rate was determined from knowing the number of caps and cans per shift or per hour and the number of gallons in each bath. The cap/can line cleaned the aluminum caps and cans used in slug production. The line involved nitric acid etch tanks, phosphoric acid etch tanks, degreaser, aluminux etch, and a number of hot and cold water rinses. A capacity of 165 gal of 1,1,1-trichloroethane per bath was assumed. Degreasing emissions were estimated using the average and maximum production rate, the tank volumes, and approximate amount of raw materials in each tank. Solvent emissions were assumed proportional to the surface area of the tanks. Solvent dragout onto parts and fugitive losses from holding tanks and drips were included. The standing losses because of the tanks ‘breathing’ and working losses because of vapor displacement during filling were also added to the emissions. These loses are most important for the aboveground tanks that were not insulated and expanded and contracted daily because of temperature changes.

The 1,1,1-trichlroethane used in 321-M and 313-M was recycled by a solvent distillation unit in each building in 1985. Trichloroethane was heated to its boiling point; evaporated solvent rose to the top of the still and then was condensed by cooling coils. The condensate went through a water separator and into a solvent cleaning tank. The oils and grease were removed as sludge by heating the solution. The still was considered a closed loop system.

Emission estimates for equipment were not made because the degreaser emissions were estimated using the difference of the use and disposal information. The estimates also accounted for fugitive emissions from the distillation units or hold tanks and losses to sumps (Radian 1992a).

Use of 1,1,1-trichloroethane in M-Area seemed to be a function of the production of caps and cans. The cores were plated then put inside a can, so there was a one to one correspondence between the number of caps and cans cleaned and the number of parts plated. Degreasers were completely changed out from one to four times each year. Engineers calculating the emissions for

1996 permit plotted the trichloroethane use as a function of production for each month from November 1986 to August 1987 and found that the use and production were correlated as expected. In 1992, Radian tried to correlate the use of trichloroethane reported in degreaser and solvent disposal logs to production from November 1986 to August 1987. After accounting for tank cleaning and changeout of degreasers and the number of parts from storage or recovery, which inflated the number of caps and cans actually cleaned, a correlation was found (Radian 1992a) (see Table 17-18).

Table 17-18. Estimates of the Production and Use of Trichloroethane Cleaning Solvent from November 1986 To August 1987[a]

Month and year	Number of parts produced	Gallons of 1,1,1- trichloroethane used
11/86	11881	170
12/86	15795 [a]	258
01/87	11743	255
02/87	7744	163
03/87	12427	532
04/87	12739	113 (278)[b]
05/87	5887	256 (91)[b]
06/87	2507	72
07/87	5468	338
08/87	713	129

[a] In December 1986, 32,652 parts were produced, but 16,857 parts were thought to have come from storage or recovery and were not reflective of the number actually cleaned that month.

[b] Degreaser records for April 1987 suggested than only 113 gal of trichloroethane were added for 12,739 parts plated. The tank was cleaned on May 7. Radian attributed the cleanout volume to April and subtracted the amount for May, averaging the two points. Adjusted values are in parenthesis.

Source: (Radian 1992a).

Emissions from the cap/can line were listed for 61 sources. A maximum production rate of 110 cans h^{-1} and an average rate of 528 caps and 528 cans per shift was used for production of all types of caps and cans. The degreasers held 165 gal of trichloroethane each, and evaporation and dragout emissions were exhausted out one of several hoods along the line (Radian 1992a).

The calculations given in the notebook were for documentation of general methods and do not describe how emissions for each source were calculated. It seems that the methods required more detailed information than we have been able to collect for the dose reconstruction. The methods also seem to be conservative. The estimates are probably less uncertain than any we could calculate based on information available to us now. In their 1992 recommendations for improving the emissions estimates, Radian suggested that better use data, tank throughput data, waste generation and solvent charge data records should have been kept, which suggested that these records were incomplete or inadequate. Used solvent was shipped to offsite solvent recovery, and although the shipping records were not compiled in an areawide useful format, it appears that Radian used these types of records to estimate solvent disposal.

The disposal rate was used to determine the production in comparison to other years. Radian reported that interviewees indicated that the tool degreaser operated about 4 hours per day and was covered when not in use. Because the use and disposal data for this equipment were not available, Radian (1992a) used the production for caps/cans and assumed that emissions were a function of the production rate.

Radian (1992a) estimated actual emissions for 1985–1990 using use and disposal logbook data obtained during interviews. Fugitive emissions (dragout, recycling still losses, holding tanks, floor sumps, etc.) were calculated as the difference between use and disposal volumes. As *RAC* has also found, these data were not available for much of the time period. However, data on trichloroethane use from November 1986 to August 1987 were plotted against production through 313-M (see Table 17-18). After making corrections for cleaning out the tanks and adding stored parts into the production, the two correlated well. Therefore, the use data for this 1 year, where it seemed most complete, was scaled based on production for other years. Although the degreasers have cooling coils that might be considered a pollution reduction device, no other abatement equipment was in place and all emissions were assumed to have been uncontrolled. The operating procedures (DPSOL313-230) specified that the heat was to be turned off and the degreaser cover was to be kept closed when the degreaser was not in use. If this was done, the downtime releases should have been minimal. The tool degreaser was assumed to have been used 4 hours each day and was covered when not in use. The cap/can degreaser operated 24 hours each day in 1986. The ultrasonic cleaners in 321-M used Freon, so they were not included.

Peak production for Building 313-M was said to have been in 1985 and 1986, ‘ramping down’ in 1987, and stopping in 1988. The emissions rate, based on maximum solvent use during this peak production period for the degreaser, was reported to have been about 96 ton y^{-1} in 1985 and 1986. Degreaser emissions for 1987 totaled 43.3 ton (Westinghouse 1996a).

For 1985, 1986, and 1987, a materials balance approach was also used, calculating the amount emitted as the difference between the amount used and the amount reported to have been disposed. The use and disposal data for 1986–1987 were scaled to production for other years and provided the basis for the annual actual estimates for 1985–1990.

Emissions for 1987 and 1988 were calculated by comparing production rates for 1986 with years for which log information was missing. The production in 1987 was said to be 200,000 parts, resulting in 19 ton y^{-1} of degreaser solvent from a Freon degreaser. Production in 1988 was said to have been 175,000 parts, corresponding to 16.6 ton y^{-1} based on the 1987 degreaser log data.

Calculations of trichloroethane release from degreasers involved using a measured emissions rate for a Freon degreaser bath, then applying this value to other degreasers by relating the surface areas and vapor pressures of other solvents in various tanks and baths. The design annual emission rate was obtained by converting pounds per hour to pounds per year, assuming 8760 h y^{-1}.

Emissions test data from 321-M for Freon were applied to the degreasers in 313-M by assuming that solvent releases were proportional to the tank surface area. Because the monitoring data were for Freon, Radian (1992a) used the ratio of the vapor pressure of trichloroethane to Freon to estimate trichloroethane emissions.

For example, the 321-M degreaser for which stack test data was available was 10,080 in^2 and used Freon. The 313-M cap/can degreaser of interest was 2520 in^2 and used trichloroethane.

$$65.4\,\text{lb}\,\text{h}^{-1} \times \frac{263\,\text{mmHg vapor pressure trichloroethane}}{602\,\text{mmHg vapor pressure Freon}} \times \frac{2520\,\text{in}^2}{10080\,\text{in}^2} = 7.14\,\text{lb}\,\text{in}^{-2}\ \text{trichloroethane}$$

$$7.14\,\text{lb}\,\text{h}^{-1} \times 8760\,\text{h}\,\text{y}^{-1} = 62{,}546\,\text{lb}\,\text{y}^{-1} = 31.3\,\text{ton}\,\text{y}^{-1}\ \text{trichloroethane maximum design emissions}$$

This kind of calculation for maximum design emissions was done for all of the degreasers in M-Area.

The degreaser in 313-M was not used in 1988, 1989, or 1990, so the actual emissions estimates were zero. Emissions estimates for the specific degreasers were obtained from the worksheets and are summarized in Table 17-19.

Table 17-19. Estimates of Solvent Emissions from M-Area Degreasers for Selected Years Between 1985 and 1990[a]

Equipment	Years	Pounds per year design maximum	Pounds per year actual based on (use – disposal)	Comments
313-M cap/can degreaser	1985	62,546	40,874 (47408 – 6534)	1985, 90% production
	1986		45,413 (52673 – 7260)	1986, 100% production
	1987		13,624 (15802 – 2178)	1987, 30% production
	1988	62,546	0	degreaser not in use,
	1989		0	degreaser empty
	1990		0	
Tool degreaser	1985–1990	62,546	7569	scaled by 4 hrs/24 hrs
320-M solvent degreaser	1985	250,300	145770	actual based onsite
	1986		145770	interviews
	1987		72920	
	1988–1990	250,300	0	degreaser not in use, degreaser empty
321-M tube cleaning degreaser	1985	250,300		
	1986			
	1987			
	1988–1990		0	used Freon
321-M component cleaning degreaser	1985	85,664	5214	based on production,
	1986			related to 1986
	1987			
	1988–1990	85,664	0	degreaser not used, degreaser empty

[a] These estimates were developed by Radian (1992a) for the air emissions inventory.

Degreaser releases in Table 17-19 totaled 99.7 ton in 1985, 99.3 ton in 1986, and 47 ton in 1987. Working losses from the trichloroethane tanks were also calculated. The large tank had an

economizer vent to minimize breathing losses. Working losses for the tank were 116.4 lb y^{-1} from 1985 to 1988 and zero in 1988 and 1989 when tank was not used (Radian 1992a).

From 1987 to 1995, the TRI, reported to the EPA and SCDHEC, included estimates of the pounds per year of 1,1,1-trichloroethane released from the SRS (Westinghouse 1996b) (Table 17-20). The estimate for 1987 is much larger than the air permit application estimates (Westinghouse 1996a).

Table 17-20. Estimates of 1,1,1-Trichloroethane Released from the SRS

Year	Air emissions(lb y^{-1})	Ton y^{-1}
1987	429,800	215
1988	10,400	5.2
1989	21,300	10.6
1990	18,290	9.2

[a] Source: Westinghouse (1996b).

Summary

This section has presented many pieces of information on the use and release of chlorinated solvents from M-Area. Several approaches could be used to estimate releases of chlorinated solvents to the atmosphere based on the information collected from historical reports and the more recent air emissions inventory information. It is difficult to know which data to use and how to weight information according to its reliability or applicability to developing a source term. Data from the mid to late 1980s are most complete, but they might not apply to the 1960s when the older, leaking degreasers were in use.

We believe that almost all of the chlorinated solvents used were discharged to surface water until 1979 and most of this evaporated. In other studies of this kind, where information on site-specific environmental fate and transport are lacking, researchers have estimated that 95–100% of the chlorinated solvent was released to surface water or soil evaporates (McGavran et al. 1996; ChemRisk 1994). Unfortunately, inventory amounts did not always reflect use and are not available for all facilities or all time periods. Christensen and Brendell (1981) attempted to base their release estimates on solvent consumption estimates, which were based on purchase records. However, they found that the records for trichloroethylene were not available, and their use estimates had to be derived from “M Area personnel judgment.”

Christensen and Brendell (1981) estimated that solvent use from 1952 to 1979 totaled about 13 million lb or about 6500 ton. They estimated approximately 1850 ton of trichloroethylene, 4350 ton of tetrachloroethylene, and 335 ton of trichloroethane were used during that time period. For 1970, Christensen and Brendell (1981) estimated 89 ton of trichloroethylene and 54 ton of tetrachloroethylene were used. Monier (1970) estimated the gallons of 100% solvent released to Tim’s Branch and the settling basin, which corresponded to 58.5 ton of trichloroethylene and 66.9 ton of tetrachloroethylene, using a 10.9 lb gal^{-1} conversion. Monier (1970), in a 1-page memo, presents values for tetrachloroethylene discharged that are about 20% greater than Christensen and Brendell (1981) estimates for use. However, the Monier (1970) estimate for trichloroethylene is about 30% of the Christensen and Brendell (1981) use estimate. The Monier (1970) estimates were said to have been based on production levels, but how the estimates were made is unknown. Hardt (1970) estimated that 6 ton of trichloroethylene and 45.5 ton of tetrachloroethylene were

released to Tim's Branch in 1 year. Looney et al. (1987) estimated that 16.5 ton of trichloroethylene and 92.4 ton of tetrachloroethylene were discharged to the settling basin from 1971–1985, which averages to about 1.1 ton y^{-1} of trichloroethylene and about 6.2 ton y^{-1} for tetrachloroethylene over the 15-year period. The variability of the estimates is large, but the estimates of Christensen and Brendell seem larger than most. Their estimates seem to be the most defensible because they explained the methods used to calculate their estimates.

Essential Materials Ledgers suggest that about 2.4×10^6 kg or 2662 ton of trichloroethane was used from 1979–1986 (Westinghouse 1984, 1985, 1987a). The 1979 Essential Materials Reports suggest that 253 ton was used that year, an estimate nearly 3 fold larger than the 85-ton estimate reported in Christensen and Brendell (1981).

The largest shipments of chemicals were received by M-Area between August 1984 and September 1986 (Gary 1996). The permit application and Site workers stated that production for Building 313-M was greatest in 1985 and 1986, began ramping down in 1987, and stopped in 1988 (Westinghouse 1996a; Gary 1996). Radian determined that peak production was in 1986. They called this 100% and assumed that 1985 production was 90% and 1987 production was 30% of the production in 1986 (Radian 1992a). Radian (1992a) actual emissions estimates implied that five degreasers were operating in 1985, but only three were operating in 1986. The essential materials ledgers suggest that more trichloroethane was used in 1985 than in 1986 (Westinghouse 1987a). Based on production information compiled for the permit application (Westinghouse 1996a), we assumed that the 1987 amounts were 30%, the 1988 amounts were 26% of the 1986 amounts, and that there was no production in 1989.

If we subtract the amount of solvent estimated by Christensen and Brendell (1981), shown in Table 17-6, to have gone to the Tim's Branch and settling basin from the total amount used for the different time periods, then from 77–85% of the solvent remains. This amount might have evaporated during routine use in M-Area. If 95% of what was released to the sewers and then to the Tim's Branch or the settling basin in turn evaporated, then the total amount evaporated would be the amount evaporating during use plus 95% of what was discharged. For the values derived by Christensen and Brendell, this seems an unnecessarily complex analysis given the uncertainty of the values. As demonstrated in Table 17-21 (for trichloroethylene and the first 8 years of tetrachloroethylene use), the total amount evaporated from use and discharge to surface waters equals about 99% of the amount used. From 0.9–1.2% may have remained in groundwater.

If we assume that 95–99% of the solvent used before 1980 evaporated into the air (eventually as it was used in degreasers or after being released into liquid effluent), then we might estimate that the range of amounts given in Table 17-22 might have evaporated into the air. All of the estimates in the second and third columns of Table Table 17-22 are from Christensen and Brendell (1981).

Table 17-21. Trichloroethylene and Tetrachloroethylene Evaporated during Use and Surface Water Discharge[a]

Time period	Tons used per year minus release to sewers	Tons remaining	% remaining per year	95% released to sewers	Total tons release to air each year
Trichloroethylene					
1952–1985	76.5 – 11.5	65	85	11	76
1959–1961	140 – (12.5 + 11.5)	116	83	23	139
1962–1970	89 – (12.5 + 7.5)	69	77	19	88
Tetrachloroethylene					
1962–1970	54 – (12.5 + 7.5)	34	63	19	53

[a] The amount of solvent used, minus the amount discharged to the sewers, plus the amount evaporated from sewers, equals the amount evaporated during use and surface water discharge.

Table 17-22. Use Amounts Estimated by Christensen and Brendell (1981) and 95% and 99% of These Amounts

Time period	Amount used per year	Total = amount used per year × number of years	95% of the amount used	99% of the amount used
Trichloroethylene				
1952–1958	76.5	535.5	72.7	75.7
1959–1961	140	420	133	138.6
1962–1970	89	801	84.5	88.1
1952–1970 total		1756.5	1668.7	1738.9
Tetrachloroethylene				
1962–1970	54	486	51.3	53.5
1971	185	185	175.7	183.2
1972	315	315	175.8	311.8
1973	415	415	394.2	410.8
1974	335	335	318.2	332
1975	575	575	546.2	569.2
1976	500	500	475	495
1977	560	560	532	554.4
1978	412	412	391.4	407.8
1979	412	412	391.4	407.8
1962–1979 total		4195	3985.2	4153
Trichloroethane				
1979	85	85	80.75	84.2
1980	150	150	142.5	148.5
1981	100	100	95	99
1979–1981 total		335	318.2	331.7

After 1979, trichloroethylene and tetrachloroethylene were not being used. Most of the solvent was recycled and waste solvent was barreled and buried. Eliminating most of the surface water discharges greatly reduced the amount evaporating from surface water, but trichloroethane in the degreasers continued to evaporate into the air during use and leakage of equipment or spills. After 1985, the air stripper emissions also contributed to releases to air.

The estimates in Table 17-23 are derived from the values reported in the M-Area essential materials and inventory control ledgers (Westinghouse 1984, 1985, 1987a), except for 1988, which was calculated to be 26% of the 1986 production levels according to the values compiled for the permit application (Westinghouse 1996a). We believe the 1987 ledger total of 825 ton is in error. It is more than twice a value that would be consistent with the other ledger entry totals. The handwritten 1987 entries were difficult to read and had been corrected several times. In addition, several different sources insist that production and use in 1987 was less than in 1986 or 1985 (Gary 1996; Westinghouse 1996a; Lorenz 1998). With the exception of this one entry, the largest value for each year was used. The estimate for 1987 was derived from the TRI submitted to the EPA (Westinghouse 1996b). The air permit application suggested production in 1987 was 30% of 1986, which led to an estimate of 119 ton of trichloroethane used in 1987. The TRI estimate of trichloroethane releases from the Site is 215 ton, almost twice the permit's estimate of the amount used. There are no other large sources of trichloroethane at the SRS as far as we know to account for the difference. The TRI is certainly a conservative estimate, but 119 ton may be an underestimate.

Christensen and Brendell (1981) estimates for trichloroethane in Table 17-22 were much less than those in Table 17-23 and were not used to derive totals. After 1979, when waste solvent was barreled and more care was taken to limit releases, we assume that 75–95% of the solvent used eventually evaporated from the process, resulting in the estimates in Table 17-23.

Table 17-23. Amount of 1,1,1-Trichloroethane Used, Estimated from the Essential Materials Ledgers and Operating Permit Application, and 75% and 95% of These Amounts

Time period	Amount used	75% of the amount used	95% of the amount used
1979	253	189.7	240.4
1980	143	107.2	135.8
1981	198	148.5	188.1
1982	275	206.2	261.2
1983	308	231	292.6
1984	308	231	292.6
1985	264	198	250.8
1986	396	297	376.2
1987	215	161.2	204.2
1988	103	77.2	97.8
1979–1988 total	2463	1847.2	2339.8

These estimates are larger than the air permit applications estimates for the M-Area degreasers, which total about 100, 99, and 47 ton in 1985, 1986, and 1987, respectively.

The DOE Environmental Survey team thought that 200 ton y^{-1} of trichloroethane may have been released into the air from M-Area. The survey report mentioned that 1,1,1-trichloroethane released from the M-Area was derived from measured release data (DOE 1987). However, no records or personnel recollecting any monitoring of solvents in M-Area have been found during

our study. The monitoring data from which their estimate was derived were not summarized in their report. It is interesting that the average of 75% of the use amount for 1979–1988 is 187 ton y^{-1} and for 95% it is 243 ton. The average of 75% of the use estimates for 1984–1987, when the survey was being conducted, is about 220 ton y^{-1}.

After 1985, the air strippers released trichloroethylene and tetrachloroethylene to the air. The catox unit reduction of emissions seemed to be obvious after 1989. The accuracy of estimates in the annual reports, compared to the operating permit, compared to the AIRS database, is unknown. Because they are the largest, most conservative estimates, we used the estimates in the calculations made to estimate the AIRS database emissions estimates (Radian 1992a). The estimates in the calculations notebook appear to be higher than the AIRS database estimates for the M-Area air stripper in 1985–1987, but they are lower by as much as a factor of 2.5 for 1988–1990. For the estimates in Table 17-24, we used the highest value given for 1985–1990. We assumed that 70% of the solvent released was trichloroethylene and 30% was tetrachloroethylene based on the assessment by Colven et al. (1987).

Table 17-24. Estimates of Trichloroethylene and Tetrachloroethylene Released from Air Strippers and Soil Vacuum Extraction Units

Year	Total emissions from the air strippers in tons	Trichloroethylene	Tetrachloroethylene
1985	9.8	6.8	3.0
1986	24.4	17.1	7.3
1987	22.1	15.5	6.6
1988	18.4	12.8	5.6
1989	14.2	9.9	4.2
1990	4.4	3.1	1.3
1992	8.6	6.0	2.6
1985–1992 total	102	71.3	30.6

We might refine this estimate by subtracting what was known to have contaminated groundwater, been barreled for disposal, or retained in the soil. We know that much of the tetrachloroethylene was recycled by distillation, beginning in about 1971, but eventually most of this would have evaporated. We know some of the solvent released to soil and surface water was degraded and some remained complexed in the soil. However, environmental fate and transport studies on lakes and streams suggest this was probably a small percentage of the total. We know an amount of solvent worthy of remediation has seeped into the groundwater below M-Area. We have little basis for a reasonably certain estimate of how much solvent traveled to the groundwater below M-Area. We might also assume that a large percentage of the solvents in the groundwater will eventually be removed and released to the air through the remedial air stripping program. Some of the information available about these quantities, generated by the Site for all three solvents are summarized in Table 17-25.

Table 17-25. Summary of the Use and Release and Inventory Estimates for Chlorinated Solvents in M-Area for Various Time Periods (amounts are in tons)

Inventory	Time period	Trichloro-ethylene	Tetrachloro-ethylene	Trichloro-ethane	Reference
Settling basin soil inventory	1971–1985	0.18	3.96	0.15	Looney et al. (1987)
Total amount in the groundwater below M-Area, total solvent reported, 70% reported to be trichloroethylene	1987	121	51.7		Colven et al. (1987)
In the groundwater, 70% assumed to be trichloroethylene	1992	99	42.7		Arnett et al. (1993)

Similar kinds of amounts were reported for all three chlorinated solvents combined. From 1952 to 1970, we can assume that no trichloroethane was used, but it is hard to estimate what percentage of the total is trichloroethylene compared to tetrachloroethylene. Table 17-26 summarizes the various total inventory amounts in groundwater and estimates of the amount removed from groundwater using the air strippers taken from various reports.

Table 17-26. Total Chlorinated Solvent Estimated Inventories from Various Reports

Inventory	Time period	Amount of solvent (in kg)	Reference
Total amount of solvents in groundwater in 1990	1989	225 ton	Bebbington (1990)
Amount estimated to have been removed from groundwater as of 1992	1992	143 ton	Arnett et al. (1993)
M-Area air stripper	1985–1990	176 ton for 5 years	Westinghouse (1996a)
M-Area air stripper	1985–1990	102 ton for 6 years	Radian (1992a)
M-Area and A-Area air stripper	1985–1994	121 ton for 10 years	Faugl (1996l, 1996m)
M-Area air stripper	1987	35 ton for year	DOE (1987)

An estimate of releases of all three solvents to the air could be made more accurate by subtracting the amount thought to have been retained in the settling basin soil and the amount thought to have traveled to the groundwater, then adding back the air stripper releases. However, the estimates we have for the amount in groundwater and soil are very uncertain and very small relative to the total amounts released. We added the air stripper releases because they occurred from 1985 to the present time, were based on monitoring data, and seemed fairly reliable.

Estimates of the amount of solvent in groundwater or removed from groundwater since 1985 vary from about 100 ton to more than 200 ton as shown in Tables 17-25 and 17-26. This is 2–3%

of the total amount of trichloroethylene and tetrachloroethylene that we estimate may have been used. Given the wide range of uncertainty associated with these estimates, the subtraction of the relatively small amount lost to groundwater would make little difference to the totals. These solvents are fairly soluble in water and would not be expected to be retained in settling basin soil. Looney et al. (1987) estimates of the total solvent inventory in basin soil are less than 0.07% of the total amount of the two solvents used. Subtracting these values would make the total estimates less conservative. For these reasons, subtracting the amounts did not seem justified.

We might also try to refine the estimate using what we know about the volatility of the solvents and the history of operations in M-Area. Although tetrachloroethylene is less volatile than trichloroethylene, it was used during a time when the degreasers were older, more corroded, and leaking. In 1979, we know that new degreasers were put in place, tetrachloroethylene was replaced with trichloroethane, and solvent residues and sludge were being barreled for disposal. Much less trichloroethane was discharged to the process sewers and Tim's Branch after 1979. Before waste solvents were barreled and buried in 1979, almost all of the solvent used eventually evaporated. Assuming that 95–99% of what was used eventually evaporated seems to be a reasonable but cautious assumption.

Based on the assumption that 95–99% of what was used before 1980 and 75–95% of what was used after 1980 was released to the air, the ranges total

- 1740–1810 ton of trichloroethylene: 1669–1739 ton from M-Area operations from 1952–1970 and 71 ton from the air strippers from 1985–1992
- 4016–4184 ton of tetrachloroethylene: 3985–4153 ton from M-Area operations from 1962–1979 and 31 ton from the from the air strippers from 1985–1992
- 1847–2340 ton of 1,1,1-trichloroethane from M-Area operations from 1979–1988.

The total for all three solvents is 7603–8334 ton. These ranges represent the assumption of 95–99% for 1952–1979 and 75–95% for 1980–1992. These ranges do not represent propagated uncertainty values.

These estimates are probably greater than a central value because of the conservative assumptions made for the air emissions inventory and permit application and our cautious assumption that 95–99% of what was used from 1952–1979 and 75–95% of what was used from 1979–1988 eventually evaporated. We do not know whether the estimates used for 1952–1985 overestimate or underestimate the true releases. Some people may think that because the degreasers are covered when not in use, used solvent was distilled and recycled and still bottoms were barreled after 1979, estimating 75–99% of the solvent used volatilized is an overestimate. However, we have little information on the condition of the degreasers or about leaks and spills before 1985 and studies of other degreasing operations have shown that assuming 75–99% evaporation is not unreasonable.

The uncertainty in the estimates is large. Various estimates of usage from different sources suggest that use estimates may vary by as much as a factor of 3, especially for those derived from monthly ledgers. The amount evaporated may have varied from 75–99% of what was used, depending on the retention of the material, migration in groundwater, degradation, and other parameters for which we have little or no site-specific information.

Based on a factor of three uncertainty in usage and using likely usage quantities described previously, and also utilizing ranges of percent released from volatilization, uncertainty was calculated for chlorinated solvents. Triangular distributions were assigned to the usage estimates,

with the preferred value being the most likely estimate. Uncertainty in percent volatilized was represented by uniform distributions. Total releases for the periods during which each solvent was used were estimated for comparison with the above totals.

For trichloroethylene, usage during the years 1952–1970 resulted in total releases from M-Area operations that are lognormally distributed with a geometric mean of 1700 ton and a geometric standard deviation of 1.07. This distribution increases the range of releases between the 5th and 95th percentiles to 1530–1900 ton. This 5th–95th percentile range is used to describe all subsequent uncertainty ranges. Releases from the air strippers for this solvent from 1985–1992 are also lognormally distributed with a geometric mean of 70 ton and a geometric standard deviation of 1.14. The range on these releases is from 57–87 ton.

Tetrachloroethylene releases total 4055 ton with a GSD = 1.09 for the years 1962–1979, with a range from 3520–4675 ton. For the air strippers, this solvent was released over the period from 1985–1992 in the quantity of 30 ton with a GSD = 1.2. The air stripper releases ranged from 22–40 ton.

A variety of trichloroethane release estimates were made from 1979–1988. Factor of three uncertainty bounds were placed on releases that did not have estimates with a range this great, but even larger bounds were used if the estimates of release exceeded a factor of three. Uncertainty calculations for releases of this solvent were lognormally distributed with a GM=2200 ton and a GSD=1.14, with releases ranging from 1780–2710 ton.

The Standard 8 results submitted to SCDHEC said that in 1991, trichloroethylene was emitted by 24 sources in A-Area, E-Area, F-Area, G-Area, H-Area, L-Area, and M-Area. The ambient standard was 6750 $\mu g\ m^{-3}$, and the maximum 24-hour average Site boundary concentration was calculated to be 4.77 $\mu g\ m^{-3}$. The Standard 8 results submitted to SCDHEC said that in 1991, tetrachloroethylene was released from 22 sources in A-Area, E-Area, F-Area, G-Area, H-Area, M-Area, and N-Area. The maximum 24-hour average Site boundary concentration was calculated to be 2.0 $\mu g\ m^{-3}$. The ambient air standard was 3350 $\mu g\ m^{-3}$. Since M-Area operations have stopped, tetrachloroethylene and trichloroethane releases seem to be from many different sources that use small amounts for cleaning, degreasing, or laboratory work.

Chlorinated Solvent Releases from the Reactor Areas

Chlorinated degreasing solvents were also used in the reactor areas, as a cleaner in the tritium facilities, and probably in the canyons as well.

In an analysis of M-Area use of solvents, Christensen and Brendell (1981) reported that each of the reactor areas (C, K, L, P, and R) had 1000-gal degreasers. They also noted that the 105 Buildings in the reactor areas obtained one or two barrels of solvent from M-Area storage each year (Christensen and Brendell 1981). We have been unable to find records that characterize the use of the degreasers in the 100-Areas. It is likely that each was used as long as each reactor was operating. Documentation on the rate of solvent use and how dirty solvents were disposed of has not been found. Emissions of chlorinated solvents from the reactor areas were not included in the AIRS database emissions estimates.

Trichloroethylene is also mentioned in several reports as having been a quality assurance concern in the reactor areas. A Works Technical Monthly Report from February 1953 says that a "difficulty with trichloroethylene in the 305 reactor had been substantially eliminated" (Du Pont 1953). Reactivity had been nearly restored to its initial level. However, there were still traces of

trichloroethylene in the effluent gas that was being recirculated through a purification system. They expected it to take months to remove all traces. Meanwhile, the reactor was back in normal operation. Efforts to avoid any similar problem in the 100-Areas were intensified. The Reactor Technology Section of the report for this same month reports that a means for detecting small quantities of triclene in the reactor complex and in fuel column assemblies was investigated. The GE leak detector was being used for this purpose in 305 Building, and this appeared to be a satisfactory method.

The Metallurgy Section Report added that trichloroethylene was being removed in a 3-week air purge at a rate of about 2 g h^{-1}. Helium was being discharged from the pile, purified of trichloroethylene, and recirculated through the pile (Du Pont 1953). Mention of this problem does not occur in subsequent years. Although the nature of the problem is not completely clear, it did not appear to involve environmental releases or large quantities of solvent and does not seem to be of concern for dose reconstruction. It seems that trace amounts of trichloroethylene on fuel, and perhaps other reactor components, led to trichloroethylene vapors in the blanket gas.

Chromium

Chromium is a carcinogenic metal. Hexavalent chromium (VI) is more toxic than the predominant form in the environment, trivalent chromium (III). Chromium (VI) is corrosive and causes ulcerations, liver and kidney damage, and a wide range of respiratory effects. Hexavalent chromium causes lung, nasal, gastric, and other cancers in humans, and high levels cause birth defects in laboratory animals.

Chromium may have been released to the air as a contaminant of ash from coal burning. Particulates from coal-fired power plants have been shown to contain chromium in the range of 2.3–31 ppm, reduced to 0.19–6.6 ppm by fly ash collectors (EPA1988).

Emission factors for drift losses from cooling towers described in AP-42 could be used to calculate chromium emitted as drift from the towers as water cycles through and aerosolization from cooling towers. These calculations would be based on the assumption that dissolved salts and suspended solids become concentrated in the liquid phase as a result of evaporation. However, data on the throughput through cooling towers is not readily available. Although chromium concentrations in some of the cooling and process water spills (summarized in Chapter 18) were estimated, we do not know the concentration of chromium in cooling water, nor the time periods for which chromium was used for water treatment in different areas. Lacking this information, we can not estimate chromium releases to the air from cooling water with acceptable uncertainty. A screening calculation based on information about how much cooling and process water was treated with chromium and how much of this water was run through some sort of cooling tower where it may have be subject to aerosolization might be useful. Based on chromium concentrations and throughput data, 37 kg of chromium was the maximum amount calculated to have drifted from reactor cooling water at Argonne National Laboratory-West in 1972, under worst-case conditions when the throughput was an estimated 1.8×10^7 gal y^{-1} (Argonne National Laboratory-West 1973). Although worst-case, this amount was below levels of industrial hygiene concern for workers working under the towers. SRS progress reports contain information on discharges of water to surface streams and seepage basins. From this, a maximum of 2.1×10^{11} gal y^{-1} was discharged from the reactor areas and separations areas. If all of this water was heavily treated with chromium, like the water used at the Argonne Cooling Towers,

then as much as 475 ton of chromium may have been aerosolized, for the worst case. Generally, drift from cooling towers and aerosolization of chromium-treated water from other uses may be of concern for worker exposure, but they are not of concern for offsite exposures. Emissions estimates for chromium were extracted from the AIRS database. Chromium sources for 1985 included 56 welders, 3 diesel fire protection pumps, 16 diesel generators, 21 emergency diesel generators, and 12 grinders. In 1987, chromium emissions were reported from 31 welders, 3 diesel pumps, 18 diesel generators, 21 emergency diesel generators, 5 grinders, and 1 blaster. In 1991, chromium releases were estimated for 8 grinders; 54 welding units (listed as welding exhaust, arc welders, welding machines, plasma arcs, arc torches, welding tables, pack welders, seam welders, tank welders, and other welding operations); 3 pumps; 21 diesel generators; 22 emergency diesel generators; and 1 blaster. These sources were calculated to emit less than 10^{-3} ton y^{-1} actual in 1985 and most of these emissions estimates were less than 10^{-6} ton y^{-1}.

Table 17-27. Chromium Releases of Most Concern

Year and source description	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
1985		
Coal Storage pile in H-Area	1.5×10^{-3}	9.0×10^{-5}
H-Area Powerhouse, ash sluice sump	2.2×10^{-4}	4.4×10^{-5}
H-Area Manufacturing Building 232 radiological equipment repair	7.8×10^{-3}	1.3×10^{-4}
H-Area Manufacturing Building 234 finishing operations	1.34×10^{-4}	1.43×10^{-4}
H-Area Manufacturing Building 234 inert finishing operations	1.73×10^{-5}	1.73×10^{-5}
H-Area Manufacturing Building 234 fabricated metals machining	2.60×10^{-1}	8.69×10^{-2}
H-Area Reclamation Building 238 Milling and machining hood	1.17×10^{-3}	1.70×10^{-4}
H-Area Reclamation Building 238 lathe hood, Room 1	2.35×10^{-2}	2.59×10^{-4}
H-Area, Building 241058 Maintenance and E&I shops grinding		
There are four of these:		
1. Aluminum grinder	3.13×10^{-2}	1.30×10^{-3}
2. Vertical drill press	3.13×10^{-2}	3.91×10^{-3}
3. Band saw	3.13×10^{-2}	1.30×10^{-3}
4. Abrasive Belt Grinder	3.13×10^{-2}	1.30×10^{-3}
K-Area coal pile	4.9×10^{-5}	4.9×10^{-5}
K-Area coal storage run-off containment basin	4.9×10^{-5}	4.9×10^{-5}
1987 emissions were the same as 1985, with the addition of:		
H-Area Building 232 Hood metallurgy	4.50×10^{-2}	1.08×10^{-3}
H-Area Building 232 Hood cutting	2.85×10^{-2}	1.34×10^{-3}
H-Area Manufacturing Building 232 radiological equipment repair	7.8×10^{-3}	1.3×10^{-4}
H-Area Manufacturing Building 234 finishing operations	1.56×10^{-4}	1.56×10^{-4}
H-Area Manufacturing Building 234 inert finishing operations	3.19×10^{-5}	3.19×10^{-5}
H-Area Manufacturing Building 234 fabricated metals machining	2.60×10^{-1}	8.69×10^{-2}

Year and source description	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
H-Area Reclamation Building 238 milling and machining hood	1.17×10^{-3}	1.64×10^{-4}
H-Area Reclamation Building 238 lathe hood, Room 1	2.35×10^{-2}	3.29×10^{-5}
H-Area, Building 241058 Maintenance and E&I shops		
Grinding included four sources:		
1. Aluminum grinder	3.13×10^{-2}	1.30×10^{-3}
2. Vertical drill press	3.13×10^{-2}	3.91×10^{-3}
3. Band saw	3.13×10^{-2}	7.14×10^{-3}
4. Abrasive belt grinder	3.13×10^{-2}	1.30×10^{-3}
K-Area coal pile	4.9×10^{-5}	4.9×10^{-5}
K-Area coal storage run-off containment basin	4.9×10^{-5}	4.9×10^{-5}
1990		
H-Area Building 232 hood metallurgy	4.50×10^{-2}	1.08×10^{-3}
H-Area Building 232 hood cutting	2.85×10^{-2}	1.34×10^{-3}
H-Area Manufacturing Building 232 radiological equipment repair	7.8×10^{-3}	6.50×10^{-5}
H-Area Manufacturing Building 234 finishing operations	1.55×10^{-4}	1.56×10^{-4}
H-Area Manufacturing Building 234 inert finishing operations	3.19×10^{-5}	1.43×10^{-5}
H-Area Manufacturing Building 234 fabricated metals machining	2.60×10^{-1}	8.69×10^{-2}
H-Area Reclamation Building 238 milling and machining hood	1.17×10^{-3}	1.57×10^{-4}
H-Area Reclamation Building 238 lathe hood, Room 1	2.35×10^{-2}	3.77×10^{-5}
K-Area coal pile	4.9×10^{-5}	4.9×10^{-5}
K-Area coal storage run-off containment basin	4.9×10^{-5}	4.9×10^{-5}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Table 17-28 presents the total emissions for years 1985, 1987, 1990, and 1992.

Table 17-28. Total Chromium Emissions for 1985, 1987, 1990, and 1992

Year	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
1985	4.51×10^{-1}	9.92×10^{-2}
1987	4.85×10^{-1}	1.02×10^{-1}
1990	4.89×10^{-1}	1.01×10^{-1}
1992	4.14×10^{-1}	5.37×10^{-2}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

About 125 emissions points were listed for chromium each year. The largest air emissions for chromium reported were from machining in 234-H, followed by the maintenance shops grinding and drilling operations in H-Area (Faugl 1996d). The total actual release estimate in the AIRS database was about 0.1 ton y^{-1} or 200 lb y^{-1} in the late 1980s and 1990s. The maximum release estimate in the AIRS was about 0.46 ton y^{-1}. It is likely that metal fabrication and welding operations may have been greater in the early years when many facilities were being constructed. Generator use is probably greater today than in the early years of operations. Chromium releases probably totaled from 0.1 to 0.5 ton y^{-1} for most years. There was not enough information available to do useful uncertainty calculations for the chromium release estimates. There is insufficient information available to estimate chromium releases from its use as a corrosion inhibitor in cooling and process water with reasonable certainty.

Coal

Much of the electric power distributed at the SRS was generated onsite at the power plants. These plants also provided steam for the Site. Historically, electricity and steam at the SRS has been generated by burning coal. The coal was generally moderately to low sulfur coal, which was received by rail, sprayed with water, and stored in open piles. Coal piles originally existed in A-Area, C-Area, D-Area, F-Area, K-Area, H-Area, L-Area, P-Area, and R-Area. The coal pile in R-Area was removed in 1964, the L-Area coal pile was removed in 1968, and the coal piles in C-Area and F-Area were removed in 1985. In 1991, the K-Area coal pile was removed down to a 2-in. base and 75% of the P-Area coal pile was also removed (Arnett et al. 1993).

The power facilities usually kept at least a 90-day supply of coal, which was not rotated; therefore, the coal was subjected to long-term exposure, weathering, and release to the environment. Oxidation of sulfur compounds in coal piles during weathering can result in the formation of sulfuric acid, and the acid and metals can contaminate rainwater runoff and leachate from coal piles. Releases to surface water from coal and ash are discussed in Chapter 18.

There were seven coal-fired steam plants onsite: 484-D, which has four pulverized coal-fired boilers; 784-A, 184-C, 184-K, and 184-P, which all have two boilers; 284-F with four boilers; and 284-H with three boilers (DOE 1987). The SRS power plants have been in operation since 1952. They have perhaps lasted longer than most commercial coal power plants due, in part, to the fact that only two or three of the four boilers in D-Area and one or two of the two or three boilers in the other areas operated at any one time (Faugl 1996a). The L-Area powerhouse was put out of service in about 1964, R-Area stopped operating in about 1978, and C-Area and F-Area

powerhouses were shutdown between 1985 and 1988. In 1987, the D-Area to F-Area steam line was installed. The K-Area powerhouse has burned oil since 1994 and the P-Area powerhouse has burned oil since about 1987 (Willis 1997). The powerhouses appear to have been operated independently from other facilities in each area. Mention of their operations was rarely made in the monthly progress reports. Very few historical records about the operations of the power plants were located in Phase I. Most of the information on the powerhouses came from interviews with current and former employees of the Power Department, from the recent Title V Part 70 Air Permit (Westinghouse 1996a), or the Site annual environmental reports.

Coal Consumption and Composition

The amount of coal burned before 1972 is unknown. The annual environmental reports summarized the status of power plants onsite and reported the amount of coal burned and the approximate sulfur content of the coal. Beginning in 1972, the reports gave estimates of the power plant emissions of sulfur dioxide, nitrogen dioxide, and fly ash particulates in the early and mid-1970s and carbon monoxide, and volatile organic compounds (VOCs) in recent years, for each year. The site did not conduct analyses to estimate the content other metals, such as cadmium, chromium, lead, manganese, mercury and nickel, in the coal burned. Attempts to locate more primary data on sulfur analysis of coal and coal receipt, inventory, or storage information were not successful. Records of this type were not found in the Phase I document search. Essential materials ledgers for D-Area and the other powerhouses were not located. The Bureau of Mines did much of the coal analysis in earlier years. The regional office recommended we contact the office in Washington, D.C. Neither office had an idea where we might find records of coal analysis for the SRS. In 1996, Tom Thome, Stan Smith, and H.S. (Sid) Willis, with the Power Department; Robert Garvin and Mal Schroeder, who worked in D-Area; and retired workers Henry Main, Ray Fleming, Leo Shelton, Peter Gray, W.B. Holt, and Jim King were interviewed about power plant operations, essential materials records, coal inventory and analysis records, and potential releases of chemicals to the environment.

Many of the people interviewed indicated that dispersion of coal into the air from the coal pile was not considered to have been a problem. It seems that suspension of reject coal and dust from the coal storage piles were not of concern because the moisture content of the coal was very high. At some times during the year, the piles may have been dry, but dust emissions were probably not of concern for offsite exposures. Dust pollution from the piles would probably have been much less than boiler stack releases of ash and other pollutants. Any estimates of fugitive dust emissions would be very uncertain and require a modeling effort that is probably not justified given the low magnitude of the release and the distance from the coal piles to offsite populations.

Emissions of oxides of nitrogen, sulfur dioxide, metals and particulates from the powerhouses because of coal combustion were evaluated for this study. The annual environmental reports provided data on the amount of coal burned; the coal's sulfur content; estimates of the amount of sulfur dioxide, fly ash, and oxides of nitrogen emitted; and how the emissions compared to South Carolina Emission Standards effective at the time. This information and information from other reports about the coal burned and the ash and particulate matter (PM) released was compiled and is summarized in this section. Later in this chapter, nitrogen dioxide

and sulfur dioxide emissions from coal are characterized, along with nitrogen dioxide and sulfur dioxide emissions from other sources. We also assessed the metals contained in coal and in ash emissions and address them individually in this chapter.

Powerhouses

D-Area Powerhouse. The largest power plant was in D-Area. The D-Area power plant has four units or boilers with a total capacity of 396×10^6 BTU h^{-1} (Evans and Giesey 1978). The plant was described in Savannah River Ecology Laboratory (SREL) documents published in the early and mid-1970s as a 83-MW pulverized coal-fired plant with four 38-m stacks that had been in operation since 1952 and burned approximately 2.6×10^5 metric ton of coal each year. The coal and ash disposal and runoff systems used in D-Area are described in Chapter 18.

H-Area Powerhouse. The 284-H powerhouse was southwest of the H-Canyon. The H-Area coal piles were located near the powerhouse. A coal pile runoff basin, with a maximum runoff reported to be approximately 219,400 ft^3, had an overflow pipe that drained water to an NPDES outfall. The basin was reported to be wet most of the time, and fugitive dust emissions were said to be negligible (Radian 1993). A description of the three stoker-fired steam generators at the H-Area powerhouse said that coal, received by rail in hopper-bottom cars, was dumped into a track hopper, conveyed by belt to a crusher, then to a bucket elevator, then to the silo for consumption or to the storage yard. From the silo, the coal was fed into hoppers above each boiler by conveyer. The crusher sized the coal to about ¾-in. Ash generated from this facility was said to go from cinder hoppers by water jets to a jet pump. Dust collectors removed fly ash from gases leaving the boilers. Fly ash was removed from the dust collector hoppers and the stack by a hydroveyor. The ashes were mixed in the hydroveyor with water, discharged to an air separator and then to a jet pump, which discharged to an ash basin about 1500 ft south of the boiler house (Fisk and Durant 1987). Bottom and fly ash collected from the coal-fired boilers were transported to the ash disposal basin via a wet sluice. When the ash slurry reached the basin, the suspended ash settled to the bottom and the supernatant was decanted to an NPDES outfall (Radian 1993).

K-Area Powerhouse. In 1992, two stoker boilers operated in Building 184-K. Cyclonic precipitators were used to control particulate emissions, but sulfur dioxide and nitrogen dioxide emissions were not controlled. Radian (1992b) calculated emissions from the K-Area powerhouse and from coal storage, transport, disposal, and runoff systems. Coal from rail cars was stored in piles and delivered to the crusher by conveyor. Bottom ash and fly ash were transported by wet sluice to an ash disposal (evaporation) pond.

Ash and Particulate Matter

Burning coal produces ash. Ash typically contains arsenic, cadmium, chromium, lead, mercury, and selenium. Ash is of concern as an air pollutant, contributing to particulates, and as a surface water contaminant because of contamination of runoff precipitation from ash disposal piles and leaching of toxic compounds from piles and disposal basins. The effects of ash effluents released to surface water are discussed in Chapter 18, which describes the release of chemicals to surface water.

The annual reports estimated an annual fly ash emission of 2.75 lb/10^6 BTU heat input for 1972 to 1975. It is reasonable to assume that the emissions before 1972 were similar. From 1972

to 1975, fly ash releases were said to have been within standards with the exception of fly ash in D-Area, which was reported as 2.75 lb/10^6 BTU heat input for all 4 years. The fly ash standard reported in the 1974 annual report was 0.6 lb/10^6 BTU heat input (Ashley and Zeigler 1975). In 1972, funding for equipment to reduce fly ash emissions had been obtained (Du Pont 1973a) and construction of fly ash emission control equipment was said to be underway in 1974 (Du Pont 1975). In 1973 and 1974, according to the annual reports (Du Pont 1974a, 1975), 400-D Area powerhouse was discovered to be releasing more fly ash than was then permitted from commercial electrical generating plants, so electrostatic precipitators were installed (Bebbington 1990). A 1973 report of environmental activities at the SRP compiled for press day stated that electrostatic precipitators were being installed on each of the four pulverized coal fired boilers and the major powerhouse to help comply with particulate emission regulations (Rusche 1973). In 1975, all of the standards were met except for fly ash. However, emissions from the largest source, the D-Area power plant (which burned about 65% of the coal burned at the SRS during that time), were for the first time in compliance with the standard. Electrostatic precipitators, put into operation in November 1975, were said to have reduced emissions from 2.75 lb to less than 0.03 lb/10^6 BTU heat input (Du Pont 1976). Fly ash emissions exceeded standards at the other power plants in 1976. Dust collectors were being installed at the A-Area plants, and installation was planned for plants in the other areas depending on the performance in A-Area (Du Pont 1977a). Fly ash emission estimates were reported as within standards in 1977 and 1978 (Du Pont 1978). By 1978, two-stage mechanical dust collectors had been installed on 11 of the 15 stoker-fed boilers and the remaining four were to be installed by July 1979 (Du Pont 1979). The emissions reported for 1979 were similar to 1978 estimates (Du Pont 1980). Mechanical cyclone collectors were used on all of the other boilers after 1979. In 1985, the A-Area boilers failed the test for particulate emissions. The steam input was reduced and the boilers overhauled during the next summer. They then passed State compliance tests for particulates (Ziegler et al. 1986). In 1986, the total suspended particulate standards were still set at 0.6 lb/10^6 BTU heat input (Ziegler et al. 1987). The total suspended particulates from the power plants were within applicable standards in 1987, 1988, and 1989 (Mikol et al. 1988; Davis et al. 1989; Cummins et al. 1990).

Analysis of fly ash for ^{226}Ra was an interest in 1964 after an article in the journal *Science* reported that conventional fossil fuel plants discharge relatively greater amounts of naturally occurring radionuclides into the air than nuclear powered plants of similar size (Eisenbud and Petrow 1964). The amount of ^{226}Ra in the D-Area powerhouse ash samples was measured. Coal used at the Site was said to have averaged about 9% ash. The fly ash collectors in the powerhouse were judged to be about 80% efficient, but not all of the ash formed reached the collectors. The memo stated, “The amount of fly ash discharged per ton of coal was obtained from a study made here within the past ten years by Combustion Engineering Company, acting as consultants.” This study, which was not found among the Site documents reviewed, was said to have reported 40 lb of ash per ton of coal for the D-Area Powerhouse, 10 lb ton^{-1} for F-Area and H-Area, 4 lb ton^{-1} for P-Area, 11 lb ton^{-1} for C-Area, and 11 lb ton^{-1} in the 300/700-Area (Brogdon 1964). No mention of the amount of coal burned during this time period is given. The amount of fly ash discharged appears to mean the amount discharged to the air. The amount collected by the fly ash collectors used at the time and sent to the ash basins would have likely been larger than the amounts reported. Why P-Area emissions are lower than the other areas and D-Area is so much larger was not explained (Brogdon 1964). If we weight percentages to account for the fact that most of the coal was burned in D-Area, an average of about 33 lb ton^{-1} results. The ash calculated

to have been released by applying this average estimate to the amount of coal reported to have been burned in the annual reports for the 1970s is given in the fifth column of Table 17-29.

In an effort to estimate the amount of mercury released from coal combustion, Kvartek et al. (1994) reported the amount of coal burned at the SRS each year for 1980–1993. The values are reproduced in the fourth column of Table 17-29. The source of these values or how they were derived was not explained. It is interesting that the values derived by Kvartek nearly match the annual report values, implying that perhaps they came from the same records that led to the annual report numbers.

Vendor analysis in the 1990s suggest that the coal averages about 12,180 BTU lb^{-1} or 2.43×10^7 BTU ton^{-1} (Faugl 1996a). The fly ash emissions through the 1970s were estimated to have been 2.75 lb/ BTU which corresponds to 67.1 lb ton^{-1} of fly ash.

The operating permit application implied that 2757 lb of PM were produced from coal burned in 1986. According to the annual reports, 455,000 ton of coal was burned in 1986 (Westinghouse 1996a). If we use this same relationship between coal burned and PM produced, the values in the last column of Table 17-29 result.

Table 17-29. Estimates of the Amount of Fly Ash and PM Generated Based on Estimates of the Average Amount of Coal Burned. Derived from Data in Brogdon (1964), Kvartek et al. (1994), and the Site Annual Environmental Reports

Year	Amount of coal burned reported in annual reports (ton)	Amount of coal burned reported in annual reports (kg)	Coal burned at the SRS each year estimated by Kvartek (1994) (kg)	Pounds of fly ash released per year based on Brodgen (1964) and annual reports (based on 33 lb ton^{-1})	Pounds of fly ash released per year (based on 65 lb ton^{-1})	PM emissions
1952–1971						
1972	500,000	4.5×10^8		1.65×10^7	3.2×10^7	3030
1973	500,000	4.5×10^8		1.65×10^7	3.2×10^7	3030
1974	500,000	4.5×10^8		1.65×10^7	3.2×10^7	3030
1975	500,000	4.5×10^8		1.65×10^7	3.2×10^7	3030
1976	500,000	4.5×10^8		1.65×10^7	3.2×10^7	3030
1977	500,000	4.5×10^8		1.65×10^7	3.2×10^7	3030
1978	500,000	4.5×10^8		1.65×10^7	3.2×10^7	3030
1979	500,000	4.5×10^8		1.65×10^7	3.2×10^7	3030
1980	500,000	4.5×10^8	4.0×10^8	1.65×10^7	3.2×10^7	3030
1981	500,000	4.5×10^8	4.3×10^8	1.65×10^7	3.2×10^7	3030
1982	500,000	4.5×10^8	3.9×10^8	1.65×10^7	3.2×10^7	3030
1983	460,000	4.2×10^8	4.2×10^8	1.51×10^7	3.0×10^7	2787
1984	460,000	4.2×10^8	4.1×10^8	1.51×10^7	3.0×10^7	2787
1985	455,000	4.1×10^8	4.1×10^8	1.50×10^7	3.0×10^7	2757
1986	455,000	4.1×10^8	4.0×10^8	1.50×10^7	3.0×10^7	2757
1987	453,000	4.1×10^8	4.1×10^8	1.49×10^7	2.9×10^7	2745
1988	374,000	3.4×10^8	3.4×10^8	1.23×10^7	6.3×10^7	2766
1989	227,000	2.1×10^8	2.1×10^8	7.5×10^6	1.5×10^7	1375

It is interesting to note the amounts of coal burned by the D-Area power plant reported in some of the reports on ash basin ecology. In 1975, 333,500 ton of coal, averaging 1.39% sulfur, were burned. In 1975, the ash content of the coal was said to have been 11.9%, which had increased from 9% in earlier operations (Evans and Giesey 1978). In 1978, the D-Area plant burned 451,940 metric ton of coal annually, producing 32,650 metric ton of ash, which was sluiced into a settling basin by approximately 4.5 billion L of water (Rodgurs et al. 1978).

Before 1976, or for 24 years between 1952 and 1975, fly ash was removed by mechanical cyclone separators that had a maximum efficiency of about 75%, which implied that as much as 75% of the fly ash was collected by mechanical cyclone collectors. About 40% of the ash generated was bottom ash. The electrostatic precipitators installed on each of the four stacks of the D-Area powerhouse in 1975 and 1976 were estimated to be about 99% efficient in removing fly ash from stack emissions (Weiner 1979; Evans and Giesey 1978).

Several SREL studies on the effects of ash and coal pile and basin runoff on the swamp ecosystem evaluated the deposition of ash released to the air from the D-Area power plant. Weiner (1979) noted that submicron fly ash particles can be transported considerable distances in the air and can contain many trace elements. Researchers studied 29 trace elements in soils in relation to distance from the 484-D power plant and found elevated levels of barium, beryllium, copper, mercury, manganese, selenium, and strontium near the stacks. Levels of cadmium were below the detection limit. Lead levels were not thought to be impacted by the stack emissions. Data collected in 1976 were used by Weiner (1979) and later by Evans et al. (1980) to estimate annual inputs of cadmium, copper, manganese, and lead in bulk precipitation at Skinface Pond. A total of 130.7 cm of rainfall was reported at the pond during 1976. Approximately 87% of this was sampled and analyzed. The deposition values were 0.68 mg m^{-2} for cadmium, 3.0 mg m^{-2} for copper, 3.0 mg m^{-2} for manganese, and 8.4 mg m^{-2} for lead. These levels were said to be similar to inputs reported for other rural sites, such as Lake Superior, Walker Branch in Tennessee, and a lake in Nebraska; however, proximity of these waters to power plants was not noted (Weiner 1979).

A study conducted in 1979 on bulk aerial deposition of trace elements supported a prediction of limited impact of fly ash on aerial inputs of trace metals at a distance from the power plant (Weiner 1979). The cadmium, copper, manganese, and lead levels in bulk precipitations were analyzed for this study. At a distance of 5.5 km, little of the copper, manganese, or lead deposited from air could be directly attributed to ash deposition. Researchers found no significant changes in mean concentrations after installation of electrostatic precipitators on the stacks of the power plant (previously equipped with mechanical cyclone collectors for the removal of fly ash). This led Weiner to conclude that fly ash was not a major source of airborne metals for Skinface Pond, located near Jackson, South Carolina, about 5.5 km northwest of 484-D power plant. Because the total concentration of cadmium in soil was less than the detection limit of the analytical method used, it was not determined whether the concentrations of cadmium in soil were influenced by the power plant emissions (Weiner 1979).

A similar assessment, published several years later, used the same data as Weiner (1979) and reported the aerial deposition of cadmium, copper, lead, and manganese at Skinface Pond measured for 21 months in bulk precipitation (Evans et al. 1980). The authors had already developed a model to predict cumulative deposition of fly ash as a function of distance from the power plant. They calculated a correlation coefficient of the log of the concentrations of elements in the surface soil and the log of the distance from the power plant. The soil data supported their model because it found a negative correlation between ash content and distance from the plant, but levels were very low (0.035 μg g^{-1} for mercury, 10.5 μg g^{-1} for lead, and 1.1 μg g^{-1} for arsenic). The authors concluded that based on the soil measurements and precipitation measurements, the D-Area power plant was a minor contributor of cadmium, copper, lead, and manganese to the pond. The inputs to the pond were similar or less than those measured for Lake Superior. The model had predicted that detectable elevations in metal concentrations in soils would occur only within a few kilometers of the plant. They observed a negative correlation for strontium, mercury, arsenic, and copper, which were most enriched in ash relative to background soils (Evans et al. 1980).

Evans et al. (1980) found that basin disposal was a more serious metal contamination threat to the terrestrial and aquatic ecology of the swamp near D-Area than deposition of ash released from the stacks. The deposition of copper, cadmium, lead, and manganese within 3 km of the

plant was similar to other rural areas, which suggested to Evans et al. (1980) that little of the deposition was from fly ash.

This research showed no significant changes in the concentrations of metals after installation of the new electrostatic precipitators on all four stacks of the power plant, which had previously been equipped with mechanical cyclone collectors for removal of fly ash. Evans et al. concluded that fly ash from the stack emissions was not a major source of metals to the ponds.

Ash emissions to the air from ash piles were not estimated in any of the reports because by 1985, most of the ash disposal involved water slurries and disposal basins. Dry ash piles were not mentioned as an environmental or occupational health concern in the documents we reviewed.

Taken together, the monitoring data suggest that the ash released to the air did not influence the area surrounding the D-Area power plant. Ash and coal dust release from coal and ash piles were probably not a concern for offsite exposures. Emissions of ash before 1973 were on the order of 3.3×10^7 lb y^{-1} for the entire site. After 1973, much of the ash release was controlled by precipitators. How much respirable PM was released from the power plants is difficult to estimate, but a release of about 3000 lb or 1.5 ton y^{-1} during maximum operations seems reasonable.

These estimates are quite uncertain. The D-Area power plant annual release estimates were used to predict coal burning for the entire site. This assumption introduces a non-quantifiable amount of uncertainty, which may be as large as an order of magnitude, since the D-Area power plant appears to have burned more coal than any other plant on Site.

The uncertainty that can be quantified for coal burning and therefore fly ash releases relates to the amount of ash released per ton of coal burned. The study that analyzed fly ash releases for radium predicted fly ash releases for each powerplant, with a weighted average of 33 lb ton^{-1} and a median estimate of approximately 10 lb ton^{-1}. The vendor analysis predicted fly ash releases on the order of 65 lb ton^{-1}. Because 33 lb ton^{-1} represents an actual weighted average of Site fly ash releases, this estimate for fly ash released per ton of coal burned was used as the most likely estimate for the distribution of these values ranging from 10 to 65 lb ton^{-1}, distributed triangularly.

All fly ash was assumed to be released to the environment before 1972. After 1972, the fly ash releases were assumed to be captured by pollution control equipment, which were somewhere between 75 and 99% effective at removing fly ash from the effluent stream. This distribution was assumed to be uniform.

The fly ash release estimates were calculated for each year between 1952 and 1989, based on values for amount of coal burned, estimates of fly ash released per ton of coal burned, and information about the reduction in fly ash emissions after 1972. The estimates for coal burned before 1972 were inferred from reactor production values, and estimates for coal burned from 1972–1989 were obtained from annual reports, as noted in Table 17-29. Annual estimates are shown in Figure 17-1.

Figure 17-1 shows the releases of fly ash depicted annually, along with the total amount of coal burned. It is important to note the differences in the scales for coal burned (on the right-hand side of the figure) and fly ash (on the left-hand side of the figure). With the exception of the first few years, coal use remained relatively constant over the period of study. Fly ash releases, however, decreased dramatically after 1972, when emission controls were put into place to reduce emissions from 75–99%. This had the effect of reducing the total release values, as compared to

the values in Table 17-29. The range of values represented by the 5th and 95th percentile levels reflects the uncertainty in the total amount of fly ash released per ton of coal burned.

Using these annual release estimates to calculate total fly ash releases and annual average release of fly ash over the entire period of study yields lognormal distributions. The distribution of total fly ash released from 1952–1989 had a geometric mean of 1.5×10^5 ton and a geometric standard deviation of 1.42. The average release per year had a GM of 4.2×10^3 ton y^{-1} and a GSD of 1.42. Although many of the years of operation had median fly ash releases over 8.0×10^3 ton y^{-1}, averaging all of the annual releases drops the annual average estimate to a lower level because of the inclusion of the years after 1972.

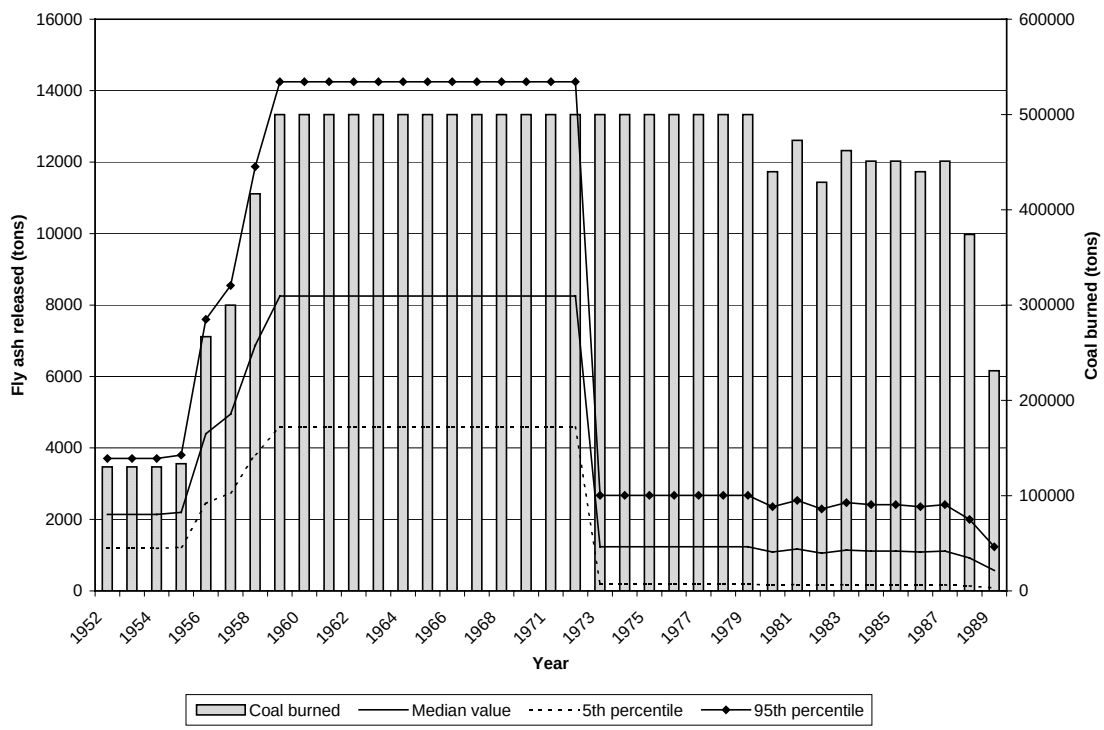

Figure 17-1. Coal burned and fly ash released calculated annually from 1952–1989. The bars in the figure represent coal burned each year in tons, with the scale on the right hand side of the figure. The lines represent the median, 5th and 95th percentile values for estimates of fly ash released annually in tons, with the scale on the left hand side of the figure.

Gasoline and Fuel Oils

Automotive gasoline typically contains more than 150 chemicals, including small amounts of benzene, xylene, toluene, and sometimes lead. Much of the health concern about gasoline has centered around the hazard from breathing vapors while filling fuel tanks. There is no evidence that exposure to gasoline causes cancer in humans, but gasoline does contain small amounts of carcinogens, like benzene and contained lead, especially before the late 1970s. Benzene is the

primary concern for gasoline emissions. Estimates of benzene releases, including those from gasoline tanks and transfer stations, are included in this chapter in the section about benzene.

The concern for this dose reconstruction study is chemicals in gasoline that might enter groundwater, surface water, or the air after being spilled or leaked from transfer tanks or storage tanks. When gasoline is spilled on soil or surface water, most of it will evaporate.

Fuels oils are petroleum mixtures used for fuel and as solvents. Examples of fuel oils are kerosene, diesel fuel, heating oil, and range oil. Fuel oil can contain varying amounts of many different hydrocarbons and additives depending on its use. Fuel oils can contain carcinogens, but for most normal uses, they do not present a carcinogenic hazard. The Occupational Safety and Health Administration (OSHA) standard for petroleum distillates is 400 ppm in air (ATSDR 1996).

Much of the gasoline and diesel fuel at the SRS has been stored in underground tanks. Leaks, spills, and transfer errors resulting in the release of gasoline, diesel, or fuel oil to surface water is discussed in the section on surface water discharges. Emissions of gasoline from storage tanks can be made using AP-42 values and methods developed by the American Petroleum Institute, involving conservative assumptions, estimates of tank throughput, and characteristics of the tanks. The vapor space is generally assumed to be 25% of the tank volume. Working losses are derived from data on fill rate. Benzene emissions from gasoline use and storage were estimated and are described in the previous section on benzene.

Diesel-powered Generators

Generators have been and are still some of the largest contributors of air pollutants from the SRS. Diesel exhaust is a pollutant that exists everywhere. It is a highly complex mixture of gases, vapors, and particulates that are mutagenic to bacteria and have irritant effects. Diesel exhaust would be important to consider for emissions of benzene and sulfur dioxides (Mauderly 1995). Diesel exhaust has been included in the AIRS database emission estimates. Many SRS facilities have diesel engines and generators for use in the field or during a power failure. These are periodically run as a part of testing and maintenance. The highest release for cadmium in 1985, 1987, and 1990 was 1.35×10^{-4} actual ton per year from the H-Area diesel house diesel generator. Followed by 2.0×10^{-5} for each of eight diesel generators in K-Area engine house 108001 and 108002. Diesel exhaust also accounts for much of the oxides of nitrogen, sulfur dioxide, and other toxic metals released from the Site.

For example, the 200-H emergency power generator is a 600-kW generator used for emergency power. Testing is performed routinely on the generator. The engine is permitted for 2080 hours of operation each year. Actual emissions calculated were 2.82×10^{-6} ton y^{-1} for lead, 1.47×10^{-1} ton y^{-1} for oxides of nitrogen, 2.93×10^{-3} ton y^{-1} for oxides of sulfur, 6.46×10^{-5} ton y^{-1} for benzene, 5.43×10^{-7} ton y^{-1} for arsenic, 6.70×10^{-7} ton y^{-1} for mercury, 1.05×10^{-6} ton y^{-1} for manganese, and 1.32×10^{-6} ton y^{-1} for nickel (Westinghouse 1996a). Emissions from most of the diesel engines at the SRS are included in the AIRS database estimates for each chemical. Uncertainty was not calculated for these emissions.

Hydrazine Mononitrate

Hydrazine compounds and ferrous sulfamate were used in the separations process for reducing plutonium to Pu (III). Hydrazine mononitrate was also used in the frames process that produced ^{238}Pu (Pickett 1996). Most of the concern about hydrazine and formation of azides and hydrazoic acids were about explosion hazard, rather than health hazard (Thompson and Hale 1972).

In 1975, the Energy Research and Development Administration (now the U.S. Department of Energy) formed a Toxic Materials Advisory Committee to advise them on the use of toxic materials in the weapons complex. The SRS requested that the committee investigate the current status of hydrazine, which had just been classified by OSHA as a carcinogen. It was noted in the Industrial Hygiene Summary report for 1975 that this request had been made and that hydrazine mononitrate was used extensively in the SRS separations areas (Harper and Croley 1976).

Hydrazine was handled as the 30% mononitrate solution in 200-F and 200-H Areas and as hydrazine hydrate (85%) solution and hydrazine sulfate (100%) powder in Building 773-A. Industrial hygiene surveys indicated the highest air concentrations that might affect workers were in the drum handling area in Building 223-F. Air samples were taken by a gas scrubber technique using a Drager multigas detector with hydrazine detector tubes. A maximum concentration of 0.5 ppm was detected in the hydrazine drum handling area in Building 223-F. The hydrazine stored at 221-H was supplied from the 223-F handling area (Harper and Croley 1976).

A computerized essential materials ledger for the Separations Department from April 1982 listed a beginning inventory for hydrazine mononitrate of 25,650 lb and an ending inventory of 24,400 lb, so presumably 1250 lb had been used that month (Du Pont 1982a). Hydrazine was not noted on other ledgers found in central records.

A 1984 document characterizing wastes, lists hydrazine as a part of the waste from the FB Line, HB Line, and the canyons, all of which went to the waste tanks (Smithwick 1984). Hydrazine was also listed as a component of the Savannah River Laboratory (SRL) liquid wastes. A memo on the SRL seepage basins said that in over 28 years of use, less than 3 lb of hydrazine had been discharged to the SRL seepage basins (Lower 1983).

A memo written in 1985 on workplace monitoring, reports that hydrazine mononitrate was no longer used in the canyon process after 1985 (Karoly 1985). A search of the AIRS database for both hydrazine and hydrazine mononitrate found no emissions estimates for H-Area or F-Area from 1985 or after. Five emissions were reported in 1985 for A-Area (700-Area) totaling less than 2×10^{-5} ton y^{-1} maximum design emissions and less than 9.0×10^{-6} ton y^{-1} actual emissions, or about 8.18×10^{-3} kg y^{-1} (Faugl 1996a). However, a set of handwritten essential materials control sheets, labeled “total for the 200-Area, 222-F, 221-H, and B-Line” (in H-Area) list hydrazine mononitrate (EM Code 107), with the monthly consumption amounts shown in Table 17-30 (Westinghouse 1987b). The amounts reported were probably in pounds. The amounts were extremely variable and represent bookkeeping rather than actual use amounts, as evident by the large negative inventories at the end of several 6-month periods. Based on these sheets, the average monthly consumption during the time period was about 1816 lb mo^{-1} (Westinghouse 1987b).

Table 17-30. Monthly Consumption of Hydrazine Mononitrate in H-Area (1986 and 1987)[a]

Month and year	Total (lb)	222-F (lb)	221-H (lb)	B-Line (lb)
Jan 1986	0	0	0	
Feb 1986	0	0	0	
March 1986	2700	0	2700	
April 1986	1800	0	1800	
May 1986	2700	0	2250	
June 1986	6032	3150	3150	
July 1986	318	0	250	68
Aug 1986	950	0	950	0
Sept 1986	2400	0	2400	0
Oct 1986	197	0	0	197
Nov 1986	3613	0	3600	13
Dec 1986	–2560	0	–2700	140
Jan 1987	1820		1800	20
Feb 1987	1350		1350	0
March 1987	2475		2475	
April 1987	2475		2475	0
May 1987	900		900	0
June 1987	5530		5250	280
July 1987		0	–1200	
Aug 1987		0	1800	
Sept 1987		0	0	
Oct 1987		0	0	
Nov 1987		0	450	
Dec 1987		0	1350	

[a] Source: Westinghouse (1987b).

A table of bulk chemical consumption in H-Area that was part of a file on waste reduction studies kept by a chemical engineer in H-Area, listed the number of pounds of hydrazine mononitrate consumed per month from July 1987 through September 1988 (Pickett 1997). The values are presented in Table 17-31.

The total is given as 4200 lb or 1909 kg, which averages about 280 lb or 127.2 kg mo^{-1} or 1526 kg y^{-1} (Pickett 1997). These same notes list components of the high activity and low activity waste and contained no information to suggest that the hydrazine was not completely reacted in the process.

The operating permit application described the frame waste recovery system in H-Area, which purified and concentrated ^{238}Pu solutions and used hydrazine. A solution of 30% hydrazine mononitrate ($N_2H_5NO_3$) was added to the solution to stabilize plutonium at the lower valence state. It acted as a reductant and also helped scavenge ions that affect column adsorption. The permit application said that the frames waste recovery received plutonium and neptunium solutions from the HB-Line. The solutions were treated with aluminum nitrate to complex any fluorides, nitric acid to form nitrates, then hydrazine mononitrate and ferrous sulfamate to reduce the plutonium or neptunium in preparation for heat kill and anion exchange steps. Heat kill involved heating the solutions to enhance the reduction to a valence state for anion exchange. The

anion exchange columns absorbed the plutonium and neptunium ions, which were removed from the column with weak nitric acid and transferred back to the HB-Line. The permit application suggests that the reaction of hydrazine mononitrate and nitric acid produced nitrogen, nitrous oxide, and water.

Table 17-31. Estimates of the Monthly Consumption of Hydrazine Mononitrate in a Waste Reduction Study[a]

Month and year	Pounds consumed per month in H-Area
July 1987	–1200
Aug 1987	1800
Sept 1987	0
Oct 1987	0
Nov 1987	450
Dec 1987	1350
Jan 1988	900
Feb 1988	900
March 1988	9000
April 1988	–300
May 1988	150
June 1988	–8850
July 1988	0
Aug 1988	0
Sept 1988	0
Total	4200

[a] Source: Pickett (1997).

Clearly, hydrazine used in the canyon solvent extraction, frames, and B-Line processes would have reacted to form gaseous products, primarily oxide of nitrogen. All of hydrazine was probably converted to nitrogen compounds and water. What was left may have remained in liquid waste; however, canyon wastes were neutralized with caustic, and the sodium nitrate should have reacted with any excess hydrazine. We have found no evidence of any spills or releases of hydrazine to the soil, seepage basin, or air. Although we can reconstruct the amount of hydrazine that may have been used in the process from the 1982 inventory, we do not know and lack the information necessary to estimate how much of what was used may have been released to the environment. Current and former employees with knowledge of separations area processes were asked about the use and release of hydrazine and they thought that any hydrazine used would have been consumed by the process.

Hydrogen Sulfide

Hydrogen sulfide is a very toxic gas. Exposure to hydrogen sulfide is not associated with long-term, chronic effects like cancer. The workplace standard (OSHA Permissible Exposure Limit) is 10 ppm. A concentration of 500 ppm in air is lethal to most people.

Hydrogen sulfide was used in large quantities to make heavy water in D-Area from 1952 to 1982. The heavy water production process, called the GS process, involved continuous countercurrent contacting of water with hydrogen sulfide, through first a cold tower then a hot

tower (much like distillation towers). This operation used large quantities of pressurized hydrogen sulfide gas. Leaks were a constant concern for worker safety. Alarmed monitoring instruments were placed in the operating areas, and personnel carried hydrogen sulfide-sensitive paper. There were a number of releases of hydrogen sulfide, none of which was reported to cause injuries to employees (Bebbington 1990).

In 1954, Clark discussed the impact of a hypothetical hydrogen sulfide or firestorm situation from burning large amount of hydrogen sulfide (Clark 1954). SRS researchers thought that an accident in the 400-Area would have most likely affected the Georgia Bank of the Savannah River at the point closest to the 400-Area. Hypothetical problems such as a fire, large leak of gas that did not burn, an attack on the facility, a hurricane or tornado, and an earthquake were discussed. This memo also described the safety measures taken in construction of the towers and operation and maintenance of the plant (Clark 1954). In a letter written to the Atomic Energy Commission in 1957, Site personnel stated that the probabilities of personnel offsite receiving a serious overdose of hydrogen sulfide were calculated to be about 10^{-6}. At that time, the Site had operated safely for the previous 3 years and they felt there was no need for an offsite warning system (Cole 1957).

Routine purges of hydrogen sulfide from the heavy water facility and minor leaks were routed to a 400-ft flare tower, where the carried hydrogen sulfide was burned (forming sulfur dioxide) (Rusche 1973). Reinig et al. (1973) described a 320-ft flare tower for controlled process venting of carried hydrogen sulfide from the extraction plant. The amount of sulfur dioxide released was said to be very small compared to the amount emitted from the powerhouse operation in 484-D (Reinig et al. 1973).

Reinig et al.. (1973) estimated that 42 ton of hydrogen sulfide per year were released from the GS process via the high and low pressure lines of the 320-ft flare tower. Some of this was burned to sulfur dioxide. The flow and hydrogen sulfide concentration in the stream vented to the flare tower was being measured on a grab-sample basis in 1973. Reinig et al. (1973) estimated that a total of about 150 ton y^{-1} was released from D-Area processes, including releases to the flare tower. Reinig et al.. (1973) recommended ambient air monitoring be conducted offsite in Georgia for sulfur dioxide and hydrogen sulfide. As far as we can determine, monitoring for hydrogen sulfide was not routinely performed until a special program to monitor for hydrogen sulfide and sulfur dioxide using monitoring equipment in mobile trailers was started in 1977. Hydrogen sulfide and sulfur dioxide were detected in ambient air most frequently at stations near D-Area. The maximum hydrogen sulfide level detected was 0.10 ppm at the D-Area 3G pumphouse. The average hydrogen sulfide concentration at all sites was about 0.004 ppm. Georgia and South Carolina had no standards for hydrogen sulfide at that time (Du Pont 1979). The worker health and safety standard, National Institute of Occupational Safety and Health's ceiling limit, is 10 ppm or 15 mg m^{-3} (NIOSH 1994).

Several accidental releases of hydrogen sulfide have been reported in Site reports and local newspapers. On March 19, 1960, a hydrogen sulfide gas release and fire occurred in GS Unit 22. The gas was released from a pipe connection at the bottom of a condenser, which pulled apart because of insufficient thread engagement. The pipe joint, 16 in. in size, opened several inches allowing about 40 ton of hydrogen sulfide to be rapidly released. The gas immediately caught fire. The area was evacuated. According to the incident report, the fire burned for 46 minutes and the force of the pressurized gas release was felt throughout the 400-Area. Five "very minor

injuries" were sustained by maintenance personnel in the area. After this incident, all of the 16-in. screwed flanged connections were replaced with welded connections (Ellett 1960).

Another leak occurred over May 19–30, 1980. This incident involved a tank at Building 402-D. Hydrogen sulfide leaked from a crack in a weld of the bottom flange of storage tank 22. Another tank, 8B, was empty and undergoing a bolt inspection. This tank was filled with nitrogen in preparation for hydrogen sulfide transfer from the leaking tank 22 to tank 8B. The first transfer attempt May 20 failed because a hydrate plug had formed in the leaking line. As the crack length and leak rate increased, the 400 and CNX and TNX Areas were evacuated. Rather than transfer the hydrogen sulfide as a liquid, the hydrogen sulfide was transferred as a vapor to a shutdown extraction unit and then into Tank 8B. It took until May 29 to vaporize and transfer all of the hydrogen sulfide. About 900 lb of gas remained in the tank. This was flared and the tank was purged by steam. The hydrogen sulfide release was said to have been minimized by a hydrate that formed and sealed the crack from the inside. Ice that formed on the outside of the line may also have helped contain the leak. Nonessential personnel returned to work in the 400-Area and the roads were reopened on May 30. Smudge pots (flambeaus) were lighted and placed on the ground north and south of the tank just outside the dike to ignite any large release of hydrogen sulfide that might have occurred. The concentration of hydrogen sulfide 0.5 m from the leak was less than 1 ppm between May 21 and May 29, with the exception of a 100-ppm level on May 21 when the leak first started and several levels of 2.0 and one of 10 ppm when the last of the hydrogen sulfide was purged from the tank (Haywood 1980). A Public Affairs report and numerous newspaper articles on this incident were also found in the Phase I document search. These had been photocopied and were added to the 1980 investigation and incident reports (referenced here as Haywood 1980). The evacuation and stopping of road, river, and rail traffic were the major points of most of the articles. The leak was said to be small enough that it posed no threat to the offsite public. The total amount of hydrogen sulfide estimated to have been released as a result of the leak was 0.5 ton (Haywood 1980).

The incident report mentions that previous dispersion studies had predicted that hydrogen sulfide concentrations of 500 ppm or greater could exist several miles away from the 400-Area if 1 ton of hydrogen sulfide was released and the wind velocity was low (Haywood 1980).
Table 17-32 compiles hydrogen sulfide release estimates from different sources.

Table 17-32. Hydrogen Sulfide Release Estimates

Year	Amount (ton y^{-1})	Basis of the estimate	Reference
1960	40	Incident report	Ellett (1960)
1973	150	General process losses and flare tower	Reinig et al. (1973)
1980	0.5	Incident report	Haywood (1980)

Although the heavy water production plant was shutdown permanently in 1981 (DOE 1987), D-Area continued to do heavy water reprocessing, called heavy water rework in many documents (Arnett et al. 1993). Long before the first air emissions inventory was conducted, the GS process was no longer in operation. Recent release estimates from the AIRS database will not be useful for estimating releases in the early years when large amounts of hydrogen sulfide were being used to make heavy water.

In recent years, the largest source estimated in the AIRS database for hydrogen sulfide was the degassifiers at the water treatment plant, used to remove sulfur and other compounds from the water. Emissions estimates for these facilities in 1985 are shown in Table 17-33. Routine releases of hydrogen sulfide from groundwater treatment were included in the AIRS database. For example, emissions estimates for the 183-K Buildings that make up the groundwater treatment plant were described in the K-Area calculations. Groundwater was treated for steam generation, reactor cooling, and domestic use, which involved degassification, clarification, pH adjustment, and treatment with biocides (primarily sodium hypochlorite). The water contained sulfides at a concentration of about 0.9 mg L^{-1}, which were removed by the degasifiers that were designed to remove carbon dioxide. Hydrogen sulfide emissions from the degasifiers were calculated from the flow of groundwater and concentration of sulfides per gallon, assuming all of the sulfides were liberated as hydrogen sulfide. The emission rate calculated for 183-K was 0.09 lb h^{-1} hydrogen sulfide, which equals a maximum emission of 0.39 ton y^{-1} or 0.197 ton y^{-1} for each unit (Radian 1992b). The actual emissions estimate for all nine units in 1985 totaled 3.05 ton y^{-1}.

Table 17-33. Emissions Estimates for Nine Water Degasifiers Operating in 1985

Degasifier	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
Each of two 183-C filter and softener plant degasifier	8.20×10^{-2}	5.60×10^{-2}
282-H Powerhouse degasifier	2.60	2.60
Each of two 183-K filter and softener plant degasifier	1.97×10^{-1}	1.97×10^{-1}
Each of two 183-L filter and softener plant degasifier	1.25×10^{-1}	8.50×10^{-2}
Each of two 183-P filter and softener plant degasifier	1.60×10^{-1}	1.10×10^{-1}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

The Standard 8 results submitted to SCDHEC said that in 1991, hydrogen sulfide was released from 30 sources in A-Area, B-Area, C-Area, F-Area, H-Area, K-Area, L-Area, P-Area, and S-Area. The maximum 24-hour average Site boundary concentration was calculated to be 0.201 μg m^{-3}. The ambient air standard was 140 μg m^{-3} (Dukes 1993). In summary, Reinig et al. (1973) estimated that 150 ton y^{-1} may have been routinely released from D-Area from 1952–1973. Two accidental releases released an additional 40 and 0.5 ton, in 1960 and 1980, respectively, most of which burned (Ellett 1960; Haywood 1980). From 1952–1982, a total of 4690 ton of hydrogen sulfide may have been released based on the assumption that 150 ton were released each year from 1952–1982. After 1982, emissions were probably less than 3.5 ton y^{-1}, primarily from the aeration of groundwater. Although the uncertainty in these release estimates is great, there was not enough information to quantify the uncertainty associated with these release estimates.

Lead

Most of the lead used onsite was probably used as shielding material. Leaded oil was used as a lubricant for the uranium extrusion press and lead was emitted into the air from this process. A three to one mixture of oil and lead powder was used as a lubricant for the extrusion process in 321-M. About 9 qt of leaded oil was used each day. About 6 ton of lead was used in the year 1985, 4.5 ton in 1986, and approximately 1.7 ton in 1987 (Westinghouse 1987a). Lead fume emissions from these processes have not been well characterized, and the potential for their emissions has not been assessed (Faugl 1996a). Industrial hygiene records indicate that worker exposures to metal fumes from the 321-M extrusion process were minimal and the process was not an occupational hazard. The air around the extrusion press was vented outside. Lead concentrations in workroom air were less than 5.3 $\mu g\ m^{-3}$ in 1985 (DOE 1987).

In 1967, an air sampling program for lead was initiated in M-Area. Measurements of lead and carbon monoxide were taken twice daily for 18 days. The average concentration was reported to be 0.16 $mg\ m^{-3}$ for lead with a footnote that said, "only two positive samples, indicating a very clean atmosphere." The report section about monitoring was titled "air purity," but the text did not state whether the sampling was done out of concern for industrial hygiene or environmental releases. The report did not indicate where the samples were taken and it was not clear whether the samples were from the stack or ambient air (Du Pont 1967). A 1987 report describing ambient air quality monitoring onsite and offsite said that lead was not monitored because the potential release was insignificant compared to the standard (Mikol et al. 1988).

There is little information on lead released to the air before 1985. The emissions estimates in the AIRS database included lead released from M-Area and smelting, lead pots, and small welding and cutting operations at various locations onsite (Faugl 1996g). Welding and painting operations were estimated to release less than 10^{-3} ton y^{-1} total. About four times as many emissions points are listed for 1995 than for 1985, 1987, and 1990, and almost all of these were welding machines, welding booths, and welders. Table 17-34 presents the lead releases in the AIRS Database and Table 17-35 lists lead emissions by source.

The largest emission estimated in 1990 was lead oxide from the tank farm, at 2.40×10^{-3} ton y^{-1} actual estimated emissions. These emissions were probably from maintenance work that involved the removal or application of lead paint.

Lead has also been released from the combustion of fuels containing lead. The emissions estimate for the large 200-H emergency power generator, actual emissions assuming 2080 hours of operation each year was 2.82×10^{-6} ton y^{-1} for lead (Westinghouse 1996a).

The air quality permit application estimated that the two 784-A boilers released a maximum total of 3.87×10^{-3} ton y^{-1} of lead, based on each boiler burning 8500 ton of coal or 17,000 ton y^{-1} for the two boilers (Westinghouse 1996a). If we assume that coal with a similar lead content was burned Site-wide, as much as 0.114 ton y^{-1} of lead could have been released during the years when about 500,000 ton y^{-1} of coal was being burned. The coal crushing operation at 784-A was also evaluated for the air quality permit. The maximum emissions for several different pieces of coal-handling equipment and the coal pile, assuming operating at capacity of 66 ton h^{-1} totaled 1.05×10^{-3} ton y^{-1} for lead, based on the amount of coal burned (Westinghouse 1996a). Actual emissions estimates for metals from the fuel oil-fired package boilers in K-Area included lead with 1.65×10^{-4} ton y^{-1} (Westinghouse 1996a).

Table 17-34. Key Lead Releases in the AIRS Database

Year and source description	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
1985 total	9.97×10^{-3}	4.68×10^{-3}
F-Area tank farm, lead oxide gas	2.40×10^{-3}	2.40×10^{-3}
H-Area coal storage pile	1.50×10^{-3}	8.70×10^{-5}
H-Area Building 284 powerhouse ash sluice sump	1.67×10^{-4}	3.65×10^{-5}
H-Area Manufacturing Building Number 3, 234-H Fabricated metals machining	5.20×10^{-3}	1.74×10^{-3}
K-Area coal pile	4.70×10^{-5}	4.70×10^{-5}
K-Area coal pile runoff basin	4.70×10^{-5}	4.70×10^{-5}
N-Area Lead Pouring Building 711-04 lead melting pot	0	5.36×10^{-8}
1987 total	9.97×10^{-3}	4.68×10^{-3}
F-Area tank farm, lead oxide gas	2.40×10^{-3}	2.40×10^{-3}
H-Area coal storage pile	1.50×10^{-3}	8.70×10^{-5}
H-Area Building 284 powerhouse ash sluice sump	1.67×10^{-4}	3.65×10^{-5}
H-Area Manufacturing Building Number 3, 234-H Fabricated metals machining	5.20×10^{-3}	1.74×10^{-3}
K-Area coal pile	4.70×10^{-5}	4.70×10^{-5}
K-Area coal pile runoff basin	4.70×10^{-5}	4.70×10^{-5}
1990 total	9.97×10^{-3}	4.68×10^{-3}
1990 values are the same as 1987 with the addition of		
H-Area Building 284 powerhouse ash sluice sump	1.67×10^{-4}	3.83×10^{-5}
N-Area Lead Pouring Building 711-04 lead melting pot	0	2.68×10^{-8}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

The amendment to the air quality permit application included handwritten calculation sheets for lead emissions for the lead melter in Building 711-4N. All particulate from the pot was assumed to be lead and the emission included melting and pouring. Estimates were based on the amount of lead melted and an emission factor. The lead melter held about 16,000 lb of lead in two pots (Westinghouse 1996a). Lead has been melted and molded as needed. Apparently, no operating records were kept before July 1991. Records for 1994 were compiled by International Technology Corporation for the operating permit application; they suggested that monthly quantities of lead melted ranged from 0 to 15135 lb. The total amount melted in 1994 was estimated to have been 45,005 lb. The operating procedures indicate that lead was heated for about 1 hour until it was 700°F, then it was poured through a pipe into a mold. Exhaust was pulled from the enclosure for each pot, combined, ducted through a high-efficiency particulate air (HEPA) filter and exhausted out the stack. The maximum emissions estimate for the N-Area melting pot was 3.5×10^{-5} ton y^{-1} (Westinghouse 1997c). This emissions estimate is about 600 times higher than the emissions estimate reported in the AIRS database (Faugl 1996g). Further information on the lead melter has not been found.

The Standard 8 results submitted to SCDHEC said that in 1991, lead was emitted from 26 sources in A-Area, D-Area, H-Area, K-Area, N-Area, and S-Area. The estimated maximum quarterly Site boundary concentration was calculated to be 0.0015 μg m^{-3}. The ambient air standard was 1.50 μg m^{-3} (Dukes 1993).

Table 17-35. Sources of Lead Emission Estimates for Selected Years[a]

Emission source	Year	Actual[b] emission estimate (ton y^{-1})
H-Area coal storage pile	1985	8.70×10^{-5}
	1987	8.70×10^{-5}
	1995	1.77×10^{-5}
H-Area powerhouse	1985	3.65×10^{-5}
	1987	3.65×10^{-5}
H-Area manufacturing building	1985	1.74×10^{-3}
	1987	1.74×10^{-3}
	1995	1.74×10^{-4}
K-Area coal pile	1985	4.70×10^{-5}
	1987	4.70×10^{-5}
K-Area coal pile storage runoff	1985	4.70×10^{-5}
	1987	4.70×10^{-5}
F-Area tank farm	1985	2.40×10^{-3}
	1987	2.40×10^{-3}

[a] Source: Faugl (1996g).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

From 1987 to 1995, the TRI, reported to the EPA and SCDHEC, included estimates of the pounds per year of lead released from the SRS, shown in Table 17-36. We do not know the reason for differences between years or why the release reported in 1995 was so low. The emissions from coal consumption do not appear to be included in the TRI estimates.

Table 17-36. Toxic Release Inventory Release Estimates for Lead (per year)[a]

Year	Air emissions (lb y^{-1})
1989	120
1990	12
1992	29
1993	76
1994	8
1995	1

[a] Source: Westinghouse (1996b).

It is likely that in years with no amount reported, some amount below the reporting limit was released (Westinghouse 1996b).

In summary, we did not obtain sufficient information to determine lead releases from the M-Area extrusion process or gasoline combustion. The annual total in the AIRS database for 1985, 1987 and 1990 was 4.68×10^{-3} ton y^{-1} and accounted for welders, lead melting pots and metal machining (Faugl 1996g). Lead emissions from coal burning may have ranged from 0.045 to 0.114 ton y^{-1} depending on the year. The lead melting pot emission estimate used for the operating permit application was 3.5×10^{-5} ton y^{-1}. These emissions total about 0.05–0.119 ton y^{-1} or 238 lb y^{-1} compared to 120 lb reported to the EPA for the 1989 TRI.

An attempt was made to calculate uncertainty for the lead releases, based on the information available in the AIRS database, the TRI release estimates, and the permit application estimates. Very little information was provided about the basis of release estimates, making it quite difficult to quantify the uncertainty. Although we have had much success providing uncertainty estimates for releases from coal burning, only one estimate of lead released per ton of coal burned was found, and the uncertainty related to that estimate was unknown. The distribution of uncertainty in the release estimates produced from using only the ranges of estimates was quite narrow and misleading in terms of the total uncertainty in these releases. The lack of information precluded any calculations of uncertainty and the range of releases presented is provided as the best available information on lead emissions.

Manganese Compounds

Manganese dioxide was used in the separations area head end processes. Most of the manganese was discharged to the liquid waste tanks. The separations area processes did not release notable amounts of manganese to the air. Potassium permanganate solutions were used in D-Area and in small amounts in many laboratories. Releases to surface water are described in Chapter 18. Manganese was measured in ash basin effluents, but manganese releases to the air do not appear to have been monitored.

Manganese emissions estimates were entered into the AIRS database for a number of grinding and cutting operations in A-Area, F-Area, H-Area, N-Area, S-Area, and T-Area; H-Area and K-Area coal storage and handling; water filtration and treatment; F-Canyon and H-Canyon stacks; and combustion sources. Table 17-37 lists the total emissions for several years. Some of the highest emissions estimates were for machining in H-Area manufacturing (Faugl 1996f).

Table 17-37. Key Sources of Manganese Emissions Estimates in the AIRS Database [a]

Emission source	Year	Actual[b] emission estimate (ton y^{-1})
A-Area boiler house	1995	1.22×10^{-4}
A-Area ash pile	1995	5.21×10^{-4}
H-Area coal storage pile	1985	5.60×10^{-5}
	1987	5.60×10^{-5}
	1995	5.72×10^{-5}
H-Area powerhouse	1985	4.63×10^{-5}
	1987	4.63×10^{-5}
H-Area manufacturing building	1985	1.73×10^{-4}
	1987	4.95×10^{-4}
K-Area coal pile	1985	3.00×10^{-5}
	1987	3.00×10^{-5}
K-Area coal storage runoff area	1985	3.00×10^{-5}
	1987	3.00×10^{-5}

[a] Source: Faugl (1996f).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Most of the emissions points in the AIRS database for manganese were for grinding, cutting, torch cutting, belt sanders, metal machining, turning, and drilling, and other similar maintenance facility and machine shop operations. Estimates for the coal operations in D-Area and A-Area were also listed and are shown in Table 17-38.

Table 17-38. Manganese Releases to the Air in the AIRS database for 1985, 1987 and 1990[a]

Year and source description	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
1985		
H-Area manufacturing Building 232		
232-H hood metallography		0
234-H hood cutting		0
234-H radiological equipment repair	1.05×10^{-2}	1.73×10^{-4}
234-H finishing operations	1.43×10^{-5}	1.43×10^{-5}
234-H inert finishing operations	1.73×10^{-6}	1.73×10^{-6}
234-H fabricated metals machining	2.62×10^{-2}	8.83×10^{-3}
238-H milling and machining hood	2.35×10^{-3}	1.70×10^{-5}
238-H lathe hood	2.35×10^{-3}	2.59×10^{-5}
K-Area coal pile	3.00×10^{-5}	3.00×10^{-5}
K-Area coal pile runoff basin	3.00×10^{-5}	3.00×10^{-5}
1987 and 1990		
H-Area coal storage pile	9.50×10^{-4}	5.60×10^{-5}
H ash sluice sump	2.12×10^{-4}	4.63×10^{-5}
H-Area manufacturing Building 232		
232-H hood metallography	6.0×10^{-4}	1.44×10^{-4}
234-H hood cutting	3.80×10^{-3}	1.78×10^{-4}
234-H radiological equipment repair	1.05×10^{-2}	1.73×10^{-4}
234-H finishing operations	1.56×10^{-5}	1.56×10^{-5}
234-H inert finishing operations	3.19×10^{-6}	3.19×10^{-6}
234-H fabricated metals machining	2.64×10^{-2}	8.83×10^{-3}
238-H milling and machining hood	2.35×10^{-3}	1.64×10^{-5}
238-H lathe hood	2.35×10^{-3}	3.29×10^{-5}
Coal transfer operation	1.11×10^{-4}	3.30×10^{-6}
Coal storage pile	3.29×10^{-5}	1.23×10^{-5}
Coal crushing operation	3.24×10^{-3}	0
788-A ash pile	6.10×10^{-4}	5.21×10^{-4}
484-D coal storage pile-coal transfer	4.42×10^{-2}	3.29×10^{-3}
484-D coal storage pile	4.44×10^{-2}	3.48×10^{-3}
484-D coal storage pile coal reject pile	1.64×10^{-6}	5.79×10^{-7}
488-D ash basin - disposal basin	9.41×10^{-3}	8.02×10^{-3}
488-D ash basin - coal reject handling	1.63×10^{-4}	1.62×10^{-4}
488-D ash disposal basin	3.66×10^{-3}	0
H-Area coal transfer	9.27×10^{-5}	3.09×10^{-6}
K-Area coal pile	1.86×10^{-5}	1.86×10^{-5}

[a] Source: Faugl (1996f).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

The highest emissions estimates for all years were for the C-Area, Building 717 grinders and dust collectors used in the contaminated maintenance facility at 0.029 ton y^{-1}. Followed by 0.0088 ton y^{-1} for fabricated metals machining in 234-H (Faugl 1996f).

Table 17-39. AIRS Database Manganese Total Emissions[a]

Total	Maximum	Actual[b]
1985	6.05×10^{-2}	9.79×10^{-2}
1987	6.52×10^{-2}	6.95×10^{-2}
1990	6.56×10^{-2}	6.95×10^{-2}
1992	2.84×10^{-2}	5.61×10^{-3}

[a] Source: Faugl (1996f).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

There appears to be an error in the total maximum emissions estimates, which are less than the actual, because actual values were given, but the maximum values were zero, for several of the metal and pipe cutting and grinding operations. The manganese emissions estimate for the 200-H emergency power generator, actual emissions, was 1.05×10^{-6} ton y^{-1}, assuming 2080 hours of operation each year. Actual emissions estimates for metals from the fuel oil-fired package boilers in K-Area included manganese with 2.59×10^{-4} ton y^{-1}. The coal crushing operation at 784-A was also evaluated for the air quality permit. The maximum emissions for several different pieces of equipment and the coal pile, assuming operating at capacity of 66 ton h^{-1}, totaled 3.38×10^{-3} ton y^{-1} for manganese based on the amount of coal burned (Westinghouse 1996a). This suggests that 0.099 ton y^{-1} may have been released during the years when 500,000 ton y^{-1} of coal was burned.

At the time of the 1996 permit, the F-Canyon was not operating and actual emissions were reported as zero. The combined F-Canyon stack maximum controlled emission estimates assuming 24 h d^{-1} releases of manganese oxide totaled 2.38×10^{-6} ton y^{-1} based on engineering calculations (Westinghouse 1996a). We have no information about releases from the use of permanganate in laboratories and we have no evidence that any releases occurred. From 1987 to 1995, the TRI, reported to the EPA and SCDHEC, included estimates of the pounds per year of manganese compounds released from the SRS.

Table 17-40. Manganese Compound Release Estimates from the Toxic Release Inventory

Year	Air emissions (lb y^{-1})	Ton y^{-1}
1988	190	0.095
1989	190	0.095
1990	150	0.075
1993	42	0.021
1994	31	0.015

It is likely that in years with no amount reported, some amount below the reporting limit was released (Westinghouse 1996b).

Most of the routine releases of manganese came from metal cutting, grinding, machining, and finishing operations and coal burning operations. Releases were probably less than 0.099 ton y^{-1}, or less than 200 lb y^{-1}. They may have ranged from about 0.07 ton y^{-1} to 1.9 ton y^{-1}. Again, the combination of the AIRS database information, release estimates from coal burning and the TRI release estimates did not provide enough information to effectively quantify uncertainty for manganese releases. The estimates presented are our best estimate.

Mercury

Mercury is a naturally occurring metal released into the atmosphere from deposits in soil and rock and from combustion of coal. Mercury in the environment can exist in three valence states in inorganic and organic compounds and the elemental or liquid mercury form. Elemental mercury can volatilize from soil, water, and waste.

Inhalation, ingestion, or skin absorption of mercury can cause neurological damage. Inorganic mercury, such as mercury salts, also damages the nervous system. Chronic exposure to mercury salts and mercury vapor may cause kidney damage, and neurological effects. Epidemiological studies suggest that mercury is not a carcinogen. Mercury can cause birth defects. Organic mercury is readily transported across the placental barrier. Mercury can cause damage to the central nervous system, causing impaired learning and behavior disabilities.

Mercury vapor and mercury bound to particulates is readily transported through the air. In the environment, mercury can be transformed by microorganisms in sediments to organic methyl mercury, which bioaccumulates up the food chain. Methyl mercury in fish is often the environmental exposure pathway of greatest concern for exposures to the public. Methyl mercury causes nervous system toxicity. Unborn children and children under the age of 6 are most sensitive.

Atmospheric mercury pollution has emerged as one of the leading environmental issues of the 1990s. Mercury in the atmosphere occurs almost exclusively as Hg(0), elemental mercury, but it may be oxidized and deposited as mercury ions and salts. The Hg^{2+} and Ch_3Hg^{+} ions are complexed by hydroxide, chloride, sulfide, and humic acids in surface water and sediments. The cycling, transformation, and bioaccumulation of mercury are complex. Understanding some of the Site-specific environmental fate characteristics of mercury will be important if mercury is evaluated further in later phases of the dose reconstruction project.

This section summarizes what has been documented about the use and release of mercury at SRS facilities and addresses the releases to air from each facility or process.

Overview of Mercury Use and Release

All plant areas, except possibly the reactor areas, have released mercury to the ground or surface streams during use in the aluminum dissolution process, in laboratories as a chemical reagent, and as a necessary component of process equipment (Horton 1974a). Mercury has been excluded from all reactor areas to prevent reactions with aluminum in fuel and other components (Du Pont 1972a).

Mercury was used in the separations areas as a process catalyst for aluminum dissolution (as a catalyst to increase the rate at which aluminum dissolves in nitric acid) in H-Area and to much less of an extent in F-Area. It also served to remove chlorides that, if not removed, corrode stainless steel in tanks and processing equipment (Kvartek et al. 1994; Pickett et al. 1989). The addition of mercuric nitrate was also reported to decrease the releases of ^{131}I (Ice 1973; Dodgen and Sykes 1971; Du Pont 1967). The separations areas also used large quantities of sodium hydroxide (caustic), of which mercury was an impurity (Horton 1974b).

Mercury has been used in mechanical and diffusion pumps to handle tritium gas in the tritium facilities. Before 1968, old pumps and any mercury drained from the tritium facility pumps were buried in the burial grounds. Various Site researchers have estimated that the tritium facility disposed of approximately 10,000 kg of mercury in the burial grounds (Kvartek et al. 1994; Horton 1973; Westinghouse 1992). Mercury has also been used in many kinds of equipment in laboratories and other facilities onsite. The use and potential for mercury release from each of these facilities is discussed in more detail in the following sections.

Mercury discharged in liquid effluent went primarily to underground storage tanks and the seepage basins before the Effluent Treatment Facility (ETF) began operation in 1988. Mercury was also released to the air through the separation's process stacks. Mercury in separations effluent discharged to the seepage basins and contaminated groundwater underneath the basins, which outcrop to Four Mile Creek and flows to the Savannah River. Mercury releases to surface water are described in Chapter 18. The results and interpretation of monitoring of water, sediments, and fish for mercury is discussed in Chapter 20.

Mercury Use for Separations in the 200-H and 200-F Areas

Mercury was first used in H-Area for the HM process used to recover enriched uranium from spent aluminum-uranium reactor fuel in 221-H in 1959. About 7 kg of mercury was added to the large dissolver, and about 5 kg of mercury was added to the small dissolver in 221-H for each run (Kvartek et al. 1994). Mercury was first used to recover ^{238}Pu and ^{237}Np in the 221-H frames process in 1961. Mercury was initially used in 221-F to dissolve plutonium-aluminum targets and scrap in 1965. Mercury was also used during the recovery of plutonium from scrub-alloy received from Rocky Flats in 1984. About 105 kg of mercury was used to dissolve aluminum and precipitate chlorides in a typical charge.

Kvartek et al. (1994) summarized the concentrations of mercury charged to the H-Area dissolver for various campaigns from 1959–1985. These data were derived from a report titled, *Mercury Requirement for Separations Processes by* Pickett et al. (1989). Carlisle Pickett, the author of the report, and a chemical engineer in H-Area was interviewed by *RAC* researchers on two separate occasions. He provided us with copies of information from a waste reduction study, tables of flowsheets, chemical makeup tank data, and other estimates similar to those summarized in the 1989 memo. Mercury concentrations that were used ranged from 0.0025 to 0.04 moles/liter (M). The highest concentrations were used before 1962 to dissolve plutonium-aluminum targets and Mark VI-J fuel. In March 1985, the mercury concentration in the dissolvers was reduced to 0.002 M to minimize the amount of mixed hazardous waste generated (Kvartek et al. 1994), but no suitable replacement for mercury in the process has been found.

Kvartek et al. (1994) also published a table of estimates of the amounts of mercury used in the chemical separations processes from 1974–1980 based on information in essential materials

transaction reports. A total of 2500 kg was used in F-Area in 1985, 1986, 1987, 1988, 1989, and 1992. H-Area use is summarized in Table 17-41. No mercury was reported to have been used in H-Area in 1978, 1980, 1981, 1991, and 1992. Inventory adjustments resulted in a net gain in the inventory of mercury (negative amounts used) in 1990 and 1993.

Table 17-41. Mercury Use (kg) in the F-Area and H-Area Canyons by Year (Kvartek et al. 1994)

Year	221-H	221-F	Total	1% (emissions/ consumption)
1974	3600	No data available	3600	36
1975	4300	No data available	4300	43
1976	3300	No data available	3300	33
1977[a]	2700	No data available	2700	27
1978	No data available	No data available	-	
1979[a]	240	No data available	240	2.4
1980	No data available	No data available	-	
1981	No data available	No data available	-	
1982	2600	No data available	2600	26
1983	2900	No data available	2900	29
1984	4800	No data available	4800	48
1985	4800	620	5420	54.2
1986	1500	280	1780	17.8
1987	1100	300	1400	14
1988	0	680	680	6.8
1989	0	560	560	5.6

[a] partial year inventories

An average of 2449 was used in H-Canyon during the 13 years for which data was reported, and an average of 488 kg was used in F-Canyon after 1985 when mercury was used in the dissolution and head end processes. Less mercury would have been used from 1960 to 1964.

Mercury was used in the H-Canyon process as mercuric nitrate catalyst in the dissolution process. The catalyst destroyed the passive aluminum oxide surface that forms on fuel and can hinder dissolution. Some of the mercury added to the dissolvers as a catalyst was released as a vapor through the 292-H and 292-F stacks. A 1985 description of the dissolving process said that mercury was boiled for 1 hour in nitric acid in a tank to make up the catalyst. The fuel and nitric acid were preheated in the dissolvers, then the mercuric nitrate was added. It immediately reacted and mercury was vaporized. The solution in the dissolvers was then heated for about 22 hours after the catalyst was added. After dissolution was complete, the solution went to the head end system in preparation for the first cycle of solvent extraction. The 11.3E evaporator was part of the head end system. The aluminum, fission products, mercury, and other waste materials remained in the aqueous waste stream, the volume of which was decreased using the 9.1E and 9.2E evaporators. Most of the mercury in the process liquid waste was eventually sent to the high activity waste tanks. Mercury in evaporator overheads was sent to the seepage basins (Franklin 1985).

Information on the amount of mercury used, discharged to the seepage basins, or routed to the waste tanks does not seem to be adequate for using a materials balance approach to estimate releases.

Mercury vapors were released from the dissolvers, process waste evaporators, and the catalyst makeup tank. Most of the mercury emissions exhausted through the 292-H or 292-F stack. The 1987 DOE Environmental Survey states that mercury was released to the air from the H-Area separations and tritium facilities 292-H main stack, 232-H stacks 1 and 2, 234-H stack, 242-H evaporator stack, 241-H as well as the 241-F stacks and 242-F stack. The survey team thought there was potential for stack emissions of mercury to be inhaled by the surrounding population (DOE 1987).

H-Area Mercury Monitoring Program

A characterization program for the H-Canyon stack emissions was initiated in 1984 to identify mercury sources and to determine the amount of mercury being released from the stack.

A monitoring program, described in Franklin (1985), was established to measure emissions to the 292-H stack and to correlate the emissions to 221-H canyon operations. Six potential sources of mercury emissions from the HM process were identified: the catalyst makeup tank; the 6.1D and 6.4D dissolvers; and the 9.1E, 9.2E, and 11.3E evaporators. All of the six pieces of processing equipment that could potentially emit mercury vapors were exhausted by the vessel vent system. The air in the vessel vent system was passed through a reactor to remove iodine and a glass wool filter to remove particulates before passing through the sand filter and exhausting out the 292-H stack (Franklin 1985).

Mercury released through the stack during routine operations of the 6.1D and 6.4D dissolvers and the 9.1E, 9.2E, and 11.3E evaporators was monitored using a model 411 gold film mercury vapor analyzer manufactured by the Jerome Instrument Corporation modified for continuous monitoring. The mercury vapor monitor was installed in the 292-H stack sampling room, and the sample point was located at the 50-ft elevation point. Samples were taken at 15-min intervals at various times from January–April 1985. The milligram per cubic meter amounts were converted to pounds per hour based on the average flow rate of 217,000 ft^3 min^{-1} through the stack. Hourly averages were calculated and graphed. The data were presented as a series of graphs of daily data. The spikes and dips were not labeled and we cannot correlate concentrations with process activities. Maximum and minimum values for the entire study are difficult to determine. The report of the study by Franklin (1985) said that a good correlation between mercuric nitrate catalyst addition to the 6.4D dissolver and mercury emission levels was observed. He said there was generally a significant spike visible on the graph at the time of the catalyst addition, while startup of evaporators and makeup tank did not result in a spike (Franklin 1985). The 6.1D dissolvers operated on average only four times per month, so they contributed much less to the total release. The catalyst makeup tank and evaporators had little effect on the total release. Franklin (1985) estimated a yearly emission of 64.5 lb (29.3 kg) based on 15 dissolutions per month for the first quarter of 1985. This emission rate was calculated from data obtained during a period of relatively high production. The maximum theoretical capacity of the 221-H canyon during that time was 24 dissolutions per month, which would have resulted in a mercury emission rate of 104 lb y^{-1} (47.3 kg y^{-1}) (Franklin 1985). Franklin (1985) noted that previous data reported had indicated a release of about 38 lb y^{-1}, but no reference for this value was given.

The monitoring study was used to demonstrate compliance with air pollution regulations. The South Carolina regulatory limit for mercury emissions was said to have been 0.1 ton y^{-1} or 200 lb y^{-1} in 1985 (Franklin 1985).

The H-Canyon started a program to reduce mercury in March 1985. The amount of mercuric nitrate added to the dissolvers was decreased by 60% (Franklin 1985).

The mercury monitor has been out of service since the 1985 study (DOE 1987). Mercury consumption in H-Area was about 5000 lb during the first quarter of 1985. The measured emissions to the air were less than 50 lb during this time (DOE 1987). These measurements suggest that nearly all of the mercury in the process was incorporated into liquid waste and about 1% (50/5000) was released into the air. If we apply this factor to the mercury reported to have been used in the canyons (Kvartek et al. 1994), then we can predict the kilograms per year emitted shown in Table 17-41. Assuming 2.5 kg y^{-1} was released in 1978, 1979, 1980, and 1981; 1 kg y^{-1} was released from 1954—1958; and 36 kg was released each year from 1954–1973, a total of 895 kg (1% of the total amount used) is the predicted release. This averages to about 29 kg y^{-1} over 31 years. The uncertainty in such an estimate is very large. The uncertainty in applying 4-months of sampling data to 20 years of operations and the uncertainty in the estimates of the amount of mercury used are difficult to quantify.

The DOE Environmental Survey Report from 1987 mentions a mercury monitor that had been removed from 292-H stack for repair, but it was suppose to have been reinstalled when it was repaired (DOE 1987). As far as we know, monitoring data have not been collected since this study. However, in 1996, plans were made to collect stack monitoring data, including mercury concentrations from F-Area, which recently begun processing. As of September 1997, funding to perform stack monitoring for nonradiological emissions had not been obtained (Villa 1997).

Kvartek et al.. (1994) estimated that in 1988 an average of 2.7 kg (5.9 lb) of mercury per year was released from the 292-H stack, and a maximum theoretical discharge of 4.8 kg y^{-1} (106 lb y^{-1}) was released (Kvartek et al.. 1994). At this time, the amount of mercury used for dissolving aluminum had been reduced. Estimates of mercury release before the reduction were 29 kg y^{-1} (63.8 lb y^{-1}) with a maximum theoretical discharge of 47 kg y^{-1} (103 lb y^{-1}) (Kvartek et al.. 1994).

Mercury was also collected by the separations area waste evaporators. There were two evaporators in both the F and H Waste Management Areas. Each evaporator had two air emission points: the vessel vent and the building exhaust. DOE (1987) reported that air emissions from the F-Area and H-Area evaporators had not been characterized. Franklin (1985), which included the three H-Area evaporators, was not mentioned.

Operating Permit Release Estimates for H-Area

Mercury vapor emissions occur as a result of the addition of mercuric nitrate catalyst $Hg(NO_3)_2$ to each dissolver batch. The mercury emissions for the Part 70 permit application were calculated for the time period when the mercury reduction program initiated after 1985 was in place. The calculations were based on actual monitoring of the 291-H stack for mercury emissions. According to the permit, mercury emissions from the 291-H stack were monitored on March 19 and 20, 1987. Emissions included in the permit application used this monitoring data and referenced two memos from M.G. Franklin to T.G. Cambell and to J.G. McKibbin, dated March 1988 and March 1985. A report dated August 1985 was located and is included in the

Phase I database (Franklin 1985), but the memo from Franklin in 1988 or any other reports about the 1985 or 1988 monitoring results were not found.

Based on the amount of fuel run through the dissolver during the time of the sampling, Radian estimated that for Mark 16B fuel with 227 kg of aluminum and 0.0021 M mercuric nitrate in 14,000 L of dissolver solution, at a use rate of 9.54 kg run^{-1}, 14 runs mo^{-1}, 12 mo y^{-1}, 1603 kg of mercuric nitrate catalyst was added, which corresponds to the measured emission of 6.3 lb y^{-1} (2.9 kg y^{-1}). Other charges to the dissolver were analyzed. For other fuels, at 370 kg aluminum per run, 10.5 kg of mercuric nitrate per run was used for 20 runs mo^{-1} and 12 mo y^{-1}, resulting in an actual use of 2527 kg y^{-1}. Assuming the same emission rate as for Mark 16B fuel, the estimated uncontrolled emission was

$$\frac{6.3 \text{ lb y}^{-1}}{1603 \text{ kg y}^{-1} \text{ used}} \times 2527 \text{ kg. y}^{-1} \text{ used} = 9.9 \text{ lb y}^{-1} = 4.95 \times 10^{-3} \text{ ton y}^{-1} \ .$$

The maximum uncontrolled emission from the dissolver was calculated by extending the estimate from 7200 hr y^{-1} actual operation to 8760 hr y^{-1}, resulting in 12 lb y^{-1} (6.05×10^{-3} ton y^{-1}).

In their 1993 H-Area air emissions calculations notebook, Radian assumed that no emissions of mercury occurred from the dissolution step of the process because mercuric nitrate "forms a large nitrate complex that remains in solution. On aluminum surfaces Hg^{+2} is reduced to Hg^0 which forms an amalgam with Al^{+3} in strong acid solutions which says that the *in situ* retention of Hg^{+2} as a catalyst in the acid solution is a contributing factor in the formation of NO gas which is vented from the dissolvers." Radian (1993) referenced Cotton and Wilkinson's Advanced Inorganic Chemistry, 2nd edition, pp. 520-521 and WSRC-TR-91-606, Purex Process Chemistry, 1991, which *RAC* was unable to locate, in support of this assumption. A special stack sampling study conducted in 1985 suggests mercury was released from the evaporators and the dissolvers (Franklin 1985). A study of emissions from the Idaho Chemical Processing Plant, which used a similar dissolution process, suggests that most of the mercury remains in the liquid waste (Herbst 1979).

Radian (1993) described the use of mercury in the dissolving process. Mercury was gravity fed to the mercuric nitrate storage tank and dissolved in the tank with concentrated HNO_3, forming $Hg(NO^3)_2$ and H_2. Concern about hydrogen emissions from the reaction was expressed, but the mercury nitrate complex was thought to be nonvolatile and there was thought to be no mercury above the solution in the tank. The mercury charging station for mercuric nitrate production was also described. Elemental mercury was poured from a flask into the charging station, which gravity fed into the mercuric nitrate storage tank. For the air inventory, Radian estimated actual mercury vapor losses from the charging operation using the EPA's AP-42 equation for organic liquid transfer losses. Radian estimated the vapor pressure of mercury under these conditions to be 1.26×10^{-5} atm at 25ºC and calculated emissions using the dissolver 'demand data' for mercuric nitrate (Radian 1993). They estimated 0.00125 lb of mercury was released per 10^3 gal transferred. The mass (m) of mercury processed was calculated from dissolver charge records using the equation:

$$m\ 100\%\ Hg = M_{Hg} \times (lb/453.6\ g) \times \Sigma(V_{dis} \times M_{Hg}) \quad (17\text{-}1)$$

where
M_{Hg} = molecular weight of mercury (200.16 g mol^{-1})
M_{Hg} = molarity of mercury in mol L^{-1} for each batch
V_{dis} = volume of dissolver solution in L for each batch.

The molarity of mercury charged to the dissolver varied with the type of target dissolved. A table in the Radian report indicates that for the 1985 estimate, they had obtained information on 13 different types of targets dissolved, the V_{dis} for each target, the number of batches run for each type of target, and the M_{Hg} for the dissolving solution used for each target. The total amounts were summed to 9026.8 lb of 100% mercury processed into mercuric nitrate or 79.58 gal of mercury, resulting in a transfer loss of (0.00125 lb 10^{-3} gal) × 79.58 gal mercury or 9.948×10^{-5} lb of mercury emitted from transfer operations during 1985. The hourly emission rates reported in the AIRS database were calculated from this value using the assumption that that mercury was transferred into the mercuric nitrate tank a maximum of 16 hr y^{-1} (Radian 1993).

The 242-25H evaporator process accounted for mercury emissions of 1.55×10^{-4} ton y^{-1}, based on airflow, capacities, and throughputs. Emission estimates for the mercury mixing station in 222-H, also called the Canyon Bulk Chemicals Facility, were 1.96×10^{-8} ton y^{-1}, maximum. This was based on a maximum use of mercuric nitrate of 3.8 lb h^{-1} for dissolving (Westinghouse 1996a).

Operating Permit Releases Estimates for F-Area

Mercury vapor was released from the F-Area dissolver because mercury catalysts were added to process the Rocky Flats scrub alloy fuel. The permit application stated that actual measurements of mercury from the H-Canyon 292-H stack on March 19 and 20, 1987, were used to estimate releases from F-Canyon processes. The dissolver batch corresponding to the measurements was characterized as containing 227 kg of aluminum and 0.0021 M mercuric nitrate in 14,000 L of dissolver solution running at an average of 14 dissolutions mo^{-1}. The amount of mercury released was proportional to the amount used in the dissolver and the amount of aluminum present because mercury amalgamates with aluminum. The HM process was estimated to use 9.5 kg of mercury per HM run or 1603 kg y^{-1}. About 38,173 kg of aluminum was processed each year. The actual, measured amounts of mercury emitted under these conditions was 6.3 lb y^{-1} (2.9 kg y^{-1}) (Westinghouse 1996a).

In the HM process, mercuric nitrate was not used for the head end strike. However, processing of the Rocky Flats scrub alloy involved adding mercuric nitrate to the dissolver and mercury vapor emissions from the head end process. In F-Area, 384 lb of mercury was added per dissolver run and 200 lb per strike in the head end process for similar processing. Processing of the Rocky Flats fuel was different because of concerns about plutonium criticality safety. Based on fuel composition and operating procedures limiting the amount of plutonium processed, the amount of catalyst used was calculated to be 109 kg per run for the dissolver and 57 kg per run for the head end. Correlated with measured emissions from the Mark 16B fuel, an actual emission of 16 lb y^{-1} from the dissolver and 8 lb y^{-1} from the head end was calculated, or 24 lb or 0.012 ton y^{-1} combined. The maximum emission from both, based on 24 h day^{-1} operation, was

estimated to be 59 lb y^{-1} or 2.95×10^{-2} ton y^{-1}. At peak production, about 1125 lb of mercury was added to the dissolver and head end for each run of Rocky Flats scrub alloy fuel. The dissolver was calculated to emit 72 lb y^{-1} based on the amount used. Emissions estimated based on the amount of aluminum dissolved yielded a much lower estimate, 1.13 lb y^{-1}. Therefore, the more conservative method was chosen and 72 lb y^{-1} (3.6×10^{-2} ton y^{-1}) is the reported actual uncontrolled emissions for the process. This was based on three runs per month, 12 mo y^{-1}, 30 h run^{-1}, and 1080 h y^{-1} (Westinghouse 1996a).

F-Area and H-Area Evaporators

The permit application reported an emission rate of 1.59×10^{-7} ton y^{-1} for mercury from the 242-1F evaporator. The permit listed all of the evaporators and overhead receiver tanks in F-Area and H-Area, a total of 22 facilities. Based on feed rates, airflow rates, tank volumes, and vapor pressures, actual emission estimates for mercury totaled 2.17×10^{-5} ton y^{-1} for F-Area and 8.66×10^{-5} ton y^{-1} for H-Area. The total for both areas was 1.08×10^{-4} ton y^{-1}. Included in these estimates are all of the 242-F and 242-H evaporators, all of the receiver tanks, the 241-H diversion box and pump tank, the 241-H-100H waste transfer facility tanks, 242-25H evaporators, 242-3F CTS, 242-21H CTS, 241-70H pump tank and Pits 5 and 6, and 241-18H CTS pump tank (Westinghouse 1996a).

Releases from the ETF stack from chemical mixing tank, filter cleaning, and waste concentrate tank were estimated to total 3.48×10^{-10} ton y^{-1} of mercury.

The emissions estimate for the 200-H emergency power generator, actual emissions assuming 2080 hours of operation each year, was 6.70×10^{-7} ton y^{-1} for mercury (Westinghouse 1996a).

Air Emissions Inventory Estimates for F-Canyon and H-Canyon

Mercury estimates for the canyon stacks were discussed in the permit application and the calculational document for H-Area, but they were not in the AIRS database for the years examined (Faugl 1996h). *RAC* asked Westinghouse to search the AIRS database again, specifically for canyon stack releases. A search of the AIRS database entries for mercury found mercury emissions were estimated for the separations area in 1994 and 1995. Emission estimates for the F-Canyon stacks were first seen in 1994. All of the actual emissions were reported to be zero, which was expected because the facility was not operating at the time. The emission estimates for maximum design capacity for F-Canyon were 0.099 ton y^{-1} for the dissolver and 0.192 ton y^{-1} for the head end exhaust in 1994 and 1995 (Faugl 1996e). The AIRS database maximum design capacity emissions estimates for 1995 for H-Canyon were 0.0195 ton y^{-1} for the dissolver and 0.030 ton y^{-1} for the head end operations (Faugl 1996e). Based on process history, we would expect that the emissions from H-Area would have been greater than from F-Area. The AIRS database estimates of 0.0495 ton y^{-1} for H-Area and 0.29 ton y^{-1} for F-Area do not seem to be representative for most years. It could be that the F-Area emissions were calculated using the assumption that the plant was processing Rocky Flats scrub alloy fuel at full capacity.

Air Emissions Inventory Estimates for Other Sources

The AIRS database contained emissions for the H-Area and K-Area coal piles and many diesel generators and pumps with a total of less than 4×10^{-5} ton y^{-1} actual emission in 1990 (Faugl 1996h). Some of the key sources are listed in Table 17-42.

Table 17-42. Mercury Emission Estimates in the AIRS Database

Year and source description	1985 and 1987		1990	
	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
1985 and 1987				
F-Area seepage basin	2.14×10^{-2}	2.14×10^{-2}	2.14×10^{-2}	2.14×10^{-2}
Old F-Area seepage basin	2.31×10^{-3}	2.31×10^{-3}	2.31×10^{-3}	2.31×10^{-3}
H-Area seepage basins	1.47×10^{-10}	1.47×10^{-10}	1.85×10^{-10}	1.85×10^{-10}
723-A met lab basin	1.00×10^{-6}	1.00×10^{-6}	1.00×10^{-6}	1.00×10^{-6}
H-Area coal storage pile	5.70×10^{-5}	5.70×10^{-5}	5.70×10^{-5}	5.70×10^{-5}
H-Area powerhouse ash sluice sump	4.79×10^{-6}	4.79×10^{-6}	4.79×10^{-6}	4.79×10^{-6}
K-Area coal pile	1.80×10^{-6}	1.80×10^{-6}	1.80×10^{-6}	1.80×10^{-6}
K-Area coal pile runoff basin	1.80×10^{-6}	1.80×10^{-6}	1.80×10^{-6}	1.80×10^{-6}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Table 17-43 presents the totals in the AIRS database, which did not appear to include the H-Canyon and F-Canyon stack releases.

Table 17-43. Total Mercury Releases from the AIRS Database

Totals	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
1985	2.54×10^{-2}	2.38×10^{-2}
1987	2.54×10^{-2}	2.38×10^{-2}
1990	3.17×10^{-2}	7.26×10^{-3}
1992	9.20×10^{-2}	2.36×10^{-3}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

In the AIRS database, the H-Canyon stack maximum design capacity emissions estimates for 1995 were 0.0195 ton y^{-1} for the dissolver and 0.03 ton y^{-1} for the head end. The F-Canyon estimates were 0.0997 ton y^{-1} for the dissolver and 0.192 ton y^{-1} for the head end. These are the largest emissions estimates reported.

Seepage Basins

Mercury was discharged to the seepage basins with low activity waste and evaporator overheads. Some of the mercury probably evaporated into the air. We did not find reports of monitoring or investigations about mercury released to the air from the H-Area or F-Area seepage basins.

The Old F-Area seepage basin was also included in the Part 70 Operating Permit Application. The 1.02-acre area, in place since 1954, was estimated to have released 2.31×10^{-3} ton y^{-1} mercury based on a concentration of mercury of 0.32 ppm, the highest concentration detected, and data on wind speed, depth, area, and temperature. The model CHEMDAT7 was used. The emission rate was estimated for 1993 and 1994. This estimate corresponds to that listed for the Old F-Area basin in the AIRS database for 1985 and 1987 (Faugl 1996h). The Ford-Area seepage basin was treated similarly, but a mercury emission estimate of zero was given (Westinghouse 1996a). Emissions for the H-Area seepage basins were listed in the AIRS database printouts (Faugl 1996h). The H-Area calculations did not describe how the values for the seepage basins were calculated (Radian 1993). The estimate was likely based on surface area and was calculated using an approach similar to that described in the permit application for the Old F-Area seepage basin. The 1985 actual emissions estimate of 1.4×10^{-10} ton y^{-1} presumably corresponds to a time period when the canyons were operating at near maximum capacity. If this amount was released each year from the H-Area seepage basin, then about 5.3×10^{-9} kg total might have been released over the 36-year period between 1954 and 1989.

Tritium Facilities

The tritium production facilities, 232-H and 234-H, used mercury in several types of process pumps such as Sprengel pumps, Edwards mercury diffusion pumps, and CEC mercury booster ejection pumps. These mechanical and diffusion pumps were used as a part of the tritium facilities gas handling system. The mercury generated a vacuum used to transfer tritium gas and evacuate process equipment. Several different types of tritium pumps have been used over the years. They all have reservoirs containing 3 to 105 kg of circulating mercury. The mercury deteriorates from oxidation and reaction with pump components and must be changed occasionally. The waste mercury may have contained a small amount of tritium and most was apparently buried in the radioactive burial ground (Horton 1973). In the 1980s and 1990s, some of the mercury pumps were replaced with oil diaphragm-type pumps (DOE 1987).

In a memo justifying discontinuation of biological monitoring in H-Area, Karoly (1985) reviewed potential sources of mercury exposure for workers and estimated the mercury inventory in equipment at various facilities. He reported that diffusion pumps at 232-H, contained from 6 to 300 lb of mercury per pump. Four diffusion columns in 232-H contained 30 lb per column and were changed out once every 2 years. Seal pots in the inert gas system for 232-H and 234-H contained 200 lb per pot and required little or no maintenance. Nine diffusion pumps in 234-H contained from 6 to 120 lb per pump, and four mechanical pumps in 234- H contained 160 lb each. Merrick scales in the 184 C, K, and P powerhouses and in 3/700-Areas contained about 0.5 L of mercury per scale. These were closed systems that should not have released mercury to the environment. In addition, about 1700 lb of mercury was stored at 234-H in polyethylene bottles inside a hood and about 1.5 ton was stored in 232-H in 55-gal drums in a chemical storage

area (Karoly 1985). Mention of 232-H maintenance operations in logbooks and other documents refers to changing mercury in pumps during pump maintenance. Deteriorated mercury was drained and the pump refilled in an air supply hood in the 1970s and 1980s. Waste mercury was bottled for disposal. Workers did not measure how much was released to the air from these activities. Shift logbooks and special work permits also mentioned leaks in pumps and maintenance of mercury-containing equipment. The Spengle pumps were drained and repaired in hoods in a contained area (Du Pont 1959). Although most of the concern centered on radiological contamination, reports suggested that mercury was occasionally monitored for industrial hygiene purposes and it was not reported to be a problem in any of the reports reviewed.

According to Kvartek et al. (1994), Schmitz 1989 reported the mercury inventory in the tritium facilities in 1989 was 2200 kg of contaminated mercury in storage, 860 kg in pumps, 110 kg uncontaminated fresh mercury in storage, and 140 kg of mercury in other sources and in 1992, Rowan (in a personal communication with Kvartek et al. (1994), reported 28 pumps, with a total capacity of 241 kg of mercury, were operating in 232-H.

In a technical memorandum given to RAC by Ed Kvartek, Johnson (1983), reported the 1983 Mercury Inventory Record for the Tritium Facilities, compiled from essential materials transaction reports. After 1968, when the burial of mercury waste was stopped, the amount of mercury used in the facilities dropped to 7530 pounds (3200 lb from 1968–1972 and 4330 lb from 1973–1983). The decrease was attributed to reuse of unoxidized mercury drained from pumps. Before 1968, all of the mercury drained from pumps (about 20,000 pounds) was buried as waste (Johnson 1983).

Data compiled by Kvartek et al.. (1994) suggest that about 10,200 kg of mercury was used by the tritium facilities from 1960 to 1987. The highest amount reported was 1300 kg in 1963, followed by 1000 kg in 1982. Zero use was reported for 1979. No amount was reported for 1978, 1980, 1981, and 1986 because “no information was available”(Kvartek et al. 1994). Excluding these 4 years, the average annual use appears to have been about 424 kg y^{-1} for the 28-year period. In an attempt to estimate the amount of mercury buried in the burial ground, Horton (1973) compiled the “amounts consumed since 1960 . . . recorded in the Essential Materials Transactions Report in the Separations Department,” referencing Du Pont reports DPSP-60-19 through DPSP-72-19 for 1960 through 1972. The total kilograms used each year is reproduced in Table 17-44. The records for 1966 and 1967 were said to have been destroyed, and consumption estimates for these years were made using an interpolated average from 1965 and 1968 values (Horton 1973).

Table 17-44. Mercury Reported to Have Been Used By the Tritium Facilities (kg)

Year	Mercury use in the tritium facilities reported by Kvartek et al. (1994)	Mercury use in the tritium facilities reported by Horton (1973)
1954–1960	Not reported	Not reported
1960	600	604
1961	900	895
1962	960	959
1963	1300	1259
1964	730	727
1965	530	525
1966	500	502
1967	500	502
1968	480	480
1969	240	239
1970	200	204
1971	150	148
1972	380	384
1973	130	Not Reported
1974	77	Not Reported
1975	790	Not Reported
1976	14	Not Reported
1977	68	Not Reported
1978	No data available	Not Reported
1979	0	Not Reported
1980	No data available	Not Reported
1981	No data available	Not Reported
1982	1000	Not Reported
1983	450	Not Reported
1984	180	Not Reported
1985	32	Not Reported
1986	No data available	Not Reported
1987	36	Not Reported

Kvartek et al. (1994) thought releases to the air from the tritium facility process stacks were small. Releases from tritium facilities were not routinely monitored. Engineering calculations in 1988 estimated that stack emissions from the facilities were less than 0.2 kg of mercury per year (Kvartek et al. 1994).

The release of mercury vapors from mercury-containing process pumps used to transport tritium and other gases through the process and metal fumes from the target furnace in the H-Area tritium facilities were not measured. Pumps in both 232-H and 234-H released mercury into the process streams through volatilization and entrainment (DOE 1987). Mercury was also present in the tritium gas distribution piping. Mercury was removed at various points using gold-foil traps designed to collect mercury in the piping. About 3 to 10 traps were removed each year. The gold traps could contain 3 to 6 g of mercury, but traps and old piping were not a significant source of mercury from the tritium facilities (Kvartek et al. 1994).

No controls for mercury were applied to the process stacks. Although stack testing for mercury has been proposed, it does not appear to have ever been conducted and mercury emissions from the facilities were not measured. The DOE Environmental Survey team felt the tritium facilities had the potential to release more than 200 lb y^{-1} (0.1 ton y^{-1}) (DOE 1987).

Emissions from the tritium facilities were not described in detail in the unclassified version of the permit application, but they were included in the totals from the AIRS database.

Separations Area Waste Tanks

Most of the mercury put into the H-Area processes came out in the radioactive liquid waste, which was said to contain about 0.5 g L^{-1} mercury (DOE 1982, 1987). Most of the mercury used in the dissolvers remained in the dissolver waste solutions and was transferred to the underground waste tanks. It is estimated that in H-Area, where the majority of the aluminum dissolving was done, 80 ton of mercury had been discharged to the waste tanks as of 1971 (Du Pont 1971a). An assessment in 1988 and 1989 estimated that the underground waste tanks contained from 73,000 kg to 91,000 kg (or about 100 ton) of mercury from separations area wastes (Kvartek et al. 1994). Another evaluation published in 1994 stated that waste solvent tanks at the Solid Waste Disposal Facility (SWDF) contained spent canyon solvents that contained up to 13 mg L^{-1} mercury, and approximately 80,000 kg of mercury was contained in high level waste tanks (Kvartek et al. 1994). The underground tanks were continuously exhausted to prevent the buildup of flammable gases. Mercury in the vapor space above the liquid waste would have been released in this exhaust, but how much mercury may have been released is unknown. As far as we can determine, mercury vapor in the waste tank exhaust has not been measured. Mercury can evaporate into the air space in waste tanks and be vented into the air through the tank ventilation systems. In answers to questions asked in the 1980s about mercury release from the tanks, Site engineers reported that the potential for emissions of mercury from the tank ventilation systems was not known (Albenesius 1992). Studies have shown that most of the mercury settled to the bottom of the sludge in the tanks and very little has been released (Kvartek et al.. 1994). The sludge in the tanks was reported to contain about 1.6 weight percent mercury hydroxide (Ebra and Wiley 1984).

In the *Action Plan for the Resolution of findings in the DOE-HQ Environmental Survey of SRS*, W.C. Reinig (1989) noted the survey finding that " atmospheric mercury emissions from the Tritium Facility and waste tank farms have not been characterized and may represent significant sources of environmental contamination and a potential human health risk." No further action was recommended because the SRS believed that the only significant source was from the H-Canyon dissolvers. Measured to be about 6 lb/yr. They reported that that H-Canyon was using about 16 per cent of the mercury used in previous years and the air emissions had been reduced from 64.5 lbs (30 kg)/yr in 1986 (maximum theoretical capacity of 104 lbs/yr) to 8.3 lbs (3.8 kg)/yr in 1987 (maximum theoretical capacity of 10.8 lbs/yr). Reinig (1989) stated that a worse case engineered calculation for the Tritium Facility demonstrated that less than 0.5 lb/yr could be released and that other mercury sources were considered insignificant (Reinig 1989).

The operating permit application contained estimates of mercury released as a result of waste tank purges in F-Area and H-Area. The total estimate from all 22 of the F-Area tanks listed was 4.48×10^{-4} ton y^{-1}. The total for the 29 H-Area waste tanks was 4.05×10^{-4} ton y^{-1} for mercury, actual emission estimates. The total for both areas was 8.53×10^{-4} ton y^{-1} (Westinghouse 1996a).

Mercury in the Burial Grounds

Until the end of 1968, all of the mercury drained during pump maintenance and replacement was buried as waste (Johnson 1983). It was usually buried in 1-L polyethylene bottles that were placed in metal cans and buried in 643-E, of the SWDF, formerly called 643-G, and also referred to as the burial ground (Orebaugh and Hale 1976).

Beginning in 1968, metallic impurities and oxidized mercury in the mercury were removed through decanting and the mercury was reused. As a result of this recycling, the amount of mercury buried was reduced and no new mercury has been added to the tritium facilities inventory since 1987 according to Kvartek et al.. (1994).

Several reports estimate that between 1956 and 1968, before recycling mercury drained from the pumps was begun, the tritium facility disposed of about 10,000 kg of mercury in the 643-E SWDF (Westinghouse 1992). After 1968, mercury trapped in the gold traps, collected from leaks or spills, or that could not be drained from equipment, continued to be buried until 643-E was filled in 1972 (Kvartek et al.. 1994).

In 1973, Horton estimated that 10 ton (20,000 lb or about 9100 kg) of mercury had been buried since startup. Horton (1973) explained that the quantities buried had not been recorded and his estimate was an upper limit estimate based on mercury use. He considered that of the 3200 lb used from 1968–1972, about 1000 lb was recovered from spills and was in storage. The remaining 2200 lb might have existed as unmeasured increased inventory in the process facilities or may have been buried in discarded process equipment (Horton 1973). Horton (1973) reported that mercury burial was stopped in 1974. The burial containers were expected to remain intact for several decades and routine monitoring to detect transport of mercury from the burial ground was not recommended at the time (Horton 1973). In the years after 1974, the mercury burial estimate determined by Horton was used by others and no additional information to improve the estimate was offered (Kvartek et al.. 1994; Orebaugh and Hale 1976; Albenesius 1992).

Waste liquid mercury was placed in a 1-L polyethylene bottle and surrounded by two polyethylene bags. Two or three bottles in bags were then placed in a 5-gal steel can. These were buried throughout 44 acres of the low-level beta gamma waste trenches in 643-G burial ground. In 1973, Hale recognized that volatilization of metallic mercury (Hg^{o}) from the burial ground into the air would be a concern if the containers in which the mercury was placed failed, and he noted that the depth of soil cover required so that this would not be a concern was unknown (Hale 1973).

Orebaugh and Hale (1976) mathematically modeled transport using 9000 kg of mercury as the source term corresponding to the 10 ton of mercury said to have been buried in trenches by Horton (1973). They examined the soil redox potential, pH characteristics, and inorganic chloride and sulfate concentrations and concluded that mercury in the soil was in a stable complex. The modeling took into account the soil moisture, mercury solubility, soil composition, burial depth, soil porosity, and permeability to water and gases of the trench fill soil and surrounding and underlying soil. The soil had been studied extensively over the years because of a concern about leaching of buried radioactive materials. Experiments were conducted to measure mercury vaporization from soil and the diffusion of mercury vapor through soil with varying moisture contents. Mercury solubility was also determined experimentally. In the course of these experiments, the researchers determined that mercury on colloids could be an important transport mechanism for mercury in soil. They found that transport could occur through vaporization into

the air or transport in water of both dissolved mercury in water percolating through the soil and mercury on oxide colloids of iron and silicon in soil water. They concluded that colloidal suspension was the dominant mode for transport of mercury from the burial grounds. A worst case estimate of possible vapor transport to the atmosphere (which assumed that all of the mercury in bottles, bags, and steel cans was released to the soil) was 5 mg h^{-1}, a flux considered by the authors to be insufficient for any environmental impact (Orebaugh and Hale 1976; Kvartek et al. 1994).

As far as we know, waste mercury packaged in bottles, bags, and cans has not been released to the soil or to the air from the SWDF. We have characterized the burial of mercury to the extent possible. At this time, developing a better source term for releases to the air from the burial grounds is not feasible because of the lack of accurate burial records, releases are not likely to have been large, and mercury buried appears to have remained in the burial ground.

Mercury from Coal Burning

Coal contains varying amounts of mercury that are released as a vapor when coal is burned. Coal-fueled boilers have been used at the SRS to generate steam to heat buildings, operate process equipment, and produce electricity. The Site burned large amounts of coal over the years. Mercury is of concern for release from coal burning plants because, unlike many of the other toxic metals, it is not effectively removed by common particulate control measures. A summary of estimates of the coal consumed since 1972 is provided in the section on coal in this chapter.

Mercury releases from coal have not been measured, but they were estimated by Horton (1974a) and also by Kvartek et al. (1994). Horton (1974a) estimated average mercury releases from the Site powerhouses based on a concentration of mercury in coal of 1 ppm. He estimated that the D-Area powerhouse burned 450,000 ton of coal and released 900 lb (410 kg) of mercury per year, and that all other powerhouses at the SRS burned 215,000 ton of coal and released 430 lb (195 kg) of mercury each year. He compared this to the South Carolina Electric and Gas Company's Urquhart Station, which burned 587,000 ton of coal each year and released 1170 lb (532 kg) of mercury.

Kvartek et al. (1994) used a concentration of 0.12 ppm of mercury in coal and calculated the amount of mercury released from 1980–1993 using the assumption that all of the mercury in the coal volatilized and was released. Interestingly, the amount of coal burned reported by Kvartek et al. (1994) for these years was higher than the amounts reported in the annual reports. Table 17-45 presents the estimates of mercury releases that Kvartek et al. (1994) and Horton (1974a) compiled.

Kvartek's estimates are about 10 fold less those based on Horton's estimate because Horton (1974a) estimated that 9.1×10^{-4} kg of mercury was released for each ton of coal burned and Kvartek et al. (1994) estimated that 1.08×10^{-4} kg ton^{-1} was released. Horton (1974a) said that coal in the U.S. averaged 1 ppm mercury, as opposed to Kvartek's estimate of 0.12 ppm said to be a coal vendors estimate. We can speculate that Kvartek's estimates might be more accurate because more was understood about mercury emissions from coal in 1994 than in 1974 and emissions factors may have been more refined. However, we know nothing about the quality of the coal burned in various years and how much mercury it contained. The EPA's emissions factors for mercury released from coal burning range from 9.7×10^{-5} to 1.3×10^{-4} lb ton^{-1} burned, much closer to the Kvartek et al. lower estimates than to the Horton larger estimates.

Unfortunately, mercury emissions from the powerhouses were not evaluated for the operating permit application nor included in the AIRS database. We have no information on coal burned between 1952–1972. We know that less than 500,000 ton of coal was probably burned each year before 1972, probably much less in the early years when the reactor areas were still under construction.

Table 17-45. Mercury Emission Estimates for the Powerhouses

Year	Coal burned (ton)	Mercury release estimates reported by Kvartek et al. (1994) (kg)	Mercury release estimates calculated using EPA AP-42 emission factors[a] (kg)	Mercury release estimates calculated using Horton's (1974) data (kg)[b]
1972	500,000		21.8–29.5	455
1973	500,000		21.8–29.5	455
1974	500,000		21.8–29.5	455
1975	500,000		21.8–29.5	455
1976	500,000		21.8–29.5	455
1977	500,000		21.8–29.5	455
1978	500,000		21.8–29.5	455
1979	500,000		21.8–29.5	455
1980	500,000	48	21.8–29.5	455
1981	500,000	51	21.8–29.5	455
1982	500,000	47	21.8–29.5	455
1983	460,000	50	20.3–27.2	418
1984	460,000	49	20.3–27.2	418
1985	455,000	49	20.0–26.9	414
1986	455,000	48	20.0–26.9	414
1987	453,000	49	19.9–26.8	412
1988	374,000	41	16.5–22.1	340
1989	227,000	25	10.0–13.4	206

[a] EPA emissions factors range from 9.7×10^{-5} to 1.3×10^{-4} lb ton^{-1} or 4.41×10^{-5} to 5.91×10^{-5} kg ton^{-1} of coal burned.

[b] Estimated mercury released based on annual report coal consumption and the factor used by Horton (1974a) of 1×10^{-4} kg released per ton of coal burned.

If we assume that 500,000 ton of coal were burned each year from 1952–1972 (a conservative assumption), and use the EPA's range of emission factors from the AP-42 publication (EPA 1988), then we can estimate that a total of about 1816–2455 kg (2.0–2.7 ton) of mercury was released from coal burning from 1952–1989, for an average of about 46.5–63 kg y^{-1} (0.05–0.07 ton y^{-1}). The emission factor used by Horton (1974a) would result in release estimates ten fold greater than these.

In 1974, Spanish moss along the Savannah River was collected and the mercury content was measured. Horton (1974a) concluded that the effects of the SRS powerhouses on moss concentrations seemed small when compared to moss concentrations adjacent to Bush Field (the Augusta airport) and the stretch of industrial plants along the river known as the 'miracle mile'.

The coal crushing operation at 784-A was evaluated for the 1996 air quality permit application. The maximum emissions for several different pieces of equipment and the coal pile, assuming operation at a capacity of 66 ton h^{-1}, totaled 1.09×10^{-5} ton y^{-1} for mercury based on the amount of coal processed. Actual emissions estimates for metals from the fuel oil-fired package boilers in K-Area also included a value for mercury at 5.55×10^{-5} ton y^{-1} (Westinghouse 1996a).

Mercury Used in Laboratory, Experimental, and Other Support Facilities

Mercury has been and still is a commonly used laboratory chemical, used as a standard and reagent for many applications. Mercury is found in thermometers, manometers, barometers, switches, relays, pressure monitors, lights, batteries, and other equipment. In the 1993 inventory, 11 facilities reported having from 0.5 to 5000 kg of mercury onsite (Kvartek et al. 1994). The Site reported that discharges from experimental, laboratory, and process support facilities were very low. The 1990 Air Emissions Inventory lists 98 emission points for mercury including storage tanks, process stacks, ash disposal, coal boilers, and an incinerator (Kvartek et al. 1994).

Releases from Other Sources

Other instances of mercury mentioned in industrial hygiene or other reports have been noted. "A mercury vapor problem in 484-D was recognized by the Instrument Department and Health Physics" was noted in the June 1955 Monthly Report for the Explosives Department (Du Pont 1955). This was likely to have been an industrial hygiene concern. A Works Technical Report from April 1977 mentions bioassay of D-Area employees who routinely handled mercury. All of the employees were negative, but bioassays were to be done annually (Du Pont 1977b). This suggests that mercury was used in D-Area in large enough quantities and routinely enough to be of concern for worker exposure (Du Pont 1977b). Other records that explain how mercury was used or released in D-Area have not been discovered. Several workers with knowledge of historical operations in D-Area could not recall a use for mercury. They speculated that mercury in river water taken by the 400-Area to use for the powerhouse and heavy water processes may have been a concern for workers working in the water treatment plant.

Another small source of mercury emissions, noted in the Part 70 Operating Permit Application Amendment No. 3 from 1997, was a fluorescent lamp disposer with a maximum emission estimate of 0.00313 ton y^{-1} for mercury (Westinghouse 1996c).

Summary of Mercury Releases to Air

In summary, both Frankin (1985) and Kvartek et al. (1994) estimated that an average of 29 kg y^{-1} (0.03 ton y^{-1}) and a maximum of 47 kg y^{-1} (0.05 ton y^{-1}) was released from 292-H before 1985. Kvartek estimated an average of 2.7 kg y^{-1} (2.9×10^{-3} ton y^{-1}) and maximum of 4.8 kg y^{-1} (5.3×10^{-3} ton y^{-1}) was released from 292-H after 1988. In 1987, the DOE Environmental Survey team believed that the tritium facilities had the potential to release 0.1 ton y^{-1} (DOE 1987). Kvartek et al. (1994) predicted a release of less than 2.5×10^{-3} ton y^{-1} for the tritium facilities. There is no evidence that mercury in the burial grounds has been released to the air.

The air permit estimates for the H-Area and F-Area waste tank purges totaled 8.5×10^{-4} ton y^{-1}. We might add 0.05–0.07 ton y^{-1} from coal burning for most years.
Release estimates are summarized in Table 17-46.

Table 17-46. Summary of Estimates for Mercury Releases to Air

Source of releases	Source of estimate	Actual estimate or lower estimate (ton y^{-1})	Maximum estimate or higher estimate (ton y^{-1})
F-Area and H-Area	AIRS database (Faugl 1996h); Permit Application (Westinghouse 1996a); Kvartek et al. (1994)	0.03	0.3395
Tritium facilities	Kvartek et al. (1994) and DOE Survey (DOE 1987)	0.0025	0.1
Waste tanks	Permit Application (Westinghouse 1996a)	8.5×10^{-4}	8.5×10^{-4}
Coal burning	EPA 1988	0.05	0.07
Total		0.083	0.51

The uncertainty and error in the estimates derived from essential materials and coal inventories, estimates of mercury content of coal, waste tanks, seepage basin sediments, and canyon use and release data are not readily quantifiable. The variability of calculated values are at least an order of magnitude. There is a 10-fold difference between different reported estimates for mercury releases from coal burning. There is a similar difference between estimates on the air emissions from the canyon**.**

In an attempt to quantify uncertainty in releases of mercury, we utilized the combined information from releases of mercury from stacks, tritium facilities, waste tanks, coal burning, and other miscellaneous sources. The AIRS database and permit applications yielded information on releases from the stacks and miscellaneous sources. For use in uncertainty calculations, we created triangular distributions of the AIRS database estimates based upon the actual release, used to represent the most likely value, and the maximum release, used to represent the boundary of the triangular distribution.

Uncertainty in releases from the tritium facilities was large as a result of two release estimates from independent sources that were nearly three orders of magnitude different from one another. The DOE estimate was assumed to be the least likely, and was used as a boundary to the triangular distribution, with the release developed from engineering calculations used as most likely.

Separations area waste tanks had two different release estimates quoted in available literature, one from Kvartek and one from the operating permit application.

Releases from coal burning were estimated by multiplying the estimated coal burned at the Site each year by a distribution of estimates for total mercury released per ton of coal burned. This distribution of estimates was derived from the total percentage of mercury in coal reported by the Site throughout the years as well as some of the AP-42 estimates for releases per ton burned.

The average mercury release per year was fitted to a lognormal distribution with a geometric mean of 0.30 ton y^{-1} and a geometric standard deviation of 1.38. The 5th and 95th percent estimates of this distribution were 0.18 ton y^{-1} and 0.51 ton y^{-1}, respectively. The 95th percentile

matches the maximum release estimated by simply adding together the maximum release estimates.

This estimate for uncertainty in mercury releases is, however, based only on available release information. There is non-quantifiable uncertainty associated with using release estimates for a few years to estimate releases for all years, as well as the uncertainty of operations in the 1950s and 1960s as they compare to the 1980s.

No ambient air monitoring near the SRS was conducted for mercury as far as we know. A limited amount of modeling, needed for compliance with air quality regulations, has been done. In 1993, the maximum 24-hour average concentration of mercury in the air at the Site boundary, modeled using the EPA's Industrial Source Complex Short Term (ISCST) model, was estimated to average about 0.0024 $\mu g\ m^{-3}$, considerably below the SCDHEC ambient air standard of 0.25 $\mu g\ m^{-3}$ (Dukes 1993). However, 1993 was certainly not a year of maximum or even moderate production for the canyons.

In their report, *Assessment of Mercury in the SRS Environment*, Kvartek et al. (1994) concluded that no significant releases of mercury to the air were likely to have occurred. They believe that any releases would have been well below the SCDHEC ambient standard based on process knowledge. Kvartek et al. (1994) published an estimate of the predicted a maximum concentration of mercury at the Site boundary of 0.014 $\mu g\ m^{-3}$, also below the standard. The calculated daily amount inhaled did not exceed the EPA's reference concentration for mercury. The summed oral and inhalation hazard index for an offsite individual was 0.03 (Kvartek et al. 1994). A hazard index $\geq$1 indicates a concern for potential health effects.

Nickel

Three nickel electroplating tanks in Building 313-M used a plating solution of nickel sulfate, nickel chloride, and boric acid. Nickel in air in M-Area has not been monitored and did not seem to be an industrial hygiene concern. M-Area calculations done for the air emissions inventory included graphite from lubricants, aluminum and lithium from various processes, and ammonia from aquadag; they did not include nickel or lead from lubricant oils or nickel plating. The nickel electroplating process was said to result in negligible aerosol emissions because of the high efficiency of the plating process. Reports from the U.S. Air Force and American Airlines studies were cited as the basis for the assumptions used for calculating emissions. A worst case emission for the process was estimated to be 5.8 lb y^{-1} for 1985–1990 based on estimates from a less efficient chromium-plating process (Radian 1992a).

Nickel emissions reported in the AIRS database were primarily from welders and diesel generators and pumps. The AIRS database included nickel emissions from generators; coal operations; and many metals fabricating, machining, and finishing operations. Nickel releases were calculated for the H-Area and K-Area coal piles and 232-H manufacturing processes. The coal crushing operation at 784-A was also evaluated for the air quality permit. The maximum emissions for several different pieces of equipment and the coal pile, assuming maximum operating at capacity, totaled 1.27×10^{-3} ton y^{-1} for nickel based on the amount of coal burned (Westinghouse 1996a). Using the relationship assumed for the A-Area boilers, burning 500,000 ton of coal per year site-wide would correspond to a nickel emission of 0.037 ton y^{-1}. Actual emissions estimates for metals from the fuel oil-fired package boilers in K-Area included nickel

with 3.3×10^{-4} ton y^{-1} (Westinghouse 1996a). Some of the key emissions estimates are summarized in Table 17-47.

Table 17-47. Emissions Estimates for Nickel in the AIRS Database

Year and source description	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
1985		
Coal storage pile H-Area	1.30×10^{-3}	7.70×10^{-5}
H-Area 284 powerhouse ash sluice pump	2.54×10^{-4}	5.53×10^{-5}
H-Area manufacturing Building 232		
232-H hood metallography	Blank	0
234-H hood cutting	Blank	0
234-H radiological equipment repair	1.97×10^{-3}	3.25×10^{-5}
234-H finishing operations	8.59×10^{-5}	8.59×10^{-5}
234-H inert finishing operations	1.04×10^{-5}	1.04×10^{-5}
234-H fabricated metals machining	1.56×10^{-1}	5.21×10^{-2}
238-H milling and machining hood	1.41×10^{-2}	1.55×10^{-4}
238-H lathe hood	1.41×10^{-2}	1.55×10^{-4}
H-Area tank farm field welding operations	Blank	Blank
H-Area diesel generators	Blank	Blank
K-Area coal pile	4.20×10^{-5}	4.20×10^{-5}
K-Area coal pile runoff basin	4.20×10^{-5}	4.20×10^{-5}
1987		
Coal storage pile H-Area	1.30×10^{-3}	7.70×10^{-5}
H-Area 284 powerhouse ash sluice pump	2.54×10^{-4}	5.53×10^{-5}
H-Area manufacturing Building 232		
232-H hood metallography	3.30×10^{-3}	7.92×10^{-4}
234-H hood cutting	2.09×10^{-2}	9.79×10^{-4}
234-H radiological equipment repair	1.97×10^{-3}	3.25×10^{-5}
234-H finishing operations	9.38×10^{-5}	9.38×10^{-5}
234-H inert finishing operations	1.91×10^{-5}	1.91×10^{-5}
234-H fabricated metals machining	1.56×10^{-1}	5.21×10^{-2}
238-H milling and machining hood	1.41×10^{-2}	9.86×10^{-5}
238-H lathe hood	1.41×10^{-2}	1.97×10^{-4}
K-Area coal pile	4.20×10^{-5}	4.20×10^{-5}
K-Area coal pile runoff basin	4.20×10^{-5}	4.20×10^{-5}
1990		
H-Area manufacturing Building 232		
234-H finishing operations	9.38×10^{-5}	9.32×10^{-5}
234-H inert finishing operations	1.91×10^{-5}	8.60×10^{-6}
234-H fabricated metals machining	1.56×10^{-1}	5.21×10^{-2}
238-H milling and machining hood	1.41×10^{-2}	9.42×10^{-5}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Table 17-48 presents nickel emission estimates in the AIRS database by source. The totals are shown in Table 17-49.

Table 17-48. Emissions Estimates for Nickel in the AIRS Database, by Source[a]

Emission source	Year	Actual[b] emission estimate (ton y^{-1})
H-Area coal storage pile	1985	7.70×10^{-5}
	1987	7.70×10^{-5}
	1995	2.15×10^{-5}
H-Area powerhouse	1985	2.54×10^{-4}
	1987	2.54×10^{-4}
H-Area manufacturing building	1985	5.21×10^{-1}
	1987	5.21×10^{-1}
	1995	1.61×10^{-2}
H-Area reclamation building	1985	2.82×10^{-2}
	1987	2.82×10^{-2}
	1995	2.51×10^{-2}
K-Area coal pile	1985	4.20×10^{-5}
	1987	4.20×10^{-5}
K-Area coal pile runoff area	1985	4.20×10^{-5}
	1987	4.20×10^{-5}

[a] Source: Faugl (1996i).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Table 17-49. Total Nickel Releases[a]

Year	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
1985	3.49×10^{-1}	7.03×10^{-2}
1987	3.76×10^{-1}	7.22×10^{-2}
1989	3.86×10^{-1}	6.94×10^{-2}

[a] Source: Faugl (1996i).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

The highest emissions for 1985 were from the fabricated metals machining in 234-H, followed by field welding operations in the H-Area tank farm and diesel generators in the H-Area diesel house. The air quality permit emissions estimate for the 200-H emergency power generator, actual emissions assuming 2080 hours of operation each year, was 1.32×10^{-6} ton y^{-1} for nickel (Westinghouse 1996a).

Nickel plating in M-Area was not a significant source of nickel releases to the air. Nickel releases from metal cutting, machining and finishing operations, welding, and combustion sources were probably less than 0.4 ton y^{-1}. We estimate that total emissions may have ranged from actual to maximum estimates of 0.11 – 0.423 ton y^{-1}. These ranges are the only information available for nickel releases. Uncertainty could not effectively be calculated from the point values.

Nitric Acid

Nitric acid was used in large quantities in H-Area, F-Area, M-Area, CNX, TNX, and other areas as a process chemical. Many of the reactions involving nitric acid produced nitrates, which were discharged in liquid effluents, and oxides of nitrogen, which were discharged to the air. Some of the process exhaust was subject to control devices that reduced emissions, such as an acid scrubber, condenser, or nitrogen oxides absorption column. Much of the nitrogen oxide emissions to the air were uncontrolled. Many processes released both nitric acid fumes and nitrogen oxides and often the magnitude of the release of one was correlated to the other. Nitric acid and nitrogen dioxide releases from the SRS have been determined based on process data, opacity readings, and stack monitoring. Ambient air monitoring data, described in Chapter 19, is useful for determining how nitrogen dioxide produced onsite may have affected concentrations offsite.

Inhaled nitric acid reacts in the upper respiratory tract. Exposure to high doses of nitric acid causes lung irritation and may exacerbate lung diseases like asthma.

Air Emissions Estimates

Nitric acid emission estimates were included in the AIRS database (Faugl 1996j). Process emissions are shown in Table 17-50, and key emissions are shown in Table 17-51. The highest estimate was for a nitric acid storage tank used in 245-H for regeneration of resin, with an actual emissions estimate of 8.76×10^{-2} ton y^{-1}. In 1985, 1987, and 1990, 20 laboratory hoods and two glove boxes for the A-Area Radiological and Environmental Laboratory Process and Production, Building 735, were listed with an actual emissions estimate of 2.05×10^{-2} ton y^{-1} each. Nine laboratory hoods were listed with an emissions estimate of 1.56×10^{-2} ton y^{-1} each, totaling 31 hoods with a total actual emission estimate of 0.59 ton y^{-1}. In 1992, the number of laboratory hoods for A-Area decreased to 13, with actual emissions estimates totaling 6×10^{-2} ton y^{-1}. Ten emission points were listed for the F-Canyon, but all of the estimates given were zeros. In 1990, the M-Area emission points were listed with the same maximum value as the years before, but they were given an actual value of zero (Faugl 1996j).

Table 17-50. Process Emissions Estimates for Nitric Acid in the AIRS Database[a]

Year and source description	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
1985		
H Bldg 245 Resin regeneration building nitric acid tank	8.76×10^{-2}	8.76×10^{-2}
M-Area: Canning Building 313 core recovery HNO3 etch	2.05×10^{-7}	1.74×10^{-7}
Canning Building 313 slug nitric etch	6.48×10^{-3}	5.49×10^{-3}
Canning Building 313 Apl station 6 post etch nitric	3.22×10^{-3}	0
Canning Building 313 Apl station 7 post etch nitric	3.22×10^{-3}	0
Canning Building 313 Apl station 8 post etch nitric	3.22×10^{-3}	0
Canning Building 313 Apl etch line station 13	6.15×10^{-7}	0
Canning Building 313 Apl etch line station 14	6.15×10^{-7}	0
Canning Building 313 Apl etch line station 15	3.11×10^{-2}	0
Alloy Building 320 nitric acid tank	1.00×10^{-4}	1.00×10^{-4}
T-Area Semiworks Building 678 nitric acid storage tank	4.0×10^{-4}	4.0×10^{-4}
1987		
F-Area Canyon Stack Acid Recovery 607 Building 291	2.50×10^{-3}	2.50×10^{-3}
F-247-F Naval fuels glove boxes	6.0×10^{-4}	6.0×10^{-4}
H Bldg 245 resin regeneration building nitric acid tank	8.76×10^{-2}	3.76×10^{-4}
M-Area: Canning Building 313 core recovery HNO3 etch	2.05×10^{-7}	9.51×10^{-8}
Canning Building 313 slug nitric etch	6.48×10^{-3}	3.01×10^{-3}
Canning Building 313 Apl station 6 post etch nitric	3.22×10^{-3}	8.24×10^{-3}
Canning Building 313 Apl station 7 post etch nitric	3.22×10^{-3}	8.24×10^{-3}
Canning Building 313 Apl station 8 post etch nitric	3.22×10^{-3}	8.24×10^{-3}
Canning Building 313 Apl etch line station 13	6.15×10^{-7}	1.57×10^{-8}
Canning Building 313 Apl etch line station 14	6.15×10^{-7}	1.57×10^{-8}
Canning Building 313 Apl etch line station 15	3.11×10^{-2}	7.94×10^{-4}
Alloy Building 320 nitric acid tank	1.00×10^{-4}	1.00×10^{-4}
1990		
F-Area Canyon Stack Acid Recovery (unit) 607, 291-F	2.50×10^{-3}	2.50×10^{-3}
F-247-F Naval fuels glove boxes	6.0×10^{-4}	6.0×10^{-4}
H Bldg 245 resin regeneration building nitric acid tank	8.76×10^{-2}	3.49×10^{-4}
H-Area acid storage tank	1.13	2.65×10^{-2}
M-Area Alloy Building 320 nitric acid tank	1.00×10^{-4}	1.00×10^{-4}
M-Area: Canning Building 313 core recovery HNO3 etch	2.05×10^{-7}	0
Canning Building 313 slug nitric etch	6.48×10^{-3}	0
Canning Building 313 Apl station 6 post etch nitric	3.22×10^{-3}	0
Canning Building 313 Apl station 7 post etch nitric	3.22×10^{-3}	0
Canning Building 313 Apl station 8 post etch nitric	3.22×10^{-3}	0
Canning Building 313 Apl etch line station 13	6.15×10^{-7}	0
Canning Building 313 Apl etch line station 14	6.15×10^{-7}	0
Canning Building 313 Apl etch line station 15	3.11×10^{-2}	0

[a] Source: Faugl (1996j).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Table 17-51. Key Emissions of Nitric Acid in the AIRS Database by Source[a]

Emission source	Year	Actual[b] emissions estimate (ton y^{-1})
F-Area Canyon stack acid recovery	1985	2.50×10^{-3}
F-Area Canyon stack acid recovery	1987	2.50×10^{-3}
F-Area Canyon stack - second cycle	1992	6.4×10^{1}
F-Area Canyon stack - feed prep	1992	7.7×10^{-4}
H-Area Canyon resin regeneration	1992	1.24×10^{-4}
H-Area chemical storage tanks	1992	9.97×10^{-1}
H-Area tanks	1992	2.62×10^{-2}
T-Area Chemical Semiworks and Pilot Plant, five emission points	1985	2.0×10^{-3}
	1987	2.0×10^{-3}
	1990	2.4×10^{-3}
TNX Chemical Semiworks	1992	1.0×10^{-3}

[a] Source: Faugl (1996j).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

The total nitric acid emissions reported in the AIRS database are shown in Table 17-52. One of the largest single estimates for 1992 was the M-Area Alloy Building acid tank with 2.0×10^{1} ton y^{-1} for actual and maximum emissions. This source had an emissions estimate of 1×10^{-4} ton y^{-1} in the AIRS database for 1985, 1987, and 1990. The increased emissions estimate for this source and the estimates for the F-Canyon second uranium cycle, which totaled 6.4×10^{1} ton y^{-1}, accounted for the larger estimate in 1992. The H-Canyon stack emissions were not included. The estimates for 1985 and 1987 might have been larger than the 1992 estimates if these emissions points had been included because the production for the canyons and M-Area was greater in 1987 than in 1992.

Table 17-52. Total Nitric Acid Emissions Estimates[a]

Totals oxides of nitrogen	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
1985	1.10	6.03×10^{-1}
1987	1.10	6.26×10^{-1}
1990	2.20	6.22×10^{-1}

[a] Source: Faugl (1996j).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

M-Area Nitric Acid Releases. M-Area Nitric Acid Releases were included in the AIRS database estimates. Emissions included in the operating permit application applied to storage tanks but not operations.

Worksheets contained in Radian (1992a) for calculating nitrogen oxide emissions included complicated calculations for nitric acid releases that assumed a process rate of 75,000 gal y^{-1} for the plating line acid scrubber. The process used caustic etch tanks and nitric etch baths. To estimate potential releases, Radian (1992a) determined the number of cores per shift and the number of cores processed per month and used the concentration of materials, volumes in the tanks, tank surface areas, and material balance information (found in the degreaser logbook, DPSOLs, and material balance or essential materials logbooks for 1983). The emissions from the hot and cold water rinse tanks (used before and after cleaning, etching, and plating processes) were assumed to be negligible. Nitric acid tank emissions were said to have been very small because of the low vapor pressure of the solutions.

Storage tank emissions, calculated using AP-42 equations for working and breathing losses, were calculated using information on the throughput, temperature, tank design, and vapor pressure of contents. The standing losses because of tanks breathing and working losses because of vapor displacement during filling were added to the emissions. These loses are most important for aboveground tanks that were not insulated and expand and contract daily because of temperature changes (Radian 1992a). This was applicable because nitric acid was stored in fixed roof storage tanks in M-Area. The throughput for these tanks was reported to be 27,000 gal in 1985, 30,000 gal in 1986, 25,000 gal on 1987, 44,000 gal in 1988, 20,000 gal in 1989, and 28,000 gal in 1990. Using data on the fill rates, transfer rates, venting, and temperature changes, a maximum working loss for the tanks was calculated to be 1.73 lb y^{-1} in 1985. The breathing losses, calculated assuming the tanks were one-half full, totaled 10.80 lb y^{-1}. Another spreadsheet in the documentation for M-Area air emissions contained maximum working and breathing losses for five nitric acid and nitric acid waste tanks. These totaled 6.8 lb y^{-1} for 1985, 7.8 lb y^{-1} for 1986, 7.05 lb y^{-1} for 1987, 6.9 lb y^{-1} for 1988, and 6.58 lb y^{-1} for 1989. Calculations of throughputs for acids and caustic were handwritten and seem to be based on use records, monthly receipts, and information obtained from employee interviews.

H-Area Nitric Acid Releases Calculated for the Operating Permit Application. Nitric acid (50% HNO_3) is used to dissolve aluminum-clad fuels. The nitric acid and mercuric nitrate catalyst were heated to boiling, or a maximum of 110°C, to increase the dissolution rate. Nitrogen dioxide generated in the dissolver reacted with water in the off-gas condenser to form nitric acid, which returned as a condensate to the dissolver. Not all of the nitrogen dioxide generated was converted to nitric acid. The nitric acid emissions from the dissolver were calculated assuming (a) the off-gas was 50°C, (b) dissolving accounts for 20 hours of a 30-hour dissolving cycle, (c) 10% of the nitric acid added to the dissolver was excess and was available for emission, and (d) 20 runs per month were conducted. For calculating nitric acid emissions, Radian (1993) assumed that 45 ton y^{-1} of nitrogen dioxide was emitted from the dissolver. The iodine reactor, which used silver nitrate to convert iodine in the off-gas to silver iodide or iodate, produced nitric acid. Based on stoicheomtery, one iodine reactor was estimated to produce about 0.026 lb y^{-1} of nitric acid. The actual emissions for nitric acid from the dissolvers in H-Area totaled 1.05×10^{-5} ton y^{-1} for the iodine reactor and 0.036 ton y^{-1} for the dissolver (Westinghouse 1996a).

Emissions of nitric acid from the head end process (based on tank capacity, throughput, and vapor pressure) were said to have been negligible. Maximum emission of nitric acid from the first cycle extraction was calculated to be 0.822 ton y^{-1}. Based on flow rates, tank working volumes, and vapor pressures, Radian (1993) estimated that a maximum of 720 lb y^{-1} of nitric acid was emitted from process vessels associated with the first cycle. Second uranium cycle losses were estimated to be 1.72 ton y^{-1} maximum, and the second neptunium/plutonium cycle emissions of nitric acid were calculated to be 1.43 ton y^{-1} maximum. Solvent recovery processes were estimated not to release nitric acid or nitrogen dioxide (Radian 1993). The solvent recovery processes consisted of three separate solvent recovery systems, one for each extraction cycle, which removed radioactive contaminants and chemical degradation products from solvent using alkaline and acid washes.

The frame waste recovery system in H-Area, which purified and concentrated ^{238}Pu solutions, used nitric acid. The maximum throughput was reported to be run in 1984. The 1984 throughput was used to determine emissions estimates for the Title V Permit Application. Based on stoichiometry, throughputs, column feed rates, and maximum operating temperatures allowed for worker safety concerns, a release estimate 1.77 ton of nitric acid maximum emissions was estimated based on 1984 throughputs (Westinghouse 1996c).

The maximum nitric acid emissions from the acid recovery unit in 221-H were estimated to be 0.019 ton y^{-1} based on pump, tank, and column capacities. H-Canyon rerun process nitric acid releases from all of the process vessels were estimated to be 1.72 ton y^{-1} maximum (Westinghouse 1996c). The H-Canyon general purpose evaporator was not thought to have emitted nitric acid or nitrogen dioxide. Maximum nitric acid emissions from the Segregated Solvent facilities were estimated to be 0.012 ton y^{-1} because of solvent washing. No emissions were found to be worth reporting for the permit application for the H-Area sumps and water handling facility.

The emissions calculation for the third-level cold feed process, old HB-Line facility decontamination and decommissioning activities, and the enriched uranium system are Unclassified Controlled Nuclear Information and were not copied. They were available for *RAC* review and the emissions were included in the AIRS database values that *RAC* compiled.

The acid recovery unit in Building 221-H concentrated dilute nitric acid that had been used as a scrubbing or solvent wash solution in the H-Canyon. The process components that could have contributed to emissions were the nitric acid receiving tank, the recovered nitric acid storage tank, recovered nitric acid run tanks, overhead condensate tanks, and the nitric acid evaporator column. The evaporator column is sealed and vapor losses from dilute nitric acid tanks were assumed to be nearly zero. Transfer losses and breathing losses from the tanks were calculated for each tank based on the target class data for 13 different types of targets dissolved in 1985 and using the AP-42 equations for liquid storage tanks, tank dimensions, and nitric acid vapor pressure of 0.039 mm Hg. The total losses were estimated to be 40.92 lb for the nitric acid tanks in 1985 (Radian 1993).

Estimates of releases from the cold feed preparation area in 222-H, where reagents were formulated for use in the canyons, were made for 1985. The area operations were vented through the process vessel vent system to the 291-H stack. The cold feed preparation area housed four dilute nitric acid tanks. Radian (1993) estimated that the 1000-gal nitric acid tank at 245-H had an estimated maximum emission of 1.15×10^{-4} ton y^{-1} nitric acid. The four 17,700-gal nitric acid tanks in place since 1953 in 211-H had maximum emissions totaling 4.06×10^{-1} ton y^{-1}. The

permit application for the H-Canyon from 1995 reported maximum controlled emissions for nitric acid to total 12.1 ton y^{-1} based on engineering calculations (Westinghouse 1996c).

F-Area Nitric Acid Releases Calculated for the Operating Permit Application. Actual uncontrolled working volume losses of nitric acid were calculated to be 0.516 ton y^{-1} for the F-Area dissolver and head end processes. The nitric acid releases from the dissolver were calculated accounting for the condenser, which returns most of the nitric acid vapors to the dissolver. The condenser also converts some of the nitrogen oxides to nitric acid, but this was not considered to reduce the nitrogen dioxide estimates, which were meant to be conservative. However, to maximize the nitric acid release estimates, it was assumed that all of the nitrogen oxides emitted were converted to nitric acid. Nitric acid releases from the iodine reactor, the makeup tanks, and the B-Line waste tanks were included in the permit application estimates. Controlled emissions for nitric acid were calculated assuming the F-8 column had a 90% efficiency. The engineers thought that sufficient water was used so that the F-8 absorber could absorb 100% of the nitric acid released from the dissolvers and the iodine reactor, and 90% was thought to be a conservative value. The actual, controlled emission estimate for nitric acid from these processes was estimated to be 0.052 ton y^{-1}. Emissions for the head end process were calculated using Raoult's Law and throughputs through the process vessels. The maximum air emissions were estimated to be 10.1 ton y^{-1}. The first cycle maximum emissions for nitric acid were 2.37 ton y^{-1}. The second uranium cycle losses of nitric acid were estimated to be 0.73 ton y^{-1}. The second plutonium cycle losses of nitric acid were estimated to be 1.88 ton y^{-1}. Emissions from the solvent recovery system were estimated to be 0.45 ton y^{-1}; the low activity and high activity waste systems for 221-F were not thought to have released nitric acid into the air.

FB-Line, in Building 221-F, made plutonium buttons from a dilute nitrate solution. FB-Line tanks and vessels were enclosed in cabinets or glove boxes. The line included four major processes and four vacuum venting systems. The vessel vent vacuum system was preceded by Teflon filters and used a venturi scrubber to scrub the off-gas, which contained nitrous oxides and nitric acid. This system also used a caustic scrubber to neutralize the nitric acid vapors. Actual emissions for nitric acid from the FB-Line were estimated to be 1.94×10^{-5} ton y^{-1}. Estimates of maximum emissions from the cold feed nitric acid tanks used for the FB-Line totaled 4.2×10^{-2} ton y^{-1} (Westinghouse 1996c).

Maximum nitric acid emissions from the acid recovery unit in 221-F, which concentrated nitric acid evaporator overheads by vacuum distillation to 50% for reuse, were reported to be 0.0198 ton y^{-1}. The five 221-F nitric acid storage tanks maximum emissions estimates totaled 6.05×10^{-1} ton y^{-1}. Emissions estimates for tanks in 222-F totaled 1.77×10^{-2} ton y^{-1}. All of these exhausts flowed into the wet cabinet exhaust, which went through a series of prefilters, HEPA filters, and a sand filter then out the 291-F stack.

Other emissions for F-Area included those for the segregated solvent facilities, with a maximum nitric acid emissions estimate of 0.0556 ton y^{-1} nitric acid (Westinghouse 1996a).

At the time of the 1996 air permit application, the F-Canyon was not operating and actual emissions were reported as zero. The combined F-Canyon stack maximum emission estimates, assuming 24 hour a day releases of nitric acid, totaled 16.3 ton y^{-1} based on engineering calculations (Westinghouse 1996a). This compares to a maximum design capacity controlled

emission estimate total for nitric acid from F-Area, including the canyons of 9.89 ton y^{-1} in the AIRS database for 1995 (Faugl 1996e).

Other Areas. In the AIRS database for 1985, 1987, and 1990, 29 laboratory hoods and two glove boxes were listed as emission points for nitric acid for the Radiological and Environmental Laboratory Process and Production, Building 735. Actual emissions estimates totaled 0.59 ton y^{-1} (Faugl 1996j). A nitric acid maximum emission of $4.6\ 0 \times 10^{-6}$ ton y^{-1} was given for the Par Pond Laboratory in 1994 (Westinghouse 1996a).

From 1987 to 1995, the TRI, reported to the EPA and SCDHEC, included estimates of the pounds per year of nitric acid released from the SRS. The TRI estimates shown in Table 17-53 are lower than the actual AIRS database emission estimates.

Table 17-53. Toxic Release Inventory Release Estimates for Nitric Acid[a]

Year	Air emissions (lb y^{-1})	Ton y^{-1}
1987	71000	35.5
1988	54100	27
1989	19000	9.5
1990	8000	4
1991	3601	1.8
1992	3601	1.8
1993	37000	18.5
1994	32050	16
1995	224	0.1

[a] Source: Westinghouse (1996b).

Obviously, there is a big difference between actual emission and maximum design emission estimates for nitric acid. Estimates calculated for the AIRS database and permit application suggest that 30–150 ton y^{-1} were released in 1985–1992. Thirty ton is the total of the lowest actual emissions in the permit application (primarily 16.8 ton for H-Area, 12.1 ton for F-Area, and the total actual emission estimates for M-Area in the AIRS database of 2.85×10^{-2} ton y^{-1} plus other smaller emissions in the AIRS database). The 150-ton estimate is the highest annual estimate in the AIRS database, which was 86 ton y^{-1} for 1992 and included an emissions estimate for the F-Canyon stack but not the H-Canyon stack. If the H-Canyon stack releases were similar to those predicted for the F-Canyon stack, as much as 64 ton y^{-1} should be added to the emissions estimate, for a total of 150 ton y^{-1}. It is likely that releases in the past were similar and may have been less because 1985 was one of the highest years for M-Area as well as the canyon's production.

Total nitric acid releases had such a small range as to make uncertainty calculations from the release estimates misleading. Not all sources of uncertainty could be quantified well enough to make this a worthwhile calculation.

The Standard 8 results submitted to the SCDHEC said that in 1991, nitric acid was emitted by 47 sources in A-Area, B-Area, F-Area, H-Area, M-Area, S-Area, and TNX. The maximum

24-hour average Site boundary concentration was calculated to be 50.95 μg m^{-3}. The ambient air standard was 125 μg m^{-3}.

Oxides of Nitrogen

Because various forms of nitrogen oxides can occur together in the air and many are chemically convertible, the compounds (which include nitric oxide [NO], nitrogen dioxide [NO_2], nitrous oxide [N_2O], nitric acid [HNO_3], and other nitrogen oxides) are generally designated NO_x or oxides of nitrogen.

Most of the oxides of nitrogen are produced from combustion sources and is initially produced as nitric oxide, which is generally rapidly oxidized to nitrogen dioxide. Nitric oxide and nitrogen dioxide are chemically reactive and of public health interest. Many of the emissions estimates in the Title V Permit Application and in the AIRS database are given for nitrogen oxides or oxides of nitrogen that may include nitric oxide and nitrous oxide as well as nitrogen dioxide, which is the compound of most concern. Most of the nitric oxide released is oxidized to nitrogen dioxide. Also, several reports suggest that the canyon operators did air sparging of the evaporator to improve retention of acid in the dissolver by oxidizing nitric oxide to nitrogen dioxide (Du Pont 1974b). Since nitrogen dioxide is the toxic pollutant of most concern, we will assume that all of the oxides of nitrogen are nitrogen dioxide unless otherwise indicated.

Depending on the concentration, the presence of nitrogen dioxide in air is indicated by a light yellow to reddish brown color. Inhaled nitrogen dioxide affects the lungs at low levels of exposure, possibly decreasing pulmonary protective mechanisms. Nitrogen dioxide does not appear to be a carcinogen. In general, studies on humans suggest that levels <1 ppm do not cause significant changes in pulmonary function in normal, healthy adults. Epidemiological studies of people exposed to nitrogen dioxide in indoor and outdoor air have found both positive and negative associations with a number of acute respiratory conditions. No definitive conclusions about health effects caused by nitrogen dioxide have been drawn from such studies.

Oxides of nitrogen were released from many facilities at the SRS that used nitric acid. The facilities that used the largest amounts were the 200-F and H-Canyons and B-Line processes, the 300-M Area fuel fabrication facilities, and the TNX and CMX pilot plants. Nitrogen dioxide was also released from the power plants, the Naval Fuels Manufacturing Facility, and many small combustion engines.

The AIRS database releases for oxides of nitrogen were compiled for 1985, 1987, 1990, and 1992 and are presented in Table 17-54 (Faugl 1996k).

Table 17-54. Total Oxides of Nitrogen Emissions Estimates[a]

Totals oxides of nitrogen	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
1985	2.73×10^3	3.55×10^2
1987	2.92×10^3	4.69×10^2
1990	3.06×10^3	3.64×10^2

[a] Source: Faugl (1996k).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Process releases and releases of most concern are listed in Table 17-55.

Table 17-55. Process Releases of Oxides of Nitrogen in the AIRS Database

Year and source description	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
1985		
F-Area canyon stack, Bldg 291 second uranium cycle	4.93×10^{1}	4.93×10^{1}
F-Area canyon stack, Bldg 291 solvent recovery	2.5×10^{-3}	2.5×10^{-3}
L-Area reactor building, 105 reactivity/model	2.37×10^{-6}	1.04×10^{-5}
M-Area		
Alloy Building 320 diesel generator	3.6×10^{1}	1.00×10^{-1}
Canning Building 313 core recovery HNO3 etch	6.60×10^{-1}	5.60×10^{-1}
Canning Building 313 slug nitric etch	1.27	1.08
Canning Building 313 Apl station 6 post etch nitric	1.36	0
Canning Building 313 Apl station 7 post etch nitric	1.36	0
Canning Building 313 Apl station 8 post etch nitric	1.36	0
Canning Building 313 Apl etch line station 13	2.76	0
Canning Building 313 Apl etch line station 14	2.76	0
Canning Building 313 Apl etch line station 15	1.36	0
Alloy Building 320 Nitric acid tank	2.19	5.20×10^{-1}
N-Area Central Shops burn pit, open wood burning pit	2.46	2.46
T-Area		
Pilot Plant Building geometrically favorable dissolver	0	2.7×10^{-1}
Pilot Plant Building incinerator	2.83×10^{-1}	3.55×10^{-4}
DWPF Semiworks Building process tank	8.10	1.40×10^{-1}
Glass Melter Building stack	Blank	1.90×10^{-1}
1987		
F-Area canyon stack, Bldg 291 second uranium cycle	4.93×10^{1}	4.93×10^{1}
F-Area canyon stack, Bldg 291 solvent recovery	2.5×10^{-3}	2.5×10^{-3}
Glove boxes, Naval Fuels F-Area Building 247	9.90×10^{1}	9.90×10^{1}
H-Area hood metallography. 232 manufacturing building	9.40×10^{-3}	1.65×10^{-3}
L-Area reactor building, 105 reactivity/model	2.37×10^{-6}	1.04×10^{-5}
M-Area		
Alloy Building 320 diesel generator	3.6×10^{1}	1.00×10^{-1}
Canning Building 313 core recovery HNO3 etch	6.60×10^{-1}	3.00×10^{-1}
Canning Building 313 slug nitric etch	1.27	5.90×10^{-1}
Canning Building 313 Apl station 6 post etch nitric	1.36	3.50×10^{-2}
Canning Building 313 Apl station 7 post etch nitric	1.36	1.36
Canning Building 313 Apl station 8 post etch nitric	1.36	3.50×10^{-2}
Canning Building 313 Apl etch line station 13	2.76	7.10×10^{-2}
Canning Building 313 Apl etch line station 14	2.76	7.10×10^{-2}
Canning Building 313 Apl etch line station 15	1.36	3.50×10^{-2}
Alloy Building 320 nitric acid tank	2.19	3.40×10^{-1}
T-Area		
Pilot Plant Building geometrically favorable dissolver	0	0
Pilot Plant Building incinerator	2.83×10^{-1}	3.19×10^{-3}
DWPF Semiworks Building process tank	8.10	1.40×10^{-1}
Glass Melter Building stack	Blank	1.17

Table 17-55. Process Releases of Oxides of Nitrogen in the AIRS Database

Year and source description	Maximum (ton y^{-1})	Actual[a] (ton y^{-1})
Building 682		
Manufacturing building precipitate reactor tank	3.70×10^{-2}	0
Manufacturing building organic evaporator tank	3.70×10^{-2}	0
1990		
F-Area canyon stack, Bldg 291 second uranium cycle	4.93×10^{1}	4.00×10^{1}
F-Area canyon stack, Bldg 291 solvent recovery	2.5×10^{-3}	0
Glove boxes, Naval Fuels F-Area Building 247	9.90×10^{1}	9.90×10^{1}
H-Area hood metallography. 232 Manufacturing Building	9.40×10^{-3}	1.65×10^{-3}
M-Area		
Alloy Building 320 diesel generator	3.6×10^{1}	1.00×10^{-1}
Canning Building 313 core recovery HNO3 etch	6.60×10^{-1}	0
Canning Building 313 slug nitric etch	1.27	0
Canning Building 313 Apl station 6 post etch nitric	1.36	0
Canning Building 313 Apl station 7 post etch nitric	1.36	0
Canning Building 313 Apl station 8 post etch nitric	1.36	0
Canning Building 313 Apl etch line station 13	2.76	0
Canning Building 313 Apl etch line station 14	2.76	0
Canning Building 313 Apl etch line station 15	1.36	0
Alloy Building 320 nitric acid tank	2.19	1.60×10^{-1}
N-Area Central Shops burn pit, open wood burning pit	2.46	2.46
DWPF Semiworks		
IDMS feed preparation	1.60	1.40×10^{-1}
IDMS melter and off-gas system	1.40	2.50×10^{-1}
Manufacturing Building 682 precipitate reactor tank	3.70×10^{-2}	3.46×10^{-3}
Manufacturing Building 682 organic evaporator tank	3.70×10^{-2}	3.46×10^{-3}

[a] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Many of the emissions estimates for the generators were higher than the estimates for production processes listed above. The highest process emission in 1985 was the second uranium cycle discharge to the 291-F stack at 49 ton y^{-1}. Emissions estimates were not given for the H stack or any H-Canyon process except the diesel generators for the canyon exhaust fan house and in the HB-Line. This is because different contractors were responsible for reporting emissions from F-Area, and although the processes are similar, emissions estimates were often different. In this case, H-Area canyon process emissions may not have been submitted. Estimates for key sources were compiled from AIRS database excerpts for 1985, 1987, and 1995 and are presented in Table 17-56.

Table 17-56. Nitrogen Dioxide Emission Estimates for Key Sources from AIRS Database

Emission source	Year	Actual[b] emission estimate (ton y^{-1})
A-Area boiler house	1985	177.
	1987	219.
	1995	121.
D-Area powerhouse	1985	886.
H-Area powerhouse boiler 1	1985	128.
	1987	64.5
	1995	15.6
H-Area powerhouse boiler 2	1985	63.9
	1987	63.9
	1995	14.7
K-Area powerhouse	1985	256
	1987	354
K-Area package boiler	1995	1.87
Central Shops burning pit	1985	2.46
	1995	3.64
F-Area canyon stack	1985	49.3
	1987	48.6
	1995	0.00000199
H-Canyon exhaust fan	1985	0.507
	1987	0.507
	1995	1.35
H-Canyon HB-Line	1985	1.04
	1987	1.04
	1995	0.367
H-Canyon HB-Line (another emission point)	1985	0.0299
	1987	0.0299
	1995	0.305
H-Area manufacturing building	1987	0.00165
	1995	0.000293
M-Area canning building	1985	1.64
	1987	2.5
M-Area alloy building	1985	0.520
	1987	0.340
T-Area Pilot Plant building	1985	0.000761
	1987	0.000756

[a] Source: Faugl (1996k).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

M-Area Releases of Oxides of Nitrogen

Large quantities of nitric acid were used in M-Area. Although several reports estimate the amount of nitric acid and nitrates released to the M-Area process sewers in liquid effluent, nitrogen dioxide releases from the stack because of the nitric acid processes were not estimated until the air emissions inventory was taken in the late 1980s. The primary releases to the air from processes in building at M-Area were nitric acid (mist) and oxides of nitrogen, primarily from the nitric acid etching and cleaning (Du Pont 1971b). Air exhaust was subjected to an acid scrubber, which may have been put into place in the mid-1970s. A Du Pont report from 1973 stated that these emissions were recently reduced to one-tenth of their former quantity through process changes (Du Pont 1973b).

Nitric acid was used for etching in 321-M and 313-M processes. The DOE Environmental Survey (DOE 1987) said that nitric acid and nitrogen oxide from the 321-M cleaning room were estimated to be about 8 lb h^{-1} based on a stack emission test. They also said that nitric acid and nitrogen oxide emissions from the 313-M plating line were uncontrolled until 1987, when a water scrubber was to be used in the new plating line. The emissions from etching and core recovery processes in 313-M were controlled by the water scrubber and were estimated to be 14 lb h^{-1} maximum based on stack tests (DOE 1987). The scrubber efficiency was thought to be about 50%, but it was not accounted for in the 1983 estimates (DOE 1987). The stack emission testing was attributed to Clayton Environmental in 1983. We found and contacted a company called Clayton Environmental. Current employees thought the company previously had an office in Atlanta, but their library in Michigan did not contain copies of any studies conducted for the SRS in 1983. M-Area and Environmental Protection Department personnel did not recall or have copies of these testing reports. The actual maximum emissions estimates, including scrubber controls were 7.94 lb h^{-1} for 321-M and 5.57 lb h^{-1} for 313-M. Average emission estimates were 2.6 lb h^{-1} and 3.0 lb h^{-1}. The nitrogen oxides emissions from 300-Area were said to have been low compared to those from separations or the powerhouses.

M-Area emissions estimates in the AIRS database included 2 ton from M-Area canning and 0.34 ton from the M-Area alloy building in 1987. Oxide of nitrogen emissions were calculated for 17 nitric acid process tanks in M-Area. A stack test of the Building 321-M cleaning line detected nitrogen dioxide at 4.0 ppm, which Radian (1992a) thought corresponded to about 0.81 lb hr^{-1}. Nitrogen oxides were evolved from the nitric acid etch tank during the reduction of nitric acid or from the oxidation of the metal parts. Emissions from other tanks were scaled to match the cleaning line tank based on the amount of nitric acid used in the tank for which the stack test was done. The total mass of nitric acid was used to account for variations in nitric acid concentrations and tank volumes. The cleaning line operated at the highest temperatures of any process, so the estimates were thought to be worst case (Radian 1992a). The actual emission estimates for nitrogen oxides from the worksheets for 17 emission points in M-Area were summed and are shown in Table 17-57. The maximum design emission estimates for 1985–1990 totaled 20.97 ton y^{-1}.

Table 17-57. Actual Emission Estimates for Nitrogen Oxides in M-Area from the Air Emissions Inventory Worksheets[a]

Year	Actual[b] emission estimate (ton y^{-1})
1985	5.64
1986	6.02
1987	3.86
1988	1.81
1989	0.194

[a] Source: (Radian 1992a).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

The largest emission was from the nitric acid etch tank, followed by the core recovery nitric etch, both of the 313-M old plating line (Radian 1992a).

In 1967, an air sampling program for nitrogen oxides was initiated in M-Area. Nitrogen dioxide was continuously monitored over an 18-day period, and the average concentration was 0.06 ppm. The location where the samples were taken was not indicated, and it was not clear whether the samples were from the stack or ambient air (Du Pont 1967). The annual ambient air quality standard is currently 0.053 ppm.

F-Area and H-Area Release of Oxides of Nitrogen

Both dissolving and denitration processes in the separations areas produced visible emissions of nitrogen oxides. Concentrations of nitrogen oxides released from the dissolver were sufficient to produce a visible yellowish-brown plume from the stack. Emissions from the dissolvers were not continuous, but they peaked during dissolution of each charge to the dissolver.

The dissolver off-gas was set up to be exhausted through a water scrubber column, but materiel could be steam-jetted to the stack if the dissolver overflowed. The dissolver could overflow if dissolving proceeded at a greater rate than recovery of the nitric acid. When this happened, the off-gas was routed to the stack and a brown plume of nitrogen dioxide could be seen. Everett Sheldon (Sheldon 1996) estimated that perhaps 20% of the time the off-gas was diverted directly to the stack.

Reinig et al. (1973) studied the emissions and reported that dilution was sufficient to keep air concentrations far below standards except during extreme inversions. Compliance with air quality standards was generally achieved by using opacity meters or visual opacity readings.

A reviewer at CDC noted that many of the equations used to explain the nitric acid and nitrogen dioxide emission estimates submitted for the Operating Permit Application were not balanced. We are reluctant to modify the equations for fear this might mislead the reader because the equations, as written, were used to calculate the emissions estimates. The stoichiometry appeared to be correct for the relevant products.

H-Area Releases of Nitrogen Oxides

The 221-H uranium aluminum separations used the HM process to recover irradiated uranium (^{235}U) from aluminum clad-uranium targets using chemical dissolving in nitric acid, chemical precipitation of silica and other impurities, and concentration of metals (called the head end treatment), which was followed by two-cycle extraction of the dissolved uranium. Nitrogen oxide was generated during dissolution. Dissolving was aided by a mercuric nitrate catalyst ($Hg[NO_3]_2$). The reactions result in the evolution of nitrogen, oxides of nitrogen, nitrous oxide (N_2O), and traces of hydrogen:

$$Al + 3.75HNO_3 \rightarrow Al(NO_3)_3 + 0.225NO + 0.15N_2O + 0.11\ N_2 + 1.9H_2$$
$$U + 4HNO_3 \rightarrow UO_2(NO_3)_2 + NO_2 + 2H_2O$$

The potential amount of N_2O formed and lost to the air per pound of aluminum discharged to the dissolver, calculated using a stoichiometric mass balance, was approximately 0.2447 lb of N_2O per pound of aluminum. All N_2O formed was assumed to have been vented to the 291-H stack. The off-gas condenser was thought to convert some of the nitrogen oxide to nitric acid, which condensed and was recycled to the dissolver. The conversion of nitrogen oxide to nitric acid was probably variable and would have been affected by the reaction rate, the temperature of the off-gas, presence of oxidation catalysts, residence time, and other parameters. Because the conversion was very uncertain and the amount converted was unknown, a conservative approach would be to assume that all of the NO formed was vented to the process vessel vent system. A stoichiometric mass balance based on reaction above and the 1983 technical manual data, predicted that 0.25 lb of NO was lost to the exhaust per pound of aluminum for aluminum dissolution and 0.025 lb of NO was formed per pound of uranium from uranium dissolution.

An example calculation provided by Radian (1993) for the air emissions inventory assumed 251.52 kg of aluminum and 42.24 kg of uranium per charge (Mark 22 target class), a molarity of 3.31 mole L^{-1} for HNO_3 and 0.0125 mole L^{-1} for $Hg(NO_3)_2$ L^{-1}, a total volume of 14,000 L in the dissolver, and a total dissolving time of 22 hours. Using these values and the relationships described above, they calculated 135.69 lb of N_2O were emitted per batch or 6.17 lb h^{-1}. If one batch were dissolved each day, the rate might be 5.65 lb h^{-1} for each 24-hour period, or 135.69 lb d^{-1}. Using an estimate of 50,084 kg of aluminum, they calculated that 326,859 gal of raw metal solution was put through the first cycle uranium extraction in 1985. The most annual separation activity over the period of the emission inventory occurred in 1985, so these 1985 24-hour emissions rates were used as the 'design' rates for reporting. In 1985, 164 dissolver batches were processed in H-Canyon (Radian 1993).

For the permit application, H-Area dissolver emissions were calculated using maximum fuel quantities charged per year. Of the three dissolvers, 6.4D was the largest and was used to provide the maximum emissions. The actual emissions estimates were based on 20 dissolver runs per month of fuel containing 370 kg per batch of aluminum and 50.6 kg per batch ^{235}U with a dissolving time of 20 hours per batch. The maximum case was obtained from a one-time dissolver run of an offsite fuel containing 546 kg of aluminum and 50.6 kg of uranium per batch. The aluminum content was estimated from interviews with H-Area engineers. They were conservative estimates predicted to overestimate the amount of nitrogen oxides released. The maximum dissolver capacity and 24-hour a day operation was assumed. It was assumed that all of the oxides of nitrogen were nitrogen dioxide and that the condenser that condensed nitric acid vapor and

returned it to the dissolver did not convert any of the nitrogen dioxide evolved to nitric acid. There is excess oxygen in the process, so it is reasonable to assume that all of the NO was converted to NO_2. From process stoichiometry, it was calculated that 0.244 lb of N_2O was evolved per pound of aluminum charged, 0.392 lb of NO_2 was evolved per pound of aluminum charged, plus 0.474 lb of NO_2 per pound of uranium charged. This results in actual, worst case, uncontrolled emission of 44.66 ton y^{-1} for NO_2 and 23.9 ton y^{-1} for N_2O, assuming 20 hours per batch dissolving time. The maximum uncontrolled emission assuming 8760 hours of continuous operation was 76.5 ton y^{-1} for NO_2 and 42.9 ton y^{-1} for N_2O (Westinghouse 1996c).

Calculations for the dissolution of Rocky Flats scrub alloy, which contains plutonium, was found to be less than the uranium fuels. Therefore, all calculations were performed for the MK16B, considered to be the worst or bounding case. Radian (1993) estimated controlled emissions based on controls by three devices between the dissolver and the 291-H stack: the condenser, iodine reactor, and the fiberglass filter. The oxides of nitrogen that were formed in the dissolver and were not condensed were discharged through the stack. The efficiency of the condenser for converting nitrogen oxide to nitric acid was thought to be about 20%; therefore, about 80% of the oxides of nitrogen generated in the dissolver was released. The condenser was excluded as a pollution control device, but it was considered as a part of the dissolver process. For the conservative estimates in the air permit application, controlled emissions were said to equal uncontrolled emissions.

The head end process involves a permanganate strike according to the reaction:

$$Mn(NO_3)_2 + 2\ KMnO_4 + 2\ H_2O \rightarrow 5\ MnO_2 + KNO_3 + HNO_3$$

the centrifuge cake dissolution proceeds according to

$$MnO_2 + NaNO_2 + 2\ HNO_3 \rightarrow Mn(NO_3)_2 + NaNO_3 + H_2O$$

None of the products of these reactions was said to contribute to the air emissions inventory. Evolution of nitrogen dioxide at a maximum rate of about 10 lb y^{-1} was estimated from the addition of sodium nitrite to nitric acid in the gelatin strike tank. Compared to 44 ton y^{-1} from the dissolver, this amount seems negligible (Radian 1993). Interestingly, the permit application conservatively assumed that 5% of the dissolver off-gas was due to the head end process; therefore, 0.05×44.66 ton = 2.23 ton y^{-1} of oxides of nitrogen that was attributed to the head end. The Title V operating permit calculations led to actual emission estimates for the dissolver of 44.6 ton y^{-1} of nitrogen dioxide (Westinghouse 1996c). A weekly progress letter from October of 1959 said that analytical data were being collected on buildup of nitric acid in 291-H stack catch tank. The acid was formed by absorption of nitrogen oxides in condensing moisture in the stack plenum, which drains to the catch tank. The acid in the tank contributed about 24 gal of waste volume per day. It was hoped that operation of an acid absorber might reduce the amount of acid and decrease the waste volume (Martens et al. 1959). The dissolver off-gases from dissolution were being processed through the acid absorber by 1959, but another report suggests that before 1959, the dissolver off-gases were discharged directly to the stack (Du Pont 1959b). The acid buildup in the stack catch tank was compared with buildup when the dissolver off-gases were discharged directly to the stack. The data obtained indicated that the buildup in the stack catch tank was approximately 850 lb of 12% nitric acid per day, or 100 lb of 100% nitric acid per day

(Martens et al. 1959). This report suggests that about 100 lb d^{-1} or about 18 ton y^{-1} of nitric acid could have been removed from the air exhaust by the acid absorber.

The frame waste recovery system in H-Area, which purified and concentrated ^{238}Pu solutions, used nitric acid. The maximum pounds per run was reported in 1984. The 1984 throughput was used to determine emissions estimates for the Operating Permit application. Based on stoichiometry, throughputs, column feed rates, maximum operating temperatures allowed for worker safety concerns, a release estimate of 17.2 ton of nitrous oxide, and 0.39 ton of nitrogen dioxide and maximum emissions were estimated, using the 1984 throughputs (Westinghouse 1996c).

A conservative estimate of the nitrogen dioxide emissions from the H-Canyon rerun process, based on the amount of ferrous sulfamate used in the frames waste recovery process (a maximum of 334.3 ton y^{-1}), was calculated to be 20.6 ton y^{-1}. This upper bound limit assumes maximum processing of 24 hours per day and that all the NO formed was converted to nitrogen dioxide (Westinghouse 1996a).

The H-Canyon high and low activity waste processes that result in discharges to the canyon stack were also considered for the operating permit application. These processes included evaporation and waste neutralization. Nitrogen dioxide was evolved from sodium nitrite added to the LAW (Low Activity Waste) and HAW (High Activity Waste) evaporator feed tanks to eliminate the ferrous sulfamate. The H-Area LAW evaporators, combined were estimated to emit a maximum of 6.32 ton y^{-1}, and the HAW evaporators emission estimates totaled 2.64 ton y^{-1} for nitrogen dioxide (Westinghouse 1996c). The H-Canyon general purpose evaporator was not thought to have emitted nitric acid or nitrogen dioxide.

The emissions estimate for the 200-H emergency power generator, actual emissions assuming 2080 hours of operation each year, was 1.47×10^{-1} ton y^{-1} for oxides of nitrogen (Westinghouse 1996a).

The HB-Line converted aqueous neptunium and plutonium solutions from the canyon processes into dry oxide powders through ion exchange removal, acid oxidation, and dehydration and crystallization in a calcining furnace. In 1993, Radian estimated the amount of nitrogen dioxide emitted per pound of neptunium processed in 1985. The exact process chemistry was not available to Radian in the unclassified documentation, but a probable reaction stoichiometry was assumed, and solution volumes and throughputs were used to make the calculations. Actual annual emissions were estimated to be 0.0472 ton y^{-1} of nitrogen dioxide (Radian 1993).

The permit application for the H-Canyon from 1995 reported maximum controlled emissions for oxides of nitrogen to be 89.7 ton y^{-1}.

F-Area Releases of Nitrogen Oxides

The F-Canyon cycles, solvent recovery system, waste, acid recovery, evaporators and other process equipment were similar to those in H-Area. The recycle vessel vent system, process vessel vent system venting exhausted through the sand filters then the stack were the same as described for H-Area.

The overall profile of nitrogen oxide emissions for the F-Area separations process main stack (291-F) was evaluated in 1983. The approximate ratio of emissions of nitrogen oxide from the dissolution and denitration parts of the process was said to have been 8:1. Nitric acid fumes from both processes were recovered in the acid recovery unit and recycled as 50% nitric acid. The

nitrogen oxide emissions from 291-F were subject to a State permit limit of 180 lb h^{-1} annual average. Releases were estimated using mass balance calculations and facility production schedules. A stack test was conducted in 1983 to look at increased nitrogen dioxide from a proposed process modification. The data suggested that without the acid recovery unit, maximum emissions during the dissolution process were up to twice the hourly limit of 180 lb h^{-1}, but the average emissions for the entire process cycle appeared to be below the annual emissions limit. The efficiency of the replacement acid recovery column installed at this time was estimated to be twice that of the old column, so nitrogen oxide emissions were expected to be well within permit limits. The permit conditions did not require monitoring of nitrogen oxide emissions. The uranium denitrators were equipped with evacuators that bypass the acid recovery unit and exhaust directly to the sand filter and stack during a process emergency. The facility was suppose to report the use of the evacuators for more than 6 minutes to the State because of a concern about exceedance of the opacity limitation (DOE 1987). The tests and reports about them were cited in the DOE (1987) survey, but the reports referenced for the tests, attributed to Roberts in 1984 and Clayton Environmental in 1983, were not found by *RAC*. We also searched local phone directories and asked directory assistance for Augusta, Georgia, Aiken, South Carolina, and Atlanta, Georgia, areas for the consulting firms said to have been involved in testing in 1983, and could not find any reference to Clayton Environmental. Robert and Company of Atlanta may have done the monitoring but current personnel could not locate any reports or any employees with knowledge of monitoring done at the SRS. No documentation about these tests has been found. The permit conditions apparently do not require monitoring of nitrogen oxide emissions and no recent emissions data are available.

F-Area had two dissolvers of the same size: 6.1D and 6.4D. Very rarely did they operate at the same time, and emissions calculations for the permit assumed one dissolver operated. The maximum nitrogen dioxide emissions were thought to have been associated with processing of Mark 31A and B fuels. A dissolver run of 15 metric ton of uranium Mark31A contained about 856 lb of aluminum cladding, and 15 metric ton of Mark 31B fuel contained 794 lb of aluminum. The maximum dissolver capacity was 27 buckets or 18 metric ton of uranium targets. Decladding generally took 6–8 hours and dissolving was done for 20 to 22 hours. Minimum time of 26 hours per run was used as a conservative estimate. Maximum emissions were calculated assuming 18 metric ton of uranium per run at 26 hours per run for 8760 hours per year. However, the historical data reported that the highest amount of targets dissolved in a year was in 1976, which totaled 1352.8 metric ton, less than 25% of the theoretical maximum used (8760/26 × 18 or 6064 metric ton). Processing of different fuel, like the Rocky Flats scrub alloy, was considered and different assumptions were applied (for example, the plutonium containing targets do not go through the decladding step). As with H-Area, the F-Area condenser was not assumed to convert nitrogen oxide to nitric acid; it was only assumed to condense nitric acid vapors and return them to the dissolver.

Aluminum-clad uranium fuel assemblies were loaded in to the dissolver and declad by dissolving in sodium hydroxide followed by the dissolving of uranium metal in 50% nitric acid. Gaseous emissions included ammonia and oxides of nitrogen. The stoichiometry is such that 1 mol of uranium yields 2.42 of nitrogen dioxide. The maximum evolution of nitrogen dioxide was predicted to be 9.2 ton per run at maximum dissolver capacity. The actual uncontrolled emissions estimate based on 1944 metric ton of uranium each year was 993.6 ton y^{-1}. The actual uncontrolled emissions for the Rocky Flats scrub alloy fuel was 8.33 ton y^{-1}. Because this was

much less, in amount per time than the SRS fuel, the Rocky Flats fuel was not considered further, and the conservative assumption that all fuel was like the maximum fuel was used in subsequent calculations (Westinghouse 1996a).

The head end process permanganate strike and centrifuge cake dissolution forms nitrates and manganese compounds, none of which were thought to contribute to air emissions. Ruthenium particles and oxides of nitrogen from foaming during slurry dissolution in the strike tank were noted. Engineers estimated that the nitrogen oxides released from the head end process would not exceed 5% of the dissolver emissions, and the estimates reported in the tables of the air permit were simply 5% of the dissolver emissions or $0.05 \times 993.6 = 49.7$ ton y^{-1} actual uncontrolled emissions of nitrogen dioxide from the head end process.

Denitration was done at the F-Canyon A-Line. The facility converted uranium nitrate solutions to uranium trioxide powder. Uranium solution was concentrated using continuous then hydrate evaporators. The A-Line process vessels and the hydrate evaporators were vented through the recycle vessel vent system fiberglass filters then sand filters to the stack. The off-gas from the denitration pots was discharged from the stack after flowing through a series of scrubbers, coolers, and a nitrogen oxides absorption column (the same column used for the dissolver off-gas) (Westinghouse 1996a). The A-line denitration step converted uranyl nitrate to uranium trioxide by thermal decomposition, which evolved nitrogen dioxide. The A-Line nitrogen oxide removal system, also called the F-8 column, removed water soluble nitrogen dioxide from the off-gas. The F-8 column was the most effective control device in the process, with an efficiency of 45–70% reported by the vendor who sold the unit to the SRS. The air quality permit application reduced the emission estimate for nitrogen dioxide by 45% or $993.6\ (1 - 0.45) = 556.5$ ton y^{-1} for the dissolver and 5.0 ton y^{-1} from the head end to estimate actual controlled nitrogen dioxide emissions. How the emissions estimate for the head end was calculated to be 5 ton y^{-1} based on this reduction is not clear. A 45% reduction of 49.7 ton y^{-1} should result in the release of 27 ton y^{-1} (which equals 5% of 556.5 ton y^{-1}); however, 5 ton y^{-1} was the estimate reported for the F-Area head end process in the Operating Permit Application.

Based on stoichiometry of producing uranium trioxide from uranyl nitrate hexahydrate, 692.5 ton of nitrogen dioxide per year was the controlled maximum emissions estimate calculated for the denitrator facility. This assumed two denitrator pots were operating simultaneously and the F-8 column had an efficiency of 45%. The A-line evaporation and purification and hydrate evaporation systems were calculated to have released no nitrogen dioxide or nitric acid to the air.

Nitrogen dioxide was also created by adding sodium nitrate to the evaporator feed tanks, a part of the low activity and high activity waste handling systems. Maximum uncontrolled emissions were estimated to be 13.72 ton y^{-1} from the high activity waste and 10.81 ton y^{-1} from the low activity waste, for a total of 24.53 ton y^{-1}. No actual emissions were given (Westinghouse 1996a).

FB-Line, in Building 221-F made plutonium buttons from a dilute nitrate solution. FB-Line tanks and vessels were enclosed in cabinets or glove boxes. The line included four major processes and four vacuum venting systems. The vessel vent vacuum system was preceded by Teflon filters and used a venturi scrubber to scrub the off-gas, which contained nitrous oxides and nitric acid. This system used a caustic scrubber to neutralize the nitric acid vapors. Actual emissions of nitrogen oxides from the FB-Line were reported to be zero in 1996, but the maximum controlled emission estimates were 1.39 ton y^{-1} in the permit application.

A 1982, environmental assessment for the Naval Reactors Fuel Materials Facility estimated that the emission rate of nitrogen oxides to the air would be 0.1 ton y^{-1} (DOE 1982). Emissions for the Naval Fuels Manufacturing Facility were included in the AIRS database (Faugl 1996e) and are shown in Table 17-54.

F-Area emissions of nitric acid and nitrogen dioxide estimated in the AIRS database in 1994 and 1995 were reviewed. Most of the actual emissions were estimated as zero, indicating the facility was not operating. However, maximum design capacity estimates were given for 59 emission points, including 0.052 ton y^{-1} nitric acid, 42.9 ton y^{-1} nitrous oxide, and 76.5 ton y^{-1} nitrogen dioxide for the dissolver off-gas and 3.83 ton y^{-1} for the head end. The rest of the emission points for the canyon processes were totaled, including the first and second uranium cycle and all the vessel vents and other exhaust points that comprise the 50 sources that contributed to the canyon stack emissions. The maximum design capacity-controlled emissions for nitric acid totaled 9.89 ton y^{-1}. Actual emissions were listed for 11 sources, which were tanks and other equipment, and they totaled 0.76 ton y^{-1}. The maximum emissions for nitrogen dioxide totaled 13.5 ton y^{-1}, and the actual emissions totaled 0.24 ton y^{-1}. The maximum design capacity emissions for nitrous oxide totaled 2.9 ton y^{-1} and the actual emissions were estimated at 0.395 ton y^{-1} (Faugl 1996e).

In summary, the uncontrolled actual emissions for nitrogen dioxide from F-Canyon included 994 ton y^{-1} from the dissolvers and 49.68 ton y^{-1} from the head end process. Controlled actual emissions were estimated to be 557 ton y^{-1} from the dissolvers and 5.0 ton y^{-1} from the head end process. The total emissions for the individual sources described in the permit application were 73–90 ton. At the time of the 1996 permit, the F-Canyon was not operating and actual emissions were reported as zero. The combined F-Canyon stack maximum controlled emission estimates, assuming 24 hour a day releases of oxides of nitrogen, totaled 2530 ton y^{-1} based on engineering calculations (Westinghouse 1996a). By far, this estimate from the permit application, was the largest emission estimate given in any of the documents reviewed.

Nitrogen Dioxide from the Power Plants

From 1972 to 1985, estimates of nitrogen oxide emissions from the coal burning power plants, reported in the annual reports, were said to have been within standards. Emissions estimates were not given. After 1977, control of total suspended particulates and oxides of nitrogen was assessed using opacity meters in the powerhouse stacks. The 1986 annual report is the first to mention opacity standards, which were set by SCDHEC at 40%. The 1986 annual report states," the day-to-day control of total suspended particulates and oxides of nitrogen is maintained with the use of opacity meters in all of the powerhouse stacks. These measurements indicated that the SRP boilers were within limits greater than 99% of the time in 1986." Opacity was limited to 40% for stacks in existence before January 1, 1986. Stacks put in use after that date were to be held to a standard of 20% opacity. The 1986 environmental report also says that, "all facilities were within applicable federal and state standards except for occasional high opacity results from the F-Area separations process stack. The acid absorber column for the F-Area separations process stack had deteriorated and required renovation in 1986. After renovation was complete, the opacity requirement was met" (Ziegler et al. 1987). Oxides of nitrogen from the power plants were reported to have been within applicable standards in 1987 and 1988 (Mikol et al. 1988; Davis et al. 1989).

The 1989 environmental report mentions that the 291-F stack occasionally exceeded the opacity standard and renovations of the acid absorption column were underway to ensure compliance in the future. However, all of the coal burning power plant stacks were said to have been in compliance.

The 1992 annual report estimated emissions of nitrogen dioxide to be 3767 ton from the coal-fired boilers (which burned 243,627 ton of coal that year [corresponding to an emission factor of 30.9 lb ton^{-1}]; 17.2 ton from the fuel-oil fired boilers; and 142.6 ton from diesel equipment. These estimates were said to have been based on AP-42 calculations and opacity monitoring (Arnett et al. 1993).

K-Area coal consumption and emissions estimates of uncontrolled nitrogen dioxide emissions for 1985–1990 were included in the air emissions inventory and are summarized in Table 17-58.

Table 17-58. Estimates of the Coal Consumed and Uncontrolled Emissions of Nitrogen Dioxide Each Year for K-Area[a]

Year	Tons of coal consumed	Tons of nitrogen dioxide per year
1985	40886	255.5
1986	52394	327.5
1987	56638	354
1988	56449	352.8
1989	18717	117
1990	8064	50.4

[a] Calculated by Radian (1992b) for the air emissions inventory.

Nitrogen dioxide emissions from the two boilers in K-Area were not controlled. In 1992, nitrogen dioxide emissions were derived graphically from source test data compiled from KVB, Inc. in support of EPA research on the formation of nitrogen dioxide. A nitrogen content of 1.20%, which was the average content reported for 1988, was used for the emissions estimates. Handwritten calculations described stoichiometric oxygen requirements and said that the nitrogen dioxide emissions were calculated using a computer program written for this purpose. The calculation was said to use the 95% confidence level of the KVB Inc. data and to assume a worst case, 3% excess oxygen scenario. The resulting uncontrolled emissions estimate was 12.5 lb nitrogen dioxide per ton of coal burned. Radian (1992b) stated that the EPA's AP-42 value for NOx was 7.5 lb ton^{-1} of coal burned (Radian 1992b).

The two coal-fired stoker boilers in A-Area had two opacity monitors: one in the No. 2 boiler exhaust duct and one in the stack. However, these monitors were not used for compliance purposes. The units were exempted from continuos opacity monitoring because they produced less than 250 million BTUs. Analyses and calculations of the K-Area boilers suggest that 117.5 ton of nitrogen dioxide was produced from burning 17,000 ton of coal (Westinghouse 1996a) or 13.8 lb of nitrogen dioxide per ton of coal burned (Westinghouse 1996a).

Generally, site-specific data or vendor analysis-based emission factors are preferred over literature values. If we assume, based on the reported emissions, that the emissions factor for nitrogen dioxide released from coal burned at the SRS ranged from 12.5 to 31 lb ton^{-1}, the estimates of emissions from coal burning in Table 17-59 could be made.

Table 17-59. Estimates of the Nitrogen Dioxide Released Based on the Amount of Coal Burned Each Year

Year	Estimate of coal burned each year	Range of nitrogen dioxide emissions predicted (ton y^{-1})
1954–1972	≤500,000	3125–7750
1972–1982	500,000	3125–7750
1983	400,000	2500–6200
1984	400,000	2500–6200
1985	455,000	2845–7053
1986	439,700	2750–6815
1987	453,000	2831–7021
1988	373,935	2337–5795
1989	227,017	1418–3518

Other Nitrogen Dioxide Emissions

The AIRS database contained emissions estimates for oxides of nitrogen from emergency diesel generators, diesel compressors, welders, diesel engines, welders, and other equipment. Most of the diesel generator and engine emissions were estimated to be less than 1 ton y^{-1}, but the contribution from all of these sources was significant. In 1985, sources included 58 emergency diesel generators, 2 portable gasoline generators, 2 diesel engines, 55 diesel and gasoline generators, a diesel compressor, 9 diesel pumps, 1 welder, and 1 lead melting pot. In 1990, emissions were given for 76 emergency diesel generators, 3 portable gasoline generators, 3 engines, 67 diesel and gasoline generators, 1 diesel compressor, 11 diesel pumps, 1 welder, and 1 lead melting pot. In 1992, 117 welders were listed. This was probably because of the search criteria, which may have excluded welders from the printout in the earlier years. The totals reflect all of these sources. The lead melter used No. 2 fuel oil and also released notable amounts of nitrogen oxides (about 0.47 ton y^{-1}) (Faugl 1996k).

Facilities associated with the vitrification processes, operated in the 1990s, are not within the scope of this study. An example of one of these processes with relatively large releases is the F-Area rerun process. It involved processes of denitration, oxalate precipitation, oxalate kill, dissolution of the precipitate in nitric acid, steam stripping, formic acid denitration, and concentration using an evaporator. The nitric acid going through this process is used at several different concentrations, volumes, and temperatures. The emissions were considered to be uncontrolled because fiberglass and sand filters of the process vessel vent system do not remove nitrogen dioxide or nitric acid. The maximum emissions were estimated to be 784 ton y^{-1} of nitrogen dioxide and 0.039 ton y^{-1} of nitric acid in 1995 (Westinghouse 1996a).

Summary of Release Estimates for Nitrogen Dioxide

The actual emissions estimates in the AIRS database for 1987 totaled 469 ton. This included most of the key emissions from F-Area, H-Area, M-Area, and T-Area and relatively conservative estimates of releases from generators, engines, welders, and other equipment. The 1992 annual report suggested diesel equipment produced 143 ton y^{-1} (Arnett et al. 1993). The AIRS database

estimates did not include the D-Area coal burning. We might add 1418–7750 ton y^{-1} to the estimate to account for coal burning in 1987. As much as 70 ton y^{-1} of nitrogen dioxide may have been released from open burning operations (described near the end of this chapter). The permit application suggested releases of 6–21 ton y^{-1} for M-Area, 73–90 ton y^{-1} for H-Area, and 588–1068 ton y^{-1} for F-Area. A stand alone section of the permit applications also provided a very high total of 2530 ton y^{-1} for F-Area. Taken together, the range of estimates spans from (a) 2086 to 10,534 ton y^{-1} if the high of the 2530 ton y^{-1} permit application estimate for F-Area is used as a maximum or (b) 2086 to 9072 ton y^{-1} if an estimate of 1068 ton y^{-1} is used for the maximum for F-Area. For some operations, these ranges apply to releases in the 1985 to 1987 time frame. This is especially important for considering pollution control equipment that may not have been in place in earlier years. However, actual releases for all time periods, including the mid-1970s when the maximum amount of coal was burned and the canyon production was high, should fall within the range that was determined by adding together many maximum emission estimates. This represents more of a worst case than a best case scenario. A release of 2086–10534 ton y^{-1} for the 35 years between 1954 and 1989 totals 73,010–368,690 ton of nitrogen dioxide.

Uncertainty was calculated for these releases using the estimates from the AIRS database, permit applications, and engineering calculations, and coal releases. Although the very large estimate for releases from F-Area is not expected to approximate a true value, doing an uncertainty calculation allowed us to include it on the tail of the F-Area release distribution. The distribution of total oxides of nitrogen released per year was lognormal with a geometric mean of 6050 ton y^{-1} and a GSD=1.23. This distribution represents a range of releases from 4320–8480 ton y^{-1} of nitrogen dioxide. Using uncertainty calculations to estimate releases decreased the total range of possible values somewhat. Since the total range of estimated releases in the preceding paragraph is assumed to represent a worst case type scenario as a result of some very large point estimates, this distribution of values narrows the potential release estimates into what are probably more realistic estimates.

The Standard 8 results submitted to SCDHEC said that in 1991, 295 sources from all areas at the SRS released nitrogen dioxide. The estimated maximum annual average Site boundary concentration was calculated to be 125.4 $\mu g\ m^{-3}$. The ambient air standard was 100 $\mu g\ m^{-3}$. This suggested that if all operations were operating at maximum, nitrogen dioxide emissions could exceed standards at the Site boundary. The Environmental Protection Department staff submitting the report to SCDHEC felt that the exceedances were due to extremely conservative approaches being used in the modeling analysis and they planned to remodel for the standards using more accurate operating data and system configurations. They also said they planned to develop a control strategy plan for the D-Area Powerhouses (Dukes 1993).

Several accidents involving nitric acid spills and explosions have been reported. Two 'red-oil' explosions have occurred at the plant. These are described briefly, but well, in Durant (1978). In both cases, operators did not realize that tributyl phosphate was present in the uranyl nitrate solutions being processed. In January 1953, an evaporator in Building 678-G at TNX exploded during concentration of a uranyl nitrate and nitric acid solution. Tributyl phosphate and AMSCO diluent were present in the evaporator charge, and the explosion was a result of an exothermic reaction between tributyl phosphate and the nitrate or nitric acid or both. An experimental program was conducted after the explosion to learn more about how violent reactions may have been produced. The explosion destroyed the evaporator, ripping the pot into six pieces, and damaged the roof and siding of the building. Two minor injuries were said to have been

sustained. The operator present at the time of the explosion later said he saw orange colored fumes evolving from the top of the column and heard rumbling and roaring. The total capacity of the evaporator was 1300 gal in the shell and pot plus 525 gal in the column. No mention of chemical releases because of this incident was made in the report (Colven et al 1953). A later summary of the incident in Durant (1978) estimated that 1800 gal of solution was being evaporated at the time of the explosion, but that the deacidification was carried out in several batches of about 500 gal each. Three batches had been successfully processed. The fourth and final charge had contained the 70 gal heel of the original solution plus 160 gal of previously evaporated material diluted with water.

The other accident occurred 22 years later, on February 12, 1975, in an A-Line facility denitrator (calciner), which was used to convert uranyl nitrate solution to uranium trioxide powder. The A-Line incident involved fires from the ignition of gases that were evolved from the denitrator. Damage to the building required 6 months to repair (Durant 1978). The incident report does not specifically address releases of chemical or radioactive materials to the air or surface water.

Sulfur Dioxide

Sulfur dioxide was formed by combustion processes. Releases from the powerhouses, lead melting pots, incinerators, and engines all contributed sulfur dioxide to the air.

The AIRS database search for emissions of hydrogen sulfide, sulfuric acid, and sulfur dioxide were run together as sulfur compounds. The largest source of sulfur dioxide, the D-Area powerhouse stacks, were not included. Sulfuric acid emissions were listed for battery storage, sulfuric acid tanks, neutralization tanks, acid tanks, and wash tanks. Some of the larger sulfur dioxide emissions were given for the Central Shops Burn Pit, with maximum and actual emissions estimates of 0.55 ton y^{-1}, and the lead melting pot, with an actual emission estimate of 2.3×10^{-3} ton y^{-1}. In 1987, the Naval fuels glove boxes were listed with an actual and maximum emissions estimate of 1.6 ton y^{-1} (Faugl 1996n). Several fire-training pits were listed after 1990 with actual emissions of 2.1×10^{-3} ton y^{-1}. The AIRS database releases for oxides of sulfur were compiled for 1985, 1987, and 1990 and are shown in Table 17-60. Total process emissions of sulfur dioxide, sulfuric acid, and hydrogen sulfide are shown in Table 17-61.

Table 17-60. Sulfur dioxide emissions from the AIRS Database by Source[a]

Emission source	Year	Actual[b] emission estimate (ton y^{-1})
A-Area boiler house	1985	304
	1987	400
	1995	673
D-Area Powerhouse	1985	2530
H-Area Powerhouse Boiler 1	1985	306
	1987	306
	1995	86.6
H-Area Boiler House 2	1995	81.5
K-Area Powerhouse	1985	769
	1987	1060
K-Area Package Boiler	1995	6.73
Central Shops Burning Pit	1985	0.550
	1995	0.258
H-Canyon exhaust fan	1985	0.044
	1987	0.044
	1995	0.022
H-Canyon HB-Line	1985	0.010
	1987	0.010
	1995	0.00597
H-Canyon HB-Line (another emission point	1985	0.0299
	1987	0.0299
	1995	0.00401

[a] Source: Faugl (1996n).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

Table 17-61. Total Sulfur Compound Emissions Estimates Reported in the AIRS Database[a]

Total sulfur compounds	Maximum (ton y^{-1})	Actual[b] (ton y^{-1})
1985	4.34	4.05
1987	5.36	5.11
1990	6.0	5.66
1994	114.0	19.9

[a] Source: Faugl (1996n).
[b] Actual emissions are those predicted under typical operating capacities and times. Maximum design capacity emissions are calculated using maximum throughputs and capacities, assuming 24 hours per day and 365 days per year operating times.

The operating permit application included emissions of sulfur dioxide for the large generators. For example, the emissions estimate for the 200-H emergency power generator (actual

emissions assuming 2080 hours of operation each year) was 2.93×10^{-3} ton y^{-1} for oxides of sulfur (Westinghouse 1996a). The lead melter described in the permit application used No. 2 fuel oil and also released notable amounts of sulfur dioxide (7.44 ton y^{-1}) (Westinghouse 1996a).

The HB-Line converted aqueous neptunium and plutonium solutions from the canyon processes into dry oxide powders through ion exchange removal, acid oxidation, and dehydration and crystallization in a calcining furnace. In 1993, Radian estimated the maximum amount of sulfur dioxide emitted per pound of neptunium processed in 1985. The exact process chemistry was not available to Radian in the unclassified documentation, but a probable reaction stoichiometry was assumed and solution volumes and throughputs were used to make the calculations. Actual annual emissions were estimated to be 0.0167 ton y^{-1} of sulfur dioxide (Radian 1993).

Sulfur dioxide was formed from the burning of hydrogen sulfide discharged to the flare tower from the heavy water facility in D-Area (Rusche 1973). How much was never reported, but emissions were have said to have been very small compared to D-Area powerhouse emissions.

Sulfur dioxide emission from M-Area, H-Area, and F-Area were due to combustion engines, furnaces, and maintenance equipment and were not the result of sulfur compounds used in the processes. A document describing the design of the Fuel Production Facility in F-Area described potential sulfur dioxide releases from the calcining process. Uranium was to be loaded onto sulfur-bearing ion exchange resin then the resin was calcined to produce U_3O_8. Much of the sulfur would have exhausted out the stack. The maximum load, worst case emissions were calculated to be about 12,403 kg y^{-1} or about 13.7 ton. Average and actual emissions would be much less than this because the facility did not intend to operate at maximum load and it was predicted that about one-half of the sulfur in the calciner exhaust may be carbonyl sulfide rather than sulfur dioxide (Allender 1985). No other reference to this production facility was found. Site personnel in the Environmental Protection Department could not recall any operations of this type in separations or the Naval Fuel Facility. It may be that the facility was designed but not built nor operated.

In 1967, an 18-day air sampling program was conducted in M-Area; it measured an average concentration of 0.02 ppm sulfur dioxide in air. The purpose of the sampling, why sulfur dioxide was a concern, and where the samples were taken are not indicated (Du Pont 1967). The National Ambient Air Quality Standard is currently 0.03 ppm annual average concentration and 0.14 ppm over 24 hours.

Sulfur Dioxide Emissions from Power Plants

According to the DOE Environmental Survey, the sulfur dioxide emission limit of 3.5 lb/10^6 BTU heat input was met in the 1980s by the use of low-sulfur coal, and no pollution control equipment for sulfur dioxide was ever applied to any of the boilers (DOE 1987). It seems likely that in the 1950s and 1960s some of the coal purchased had higher sulfur content, and emissions would have been greater than they were in the more recent past and currently. Higher sulfur content coal may have been somewhat balanced by fewer tons of total coal burned in earlier years.

At some point after about 1970, the specifications required a sulfur content of less than 2% (Garvin 1996). The pulverized coal used at 400-D was of lower quality than the stoker coal that was crushed and blown into the boilers (Smith 1996). Sulfur contents reported in the annual reports also suggest that stoker coal was lower in sulfur than pulverized coal (Du Pont 1983).

The amount of coal burned before 1972 is unknown. The annual environmental reports have summarized the status of power plants onsite and reported the amount of coal burned and the approximate sulfur content of the coal. The reports gave estimates of the emissions of sulfur dioxide, nitrogen dioxide, and fly ash particulates in the early and mid-1970s. Beginning in 1972, each year the reports gave estimates of emissions of carbon monoxide and volatile organic compounds (VOCs). Attempts to locate more primary data on sulfur analysis of coal and coal receipt or storage information were not successful. Records of this type were not found in the Phase I database search. Essential materials ledgers for D-Area and the other powerhouses were not located. In 1996, Tom Thome, Stan Smith, and H.S. (Sid) Willis, who worked onsite in power engineering or previously for the Power Department; Robert Garvin and Mal Schroeder, who worked in D-Area; and retired workers Henry Main, Ray Fleming, Leo Shelton, Peter Gray, WB Holt, and Jim King were interviewed about power plant operations, essential materials records, coal inventory and analysis records, and potential releases of chemicals to the environment from the D-Area power plant and other power plants.

The Bureau of Mines did much of the coal analysis in earlier years. The regional office recommended we contact the office in Washington, D.C. Neither office had an idea where we might find records of coal analysis for the SRS.

Sample data sheets from 1987 on the coal analysis were included in materials submitted to supplement the K-Area powerhouse air emissions inventory. The sheets were from Mineral Laboratories, Inc. in Salyersville, Kentucky, and indicated that analysis had been done for Air Techniques, Inc. in Marietta, Georgia. The laboratories indicated that any analysis records they had generated would have been kept for 7 years or less. They had no recommendations for where to find coal analysis records before 1985 (Radian 1992b).

Compliance with the sulfur dioxide emissions standard was determined by estimating an annual emission based on the analysis of the sulfur content of the coal received and the amount of coal burned. This information was reported in the annual reports after 1972. From 1972 to 1975, sulfur dioxide emissions were said to have been within standards. In 1975, eight coal-fired plants were reported to have burned about 500,000 ton of coal that year, with an average sulfur content of 0.9% (Du Pont 1977a). In 1977, sulfur dioxide emissions ranged from 1.63 to 2.12 lb/10^6 BTU, with a weighted annual average of 1.87 lb/10^6 BTU heat input for all power plants (said to be within the emissions standards). In 1978 and 1979, seven coal-fired plants burned about 500,000 ton of coal each year, with an average sulfur content of 1.3% (Du Pont 1979). The emission rate for sulfur dioxide was given as a weighted annual average with a range from 1.55 to 2.50 lb/10^6 BTU and an average of 2.0 lb/10^6 BTU heat input. The South Carolina Emission Standard was 3.5 lb/10^6 BTU heat input in 1978. In 1980, 1981, and 1982, the same volume of coal was reported to have been burned, but the sulfur content was 1.4% (Du Pont 1981, 1983). The sulfur dioxide emissions were broken down into an emission rate for the four pulverized coal boilers, based on a sulfur content of 1.56%, of 2.17 lb/10^6 BTU and for the 15 stoker coal boilers, based on a sulfur content of 0.9%, of 1.30 lb/10^6 BTU (Du Pont 1983). In 1983 and 1984, seven coal-fired plants that burned about 460,000 ton of coal each year, with an average sulfur content of 1.06 and 1.07%, were reported to be operating (Du Pont 1984; Zeigler et al. 1985). In 1985, five coal-fired plants burned a total of 455,000 ton of coal a year with an average sulfur content of 1.05% (Ziegler et al. 1986). In 1986, five coal-fired plants that burned about 439,700 ton of coal each year were described in greater detail and are summarized in Table 17-62.

The sulfur content of the coal burned in 1986 averaged 1.01%, which yielded an average of 1.6 lb of sulfur dioxide per 10^6 BTU input. The sulfur dioxide standards were still 3.5 lb/10^6 BTU heat input in 1986. The 1986 annual environmental report said that, "the content of coal delivered to the site for burning is determined by analyses for sulfur, carbon, ash, water and BTU output." In 1987, five coal-fired plants burned 452,980 ton of coal that averaged 1% sulfur and yielded an average of 1.73 lb of sulfur dioxide per 10^6 BTU input, which was 49% of the South Carolina standard (Mikol et al. 1988). In 1988, the coal plants burned 373,935 ton of coal with an average sulfur content of 1.1%, which yielded an estimate of average sulfur dioxide emissions at 1.77 lb/10^6 BTU input (Davis et al. 1989). Three coal plants burned 227,017 ton of coal in 1989, with an average sulfur content of 2.6%, which resulted in a release estimate of 2.0 lb of sulfur dioxide/10^6 BTU input (Cummins et al. 1990). Three coal-powered plants burned 243,627 ton of coal in 1992, resulting in 7133 ton of sulfur dioxide, assuming a sulfur content of 1.06%. The D-Area power plant boilers also burned 30,922 gal of used oil for energy recovery and 4247 gal of propane for boiler startup. In 1992, the SRS had four package steam generating boilers (three in K-Area and one in P-Area) fueled by No. 2 diesel fuel. These boilers burned 1,718,764 gal of fuel oil in 1992. About 7133 ton of sulfur dioxide was estimated to have been released from the coal-fired boilers, 35.4 ton from the fuel-oil fired boilers, and 9.5 ton from diesel equipment in 1992 (Arnett et al. 1993). Generally, the calculated average emission values in the annual reports seem to be less than or equal to 2.0 lb of SO_2/10^6 BTU heat input.

Table 17-62. Power Plant Location, Number of Boilers, and Capacity[a]

Power plant location	Number of boilers	Boiler capacity 10^6 BTU h^{-1} input
A- Administration Area	2	71.7
D- Powerhouse Area	4	396.0
H-Separations Area	3	71.7
K- Reactor Area	2	194.5
P- Reactor Area	2	194.5
Total	13	928.4

[a] Source: Ziegler et al. (1986)

The coal analysis reported for 1992 suggested that the sulfur content ranged between 0.71–1.82%, averaging 1.06%. If we assume the range of sulfur content varied similarly in other years, then this could be used to approximate an uncertainty range. Radian (1992b) reported that the sulfur content of the coal delivered from 1985 to 1990 varied from 0.81% to 1.24% and averaged 0.97%.

Although the information about capacity is useful for helping to understand the size of the power plants, it is not very useful for estimating releases of pollutants. We know that the boilers did not operate at capacity. Three of four or two of three boilers were usually in operation at one time.

The AP-42 value for sulfur dioxide from coal is 37.8 lb ton^{-1} (EPA 1988). The 784-A two coal-fired stoker boilers have produced steam for A-Area heating since 1953. The capacity of each boiler was 72 million BTU h^{-1}. Particulate emissions were collected in multiple cyclone separators. The units burned about 8500 ton of coal per year each. The air quality permit application estimated that the two boilers released a total of 652 ton of sulfur oxides

(Westinghouse 1996a), which corresponds to an emissions estimate of 77 lb ton^{-1}. In 1992, uncontrolled sulfur dioxide emissions from the two boilers in 184-K were calculated for the air emissions inventory using monthly coal delivery data from 1988 and assuming 97% of the sulfur was converted to sulfur dioxide. The sulfur content of the coal in the 1980s and 1990 varied from 0.81% to 1.24% and averaged 0.97%. The uncontrolled emission estimate for sulfur dioxide from the boilers was 37.6 lb ton^{-1} of coal burned (Radian 1992b). K-Area coal consumption and emissions estimates of sulfur dioxide emissions, which were not controlled for 1985–1990, are summarized in Table 17-63.

Table 17-63. Estimates of the Coal Consumed and Uncontrolled Emissions of Sulfur Dioxide for 1985–1990 for K-Area Boilers[a]

Year	Tons of coal consumed	Sulfur dioxide emissions estimates (ton y^{-1})
1985	40886	769.4
1986	52394	986
1987	56638	1065.8
1988	56449	1062.2
1989	18717	352.2
1990	8064	151.7

[a] Calculated by Radian (1992b) for the air emissions inventory.

The 1992 annual report stated that 7133 ton of sulfur dioxide had been released from burning 243,627 ton of coal (Arnett et al. 1993). This corresponds to an emissions factor of 58.5 lb ton^{-1}. The emissions factors calculated at various time periods for Site coal seem to range from 37 to 77 lb ton^{-1}. The emission factor can also be calculated by converting the emissions reported in the annual reports in terms of lb/10^6 BTU heat input to pounds per ton. The resulting values are shown in the fifth column of Table 17-64.

The AIRS database provided only limited information for uncertainty calculations, but the ranges in the total sulfur released per ton of coal burned, the largest releases of sulfur from the site, allowed the uncertainty in sulfur releases from coal burning to be reasonable well quantified.

The AP-42 value for sulfur released from coal burning of 37.8 lb ton^{-1} is close to the site estimated value derived from the sulfur content in coal during the 1980-90s of 37.6 lb ton^{-1}. From annual reports, from 1977 through 1992, the average of the reported sulfur released per ton of coal burned is 43.5 lb ton^{-1}. In the air quality permit, sulfur releases were estimated at 77 lb ton^{-1}. Because the values from the annual reports are probably the most accurate estimates of sulfur releases for the Site, the average of these values (43.5 lb ton^{-1}) was used as the most likely estimate in a triangular distribution, with 37.6 as the lower limit and 77 as the upper limit. Estimates of releases of sulfur compounds from open burning, as described in the next section, were also added into the release estimate.

The distribution of annual releases of sulfur compounds was lognormally distributed with a geometric mean of 11000 ton y^{-1} and a GSD=1.18. The 5th and 95th percentile values of the distribution were 8470 and 14400 ton y^{-1}, respectively.

The Standard 8 results submitted to SCDHEC said that in 1991, sulfur dioxide was emitted from 282 sources in A-Area, B-Area, C-Area, D-Area, E-Area, F-Area, G-Area, H-Area, K-Area,

L-Area, M-Area, N-Area, P-Area, S-Area, T-Area, and Z-Area. The estimated maximum 3-hour average Site boundary concentration was calculated to be 2319 μg m^{-3}. The corresponding ambient air standard was 1300 μg m^{-3}.

Table 17-64. Coal Consumption, Average Sulfur Content, and Calculated Sulfur Dioxide Releases for 1952–1989

Year	Pounds sulfur dioxide/10^6 BTU heat input	Approximate amount of coal burned in tons[a]	% Sulfur content reported in annual reports	Pounds sulfur dioxide released per ton of coal burned [b]	Central estimate of the range of sulfur dioxide releases (ton y^{-1})
1952–1971	-	<500,000	-	-	<9500
1972	<3.5	500,000	-	-	9500
1973	<3.5	500,000	-	-	9500
1974	<3.5	500,000	-	-	9500
1975	-	500,000	0.9	-	9500
1976	-	500,000	-	-	9500
1977	1.8	500,000	-	45	11250
1978	2.0	500,000	1.3	50	12500
1979	2.0	500,000	1.3	50	12500
1980	1.5	500,000	1.4	37.5	9375
1981	1.5	500,000	1.4	37.5	9375
1982	1.5	500,000	1.4	37.5	9375
1983	1.5	400,000	1.06	37.5	9375
1984	1.6	400,000	1.07	40	8000
1985	1.5	455,000	1.05	37.5	9375
1986	1.6	439,700	1.01	40	8794
1987	1.73	453,000	1.0	43.2	9785
1988	1.77	373,935	1.1	44.2	8264
1989	2.0	227,017	2.6	50	5675
1992		243,627	1.06	58.5	7133

[a] Taken from the Site annual environmental reports

[b] Calculated using 38 lb ton^{-1} emission factor for 1952–1971. A value of 2.5×10^7 BTU ton^{-1} was used for the conversion from lb/10^6 BTU heat input. Vendor analysis in the 1990s suggests that the coal averages about 12,180 BTU/lb or 2.43×10^7BTU/ton (Faugl 1996a). Vendor analysis in the late 1980s suggests 2.6×10^7 BTU/ton (Westinghouse 1996a).

Emissions from Burning

Incineration, open burning, and solvent burning released particulates, sulfur dioxide, oxides of nitrogen, metals, and other chemicals in the air. Emissions of sulfur dioxide, fly ash, and oxides of nitrogen from coal burning are characterized in the sections above. The releases to

surface water from runoff from the coal piles, ash piles, and ash retention basins are addressed in Chapter 18.

The most significant combustion source of concern is the coal-fired powerhouses, the D-Area powerhouse being by far the largest source. The releases to surface water from runoff from the coal piles, ash piles, and ash retention basins are addressed in Chapter 18. Other fuel-burning facilities and equipment have included the beta-gamma radioactive waste incinerator in the 200-H Area, the propane-fired uranium oxide calciners in the 200-F Area A-Line, CMX Pilot Plant test incinerator, and more than 100 diesel generators throughout the facilities. The diesel-powered generators are periodically tested or used for emergency or standby power when electrical power is interrupted. Emissions estimated for diesel-powered generators are included in the Site-wide air emissions inventory and many were included in the AIRS database totals.

Incinerators

Incinerators onsite are a concern because of the potential release of toxic metals and compounds like dioxins. HEPA filters are very effective for controlling particulate and dioxin emissions. Fortunately, because the incinerators were designed to burn radioactive waste, they have been equipped with HEPA filters.

The Consolidated Incineration Facility, in 261-H, began cold runs in 1996 and started operations in March 1997. It consists of a rotary kiln incinerator, a fixed secondary combustion chamber and supporting solid and liquid feed, off-gas treatment, and ash and blowdown solidification systems. It is equipped with several scrubbers and HEPA filters The predicted emissions from this facility are described in the operating permit application submitted in 1996. The ash and blowdown was to be stabilized with Portland cement before disposal. The facility has a 150-ft main process stack and a 42-ft tank farm stack. The off-gas is subjected to coolers, scrubbers, and HEPA filters (Westinghouse 1996a).

The Beta/Gamma Incinerator in Building 230-H is a dual-chambered controlled air incinerator that burns liquid and solid waste, including spent solvent from the canyons, waste oil, rubber, polyvinyl chlorides, and other plastics. Waste oils are suppose to be certified by the generator as free of radioactive materials and toxic components, including lead, chlorinated hydrocarbons, and carcinogenic solvents. Because of the radioactivity of the waste, the exhaust is subject to stringent particulate controls. Pollution control devices include a water quench, baghouse, and HEPA filters (DOE 1987). This incinerator has released hydrochloric acid, nitrogen oxides, and sulfur oxides. Emissions from the incinerator are tested twice a year to comply with State permit requirements. The permit limits emissions of HCL to 30 lb h^{-1}, which was reported to have been easily met because of the limited amount of chlorinated waste and plastics burned. Sulfur dioxide and nitrogen dioxide emissions were said to have been low relative to the powerhouse emissions (DOE 1987; Faugl 1996a).

Interestingly, this facility was not operating during the DOE Environmental Survey conducted in 1986 because of an accident involving the introduction of Halon (a fire extinguisher) into the combustion air (DOE 1987). In December 1986, fumes containing hydrogen fluoride, hydrogen bromide, and bromine were released form the incinerator stack during testing of the Halon 1301 fire suppression system (Jewell 1990). The fumes were detected by construction workers working nearby. This incident demonstrated to the survey team that the incinerator could influence local air quality, and the survey team expected that inadvertent

burning of large amounts of chlorinated plastics could result in increased hydrochloric acid emissions and ground-level concentrations during stable meteorological conditions. They concluded that, "during normal operations with permitted limits the regulated/hazardous emissions from the facility are not significant with respect to other air emissions sources at the site" (DOE 1987).

A test incinerator located at TNX, called the SHIRCO incinerator, was built to help design larger scale incinerators. The incinerator throughput was small and emissions were controlled by a metal filter and three banks of HEPA filters (DOE 1987).

Fuel Burning

The D-Area boilers also burned oil. Waste oil from the reactor areas generated at the powerhouses and from diesel engines used as backup for primary reactor cooling water pumps was disposed of by burning in the 400-D Area powerhouse (DOE 1987). In the 1980s, the 484-D plant was said to have burned about 100,000 lb y^{-1} nonradioactive waste oils, including engine lubricants, kerosene, diesel, and fuel oils. Oils containing lead, chlorinated compounds, cutting oils, or other toxic components were not supposed to be burned. Generators were required to certify that the oil was free of these unsuitable materials before the waste oil could be received. The plant required that the generator provide a certification form that verified the contents of the waste (DOE 1987). Waste oils burned at the 400-D power plant were reported to have been spot checked for polychlorinated biphenyl (PCB) content. The oils were supposed to contain less than 50 ppm PCBs to be accepted for burning (DOE 1987). In 1992, the D-Area power plant boilers burned 30,922 gal of used oil for energy recovery and 4247 gal of propane for boiler startup. In 1992, the SRS also used four package steam generating boilers fueled by No. 2 diesel fuel. These boilers burned 1,718,764 gal of fuel oil in 1992 (Arnett et al. 1993). Releases of sulfur dioxide and oxides of nitrogen from these boilers are included in the AIRS database.

Open Burning

Open burning at the Site included (a) open pan burning of waste solvent at the burial grounds, (b) burning of wood and other waste at the burning and rubble pits, (c) burning of wood and tree slash by the U.S. Forest Service, (d) fire training exercises, (e) burning of scrap lumber, (f) clearing and dead brush disposal, and (g) controlled burning of underbrush by the U.S. Forest Service. Emissions from these activities were determined for the air emissions inventory using EPA emission factors.

Open burning was also addressed in the Part 70 Operating Permit Application. Estimates were calculated by making assumptions about the mass of wood burned in scrap piles, forest, and underbrush. Emission factors chosen for sulfur oxides were 0.4 lb ton^{-1} of scrap lumber and 0.8 lb $acre^{-1}$ for underbrush. Emission factors for nitrogen oxides were estimated to be 2.6 lb ton^{-1} of scrap lumber and 10 lb $acre^{-1}$ for underbrush. For 1994, the Site estimated that 52 fire training exercises were conducted each year, 2800 ton of lumber was burned, and 20,000 acres per year of forest were cleared. U.S. Forest Service-controlled burns in 1994 were also included. Using the EPA AP-42 emissions factors and the estimates of the mass of material burned described above, emissions of oxides of nitrogen were estimated to be 69.9 ton y^{-1} and emissions for oxides of sulfur were estimated to be 5.6 ton y^{-1}. A great deal of uncertainty is associated with

the estimates, but the uncertainty was not quantified. These values are probably conservative. We do not know how burning in 1994 compared with burning the past. Theoretically, 70 ton y^{-1} of nitrogen dioxide and 6 ton y^{-1} of sulfur dioxide could be added to the totals to account for open burning.

Burning and Rubble Pits

Burning and rubble pits were located in Central Shops and in D-Area, F-Area, K-Area, P-Area, R-Area, and L-Area (Westinghouse 1992). Very little documentation on the operation of the burning and rubble pits was found in Phase I. Most of the useful records were characterizations of waste areas prepared recently. The DOE Environmental Survey report (DOE 1987) contained a good summary of what the survey team found out about the pits by reviewing aerial photographs. According to DOE (1987), burnable solid and liquid waste was disposed of in 15 pits between 1951 and 1973. The pits were bulldozed open trenches 8–12 ft deep and 250–400 ft long. The waste was burned only about once a month; therefore, contamination of groundwater beneath these pits has been a concern. In 1973, the pits were no longer used for disposal of chemicals; they were used only as rubble pits until they were filled and covered with soil. Rubble included metal, concrete, lumber, railroad ties, telephone poles, glass, paper, and other solid waste. Most of the pits are located in flat areas away from Site streams, except the P-Area burning and rubble pit, which is about 100 ft from Steel Creek. Steel Creek is in a gorge 75 ft below ground-level, and seeps and springs along the bank suggest that groundwater moves into the creek. A survey done in 1987 found no radioactivity or organic vapor contamination attributable to the pits. No records of pit disposal before 1989 were kept. Estimates were made and published in DOE (1987) using groundwater modeling (PATHRAE model) said to have been done by Du Pont of measured levels of contaminants versus background levels. Table 17-65 summarizes the calculated amounts of some of the chemicals of concern that may have been disposed of in the pits.

Another report prepared for a Comprehensive Environmental Response, Compensation, and Liability Act investigation said that chlorinated solvents and metals (including mercury, zinc, cadmium, nickel, lead, and chromium) have been found in the soil of many of the pits, and solvents, chromium, lead, and manganese have been found in groundwater near many of the pits (Westinghouse 1992).

Determining air emissions from burning in these pits is not possible without more information on what, and how much was burned. The lead disposed of was probably in solid sheeting or other shielding material and may not have been released into the air as a result of burning. Because monitoring indicates groundwater contaminated by these pits is not migrating offsite, the pits were not evaluated further.

Table 17-65. Estimates of Chemicals Disposed of in Rubble Burning Pits[a]

Chemical	Pit	Estimated amount disposed of (kg)
Trichloroethylene	A-Area	1.2
	C-Area	186
	F-Area	7.4
	K-Area	1.0
	P-Area	3.4
Lead	C-Area	5530
	D-Area	204
	P-Area	3300
Chromium	C-Area	9500
Tetrachloroethylene	D-Area	0.1
	F-Area	7.6
	K-Area	3.1
	Central Shops	0.64
	L-Area	0.51
	R-Area	0.37
	P-Area	3.5

[a] Source: DOE (1987). Estimates of the amount of chemicals disposed of in burning rubble pits calculated using groundwater models.

Solvent Use and Solvent Burning

Degraded solvent (tributyl phosphate in kerosene) from the separations areas was stored at the burial ground. After allowing the short-lived fission product activity in the solvent to decay for several years, most of the solvent was burned. Most of the records about solvent burning were concerned with release of radioactivity and burial of radioactive material remaining in the burning pan as residue. Releases of large quantities of black smoke were reported, and slow burning generating less smoke was deemed necessary. A 1971 report stated that, “efforts to burn solvent at suitable rates without smoke generation in burners or incinerators have been unsuccessful because of entrained radioactivity in the combustion gases.” However, the release of radionuclides from solvent burning was said to have been negligible (Ice 1971).

Many of the monthly progress reports between 1956 and 1972 contain a section on the burial ground, which often reported the quantity of solvent transferred to the burial ground and the amount burned. Solvent was burned in an 8 × 8 × 4-ft steel pan with a rain shed over its top (Ice 1971). The pan corroded and was replaced in 1964 by a longitudinally halved 400-gal tank (Du Pont 1974b).

Several monthly reports referred to a data record that was being prepared that was to present a history of the storage and disposal of spent extraction solvent. The record was said to detail the accumulation of the present inventory along with “a history of the tanks in which it is stored and the burning process by which it is destroyed.” No such data record was located. A brief report on radioactive waste management at the Site first published in 1969, then issued as a revised document in 1972, said that 400,000 gal of solvent had been burned since February 1956. This amount probably represents burning over about a 16-year period (1956–1971). The text indicated

that the quantities stored and destroyed were shown in an attached figure, but the figure was not included in the copy that was found (Ice 1971).

Information on degraded solvent burning reported in many of the monthly progress reports was summarized as follows. In 1956, a burning rate of 80 lb d^{-1} in F-Area was reported. A total of 26,000 gal was burned and 55,000 gal remained to be disposed of at the end of the year. Airborne contamination was said to have been negligible (Du Pont 1974b). The removal of degraded solvent from the rerun facilities and transportation to the burial ground was mentioned in the 1956 monthly report because of problems from high activities in the unshielded truck and transport areas. During the month of May 1956, 830 gal of solvent was burned in 72 hours of operation at the average burning rate of 0.2 gal min^{-1}. The waste solvent inventory in the burial ground was said to be 70,000 gal (DuPont1956c). The June report states that routine burning continued and that the burning rate was increased from 50 to 250 gal d^{-1} by changing equipment. About 3500 gal was burned that month (June 1956), and a total of 5000 gal (35,000 lb) was reported to have been burned from January to June 1956 - (Du Pont 1956d). The July report indicated that the burning rate was increased to about 90 gal h^{-1}, and about 5300 gal was burned in July (Du Pont 1956b). The solvent being burned was said to have been in the burial tanks for about 1 year and was filtered through underground charcoal beds to reduce radioactivity. A May 1956 report (Du Pont 1956c) refers to solvent having been sent to the rerun station where it was stripped and washed before being sent to the burial ground for burning. A monthly report from 1957 stated that the total amount of solvent burned in 1956 was 31,700 gal (Du Pont 1957b). Some of the disagreement in values reported in different reports probably results from errors in reporting amounts sent to the burial grounds and amounts stored at the canyons or burial ground when compared to amounts actually burned.

In 1957, burning appeared to have been limited by rainy weather. A total of 12,600 gal was burned. In January 1957, a new type of burner was installed to test burning using a different fuel-to-air ratio. The burner was designed to spray the solvent at 8 gal h^{-1} into an air stream. The burner was being evaluated for smoke release and contamination of the surrounding area (Du Pont 1957b). In April 1957, the Separations Technology Section reported that 2200 gal had been burned so far that year and the inventory that needed to be burned was 59,000 gal (Du Pont 1957c).

Reports published in 1958 mention the periodic burning of waste degraded solvents, but no amounts were available. A weekly progress letter from the Separations Technology Section from December 1958 explained that solvent was being drawn off of the top of the storage tanks so that the high activity sediment was left in the tanks and not burned (Du Pont 1958).

No mention is made of solvent burning in 1959, 1960, or 1961. This probably reflects a change in the format of the reporting in the monthly reports rather than an absence of burning; however, it is unknown how much was burned during this period.

In 1962, 54,000 gal of low activity solvent was burned in the burial ground. In May 1962, 19,600 gal was burned during 1 week from May 17 through May 23. The June report explained that operating standards limited the radioactivity of the solvent that could be burned and most of the solvent in the storage tanks exceeded the limit and could not be burned (Du Pont 1962).

In 1964, as much as 7200 gal of spent solvent was burned each month during several months at the burial ground (Du Pont 1974b). Another monthly report said that from August 1955 through February 1964, 290,000 gal of spent solvent had been received at the burial ground, of which 170,000 gal had been disposed of by burning (Du Pont 1964b).

About 3000 gal of spent solvent was burned in January 1965. Other volumes reported to have been burned in 1965 were 5000 gal in February, 1800 gal in August, 9000 gal in November, and 7250 gal in December, which totaled 24,250 gal (Du Pont 1965). One separations area history reports that a total of 22,600 gal of waste solvent was burned in the burial ground in 1965 (Du Pont 1974b).

In the meeting minutes from a presentation by Albenesius, responses to questions from the audience included some interesting statements about solvent burning (Albenesius 1992). The speakers stated that 350,000 gal of solvent was disposed of by burning at the burial ground and that burning was stopped in 1972 because of air quality concerns (black smoke) rather than radiological concerns. Albenesius said, "they did burn solvent at night because people onsite were worrying about the black smoke produced." One of the presenters also said that someone mistakenly pumped much of a truckload of solvent down a well near the burial ground, mistaking it for a solvent tank. About 60 gal was pumped down the well, until the well overflowed. He also said that there were some spills associated with solvent burning and they are "they are well written up in a document produced by Elmer Wilhite" (Albenesius 1992). Several documents about the burial grounds authored by E. Wilhite were found, but we did not locate documents describing solvent burning or spills (Wilhite et al. 1989).

The monthly reports suggest that burning was not continuous and varied tremendously from month to month ranging from zero to 19,600 gal (in 1 week during May 1962). Solvent burning was discontinued in 1972. Obviously, burning rates reported for one time period cannot be applied to another time period. Taken together, the values in various reports suggest a typical solvent burning volume may have been about 25,000 gal y^{-1}, ranging from zero in later years, 12,600 gal in 1957, and 170,000 gal in 1964. A reasonable central estimate for the total amount of solvent burned over the 16 years solvents were burned is 400,000 gal or about 2.8×10^6 lb. The range that is implied by the very limited amount of monthly data available is about 320,000 to 650,000 gal or 2.2×10^6 to 4.4×10^6 lb.

Because monthly or annual values were available for 1956, 1957, 1962, 1964, and 1965 only, the total for 1956–1971 of 400,000 gal reported by Ice (1971), provides the best estimate during the 16-year period when solvents were burned. The amount burned averages to about 25,000 gal y^{-1}. Data from which to determine uncertainty are limited. The monthly estimates reported vary tremendously. More solvent was burned in the month of May 1962 than for the entire year in 1957. Ice (1971) may have had more information on which to base his estimate of 400,000 gal than is now available, but the uncertainty associated with the Ice (1971) estimate was not quantified or described.

The solvent was primarily n-parrafin, or kerosene, with tributyl phosphate. Burning of kerosene and tributyl phosphate probably resulted in emissions of carbon monoxide, carbon dioxide, water, nitrogen oxides, sulfur oxides, and trace amounts of metals. Some research on this has been done for crude oil burning in oil wells in Alaska and in Kuwait, and the EPA has established emission factors for burning of fuel oil and fuel distillates, given in lb/10^{12} BTU for metals.

The EPA AP-42 estimates that about 104 lb of total carbon compounds are released from burning 100 gal of fuel (EPA 1988). The BTU gal^{-1} value for No. 1 and No. 2 fuel oil ranges from 134,000 to 142,000. Table 17-66 summarizes the emissions factors for metals released from No. 6 fuel oil burning, the estimated releases assuming 400,000 gal of solvent was burned, and the maximum emission factors:

AP-42 lb released/10^{12} BTU × 142,000 BTU/gal × 400,000 gal burned = pounds released.

These values are conservative and very uncertain. Fuel oil is more crude than kerosene and the metal content of the solvent burned was likely to have been less than that of fuel oil. The amounts of nonradioactive materials released from solvent burning were much less than that released from the coal burning power plants. Because the combustion products of kerosene and tributylphosphate are not particularly hazardous, it seems that burning solvent in the burial ground area would not have produced sufficient quantities of criteria pollutants or metals to have resulted in concentrations of concern offsite.

Table 17-66. Emissions Estimated from the Number of Gallons Waste Solvent Burned at the SRS Burial Grounds (1956–1971)

Pollutant	AP-42 factor lb/10^{12} BTU	Estimated total release (lb)	Estimated total release (kg)
Arsenic	19–114	1–6	0.5–3
Beryllium	4.2	0.2	0.1
Cadmium	16–211	1–12	1.5–5
Chromium	21–128	1–7	5–3
Lead	28–194	2–11	1.5–5
Manganese	23–74	1–4	0.5–2
Mercury	1.4–32	0.08–2	0.04–0.8
Nickel	837–2330	47–132	22–60

Solvent releases to the air from the solvent extraction process H-Area and F-Area did not survive the screening process described in Chapter 16 and were not evaluated further, largely because the solvents are not very toxic and are less volatile than chlorinated solvents. The solvent extraction process was done in mixer-settler banks using tri-n-butyl phosphate (TBP or $[C_4H_9]_3PO_4$) in a kerosene solvent. Using the SRS safety analysis flow sheets, VOC emissions from these units were estimated by Radian using the EPA's AP-42 algorithm for organic liquid storage tanks. Solution volume and throughput data were derived from the dissolution solution feed rates in safety analysis flow sheets. Relative input and output streams for each mixer-settler unit were listed. Because temperatures in the canyon building were relatively constant, breathing losses of the tanks were assumed to be zero. The solvent feed loop was essentially a closed system. Degradation products of the extraction solvents include butanol, mono and dibutyl phosphate and butyl nitrate. Degraded solvent was replaced with fresh tributyl phosphate (TBP or $[C_4H_9]_3PO_4$) and n-paraffin. During times of greatest processing in the mid-1980s, amounts of TBP and the diluent (n-paraffin, kerosene, or n-dodecane, containing predominately 13 and 14-carbon n-paraffins, $C_{13}H_{28}$) fed to the contactors were used to calculate working losses for the solvent extraction process. The amounts totaled about 50 lb per contactor at maximum design capacity. Actual cycle times for extraction were not available, so the emission rate was calculated by averaging annual emissions over a continuous 24-hour period. This resulted in a very conservative estimate for the sum of the loss from each unit in the two cycles of 582.32 lb of total VOCs and 266 lb of TBP in 1985 (Radian 1993). This is a very small emission compared to

routine emissions from the use of gasoline and fuel and compared to the emissions of chlorinated solvents from M-Area.

The Operating Permit Application included emissions estimates from F-Area and H-Area waste tank purges for tributyl phosphate, n-paraffin, benzene, and mercury from 51 tanks (Westinghouse 1996a). Tributyl phosphate and n-paraffin do not present serious health hazards and source terms for these materials were not determined for this study.

The loss of degradation products was also calculated for the air emissions inventory. The maximum design capacity estimated totaled 3788 lb (1.9 ton) in 1985 for butanol and 6095 lb for butyl nitrate (Radian 1993). These chemicals were not included in the chemicals of concern because they are relatively nontoxic. Releases of this magnitude would not likely result in toxic concentrations at the Site boundary.

Uranium

Uranium in solution was discharged to the M-Area settling basin. Because chlorinated solvents in the waste contaminated groundwater, it is likely that uranium also could have entered the groundwater through this pathway. The DOE Environmental Survey team (DOE 1987) was concerned that the air stripper water and Liquid Effluent Treatment Facility water were potential sources of uranium contamination from M-Area. No other supporting documentation for this was found.

A series of engineering worksheets for air emissions inventory calculations done by Radian in 1992 included data on filter aid preparation in 313-M. A filter aid tank was installed in September 1985. Water previously sent to the settling basins was sent to the treatment facility after June 1985. An autoclave was used to pressure test slugs. A description of the autoclave waste treatment system said the purpose was to remove uranium dioxide solids from condensate in the autoclave collection sump. The sump water was mixed with filter aid and sent through the autoclave pressure filter, and the filtrate and the filter cake (as a slurry) were pumped to separate hold tanks. No volatile chemicals or dry solids were involved, but particulate emissions from the addition of filter aid to the top of the filter aid mix tank may have occurred. The tank has a vent with a fan exhausting to the outside of the building. The maximum, worst case emission estimates for natural uranium calculated for 313-M, DETF, CTF, and 340-M totaled 0.037 ton y^{-1} in 1985, 0.0665 ton y^{-1} in 1986, 0.0504 ton y^{-1} in 1987, 0.0212 ton y^{-1} in 1988, 0.012 ton y^{-1} in 1989, and 0.021 ton y^{-1} in 1990. More than one-half of the emissions were from the DEFT facility (Radian 1992a). Releases of uranium to the atmosphere are addressed in Chapter 4.4. Uranium releases may be best characterized by looking at uranium as a radionuclide that was routinely monitored. However, the toxicity of the uranium metal to the kidney may outweigh the carcinogenicity from radioactive decay. This should be considered in subsequent phases of the dose reconstruction study.

CONCLUSIONS

We found enough information to calculate the uncertainty associated with the release estimates for the chemicals presented in Table 17-67.

Table 17-67. Release Estimates and Uncertainty Ranges for Chemicals Released to the Air

Chemical	Median average annual release (ton y^{-1})	5th–95th percentile values on the median (ton y^{-1})
Coal ash	4200	2300–7100
Mercury	0.3	0.18–0.51
Nitrogen dioxide	6050	4320–8480
Sulfur dioxide	11000	8470–14400

Uncertainty calculations were not made for the chemicals listed in Table 17-68 because of a lack of information, but a range of releases was estimated.

Table 17-68. Range of Releases Estimates

Chemical	Range of release estimates (ton y^{-1})
Benzene	1.8–18
Lead	0.05–0.12
Manganese	0.07–1.9
Nickel	0.11–0.42
Nitric acid	30–150

Seven coal-fired steam plants have operated at the SRS. Burning coal released relatively large amounts of nitrogen dioxide and sulfur dioxide and contributed to the releases of arsenic, cadmium, chromium, lead, manganese, mercury, and nickel. After 1972, the SRS annual reports provided estimates of the amount of coal burned; the sulfur content of the coal; and emission estimates for sulfur dioxide, nitrogen dioxide, and particulates. About 500,000 tons of coal were burned each year from 1972 to the mid-1980s. We estimated the amount of coal burned in earlier years by assuming coal consumption was correlated to production by the reactors.

Not enough information is available to allow us to estimate releases of chromium from its use as a corrosion inhibitor in process and cooling water with reasonable certainty. Chromium releases from machining, construction, and maintenance operations probably ranged from 0.1–0.5 ton y^{-1} for most years. Cadmium releases from similar activities may have been about 0.19 ton y^{-1}. As much as 150 ton y^{-1} of hydrogen sulfide, much of which was released via a flare tower and was probably burned to sulfur dioxide, was released until 1982. After 1982, about 3.5 ton y^{-1} was released from the treatment of groundwater. Releases of hydrazine and emissions from the use, storage, and transport of gasoline and diesel fuels were evaluated. The release of pollutants from open burning was also described.

Almost all of the chlorinated solvents evaporated during use or after being discharged to surface water. Until 1979, waste solvent was released to the M-Area settling basin or to a stream called the Tim's Branch, where most evaporated. Trichloroethylene and tetrachloroethylene have also been released from air strippers used to remediate groundwater under M-Area. Release estimates for trichloroethylene, tetrachloroethylene, and trichloroethane are summarized in Table 17-69.

Table 17-69. Trichloroethylene, Tetrachloroethylene, and Trichloroethane Releases to the Air

Solvent	Source	Time period	Median	5th–95th% range (ton)
Trichloroethylene	M-Area use	1952–1970	1700 ton (average of 90 ton y^{-1} over 19 y)	1530–1900
	M-Area air strippers	1985–1990	70 ton (average of 12 ton y^{-1} over 6 y)	57–87
Tetrachloroethylene	M-Area use	1962–1975	4055 ton (average of 289 ton y^{-1} over 14 y)	3520–4675
	M-Area air strippers	1985–1992	30 ton (average of 3.8 ton y^{-1} over 8 y)	22–40
Trichloroethane	M-Area use	1979–1988	2200 ton (average of 220 ton y^{-1} over 10 y)	1780–2710

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CHAPTER 18

RELEASES OF CHEMICALS TO SURFACE WATER

ABSTRACT

This chapter provides information about waste disposal practices, processes, effluent, and environmental monitoring data that *Radiological Assessments Corporation (RAC)* used to characterize releases of chemicals to Savannah River Site (SRS) streams. Monitoring for chemicals was generally not conducted until the late 1980s. Fortunately, many of the processes, including the separations processes in F-Area and H-Area and the raw materials manufacturing processes in M-Area, had periods of peak production in the late 1980s when monitoring data are available. Monitoring data suggest that the releases of metals, nitrates, pesticides, and polychlorinated biphenyls (PCBs) were not detectable in the Savannah River. Concentrations measured upstream of the SRS were the same or greater than downstream concentrations or less than detection limits. Although large amounts of some chemicals were released to seepage basins and Site streams, the impacts to surface water extending beyond the Site boundary do not appear to be measurable.

INTRODUCTION

This chapter describes releases of arsenic, cadmium, chromium, coal and coal ash, gasoline, hydrogen sulfide, lead, manganese, mercury, nickel, nitrates, uranium, and zinc to surface water. It also summarizes the accidental spills to surface waters and fish kills that have been reported. Chapter 5, the Phase I Task Four 4 report addendum (Stetar et al. 1994), and several Savannah River Site (SRS) documents (including Reinig et al. 1973) contain descriptions of surface water flow and waste discharges into streams and sewers. Before about 1985, amounts were reported in pounds in historical documents. These amounts were converted to kilograms to make comparisons easier. Chapter 17 addresses releases of chlorinated solvents to surface water; because of their rapid volatility, volatilization of these compounds into the air may be more important than their concentrations in offsite surface water.

Because of the large amount of surface water and rainfall at the SRS, the U.S. Environmental Protection Agency (EPA) and state agencies have required that the SRS file a Spill Prevention Control and Countermeasures Plan. For this study, we photocopied and reviewed the Spill Prevention Control and Countermeasures Plans for 1987 and 1997, which are kept by the Environmental Protection Department. The plans listed all of the fuel, oil and solvent tanks, transformers, and transfer stations, by area. The plans contained information about tank capacity and contents and described containment features such as dikes (Graham et al. 1987; Westinghouse 1997). The total number or total capacity of the tanks has not been compiled but could be derived from these documents if releases were thought to warrant it.

Metals were released to surface streams from coal and ash and processing in M-Area, H-Area, and F-Area. The Spill Prevention Control and Countermeasures Plans characterized metals in waste effluents discharged to seepage basins and Site streams and ponds at the SRS; nitrate releases from M-Area, H-Area, and F-Area; and chlorinated solvent releases from M-Area. The Plans discuss the use of these materials, their discharge into wastewater going to the seepage

basins or to Site streams, and how the material discharged could be transported to the Savannah River.

A report justifying selecting 16 metals to be analyzed in 210 stream sediment samples was written in 1985 for a part of a Comprehensive Cooling Water Study done for the SRS. Sources of metals were thought to be (a) metals in the Savannah River water used to cool reactor and powerhouse operations, (b) 12 point source National Pollution Discharge Elimination System (NPDES) continuous flow outfalls, (c) shallow groundwater outcropping to Four Mile Creek from releases to the F-Area and H-Area seepage basins, (d) leachate from reactor heat exchanger tubes at the C-Reactor, K-Reactor, and P-Reactor, (e) and naturally occurring concentrations in sediments (Gladden et al. 1985).

Measurements of metals in Tim's Branch sediments in 1984, 1985, and 1986 were reported in Specht (1991). Concentrations in sediments at the "creek mouth" were also reported, but the location of the creek mouth is not clear. The report containing these values appeared to be a photocopy of overheads for a presentation. The values were not referenced and details about the sample collection, number of samples, the time of year they were taken, and the analysis were not included. It is likely that the concentrations presented were averages. The same data appear to have been used by Pickett (1990), who reported that in 1985 and 1986, Tim's Branch and Upper Three Runs Creek sediments were sampled monthly at six locations for metals. Eight samples were taken for each location (Pickett 1990). The source of the data for both Pickett (1990) and Specht (1990) was a study attributed to Starkel et al. (1987), with Environmental and Chemical Science, Inc. A report of this original study was not located.

The history of liquid effluent discharges in M-Area is important for characterizing releases. From 1952 through 1958, all effluents were discharged directly to Tim's Branch. It has been difficult to determine when the M-Area settling basin was first put into operation and what percentage of the liquid effluent discharged from M-Area went to the basin or was discharged directly to Tim's Branch. The discharge history of M-Area reported by Specht (1991) indicated that all effluents were discharged to Tim's Branch from 1952 to 1973 and that the settling basin was not completed until 1973. Pickett (1990) said that 313-M was not diverted to the basin until 1973. Other documents suggest that a seepage basin was put into use in 1958 (Christensen and Brendall 1981). Because many of the documents that describe the history of the discharges are concerned with uranium releases and uranium releases first went to the seepage basin from 313-M in 1973, the seepage basin was assumed to be used first in 1973 for these historical analyses. However, based on the history of uranium releases from M-Area and the history provided by Specht (1991) and Pickett (1990), we believe that effluents containing most of the solvents and metals were being discharged to the basin before 1973. In their discussion of the solvents released to the M-Area sewers, Christensen and Brendall (1981) indicate that solvents had been released to sewers leading to the seepage basins since 1958. Except for 7 months in 1982 when all discharges were diverted from Tim's Branch, some effluent was discharged to Tim's Branch until the effluent treatment plant began operating in 1985. Certainly, more effluent was diverted to the seepage basin with time. For reconstructing releases, we can assume that all chemicals in the effluent were released to Tim's Branch from 1952–1958. Some portion, perhaps one-half, went to Tim's Branch from 1958 to 1973, and less than 40% went to Tim's Branch from 1973 to 1982. After 1982, process water with fewer chemicals went to the stream, and after 1985, all effluent was diverted to the treatment facility.

All M-Area liquid effluents were probably released to Tim's Branch from 1952–1958, before the settling basin was used. Measured uranium releases were highest from 1964 to 1970 (Chapter 5), but this was thought to have been the result of process changes, including overflow from a waste holding tank and inadvertent loss of etching solution. We expect that the releases of other metals would not have occurred in the same pattern as uranium.

Upperbound estimates of the amounts of waste discharged to Tim's Branch include 1.35×10^8 gal y^{-1} in 1982 before diversion in May to the settling basin (Gordon 1982; Colven et al. 1985); 2.6×10^8 gal y^{-1} average, ranging from 1.0×10^8 to 3.6×10^8 gal y^{-1} in 1970 (Monier 1970); and 2.6×10^8 gal y^{-1} in 1970 (Hardt 1970).

Effluent monitoring for many of the chemicals of concern was reported in Merz (1982). If we assume that concentrations of metals in effluent were the same in 1982 as in previous years, we can estimate a total release for each metal for which Merz (1982) reported a concentration. Merz (1982) was concerned with process sewer discharges and resulting groundwater contamination, especially from solvents. He estimated that 32,000 lb of heavy metals were contained in the settling basin and only chlorinated organics had penetrated the settling basin to enter the groundwater. The metal concentrations in a 24-hour composite sample of sewer effluent going to Tim's Branch and to the settling basin, dated December 1981, were given in a table in Merz (1982). The source of the data is not referenced and we assume it was collected by Merz. Information about how the samples were collected and the analytical methods used are not included in the memorandum. If we assume average discharges to Tim's Branch were 2.6×10^8 gal y^{-1} or 9.84×10^8 L y^{-1} (Monier 1970; Hardt 1970) the milligram per liter concentration of metal in the effluent reported by Merz can be used to estimate the total kilograms discharged each year or the total kilograms discharged from 1952 to 1982, a 31-year period (assuming 365 d y^{-1} and 3.78 L gal^{-1}). Although effluent metal concentrations in 1982 and discharge volumes in 1970 are not representative of the entire time period of operations, this is the best monitoring data that is available to us. No other effluent monitoring data for metals in M-Area effluent were reported, as far as we know.

ARSENIC

Arsenic is a known human carcinogen. Arsenic was not a process chemical and site inventories reported amounts less than 0.25 lb, suggesting it was used as an analytical standard or in other laboratory applications. Although the ranking ratio for arsenic was very low (see Appendix C3), arsenic leached or carried in runoff from the coal piles to surface water was a concern, and this arsenic would not have been accounted for in the Site-wide inventory. Therefore, we compiled information on arsenic in surface water runoff from the coal and ash piles. We also reviewed Savannah River Ecology Laboratory (SREL) studies on arsenic levels in the swamp from the ash and coal piles and discussed them in the coal and ash section of this chapter. In 1981, Bradley estimated that about 90 kg of arsenic was in sludge and sediment of the M-Area seepage basin. Arsenic was released to Tim's Branch from M-Area in very small amounts (Bradley 1981), especially when compared to the amounts of metals like nickel, lead, and uranium that were released. The Site does not appear to contribute to arsenic levels in the Savannah River (Westinghouse 1996a).

CADMIUM

Cadmium is a carcinogenic metal. Cadmium compounds can cause lung disease, kidney damage, and skeletal and reproductive effects. Cadmium can cause cancer of the prostate and lung in humans. Cadmium may accumulate in meat, fruit, and fish. Shellfish, such as mussels, scallops, and oysters, are a major source of dietary cadmium. Cigarette smoking may be another important source of cadmium exposure. Liquid effluent from M-Area and various coal and ash pile runoff and leachate contained cadmium. In 1985, Lower estimated that although the continuous flow outfalls discharged no cadmium, the daily delivery of cadmium to Site streams totaled about 0.91 kg d^{-1} for the entire Site (Lower 1985). Coal and ash were probably much more important sources of cadmium from the SRS than process releases. The coal and ash section of this chapter contains a discussion of cadmium in coal ash and coal pile runoff basin effluents.

Separations Areas

Elevated levels of cadmium were observed in groundwater samples collected from wells adjacent to the F-Area seepage basin in 1982–1983 (Christensen and Gordon 1983b). Accounts of releases to the F-Area basin do not report cadmium and the source was said to be unknown (Lower 1985). Extensive sampling of the seepline was conducted in 1989 and 1990 and was published by Haselow et al. (1990). Cadmium was the only metal to exceed the maximum background soil value, but it was exceeded for only 1 sample out of 26. All the metals were less than the primary drinking water standard except cadmium from F-Area. Levels of cadmium at the seepline were between the proposed standard of 5 μg L^{-1} and the established standard at the time of 10 μg L^{-1}. The source of the cadmium in the Creek was said to have been the F-Area seepage basin but records did not address why and how much cadmium was discharged to the basin. Cadmium was not used in large quantities in F-Area processes.

M-Area

Cadmium was released in small amounts to M-Area wastewater. Based on effluent monitoring data from a time period when the average production rate for 313-M was 17,500 cores per month, Looney et al. (1987) estimated M-Area cadmium effluent discharges to the M-Area settling basin to have been 1 kg y^{-1}. Using this information, a total disposal mass, based on an average production rate of about 13,000 cores per month, over a 13.5 year period, was estimated to be 10 kg for cadmium, or about 0.74 kg y^{-1} (Looney et al. 1987). The estimate is probably most representative for the years 1971–1985.

In 1985 and 1986, Tim's Branch and Upper Three Runs Creek sediments were sampled monthly at six locations for metals. Eight samples were taken for each location. The most distant sampling point was in Upper Three Runs Creek, just downstream of the confluence with Tim's Branch. Mean concentrations plus or minus the standard error at this point were 0.4 $\pm$ 0.07 mg kg^{-1} for cadmium, compared to 1.5 $\pm$ 0.2 mg kg^{-1} just downstream of where the outfall enters Tim's Branch. The control or background location had levels of 0.1 $\pm$ 0.3 mg kg^{-1} (Pickett 1990). For comparison, a very conservative EPA preliminary remediation goal (corresponding to a 10^{-6} increased cancer risk for soil ingestion, inhalation, and dermal absorption) is 38 mg kg^{-1} for a residential scenario and 850 mg kg^{-1} for an industrial scenario (EPA 1996). The sediment

concentrations at the outfall was about 15 times the background levels Dilution appeared to be about 4 fold from the outfall to Upper Three Runs Creek (Pickett 1990). It seems likely that further dilution in Upper Three Runs Creek and the Savannah River swamp would result in very low concentrations entering the Savannah River. Since January 1984, cadmium has been analyzed in water samples collected from the Savannah River. Cadmium was found to be routinely undetectable. Any cadmium detected in Site stream sediments would not have come from the Savannah River; but would have been due to Site discharges. Site discharges may have been about 1 kg y^{-1} from M-Area to Tim's Branch and an undetermined amount from F-Area to Four Mile Creek. These discharges resulted in concentrations at or near the drinking water standard at the creek (Haselow et al. 1990). Discharges from the Site do not appear to have added detectable levels of cadmium to the Savannah River.

CHROMIUM

Chromium solutions were used at the SRS as corrosion inhibitors. Chromium was a component of waste solutions resulting from dissolving stainless steel and it was also used in cleaning solutions in the separations areas (Du Pont 1988). Chromium leached from coal and ash also contaminated groundwater beneath and surface water adjacent to the D-Area coal pile runoff basin.

In 1985, chromium was detected in water from four outfalls and an amount per day was calculated based on average effluent flow (Table 18-1). The total discharge was reported to be 3316 kg d^{-1} (Lower 1985).

Table 18-1. Chromium Concentration and Transport from Four Outfalls

Outfall	Concentration (mg L^{-1})	Mass transport (g d^{-1})	Receiving stream
A-001	0.16	287	Tim's Branch/Upper Three Runs
D-001	0.01	2,900	Beaver Dam Creek
F-008	0.033	126	Four Mile Creek
H-012	0.002	3	Four Mile Creek

[a] Source: Lower (1985).

D-Area

Chromium and other metals have also been of concern in surface water near D-Area. In 1985, Lower reported that measurements showed that the intake concentration of chromium at pumphouse 5G, the cooling water intake for D-Area operations, was one-fifth of the effluent concentration. The effluent measured included discharges from the D-Area powerhouse, heavy water production facilities, and ash basin (Lower 1985). Chromium release from the coal piles and ash disposal basins is discussed in the coal and ash section of this chapter.

As an Additive to Cooling Water

Chromium corrosion inhibitors, which were added to cooling water, were an important source of chromium released to surface streams. A 1973 report acknowledged that chromates used to inhibit corrosion in circulated chilled water and heating systems created pollution problems when leaks or spills of cooling water occurred. The report stated that an organic, biodegradable substitute was being evaluated for plantwide use (Du Pont 1973a).

Memorandums concerning spills occurring over the last two decades documented many spills and leaks of chromium-treated water (Jewell 1990). Some of the more significant spills are listed in Table 18-2.

Table 18-2. Spills and Leaks of Chromium-treated Process or Cooling Water

Date	Release	Reported amount	Remedial action
November 1977	Chromium released in effluent from retention basin 281-8H to Four Mile Creek in excess of NPDES	Not reported	Diluted to below detection limit
1977	Underground leak found in 241-H chromate cooling water system	Not reported	Leak repaired
May 1981	Cooling water containing chromium	9000 gal	Sent to the 281-8F retention basin
March 1987	Spill of cooling water containing chromium in concentrations up to 526 mg L^{-1}	10,000 to 15,000 gal	Released to an outfall to Four Mile Creek
April 1987	Water containing chromium to A-Area	More than 6950 gal	Contained and drummed

The largest and most well described of these spills occurred on March 28, 1987. From 10,000–15,000 gal of chromate-containing cooling water leaked to the ground from a cooling line that served tanks 29 and 32 in H-Area. The spill flowed into a nearby storm sewer and discharged through an outfall to Four Mile Creek before being contained with sandbags and pumped to H-Area seepage basins. The sodium chromate concentration in the cooling water was 526 mg L^{-1} (Jewell 1990; Durant 1994), suggesting that as much as 30 kg of chromium may have been released to the creek if all 15,000 gallons were not contained. Depending on how quickly the spill was contained, the release may have been smaller. Information needed to estimate releases from other spills is not available.

Site workers have not been able to tell us how much of the water used by the SRS was treated with chromium. It is likely that some water was treated and some was not. We have very little information on chromium use in cooling water and do not have enough information to determine whether a significant quantity of chromium was released to site streams and transported to the Savannah River. However, water quality monitoring data for the Savannah River do not suggest a measurable amount of chromium has been released from the SRS (see Chapter 20).

M-Area

In 1981, Bradley estimated that 113.6 kg of chromium was in sludge and sediment of the M-Area seepage basin. Concentrations of chromium, measured in a 24-hour composite sample of sewer effluent sampled in December 1981 when the plant was operating, were 0.003 mg L^{-1} in sewer effluent going to both the settling basin and to Tim's Branch. From these data, about 114 kg was estimated to have been in the settling basin sludge (Merz 1982). If we assume average discharges to the Tim's Branch were 2.6×10^8 gal y^{-1} or 9.8×10^8 L y^{-1} (Monier 1970; Hardt 1970; Colven et al. 1985). The chromium concentration of 0.003 mg L^{-1} reported by Merz would lead to an estimate of 2.95 kg discharged to the creek each year or 92 kg total from 1952 to 1982, a 31-year period, assuming 365 d y^{-1} and 3.78 L gal^{-1}.

In March through May 1985 the influent to and effluent from the M-Area settling basin was sampled and analyzed weekly for 10 weeks. Based on these sampling data, a removal efficiency was calculated for the basin (the influent concentration minus the effluent concentration divided by the influent concentration). The influent concentration of chromium was 0.037 $\pm$ 0.037 mg L^{-1}, the effluent concentration was 0.006 $\pm$ 0.006 mg L^{-1}, and the removal efficiency was estimated to be about 84% (Colven et al. 1985).

In 1984, a partial failure of the wooden dam at Steed's Pond, which received M-Area effluents, occurred, and samples were taken to determine if radioactive materials were migrating from the pond. The 1985 Annual Environmental Report reported the highest chromium concentration in Steed's Pond sediment was 290 μg g^{-1}, compared to an average background in the southeastern U.S. of 38 μg g^{-1}, ranging from 11–60 μg g^{-1} (Pickett 1990; Zeigler 1986a).

In 1985 and 1986, Tim's Branch and Upper Three Runs Creek sediments were sampled monthly at six locations for metals. Eight samples were taken for each location. The most distant sampling point was in Upper Three Runs Creek, just downstream of the confluence with Tim's Branch. Mean sediment concentrations plus or minus standard error at this point were 11.6 $\pm$ 2.2 mg kg^{-1} for chromium, compared to 80.2 $\pm$ 8.6 mg kg^{-1} just downstream of where the outfall enters Tim's Branch. The control location had levels of 7.2 $\pm$ 0.6 mg kg^{-1} (Pickett 1990).

For comparison, conservative EPA preliminary remediation goals, corresponding to a 10^{-6} increased cancer risk, are 210 mg kg^{-1} for a residential land use scenario and 450 mg kg^{-1} for an industrial land use scenario (EPA 1996).

Separations Areas

Chromium was discharged to the F-Area and H-Area seepage basin. Like mercury, it contaminated the groundwater beneath the basins. Some small amount of chromium outcropped to surface water at Four Mile Creek and may have eventually traveled to the Savannah River.

A 1% sodium dichromate solution was used in the Receiving Basin for Offsite Fuels (RBOF), 224-H, to clean lithium-aluminum alloy targets. The resulting liquid waste, containing about 1.25 mg L^{-1} chromium, was discharged to the H-Area seepage basin. After 1982, the chromate solutions used for lithium-aluminum target cleaning were sent to the 242-H evaporator rather than being directly discharged to the basins. The evaporator overheads were sent to the seepage basin and the bottoms were sent to waste tanks. The evaporator was effective in removing most of the chromium. In 1983, the contents of waste tank 23, which also contained

chromium, was sent to the evaporator, and this further reduced chromium releases to the seepage basin (Holcomb and Emslie 1984).

Chromium concentrations in wastewater going to the H-Area basins have been recorded since October 1980. Chromium releases before that time were not measured (Holcomb and Emslie 1984). The H-Area seepage basin received about 546 kg of sodium dichromate (about 217 kg of chromium) each year before 1982. A 1987 safety analysis report said that releases from January 1981 through July 1983 were 737 kg, which averages to about 24 kg mo^{-1} over 31 months or 285 kg y^{-1} (Hurrell et al. 1987). Based on an average flow rate of 1,261,000 gal wk^{-1} to H-Area and 311,700 gal wk^{-1} in F-Area, Holcomb and Emslie (1984) reported that in 1975, an estimated 123 kg of chromium had been released to F-Area seepage basin and 2318 kg to the H-Area seepage basin (Christensen and Gordon 1983b; Holcomb and Emslie 1984).

In 1980 and 1981, H-Area seepage basins were analyzed for chromium. Chromium releases from H-Area effluents to the seepage basins were said to have ranged from 2 to 5 mg L^{-1}. A chromium level greater than 1.5 mg L^{-1} was reported during a time when no material was being cleaned using the chromium solution at RBOF. H-Area personnel postulated that chromium could have been leaching along the lines to the seepage basins or that it could have been coming from corrosion inhibitors from waste tank cooling, although the cooling water was not supposed to be discharged to the seepage basins (Morris 1982). At times, the chromium concentration in the effluent discharged to the seepage basin was 10–30 mg L^{-1} (Clontz 1981). In 1981, a Request for Technical Assistance was completed for a chromium-free cleaning solution that was said to have been needed to comply with EPA release regulations. Methods to treat and reduce the amount of chromium waste solutions were also examined (Monson 1981).

In 1981, an analysis of effluents released to the seepage basin showed a maximum chromium concentration of 10.6 mg L^{-1} and an average concentration of 1.55 mg L^{-1}. Monitoring wells downgradient of basins showed chromium contamination exceeding the drinking water standards (Lower 1985). Looney et al. (1987) calculated inventories for the H-Area seepage basins from influent data, which included 590 kg of chromium through 1985. Based on measurements taken in 1960–1970, 1975, and 1983, the total disposal mass to the F-Area seepage basins from 1954 to 1983 was estimated to have been 47 kg of chromium, (Killian et al. 1987a) averaging to 1.5 kg y^{-1}. Killian et al. (1987b) predicted that before mid-1982, when the cleaning solutions were first sent to waste tanks, 554 kg of sodium dichromate (about 220 kg) of chromium was sent to the H-Area basins annually. Chromium concentrations in wastewater going to the basins have been measured routinely since October 1983. Eleven samples of H-Area effluent/H-Area seepage basin influent were taken from September to December 1983. The concentration of chromium averaged 0.072 mg L^{-1}. A maximum value of 0.36 mg L^{-1} was reported (Killian et al. 1987b). Table 18-3 summarizes the various estimates reported.

Table 18-3. Estimates of Chromium Discharged to the Separations Area Seepage Basins during Various Time Periods

Time period	Annual average to H-Area seepage basin (kg y^{-1})	Annual average to F-Area seepage basin (kg y^{-1})	Reference
1952–1981	217		Hurrell et al. (1987)
1982–1983	285		Hurrell et al. (1987)
1952–1975	2318	123	Holcomb and Emslie (1984)
1960–1983	220	1.5	Killian et al. (1987a)
1971–1985	590		Looney et al. (1987)
1954–1982	554		Killian et al. (1987b)

Because of the chromium releases to the separations area seepage basins and subsequent contamination of groundwater that outcrops into Four Mile Creek, chromium concentrations in Four Mile Creek were of interest. The chromium that traveled from the basins to the groundwater, subsequently outcropped into Four Mile Creek, and eventually traveled to the Savannah River seems to have been a very small amount. In 1987, the Site collected sediment samples to analyze for chromium from the Savannah River at river mile 120 and 187. Four Mile Creek enters the Savannah River at about river mile 150. The respective concentrations were 0.0193 and 0.0158 mg L^{-1}. These concentrations were more than 2 orders of magnitude less than concentrations measured in Four Mile Creek in 1987 (Mikol et al. 1988). Chapter 20 summarizes the results of additional chromium monitoring of Site stream and Savannah River sediments and water. Results of sediment, water, and fish sampling show no significant differences between chromium concentrations in onsite streams and other Savannah River locations, suggesting chromium contamination of the Savannah River is not detectable. Chromium concentrations measured in onsite streams and Savannah River water and reported in the Annual Environmental Reports from 1984 through 1991 have been well below the drinking water standards and near or below detection limits.

COAL AND COAL ASH

Steam and electricity were produced by as many as seven coal-fired power plants in the A-Area, C-Area, D-Area, F-Area, H-Area, K-Area, and P-Area. Liquid effluent from the F-Area and H-Area coal-fired power plants were diverted to coal-pile runoff or ash basins. Coal-fired steam plants in the reactor areas had floor drains that discharged to the ash and coal-pile runoff basins (DOE 1987). The largest power plant was in D-Area. The D-Area power plant released fly ash and coal pile runoff into the Beaver Dam Creek stream and swamp system. Most of the information in historical records is concerned with the D-Area power plants. Beaver Dam Creek received most of the coal and ash basin liquid effluents generated onsite. After 1982, D-Area had a coal pile runoff basin and ash basin that received powerhouse ash sluice. In addition, a filled ash disposal basin was used as a sanitary spray field for sanitary treatment plant effluent (Peralta and Lewis 1982).

Coal Storage

Coal was stored at all of the seven power plants. Surface runoff from the coal storage piles was discharged to surface streams until 1977, when the National Pollution Discharge Elimination System (NPDES) was initiated. To meet NPDES discharge requirements and also in response to SCDHEC requests, seven unlined earthen containment basins were constructed at each coal storage location to intercept, stabilize, and treat surface runoff from the coal piles (DOE 1987; Christensen and Gordon 1983a). To comply with the NPDES permit issued in 1977, coal pile runoff containment basins were constructed in D-Area and A-Area in 1978 and basins in C-Area, F-Area, K-Area, H-Area, and P-Area were completed by March 1981 (Cummins et al. 1990). Rainwater runoff from the coal piles flowed into the containment basins through ditches and sewers. The basins were designed to diminish the entry of large amounts of low pH runoff into surface streams. Releases of sulfur compounds, magnesium, chromium, cadmium, manganese, and arsenic in the runoff from the coal piles are of concern. The basins would have retained these compounds, but releases before the basins were constructed went into Site streams. Acid from oxidation of sulfur materials in the coal was washed by rain into the runoff basins. Monitoring of groundwater at the seven basins showed concentrations of some heavy metals above background levels (Christensen and Gordon 1983a). Because the groundwater beneath the piles does not flow offsite, it was not evaluated as a part of this dose reconstruction study. Runoff to surface water does flow into site streams which eventually flow into the Savannah River, so surface water contamination by coal and ash pile runoff were considered. Coal pile runoff basins in C-Area, K-Area, H-Area, F-Area, and P-Area were all active in 1982; L-Area and R-Area basins were not active (Peralta and Lewis 1982). The C-Area and F-Area basins have been inactive since 1985 when the coal piles were removed (Westinghouse 1992a).

Coal Reject Piles

One of the few studies done on metals in coal pile leachate was reported by Carlson (1990). He reported that acidic leachate from a reject coal pile in D-Area and inactive ash basin was contaminated with metals and sulfate and was stressing vegetation, presumably because of the low pH. The leachate from the coal pile had contaminated the water table with sulfate (up to 22,200 mg L^{-1}) and metals, especially iron and aluminum. The reject coal pile was located adjacent to one of the old ash basins. The volume of reject coal placed along the berm surrounding the basin and the time period over which it was deposited was unknown. The pile was about 380 m long, up to 30 m wide, and 2.5 m thick. By 1981, the pile was covered with soil. An area of stressed vegetation was noted north of the pile. Numerous seeps at the base of this coal pile were observed. These seeps combined to enter outfall D-003 and eventually flow into the Savannah River. The seeps had elevated levels of iron, aluminum, lead, nickel, arsenic, cadmium, and sulfate. Three of the seeps had levels of lead, nickel, chromium, and cadmium that exceeded hazardous concentration levels of inorganic pollutants from coal storage areas referenced by the author (Carlson 1990) as having been developed by Wachter and Blackwood in 1978. Two seep samples exceeded the EPA's hazardous waste levels for arsenic.

Ash Disposal

A large quantity of coal ash has been produced by the SRS. Coal ash typically contains the toxic metals arsenic, cadmium, chromium, lead, mercury, and selenium. Ash is of concern as an air pollutant, contributing to particulate emissions, and a surface water contaminant because of contamination of precipitation runoff from ash disposal piles and leaching of toxic compounds from piles and disposal basins (Evans et al. 1980). Chapter 17 describes the releases of ash to air. The solubility of trace elements in coal ash depends on the coal source, combustion, and chemical properties of rainfall and runoff (especially the pH because many of the metals are more soluble at low pH). Metals can leach into groundwater and contaminate surface water from coal and ash pile runoff or overflow from a basin (Evans et al. 1980). Significant quantities of inorganic salts may leach from the ash into the ground. Sluicing the fly ash to the basin and the small particle size of the fly ash makes it possible for the ash to pass through the basin entirely in suspension. Smaller particle size also increases the tendency of the metals to become dissolved in the water and be transported more readily through the drainage system (Guthrie and Cherry 1976; Monier and Bebbington 1970).

Many ecological studies have been conducted on the swamp areas influenced by ash basin effluent. Many studies, conducted primarily by the SREL, have determined levels of metals, the pH and other characteristics of effluent from the coal ash basins, and the effects of effluent on biota in the receiving swamp. The findings of many of these studies have been published in the open literature (Cherry and Guthrie 1979; Cherry et al. 1979; Guthrie and Cherry 1976; Skinner et al. 1978; Rodgurs et al. 1978; Lagnese and Dzombak 1993; Evans and Giesey 1978; Evans et al. 1980).

There were five dry ash piles onsite at the SRS: three in C-Area and two in A-Area. After they became inactive, ash piles were seeded with grass to help prevent erosion and runoff. In 1987, the two active piles were said to have been surrounded by a containment dike. The Radiological Sciences Group Monthly Report for November 1970 mentions ash having been hauled by truck and dumped on the ground at 100-C and 3/700, which is consistent with the 1987 assessment of dry ash piles (Du Pont 1970a). Ash at the 700-Area powerhouse was handled dry and placed in a 2-acre pile. At the other areas, ash was disposed of by sluicing ash and water slurries to ash basins. The ash settled out and water was discharged from the basins through permitted outfalls (Arnett et al. 1992).

In 1987, there were 6 active and 4 inactive ash basins. The ash basins, located near the coal-pile run off basins, received ash sluicewater from the powerhouses. Groundwater contaminants associated with these basins include metals and sulfates. Leachate from the ash basin in D-Area was suspected of being the cause of dead vegetation in an area immediately north of the ash basin. This was also said to be a result of surface drainage from the D-Area sanitary wastewater plant discharge. The toxic component of the leachate or surface runoff that was the cause of the dead vegetation had not been specifically identified in 1987 (DOE 1987).

Washwater from the 700-A Area powerhouse may have also been contaminating sediments and adversely affecting a wetland area below the outfall A008 (DOE 1987).

Since start-up the bottom ash and fly ash from the D-Area plant were combined and sluiced into one of two settling basins. This plant burned 451,940 metric tons of coal annually, producing 32,650 metric tons of ash that was sluiced into a settling basin by approximately 4.5 billion liters of water (Rodgurs et al. 1978). The two basins were filled with ash in about 1975, and a larger

basin to the east of the two was constructed. Overflow water from the basins drained into a 2.5-km^2 swamp that connects with Beaver Dam Creek. The creek flows through a small lake and a second swamp before reaching the Savannah River. Periodic flooding of the Savannah River caused backwater up to the junction of Beaver Dam Creek and the first swamp (Evans and Glesy 1978).

The ash and sluice water eventually emptied in the Savannah River after passing through the swamp. The basin effluent was diluted from water from Beaver Dam Creek and then from the flow of Four Mile Creek before it entered the river. The "influence from the ash" was said to "no longer be apparent" after the Four Mile Creek entrance (Rodgurs et al. 1978), suggesting ash components were not detected in Beaver Dam Creek once it merged with Four Mile Creek.

After 1975, sluiced coal ash from the D-Area power plant was pumped into a 14.6-ha settling basin, with a calculated retention time of about 39 days. The waters of this basin then emptied into a 6.1-ha basin with a calculated retention time of 22 days. The effluent of the second basin passed through a flume and into a swampy area. This water then flowed down a channel into Beaver Dam Creek. The effluent from the basin was said to have been diluted approximately 20 fold by the creek water depending on the time of year. This water then entered the Savannah River swamp and eventually the Savannah River (Newman et al. 1985).

The DOE 1987 survey reported coal ash generation by the power plants in cubic yards per year (DOE 1987). Table 18-4 summaries the annual ash generated by each area.

Table 18-4. Annual Ash Generated by Area[a]

Area	Location	Ash generated in $yd^3\ y^{-1}$ and discharged to the ash basins
700-A	788-A	5,000
400-D	488-1D and 2D	65,000
200-H	288-H	13,000
100-K	188-K	18,000
100-P	188-P	18,000
Total		119,000

[a] Source: DOE (1987).

The annual ash disposal rate was approximately 50,000 $yd^3\ y^{-1}$ from 1952 until 1983 and has been approximately 65,000 $yd^3\ y^{-1}$ in 1983 and the years after. This amounts to 1.94×10^6 yd^3 of ash from 1952 to 1983. The DOE (1987) survey did not include the 200-F Area powerhouse. In 1992, the 200-F powerhouse had been permanently shutdown. The powerhouse was said to have generated approximately 15,000 $yd^3\ y^{-1}$ when it operated, presumably resulting in a total of about 4.65×10^5 yd^3 of ash (Arnett et al. 1993).

The amount of coal ash discharged to the basins can be estimated using several different reports. The amount of metals discharged might be calculated using values of metals in typical coal ash. However, we lack the data necessary to determine the transport of metals from the basins to the streams, swamp, and the Savannah River.

Many researchers have examined the solubility and transport of metals in ash effluent, which is usually discharged to ponds, streams, or lakes. The effluent from sedimentation ponds that was

widely used to settle ash particles from fly ash and bottom ash transport streams was of particular concern. The effluent sluicing water contains a variety of inorganic chemicals from leachate of bottom ash and fly ash: metal cations such as Al^{+3}, Cd^{+2} and Ni^{+2}, and metal oxyanions, such as AsO_4^{2-}, SeO_4^{3}, and MnO_4^{2-}. A number of studies have examined the environmental fate and transport and ecological effects on plants and animals in and around ash ponds. Most of these studies have attempted to find ways to remove contaminants of concern for regulatory agencies. Sluice pond or basin water often contains metals from the ash particles and sometimes at concentrations that can exceed discharge limits. Promoting metal absorption and precipitation (pH dependent reactions) can reduce the concentration of metals in water leaving the ponds (Lagnese and Dzombak 1993).

The 1970 Radiological Sciences Division November monthly report stated that leaching of chemical elements from power plant department ash basins did not contribute hazardous concentrations of toxic chemicals to the river. Leachate samples were collected on three different dates in the fall of 1970 from ash basin effluents in P-Area and D-Area and were analyzed for metals by a spark source mass spectrometer and for mercury by an emission spectrographic technique. Concentrations of arsenic, barium, selenium, cadmium, and iron in ash basin effluent were found to be slightly higher than drinking water standards, but the authors concluded that "on-site plant streams provide a more than adequate dilution" (Du Pont 1970a). Metal concentrations were not determined in the Savannah River or Site streams as a part of this analysis.

In 1979, Guthrie and Cherry (1979) studied the accumulation of heavy metals in biota in the D-Area coal ash drainage system. They determined concentrations of metals in water and sediments of the basin and in Upper Three Runs Creek, which they considered an uncontaminated stream (Table 18-5).

Table 18-5. Concentrations of Selected Metals in Water and Sediments of the D-Area Ash Basin and Upper Three Runs[a]

Metal	Mean water concentration in mg L^{-1} in the ash basin	Mean water concentration in mg L^{-1} in Upper Three Runs	Mean sediment concentration in mg L^{-1} in the ash basin	Mean sediment concentration in mg L^{-1} in Upper Three Runs
Manganese	0.75	0.31	96.0	53.0
Zinc	0.37	2.6	6.5	2.1
Chromium	0.16	0.08	38.0	6.50
Cadmium	0.12	0.14	1.7	0.71
Mercury	0.04	0.006	0.80	0.70

[a] Source: Guthrie and Cherry (1979).

The sediments of the ash basin contained 2 to 10 times higher concentrations of metals, except for mercury. This study was not very helpful for determining the reduction of metals by sedimentation and dilution of the effluent in the creek. Zinc and cadmium were higher in the water from Upper Three Runs Creek than in water from the basin (Guthrie and Cherry 1979). No explanation for this was given, and it was not clear whether the samples contained sediment or were filtered. M-Area effluents could have been the source of the metals in Upper Three Runs Creek, depending on the sample location.

In 1978, metal removal by the seepage basin in D-Area was studied. The removal was determined to be effective because the concentration of iron in the basin effluent was reduced by 90% from the concentration in waste entering the basin. Concentrations of arsenic, chromium, and manganese were reduced by 86, 87, and 83%, respectively. Mercury concentrations were not measured (Peralta and Lewis 1982).

Skinner et al. (1978) studied cadmium in the food chain in the coal ash basin. They studied what this and what many similar journal articles call ”an island composed completely of coal ash.” The island was described as a 140 × 380-m island located in an ash basin, approximately 600 m southwest of the D-Area coal-fired plant, composed of coal ash that had accumulated since 1952. The ash island had received more than 3 kg m^{-2} of fly ash between 1952 and 1975. A good description of the plants that grow on the island and the ecosystem can be found in Skinner et al (1978). The coal ash basin was sampled for cadmium between October 20 and November 25, 1975. Coal ash generally has higher levels of cadmium than coal, often in the 12–35 ppm range. This has been a concern for using fly ash as a soil amendment or fertilizer. Cadmium inputs to the island were from coal ash via sluice water from boiler grating, stack precipitators, and aerial deposition. Samples of plants, snails, insects, spiders, and soil were taken from the island. Cadmium in soil was bound to organic matter and was held to the soil surface. Approximately 42% of the cadmium was extracted by distilled water in 1 hour. Cadmium concentrations averaged 0.12 ± 0.002 μg g^{-1} dry weight, with 0.05 ± 0.01 (standard error) μg g^{-1} of the cadmium water extractable (Skinner et al. 1978). The study documented in Skinner et al. (1978) was designed to examine concentrations in insects and other biota living on the ash, not cadmium leaving the Site.

Rodgurs et al. (1978) studied the removal of contaminants in the swamp and found approximately 1200 m^2 of swamp area was necessary to remove most of the ash influence. Selenium, cadmium, arsenic, and mercury levels varied from 0.12 to 0.04 mg L^{-1} in the swamp water southwest of the 400-D powerhouse. Sediment concentrations were about 1 order of magnitude higher.

Water, sediment, and duckweed samples were collected at six sampling stations and were analyzed using a neutron activation analysis procedure. The results of the analysis in the best available copy of this document are in very small type and are illegible. The text stated that dissipation by settling was most apparent for metals that were at the highest concentrations in the basin water and in the sediments at the stream-swamp confluence. The mean concentrations at the station furthest from the ash basin and closest to the river would be the most representative of effluent closest to where offsite exposure could occur. Levels of metals at the furthest sampling stations were reported not to have been influenced by the ash (Rodgurs et al. 1978).

Newman et al. (1985) sampled for metals dissolved in water from six different locations associated with the 400-D power plant ash basins and compared them to samples taken from Par Pond. Site A was located in the ash settling basin. Sites D and E were in the swamp about 300 m and 1 km below the confluence of the effluent stream and Beaver Dam Creek. Beaver Dam Creek goes on to empty into the Savannah River swamp and then eventually into the Savannah River, but samples below Site E were not reported. Table 18-6 summarizes the results.

These data indicate that the concentrations were decreased because of settling and dilution. A general decrease was seen in the concentrations of arsenic, cadmium, and chromium with distance away from ash basins. However, copper, zinc, and manganese concentrations seem to have been higher 1 km down the creek than at the first basin discharge point, and no explanation

was given for this. Turbulence stirring up sediments in the creek was mentioned and may have accounted for the increases (Newman et al. 1985).

Table 18-6. Concentrations of Dissolved Elements in Ash Basin Effluent and Swamp Waters into Which It Discharges at Various Locations (in mg L^{-1})[a]

Element	Effluent from the first ash retention basin		Effluent from the second retention basin		After a swampy area 250 m below the outfall from the second basin		Beaver Dam Creek, 300 m below where the effluent enters		Beaver Dam Creek, 1 km below where the effluent enters		Par Pond, control site	
	Mean	SD[b]	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Manganese	27	±22	25	±21	30	±17	86	±45	82	±44	3	±5
Arsenic	49	±32	46	±30	43.9	±30.7	2.5	±2.0	2.2	±1.7	1.3	±1.6
Cadmium	0.38	±0.17	0.39	±0.30	0.38	±0.37	0.21	±0.07	0.23	±0.27	0.07	±0.07
Chromium	0.50	±0.55	0.45	±0.47	0.31	±0.35	0.35	±0.31	0.38	±0.38	0.05	
Copper	2.6	±0.6	2.6	±0.67	2.6	±0.9	3.0	±0.9	3.0	±1.0	0.8	±0.9
Zinc	9	±6	8	±4	8	±7	12	±17	10	±16	2	±4
Magnesium	1733	±228	1723	±228	1726	±241	1333	±180	1332	±188	1070	±111

[a] Source: Newman et al. (1985).
[b] SD = standard deviation.

In a similar study conducted in 1978, Beaver Dam Creek was compared to Steel Creek, which was described as an adjacent creek not receiving any ash effluent. Steel Creek did receive production reactor cooling water and purge water from reactor fuel and target assembly basins until 1967. They sampled and subjected samples of precipitator ash from the power plant to leaching to predict how readily metals in the ash might be dissolved and enter Beaver Dam Creek in solution. Cadmium, nickel, and zinc were elevated by about a factor of 2 to 4; however, the authors concluded that determining statistically significant differences was difficult and that the trace metal contamination was minor (Evans and Giesey 1978).

Studies have suggested that many metals concentrate in biota. Chromium and arsenic can concentrate in some plants. Guthrie and Cherry (1979) studied plant uptake and found that plants contained 15% of the total heavy metal concentration found in the benthos. Their report stated that "The objective of this work was to examine mechanisms which operated to remove pollutants from the ash basin effluent received by a swamp drainage system". However, a removal rate for biological components of the system could not be determined from these studies. The major mechanism of removal was determined to be settling of particulates in the basin. As flow rates decreased father from the basin, the settling increased (Guthrie and Cherry 1976). Taken together, these studies suggest that much of the contaminants of concern in ash are removed by the basins and by the swamp area between the ash basin discharge points and the Savannah River. Most of the removal was attributed to settling (Rodgurs et al. 1978), but removal by biota also occurred (Guthrie and Cherry 1979; Skinner at al. 1978). There is a potential for migratory waterfowl and other wildlife onsite to ingest and perhaps concentrate some of these metals, and this may be a pathway of exposure to evaluate in later phases of the dose reconstruction study. River quality monitoring for these metals does not suggest that Site streams are a source of metals to the Savannah River (Westinghouse 1996a).

GASOLINE

Many gasoline tanks and several pumping stations and associated tank trailers have operated at the SRS over the years. Spills to surface water seem to have been less of a concern than leaks to groundwater. The Spill Prevention Control and Countermeasures Plan submitted by the SRS to the states and the EPA includes a list of all of the gasoline tanks in each area and a description of the tanks, their capacities, and containment features (Westinghouse 1997). The best description found for many of the tanks is in the Part 70 Operating Permit Application, which includes estimates of air emissions of gasoline and components like benzene (Westinghouse 1996b). The CIIS Database from 1994 indicated that 49 locations reported total gasoline stores of about 1.99×10^5 kg (Meyer et al. 1995).

In 1987, the SRS had about 75 underground storage tanks used to store petroleum products; they were located in D-Area, TNX, C-Area, F-Area, H-Area, CS, K-Area, L-Area, P-Area, R-Area, A-Area, and M-Area. Many of the tanks have been leak tested and those that failed were excavated and repaired in the late 1980s. The potential for these tanks leaking, the inadequacy of leak detection systems, and other problems perceived with tank and drum storage of chemicals were discussed in the Site Environmental Survey (DOE 1987). In response to a Tiger Team Assessment Information request, a list of SRS underground storage tanks and their contents and capacities were compiled in 1990. A memo (Morris 1990) listed the tanks and their location, capacity, and status (in use or abandoned). The memo stated that all of the petroleum tanks installed before 1965 or of unknown age were tightness tested and abandoned or temporarily closed in 1989. The leaking tanks were buried at the Diesel Fuel Storage Facility in N-Area. Benzene and lead have been found in groundwater in levels exceeding the drinking water standard near some of storage areas (Arnett et al. 1993). This contaminated groundwater has not moved offsite or outcropped into surface streams.

The records needed to estimate the amount of gasoline and other fuel that was released to surface waters over the years are not available. A summary of the fuel and oil spills that have been reported since 1980 is provided later in this chapter.

HYDROGEN SULFIDE

Hydrogen sulfide was used in D-Area to make heavy water as described in the Phase I Task 3 report (Meyer et al. 1995). An undated Teletype, likely from the late 1950s, reported a release of about 2 tons of hydrogen sulfide to Beaver Dam Creek on December 29. A stripper level control system malfunctioned and process waste containing hydrogen sulfide was released. The technical standard of 10 mg L^{-1} hydrogen sulfide in wastewater was thought to have probably been exceeded (French 195?). Evans and Giesey (1978) reported that 900 kg y^{-1} of hydrogen sulfide was discharged to Beaver Dam Creek in the mid-1970s. Three spills of hydrogen sulfide to surface water from D-Area were reported for the 1980s: (1) 364 kg released to Beaver Dam Creek in February of 1981, (2) 182 kg released to the process sewer in May 1982, and (3) 364 kg released to the same process sewer in December 1981 (Jewell 1990).

Hydrogen sulfide is a gas at atmospheric pressure and readily evaporates from surface water. It is also soluble in water, so it could be transported some distance. Some microorganisms oxidize hydrogen sulfide to elemental sulfur. Hydrogen sulfide oxidation occurs readily in oxygenated

surface water, like creeks. The half-life of hydrogen sulfide in soil and aquatic environments generally ranges from one to several hours (ATSDR 1997). It seems likely that the rapid oxidation of hydrogen sulfide might have limited the amounts that could have traveled to the Savannah River.

Total sulfides have been measured in the Savannah River for many years. It is not known how much of the sulfides are hydrogen sulfide, but sulfide loading from the Site have not been considered a water quality issue (Zeigler et al. 1986b, 1987).

LEAD

Groundwater analysis suggests that lead has migrated from waste disposal sites. Lead in groundwater has been found to be elevated near the Chemical Metal and Pesticides (CMP) pits, the Savannah River Laboratory (SRL) seepage basins (galvanized well casings contributing to elevated lead concentrations were noted) and the Silverton Road waste site. Lead in groundwater under the D-Area coal pile runoff basins has exceeded the drinking water standard. Groundwater contaminated by these sources does not appear to flow offsite.

Lead was also discharged to the separations area seepage basins. Lower (1985) stated that 623 kg was discharged to the F-Area and H-Area seepage basins in calendar year 1975 alone, an estimate much larger than other estimates. Looney et al. (1987) calculated an inventory for the H-Area seepage basins from influent data of 1475 kg for lead, presumably for 1971–1985, or about 109 kg y^{-1}. Based on monitoring done in 1960–1970, 1975, and 1983, the total disposal mass to the F-Area seepage basins from 1954 to 1983 was estimated to have been 436 kg of lead (Killian et al. 1987a), or an average of 15.4 kg y^{-1}. Eleven samples of H-Area effluent/H-Area seepage basin influent were taken from September to December 1983. The concentration of lead averaged 0.18 mg L^{-1}. A maximum value of 0.54 mg L^{-1} was reported (Killian et al. 1987b). No mention of measurable amounts of lead outcropping to Four Mile Creek has been noted.

M-Area

M-Area used lead powder mixed with oil as a lubricant in the process (Gary 1996) as described in the section on releases of lead to the air from M-Area (see Chapter 17). A Waste Characterization and Reduction Study estimated the annual use of lead powder, which was 99% lead, to be 1126 kg for all M-Area facilities (Lockwood Greene 1983).

In 1981, Bradley estimated that 273 kg of lead was in sludge and sediment of the M-Area seepage basin, resulting from a discharge of 0.23 kg d^{-1} (Bradley 1981). An M-Area settling basin characterization study published in 1985 estimated that the basin then contained 155 kg of lead (Colven et al. 1985); 95 kg in the sludge, 55 kg in the soil to a depth of 6 ft, and 5 kg in the liquid. In March through May 1985, the influent to and effluent from the M-Area settling basin was sampled and analyzed weekly for 10 weeks. Based on these sampling data, removal efficiency was calculated for the basin (the influent concentration minus the effluent concentration divided by the influent concentration). The influent concentration of lead was 0.407 $\pm$0.50 mg L^{-1}, the effluent concentration was 0.110 $\pm$0.31 mg L^{-1}, and the removal efficiency was estimated to be about 71% (Colven et al. 1985). Based on effluent monitoring data from a time period when the average production rate for 313-M was 17,500 cores per month, Looney et al.

(1987) estimated M-Area effluent discharges to the M-Area settling basin to have been 60 kg y^{-1} for lead.

As with many of the other metals, estimates of the amount of lead discharged to the settling basin can be determined. However, releases to the Savannah River cannot be estimated because not enough is known about dilution, adsorption, and other reactions and transport and fate parameters that influence the amount to reach the river.

In 1984, the wooden dam at Steed's Pond, which received M-Area effluents, partially failed, and samples were taken to determine if radioactive materials were migrating from the pond. The 1985 annual environmental report reported the highest lead concentration in Steed's Pond sediment was 65 μg g^{-1}, compared to an average background of 13 μg g^{-1}, ranging from 3-26 μ g g^{-1} in the southeastern U.S. (Pickett 1990). Measurements of lead in Tim's Branch sediments in 1984, 1985, and 1986 were reported to be 2.0 μg g^{-1} in sediments above M-Area and 33.7 μg g^{-1} in sediments below M-Area. A concentration of 1.7 μg g^{-1} in sediments at the creek mouth were reported (Specht 1991). The furthest sampling point was in Upper Three Runs Creek, just downstream of the confluence with Tim's Branch. Mean concentrations $\pm$ standard error at this location were 8.9 $\pm$ 3.8 μg g^{-1} for lead, compared to 33.7 $\pm$ 5.7μg g^{-1} just downstream of where the outfall enters Tim's Branch. The control location had levels of 2.0 $\pm$ 0.2 μg g^{-1} (Pickett 1990).

It is unclear how much liquid waste containing lead would have been discharged to Tim's Branch rather than the seepage basin during this time. Estimates of the amounts of waste solvents discharged to the seepage basins and Tim's Branch made by Christensen and Brendall (1981) suggest that the volumes discharged were variable but about 15–60% of the total went to the creek in the late 1970s. From this estimate, we might expect that 0.03–0.14 kg d^{-1} of lead may have gone to Tim's Branch, corresponding to 11–50 kg y^{-1}. Concentrations of lead, measured in a 24-hour composite sample of M-Area sewer effluent sampled in December 1981 when the plant was operating, were 0.003 mg L^{-1} in sewer effluent going to the settling basin and 0.008 mg L^{-1} in sewer effluent going to Tim's Branch. From these data, about 273 kg of lead was estimated to have accumulated in the settling basin sludge (Merz 1982). If we assume average discharges to the Tim's Branch were 2.6×10^8 gal y^{-1} or 9.84×10^8 L y^{-1} (Monier 1970; Hardt 1970; Colven et al. 1985) then the concentration of 0.008 mg L^{-1} reported by Merz would lead to an estimate of 7.8 kg discharged each year or 244 kg total from 1952 to 1982, a 31-year period, assuming 365 d y^{-1} and 3.78 L gal^{-1}.

Lead concentrations in the Savannah River and in Site streams have been less than 0.5 mg L^{-1}, the limit of detection reported for water quality monitoring of the Savannah River. After the first year, monitoring for lead was not continued for the Savannah River because all of the samples were less than the detection limit (Du Pont 1964). SREL data showed that lead levels in the river were less than 0.02 mg L^{-1}, using a more sensitive analytical technique. Concentrations from outfalls ranged up to 0.024 mg L^{-1}, corresponding to a lead transport of about 0.5 kg d^{-1} according to Lower (1985).

MANGANESE

Manganese dioxide was used in the separations area head end process. Much of the manganese in the wastewater was sent to the waste tanks (Du Pont 1974c), but some was discharged to the seepage basins as manganous nitrate. A 1985 memorandum stated that moderate

levels were generally not a health concern, and discharge limits were for corrosion of pipes caused by manganese oxides (Lower 1985). Eleven samples of H-Area effluent/H-Area seepage basin influent were taken from September to December 1983. The concentration of manganese averaged 0.56 mg L^{-1}. A maximum value of 3.2 mg L^{-1} was reported (Killian et al. 1987b). Extensive sampling of the seepline was conducted in 1989 and 1990 and was published by Haselow et al. (1990). Levels of manganese were greater than the secondary drinking water standard at the seepline. The highest concentration found was 0.72 mg L^{-1}, recorded at Road A-7 in 1984. Manganese was not identified in effluents at outfalls F-008, H-008, H-012, which discharge into Four Mile Creek.

Manganese was reported for four outfalls in 1981: (1) C-004, which discharges to Four Mile Creek with a concentration of 0.05 mg L^{-1} and an estimated transport of 46 kg d^{-1}, (2) D-001, which discharges to Beaver Dam Creek with a concentration of 0.14 mg L^{-1} and an estimated transport of 40 kg d^{-1}, (3) F-007, which empties into Upper Three Runs with a concentration of 0.052 mg L^{-1} and an estimated transport of 32 g d^{-1}, and (4) P-019, which runs into Par Pond/Lower Three Runs at levels of 0.05 mg L^{-1} and an estimated transport of 44 kg d^{-1}. Lower (1985) reported that in 1984, the average concentration measured in the Savannah River upstream of the SRS was 0.22 mg L^{-1}, said to correspond to an annual transport of 1.38×10^6 kg for that year or 3788 kg d^{-1}. This implies that 1.38×10^6 kg of manganese was being brought onto the Site in cooling water taken from the river.

Manganese discharges to the separations area seepage basins were not routinely measured. Wells downgradient of the 200-Area seepage basins contained elevated manganese. Drinking water standards for manganese were exceeded in 1981 at wells located at the CMP pits. Manganese was identified as a potential contaminant in the groundwater near the Silverton Road Waste and in groundwater and runoff released from ash and coal. (Lower 1985). Evans and Giesey (1978) reported that 675 kg y^{-1} of potassium permanganate and other manganese compounds were discharged to Beaver Dam Creek in the mid-1970s. An electrolytic process was conducted in D-Area for several years that involved pound per batch quantities of potassium permanganate (personal communication with Robert Garvin, retired 1996). The SRL seepage basins were estimated to have received 433 kg of manganese during 28 years of operation (Lower 1985).

During March through May 1985, the influent to and effluent from the M-Area settling basin was sampled and analyzed weekly for 10 weeks. Based on these sampling data, a removal efficiency was calculated for the basin (the influent concentration minus the effluent concentration divided by the influent concentration). The influent concentration of manganese was 0.034 ± 0.20 mg L^{-1}, the effluent concentration was 0.008 ± 0.009 mg L^{-1}, and the removal efficiency was reported to be about 83% (Colven et al. 1985). Any release estimate determined from this limited information would be extremely uncertain. Discharges of process wastewater to site streams may have been about 800 kg y^{-1} in the mid-1970s and early 1980s. The uncertainty associated with this estimate probably spans at least an order of magnitude.

Concentrations of manganese in water and sediments were not significantly different between sampling locations above the SRS and locations along and below the SRS boundary, which indicates the SRS did not contribute detectable amounts of manganese to the river (Westinghouse 1996a).

MERCURY

Chapter 17 provides a description of the use of mercury at SRS facilities and a brief summary of the toxicity and environmental fate of mercury released into the environment.

The SRS and the SREL have done a considerable amount of research on mercury in the environment. In their 1994 document, *Assessment of Mercury in the SRS Environment*, Kvartek et al. (1994) concluded that no significant releases of mercury to the Savannah River were likely to have occurred. They believe that any releases would have been well below the SCDHEC standard, based on process knowledge (Kvartek et al. 1994).

Several dozen SREL reports pertinent to mercury have been generated. Most of these report the findings of studies of bioaccumulation and the effects of mercury on the ecosystem of Par Pond, Upper Three Runs, Four Mile Creek, and areas adjacent to the Savannah River swamp (Kvartek et al. 1994; Gladden 1997).

Sources of Mercury

Bebbington (1990) acknowledged that mercury had been accumulating in the sediments of the 25-million gallon cooling water basins in the reactor areas, in Par Pond, and L-Lake. The mercury was presumed to come from Savannah River water, with the source said to be operations upstream of the Site (Bebbington 1990). Savannah River water, presumably contaminated upriver of the Site, was used onsite for cooling water and was subsequently pumped into Site streams and ponds.

In the 1970s, commercial fishing rights were suspended because of mercury contamination in the Savannah River. In June 1973, a monitoring program for mercury in water, sediment, and fish in onsite streams and Par Pond was started in order to show that SRS operations were not contributing to mercury in the Savannah River. The annual reports attribute mercury found in fish and sediments onsite to offsite sources upriver (Lower 1985; Arnett et al. 1993). Mercury can be found in Savannah River sediment and water upstream of the SRS because of discharges from a mercury-cell-type chlor-alkali plant, called the Olin Corporation plant, near Augusta, Georgia (GWQCB 1971; Lower 1985). The chlor-alkali plant was discharging large amounts of mercury into the Savannah River. From 1965 to 1970, a discharge rate of 5.5 kg d^{-1} was reported (Tilly and Wilhite 1972).

Page 15 of the 1990 Summary Pamphlet to the Environmental Report stated, “Since 1989, concentrations of mercury in fish collected at all onsite locations have been higher than those in fish collected from the Savannah River, indicating a possible onsite source of mercury. The SRS streams are not open for public fishing” (Cummins et al. 1991). None of the other annual report summaries or reports, before or after 1990, suggest that any of the mercury came from SRS operations. SREL personnel interviewed in 1996 maintained that the mercury contaminating onsite fish originated offsite and was brought onsite in the river water used for cooling.

Mercury Use in the Separations Areas

Mercury was used as a catalyst to dissolve aluminum fuel at a rate of 7.95 kg d^{-1} in H-Area where it was used routinely and at 0.18 kg d^{-1} in F-Area where it was used occasionally (Horton 1974a).

The 221-H large and small dissolvers used about 7 kg and 5 kg, respectively, for each run in the 1980s (Kvartek et al. 1994). In 1970, it was discovered that mercury was an impurity in sodium hydroxide (1.5 ppm in 50% NaOH) amounting to a total contribution of about 0.0045 kg d^{-1} in F-Area and 0.0091 kg d^{-1} in H-Area (Du Pont 1971a). Horton (1974a) estimated that through the early 1970s (presumably from 1959–1973), 1600 kg of mercury was released to the H-Area seepage basins and 380 kg to the F-Area seepage basins from this source.

About 80 tons of mercury was said to have been used in separations processes from 1954 to 1972 (Du Pont 1972a). The 80-ton amount is mentioned in several monthly reports and documents evaluating the underground waste tanks (Du Pont 1972a, 1971a). Use in 221-H and 221-F from 1974 to 1992 totaled about 3.5×10^4 kg or about 40 tons, and 11.3 tons (1.02×10^4 kg) was used in the tritium facilities from 1960 through 1987 (Kvartek et al. 1994). Horton (1973) used essential materials transaction reports to estimate consumption for the Separations Department to be 8.2 tons from 1960–1972. Routine use of mercury as a catalyst in H-Area in 1982 was about 3640 kg, which included recycled and new mercury (Holcomb and Emslie 1984). If we use the estimates for the years 1954-1972 and 1974-1992 of 80 tons and 40 tons, respectively, for the separations areas, then add 3.3 tons for these areas for 1973 and 11.3 tons for the tritium facilities from 1960-1987, a total of 135 tons results. We estimate that about 135 tons of mercury may have been used at the Site from 1954 to 1992.

Mercury Released to the Seepage Basins

Most of the mercury was discharged with the waste to underground tanks, but some of the liquid waste containing mercury went to the seepage basins. Neutralized low-activity waste sent to the separations area seepage basins was said to be 0.3% mercuric oxide (Christensen and Gordon 1983b). The February 1972 Monthly Progress Report for the separations areas said that more than 99% was sent to separations waste tanks and the rest went to the seepage basins (Du Pont 1972a). All of the evaporator overheads that contained mercury from separations processes were sent to the seepage basins. There were two evaporators in both the F-Area and H-Area (DOE 1987). In 1972, a heat exchanger was installed in H-Area to remove and recycle mercury from the 242-H evaporator overheads (Hurrell et al. 1987). This exchanger removed mercury by condensation and impingement, producing mercury that could be recycled into the dissolution process and decreased mercury releases to the seepage basin. Operational changes in F-Area in the 1970s also reduced mercury releases (Holcomb and Emslie 1984).

In 1977, the F-Area seepage basin was said to have received 150,000 gal of evaporator overheads each day when all facilities were running normally (Starks 1977). In recent years, mercury was recovered from the evaporator condensers and put into 1000-g bottles and returned to the process (Du Pont 1980). Kvartek et al. (1994) reports that technical personnel associated with waste management operations indicated that during normal operations, one bottle of mercury (about 32 kg) was recovered each month in H-Area and one bottle was recovered each year in F-Area. The SRP history for the separations area reported that mercury recovery averaged about

268 kg each year from the 241-1H and 242H evaporators (Du Pont 1988). Mercury was not recovered from the F-Area evaporators. About 25 kg of mercury was discharged annually to the seepage basins via evaporator overheads from buildings 242-F and 242-H. An additional 37 kg was collected annually at 242-H and returned to 221-H for reuse (Du Pont 1988). In 1988, evaporator overheads and other liquid effluent from the 200-Areas were sent to the Effluent Treatment Facility, which removed mercury.

Starting in September 1970, mercury discharges to the seepage basin were routinely monitored using trebler monitors (Du Pont 1971a). These samples were used to estimate releases to the seepage basins. The start-up or year that discharges began is assumed to be 1959 for H-Area, when HM processing began, and 1954 for F-Area.

About 12 different reports contained estimates of mercury releases to the seepage basins for different time periods. Different release estimates, many based on the same sampling data, are compiled in Table 18-7. Quarterly, monthly, daily, or annual estimates reported in pounds or tons were converted to kilograms per day or kilograms per year.

Table 18-7. Estimates of Mercury Discharged to the F-Area and H-Area Seepage Basins

Time period	Seepage Basin	Minimum (kg d^{-1})	Average (kg d^{-1})	Maximum (kg d^{-1})	Average (kg y^{-1})	Reference
F-Area Seepage Basin						
1959–1973	F				27	Horton (1974)
1959–1973	F				25	Kvartek, et al. (1994)
1955– 1970					24	Hurrell et al. (1987)
1955 – 1982	F				15.8	Christensen and Gordon (1983b)
Sept 1970–May 1972	F				20.5	Du Pont (1972b
Sept.–Dec. 1970	F	0.032	0.064	0.168	22.1	Du Pont (1972b); Du Pont (1973b)
Jan.–June 1971	F	0.0018	0.036	0.186	13.1	Du Pont (1972b)
July–Dec. 1971	F	0.0045	0.091	1.04	33.2	Du Pont (1972b)
Jan.–May 1972	F	0.027	0.032	0.772	11.7	Du Pont (1972b)
Jan.–June 1971	F		0.032		8.5	Bebbington (1971)
1971	F				24[a]	Horton (1974)
1971	F				23.6	Lower (1985)
1971	F				24	Hurrell et al. (1987)
1971	F	0.0018	0.063	0.92	23.3	Du Pont (1973b)
1972	F				7.2	Hurrell et al. (1987)
1972	F	<0.003	0.018	0.11	7.41	Du Pont (1973b)
1972	F				7.27	Lower (1985)
Jan.–March 1973	F	<0.009	0.0136	0.072	5.63	Du Pont (1973b)
1973	F				7.3	Horton (1974)
1973	F				5.6	Hurrell et al. (1987)
1973	F				5.68	Lower (1985)
1974	F				2.6	Hurrell et al. (1987)
1974	F				2.48	Du Pont (1974a)
1974	F				2.63	Lower (1985)

Table 18-7. Estimates of Mercury Discharged to the F-Area and H-Area Seepage Basins

Time period	Seepage Basin	Minimum (kg d^{-1})	Average (kg d^{-1})	Maximum (kg d^{-1})	Average (kg y^{-1})	Reference
1975	F				0.77	Hurrell et al. (1987)
1975	F				0.772	Lower (1985)
1976	F				2.9	Hurrell et al. (1987)
1976	F				2.91	Lower (1985)
1976—1977	F				1.7	Du Pont (1988)
1977	F		0.00136		0.25	Du Pont (1979)
1977	F				2.9	Hurrell et al. (1987)
1977	F				2.91	Lower (1985)
1987	F				0.82	Hurrell et al. (1987)
1978	F				0.818	Lower (1985)
1978	F				0.895	DuPont (1977a)
1979	F				1.36	Hurrell et al. (1987)
1979	F				1.36	Lower (1985)
1980	F				3.22	Hurrell et al. (1987)
1980	F				3.23	Lower (1985)
1981	F				0.682	Lower (1985)
1981	F				0.68	Hurrell et al. (1987)
1982	F				0.32	Hurrell et al. (1987)
1982	F				0.318	Lower (1985)
1983	F				1.32	Hurrell et al. (1987)
1984	F				7.41	Hurrell et al. (1987)
H-Area Seepage Basin						
1955—1970					96–136[c]	Killian et al. (1987b)a
1955—1970	H				102.2	Hurrell et al. (1987)
1955—1982	H				64.6	Christensen and Gordon 1983b
1959—1973	H				109	Kvartek et al. (1994)
Sept. 1970–May 1972	H				46.5	Du Pont (1972b)

Table 18-7. Estimates of Mercury Discharged to the F-Area and H-Area Seepage Basins

Time period	Seepage Basin	Minimum (kg d^{-1})	Average (kg d^{-1})	Maximum (kg d^{-1})	Average (kg y^{-1})	Reference
Sept. –Dec. 1970	H	0.018	0.38	1.19	29.7	Du Pont (1972b); Du Pont 1973b
Jan.–June 1971	H	0.009	0.091	0.382	33.2	Du Pont (1972b)
July–Dec. 1971	H	0.009	0.059	0.204	21.5	Du Pont (1972b)
Jan.–May 1972	H	0.022	0.068	1.13	24.8	Du Pont (1972b)
Jan.–June 1971	H	0.364	0.109	1.14	29	Bebbington (1971)
1971	H				28	Horton (1974)
1971	H				28.2	Hurrell et al. (1987)
1971	H				28.2	Lower (1985)
1971	H	0.009	0.077	0.38	28.1	Du Pont (1973b)
1971	H				28	Killian et al. (1987b)[a]
1972	H				21.8	Hurrell et al. (1987)
1972	H	0.0045	0.059	0.25	21.9	Du Pont 1973b
1972	H				22	Killian et al. (1987b)[a]
1972	H				21.82	Lower (1985)
Jan.–March 1973	H	0.014	0.054	0.177	19.6	Du Pont (1973b)
1973	H				15.4	Killian et al. (1987b)[a]
1973	H				15.4	Hurrell et al. (1987)
1973	H				21.8	Horton (1974)
1973	H				15.4	Lower (1985)
1974	H				7.7	Killian et al. (1987b)[a]
1974	H				7.7	Hurrell et al. (1987)
1974	H				7.73	Lower (1985)
1974	H				7.64	Du Pont (1974a)
1975	H				6.9	Killian et al. (1987b)[a]

Table 18-7. Estimates of Mercury Discharged to the F-Area and H-Area Seepage Basins

Time period	Seepage Basin	Minimum (kg d^{-1})	Average (kg d^{-1})	Maximum (kg d^{-1})	Average (kg y^{-1})	Reference
1975	H				6.9	Hurrell et al. (1987)
1975	H				6.91	Lower (1985)
1976	H				7.41	Lower (1985)
1976	H				7.4	Hurrell et al. (1987)
1976	H				7.4	Killian et al. (1987b)[a]
1976–1977	H				4.5	Du Pont (1988)
1977					8.3	Killian et al. (1987b)[a]
1977	H				8.2	Ross (1979); Fleming (1981); Smith (1981); Wilhite (1980)
1977	H				8.27	Hurrell et al. (1987)
1977	H				8.27	Lower (1985)
1978	H				6.954	Lower (1985)
1978	H				6.95	Hurrell et al. (1987)
1978	H				6.9	Killian et al. (1987b)[a]
1978	H				6.95	Du Pont (1977a)
1978	H				7	Ross (1979); Fleming (1981); Smith (1981); Wilhite (1980)
1979	H				4.4	Ross (1979); Fleming (1981); Smith (1981) (1980)
1979	H				4.36	Hurrell et al. (1987)
1979	H				4.4	Killian et al. (1987)a
1979	H				4.4	Lower (1985)
1980	H				2.36	Lower (1985)
1980	H				2.36	Hurrell et al. (1987)
1980	H				2.4	Killian et al. (1987b)[a]
1980	H				2.38	Ross (1979); Fleming (1981); Smith (1981) (1980)
1981	H				2.64	Ross (1979); Fleming (1981);

Table 18-7. Estimates of Mercury Discharged to the F-Area and H-Area Seepage Basins

Time period	Seepage Basin	Minimum (kg d^{-1})	Average (kg d^{-1})	Maximum (kg d^{-1})	Average (kg y^{-1})	Reference
						Smith (1981); Wilhite (1980)
1981	H				2.63	Hurrell et al. (1987)
1981	H				2.6	Killian et al. (1987b)[a]
1981	H				2.64	Lower (1985)
1982	H				8.95	Lower (1985)
1982	H				8.96	Ross (1979); Fleming (1981); Smith (1981); Wilhite (1980)
1982	H				8.9	Killian et al. (1987b)[a]
1983	H				24.5	Killian et al. (1987b)[a]
1983	H				24.5	Hurrell et al. (1987)
1984	H				27.6	Hurrell et al. (1987)
1984	H				27.6	Killian et al. (1987b)[a]

Table 18-7. Estimates of Mercury Discharged to the F-Area and H-Area Seepage Basins

Time period	Seepage Basin	Minimum (kg d^{-1})	Average (kg d^{-1})	Maximum (kg d^{-1})	Average (kg y^{-1})	Reference

[a] The data in Killian et al. (1987) were said to have been derived from Christensen and Gordon (1983b) and were estimated by the Health Protection Department of the SRS.

[b] The 1971 discharge to F-Area Seepage Basin was atypical and included an inadvertent loss of mercuric nitrate from 221-F in June and July (Horton 1974). Discharges were less in 1972 than the loss in 1971 because the Site began recovery of mercury in 242-H evaporator overheads.

[c] Killian's estimate was for 1630 kg from 1955 to 1972, a period of 17 years averaging out to 96 kg y^{-1}. The seepage basin probably received little mercury from 1955–1959, before the H-Area processes were running. Averaging the total of 1630 kg by the 12 years when the H-Area was contributing discharges results in an estimate of 136 kg y^{-1}.

Horton (1974a) made several estimates of mercury releases to the seepage basins; the highest estimate was 1636 kg in H-Area and 382 kg in F-Area from 1959-1974. The amount of mercury added after 1973 before the seepage basins were closed in 1988 was said to have been much less than the amount discharged from 1959–1973 (Horton 1974a, 1974b).

A Works Technical report for June 1972 reported mercury releases to F-Area and H-Area seepage basins for four, 6-month periods between September 1970 and May 1972 (Du Pont 1972b). This report summarized the results of the routine monitoring program started in September 1970. During that time (21 months), a total of 36 kg of mercury had been discharged to the F-Area seepage basins and 81 kg to the H-Area seepage basins. The weeks with the highest discharges for F-Area (the week ending July 16, 1971) and H-Area (the week ending October 9, 1970) were noted, but activities that occurred during the week that might have accounted for the higher releases were not mentioned. Table 18-7 summarizes average annual and daily average maximum and minimum concentrations based on the weekly composite samples provided in the report, Du Pont (1972b).

A monthly report from the Health Physics Section for April 1973 summarized the mercury discharges for the same time period based on the same weekly composite samples (Du Pont 1973b). The above normal 1971 F-Area discharge of 24 kg was said to have been due to the inadvertent loss of mercuric nitrate from 221-F in June and July. The H-Area discharges were also less in 1972 than in 1971 because recovery of mercury in 242-H evaporator overheads was initiated (Du Pont 1973c). The maximum, minimum, and average daily discharges and the annual averages from values in this report are summarized in Table 18-7.

A monthly report from December 1974 summarized releases through November to the F-Area seepage basin as 0.10 kg d^{-1}, for a total of 0.35 kg that month and a total of 2.28 kg for the year-to-date (Du Pont 1974a). We extrapolated Du Pont's values of 2.28 kg for 11 months to 2.48 kg for 12 months for Table 18-7. Releases to the H-Area were 0.021 kg d^{-1}, for a total of 0.741 kg for November 1974 and 7.15 kg year-to-date, correlating to a release of 7.7 kg for 1974 (Du Pont 1974a).

F-Area Seepage Basins

The first seepage basin constructed was in F-Area in 1954. Three more basins were built just south of F-Area and received effluent from 1954–1988.

Except for occasional peaks, one as high as 0.173 kg d^{-1}, the weekly composite samples for F-Area show a fairly constant discharge of about 0.045 kg d^{-1} in late 1970 and early 1971. Samples of the 242-F evaporator effluent suggest almost all of the mercury was from this source (Bebbington 1971).

Monitoring was also conducted from January through November 1985, while reprocessing of the Rocky Flats scrub alloy fuel was being done in F-Area. Mercury concentrations at the trebler leading to the F-Area seepage basin ranged between 20 and 191 μg L^{-1} (Kvartek et al. 1994).

Based on monitoring done in 1960–1970, 1975, and 1983, the total disposal mass to the F-Area seepage basins from 1954 to 1983 was estimated to have been 450 kg of mercury (Killian et al. 1987a), or an average of about 15 kg y^{-1} over the entire 30-year period.

H-Area Seepage Basins

The four seepage basins in H-Area, which were situated in series, were used from 1955 to 1988. The basins were closed in 1988 and have been capped. The water table is at a depth of about 6 m below the basins, and the basins are closer to the groundwater discharge zone along Four Mile Creek than the F-Area basins (Kvartek et al. 1994). In 1972, sediment analysis in H-Area indicated that about one-half of the estimated inventory for mercury could be accounted for in the top 6 in. of soil in the bottom of Basins 1 and 2, and most of the remaining mercury was said to have been accounted for in 14.5 ft of soil below Basin 4 (Holcomb and Emslie 1984). H-Area discharges of mercury were much greater than those from F-Area. Eleven samples of H-Area effluent/H-Area seepage basin influent were taken from September to December 1983. The concentration of mercury averaged 0.043 mg L^{-1}. A maximum value of 0.28 mg L^{-1} was reported (Killian et al. 1987b). Looney et al. (1988) calculated inventories for the H-Area seepage basins from influent data. Looney et al. estimated that 1805 kg of mercury had been discharged to the H-Area seepage basin through 1985. In 1971, a special sampling study was done to assess the mercury releases from the 242-H evaporator from the mercuric nitrate catalyst used for dissolving. Since October 1971, a trap was used to remove the mercury that settles from the overheads. The SRS experimented with other methods to remove more mercury from the overheads. During January through June 1971, 20.5 kg was removed at 242-H and 14.5 kg went from 242-H and 211-H to the H-Area seepage basin. The average mercury discharge rate to the basin was 0.109 kg d^{-1}. Before the program for mercury removal, the average amount discharged to H-Area seepage basins ranged from 0.36–1.14 kg d^{-1} (Bebbington 1971). Samples of the 211-H discharges and evaporator overheads did not account for the discharges going to the seepage basins. It was discovered that a manhole directly downstream of the evaporator (called the P-58 manhole) had pooled mercury in a low spot. The mercury was overflowing from the low spot into the sewer line that discharged into the seepage basin. The elemental mercury removed from the manhole weighed approximately 3.4 kg. A dam was then built to increase the trapping capacity and prevent continuous overflowing (Bebbington 1971). It is not clear if mercury was removed regularly after this. H-Area discharges ranged from 0.09 to 1.27 kg d^{-1}, decreasing to less than 0.16 kg after the mercury trapped in the manhole was removed (Bebbington 1971).

Horton (1974a) estimated that the mercury released to the seepage basins since startup totaled 1636 kg in H-Area (from 1959 through January 1974). In 1972, the H-Area discharges were reduced by passing evaporator overheads through a heat exchanger to remove mercury by condensation and impingement. The water discharged was also further purified with a hydroclone (Horton 1974a).

A series of memos (Ross 1979; Fleming 1981; Smith 1981; Wilhite 1980) about analysis of the seepage basins in F-Area and H-Areas for pH and mercury included a handwritten memo with ‘Bob Scaggs’ written across the top and a handwritten table that appears to tabulate the pounds of mercury that went through the trebler in H-Area for 1977–1982. These values are summarized in Table 18-7. The memo does not describe how the values were calculated from the trebler sample concentrations received from the laboratory analysis.

Analysis of the concentration and measurements of the average discharge per day led researchers in 1970 to conclude that perhaps the 242-H waste evaporators were not the principal source, as previously thought. “It also appears that some or all of the sources may be very

erratic," they said. For several days of the sampling, the 242-H evaporator was down and the source of mercury seemed to be Receiving Basin for Offsite Fuels (RBOF) waste. In September 1970, sampling was done over four, 3-day periods between September 8–25 (Bebbington 1971). Bebbington (1971) reported that from January to June 1970, 14.5 kg of mercury was discharged to the H-Area Seepage Basin at an average rate of 0.11 kg d^{-1}. During the same time period, 4.2 kg was discharged to the F-Area seepage basin at an average rate of 0.03 kg d^{-1}. (Bebbington 1971).

Clearly, the results of various studies done to characterize mercury releases to the basins are variable. Although data from longer time periods of monitoring might be considered more reliable, it is difficult to judge the quality of the estimates and rank or weight them accordingly. We might assume that about 26 kg y^{-1} was released to the F-Area seepage basins from 1955–1970 for a total of 416 kg for the 16-year period. From 22–24 kg may have been released in 1970 and 1971, decreasing to 7.3 in 1972 and 6.2 in 1974 for a total of 60 kg in those 4 years. From 1975 to 1980, releases were reported to be from 0.6 to 3 kg y^{-1}, totaling about 12 kg for the 6 years. From 1981–1984, about 10 kg may have been released. From 1985 to 1989, releases were lower, averaging less than 0.8 kg y^{-1} or less than 4 kg, and may have been close to zero because of the effluent treatment plant. The amount released to the F-Area basins from 1959–1989 totaled from these values is 501 kg. Because of the variability among the estimates (which may be 50%) and the sampling variability (approximately 15%), the true value is probably between 175 and 825 kg.

For H-Area, a similar tabulation suggests less than 1818 kg may have been released from 1959–1970. From 1970 to 1974, about 106 kg may have been released; from 1975–1980 about 37 kg; and from 1981–1985 about 95 kg. It is interesting that according to Killian et al. (1987b) (using data from Christensen and Gordon 1983a), releases from 1974 to 1979 averaged about 7.5 kg y^{-1} decreasing to 4.5 kg in 1979, less than 3 kg in 1980 and 1981, then increasing to 25 and 28 kg in 1983 and 1984. Discharges in 1971 and 1984 may have been the peak discharges. After 1985, releases were probably less than 8 kg y^{-1} but may have been close to zero because of the Effluent Treatment Facility. The true value for the amount discharged between 1959–1989 is probably between 1000 and 3000 kg. In comparison, the Olin Chlor-alkali plant may have discharged more than 9000 kg directly into the Savannah River between 1965 and 1970, an amount 3 to 4 times greater than the total amount that may have been discharged to all of the seepage basins during their operation.

Table 18-8. Estimates of Mercury Releases to the Seepage Basins for 1955–1989

F-Area		H-Area	
Date	Releases (kg)	Date	Releases (kg)
1955–1970	416	1959–1970	1820
1970–1974	60	1970–1974	106
1975–1980	12	1975–1980	37
1981–1984	10	1981–1984	95
1985–1989	3	1985–1989	45
1959–1989	501	1959–1989	2103

Kvartek et al. (1994) said that according to Horton (1974a), the chemical separations areas released 2000 kg of mercury into the seepage basins through the early 1970s.

It should be noted that mercury discharge to the seepage basin may not necessarily correlate with production. Discharge to the seepage basin is more likely related to mercury recycling and recovery efforts.

The transcript of a presentation given in 1992 by E.L. Albenesius included answers given in response to questions from the audience about mercury (Albenesius 1992). Someone, it is not clear who, estimated that the seepage basins (presumably F-Area and H-Area seepage basins) contained about 16 tons of mercury (Albenesius 1992). This estimate is much larger, nearly 10 fold higher, than any others we have found. It may be that the value may have been confused with estimates for the burial ground, which range around 10 tons.

The estimates of mercury released to the seepage basins are very uncertain. The mercury discharge estimates are based on measurements of mercury in liquid effluent entering the seepage basins. Analytical precision for the photometric method for mercury analysis in water was reported to be 10–15% for 1 to 5 $\mu g\ L^{-1}$ (ppb) mercury in water (Du Pont 1971b). The uncertainty in the source terms involves a 10–15% uncertainty in the analytical method. The sample error associated with the trebler proportional samplers was thought to be less than 3% (Wright 1955). The annual release estimates were based on limited data for short time periods of 1 week or several months; therefore, variability is high.

The Seepline and Four Mile Creek

Upper Three Runs Creek runs to the north of the seepage basins and Four Mile Creek runs to the south. Basin constituents seep to the underlying water table and then flow horizontally to the south toward Four Mile Creek. A small fraction may flow toward Upper Three Runs Creek (Haselow et al. 1990). Four Mile Creek is bordered by wetland areas and some portion of the groundwater moving to Four Mile Creek reaches the surface before entering the creek. The transition of upland to wetland vegetation defines what is called the seepline (Haselow et al. 1990). A small percentage of the water filtering through the seepage basin soil migrates into deeper groundwater but most of the groundwater reaches the surface at the seepline adjacent to Four Mile Creek. The flow of the groundwater to Four Mile Creek is estimated to be moving at a rate of about 15 cm d^{-1} (Looney et al. 1988). Simulations of flow in the system indicate that travel time for unretarded constituents from the basins to Four Mile Creek is about 10 years. Travel time from the basins to Upper Three Runs Creek is estimated to be about 70 years. A steady-state profile of the unretarded contaminant, tritium (tritated water), has developed between the basins and Four Mile Creek. Dixon and Rogers (1994) collected samples from five locations along the seepline in F-Area, five locations along the H-Area seepline, and three stream locations on Four Mile Creek. This sampling was the first in a series of three semiannual sampling events conducted to characterize the shallow groundwater outcropping to Four Mile Creek and associated wetlands. The results showed that groundwater at the seeplines was influenced by contaminates migrating from the seepage basins. Mercury levels were less than drinking water standards, but they had increased in Four Mile Creek from 1989 to 1992 along both seeplines (Dixon and Rogers 1994).

Dixon and Rogers (1994) said that studies in 1988 of the shallow groundwater outcropping at the Four Mile Creek and its associated seeplines and wetlands (published in Looney et al.

1988) were prompted by the observation that vegetation in wetland areas on the north side of Four Mile Creek seemed stressed. Both cadmium in F-Area and nitrate in both F-Area and H-Area seeplines were elevated above the drinking water standards. Haselow et al. (1990) collected soil, stream water, and seepline water samples from Four Mile Creek and its seeplines in 1988 and 1989 as a followup to the recommendations in Looney et al. (1988) and came to the same conclusions. The cadmium, nitrate, and manganese concentrations and the fact that the seepwater generally had a low pH confirmed that contaminants from the seepage basins were impacting the water chemistry along the seeplines. Discharges to the seepage basins were stopped in 1988, and the basins were capped and sealed in 1990. A quarterly tritium survey and a semiannual sampling program for metals, inorganics, and selected radionuclides have been conducted since 1992 (Dixon and Rogers 1994).

Well samples indicate that mercury has traveled through the soil below the seepage basins into groundwater in both the H-Area and F-Area. Mercury was also identified as a contaminant of soil adjacent to buried pipelines that transported liquids to the seepage basins (Kvartek et al. 1994). Groundwater monitoring wells show the groundwater contained mercury at an average concentration of 0.0035 mg L^{-1} and a maximum concentration of 0.0079 mg L^{-1} in 1993. Monitoring well data from 1994 demonstrate that the mercury plume from the H-Area seepage basins intersected the seepline along Four Mile Creek (Kvartek et al. 1994).

Three studies have analyzed the groundwater below F-Area intersecting the surface at the seepline along Four Mile Creek for mercury. Looney et al. (1988) found that dissolved mercury concentrations at the seepline downgradient from F-Area were below the detection limit. Subsequently, Haselow et al. (1990) found one sample above the detection limit, and Dixon and Rogers (1994) found a maximum mercury concentration of 0.005 mg L^{-1} at the seepline, which is within the range of background concentrations reported. The researchers concluded that mercury in the groundwater downgradient from F-Area had not yet reached the seepline at Four Mile Creek as of 1993 (Kvartek et al. 1994).

Bebbington (1971) stated that a study to determine quantities of mercury released to the environment concluded, based on analysis by the SRS and the Federal Water Administration the plant operations were not increasing the concentration of mercury in the Savannah River and that Four Mile Creek had mercury levels below the limit of detection, which at that time was 0.1 ppb. However, “significant quantities of mercury were being discharged to both the H-Area and F-Area seepage basins”. The Separations Department was asked to try to identify the source of mercury and reduce the discharge because the potential for mercury to move from the seepage basin to the creek existed (Bebbington 1971).

Studies have been done to determine how materials migrate from the seepage basins. Although most of the mercury is probably accounted for in the basin soils (Christensen and Gordon 1983b), data on mercury in soils, sediments, and suspended solids from Four Mile Creek indicate that mercury has migrated into the creek. A 1971 measurement indicated that about 0.36 g d^{-1} of mercury from 200-Area seepage basins was migrating into the creek (Lower 1985). In 1973, another analysis was said to confirm this (Du Pont 1973c). In a 1974 memo, Horton asserts that nearly all of the 3600 lb of mercury released to the H-Area seepage basins can be accounted for in the soil. However, later in the report he says, “Considering the concentrations measured and the quantities of soil they represent one can account for 99% of the mercury released to these basins but the large variation between samples make such accounting questionable” (Horton 1974a). The mercury in Basins 1, 2, and 3 was thought to be nonleachable. A laboratory leaching

study referred to by Horton (1974a) described mercury in the seepage basins as virtually immobile. The retention of mercury by the F-Area seepage basins had not been studied. Horton (1974a) recommended that the soil, vegetation, solubility, leachability, and volatility of mercury from the basins be studied further. Volatilization of mercury from the basins was not addressed.

Creek sediment sample concentrations also demonstrated an influence from the seepage basins. A high concentration (0.59 $\mu g\ g^{-1}$) in sediments of Four Mile Creek at Road C was thought to be due to either the addition of mercury to the creek by F-Area Cooling water, which enters just upstream of Road C, or to precipitation of mercury in the creek water with chemicals in F-Area discharges (Horton 1974a). The first explanation seemed most likely.

Mercury migration from the F-Area and H-Area seepage basins to Four Mile Creek is also addressed in the analysis of mercury monitoring data in Chapter 20. Mercury migration from the basins to the creek has occurred, but the rate of migration has not been quantified because the mercury concentrations in the downstream water samples and in the river have been below the limit of detection. A conservative rate of 22 $g\ d^{-1}$ of mercury migration to the creek was calculated based on the seepline concentrations and the rate of flow of creek water above and below the seepline. Using this rate, we calculated that about 240 kg of mercury may have been transported to the creek between 1959 and 1989. We recognize that mercury from the 200-Area seepage basins may not have reached Four Mile Creek until about 10 years after the seepage basins received liquid effluents containing mercury. Horton (1974b) calculated a migration rate of about 0.36 $g\ d^{-1}$, which would provide a release of 4 kg from 1959 to 1989. Based on flows of 15,900 $L\ min^{-1}$ above the basins and 30,850 $L\ min^{-1}$ below and concentrations of 0.023 and 0.020 $\mu g\ L^{-1}$ above and below, a transport of 0.53 $g\ d^{-1}$ above the basins and 0.89 $g\ d^{-1}$ below the basins was calculated. The difference, 0.36 $g\ d^{-1}$, was estimated to be the contribution from the basins (Horton 1974a). This calculation of the contribution of mercury to Four Mile Creek was based on the results of only two samples of suspended sediments.

It is unknown what fraction of the amount of mercury discharged to the seepage basins was transported to Four Mile Creek and eventually to the Savannah River. Several studies suggest that mercury in the seepage basins is not very mobile. The amount of mercury transported offsite from this source was said to be negligible, and concentrations in the River were below detection limits. Discharges to the creek in groundwater were estimated to be as much as 8 $kg\ y^{-1}$, based on monitoring data (Horton1974a; Lower1985; Kvarteketal1994). It seems unlikely that cumulative releases of mercury to the Savannah River would have exceeded 28 kg, assuming 0.27 of the 2604 kg discharged to the seepage basin reached the creek. The amount of mercury in the creek would have been reduced by sedimentation, absorption to sediments, retention by organic matter and biota in the swamp uptake by plants and animals in the creek, and other removal mechanisms that may have reduced the amount to reach the river to a very small fraction. The quantity that was transported from the seepage basins to Four Mile Creek and eventually the Savannah River seems to have never been measurable.

Chapter 5 describes calculations of cesium movement from the seepage basins to Four Mile Creek and through the swamp to the river. This research suggests that an average of 10% (ranging from 2 to 50%) of the cesium measured at Road C reached Road A, a distance of about 5 mi down Four Mile Creek. Four Mile Creek meanders about another 5 mi to the Savannah River. We might predict a similar reduction occurs for mercury so about 1% of the original activity in the seepage basins reaches the River. Horton (1974a) reported a concentration in Four Mile Creek below the H-Area seepage basins to be 0.20 $mg\ L^{-1}$. The maximum concentrations reported at the

seepline were 0.0015 mg L^{-1} reported by Looney et al. (1988). If we assume that the movement of cesium and mercury from the basins to the creek are similar, then the reduction may be similar. Ten percent of the maximum concentrations of mercury reported in Four Mile Creek for 1988 is 0.00015 mg L^{-1} or 0.15 μg L^{-1}. One percent of the maximum concentration at the seepline is 0.015 μg L^{-1}. These concentrations are below typical detection limits and less than the drinking water standard which is 0.002 mg L^{-1} or 2 μg L^{-1}.

Mercury in Waste Tanks

Most of the mercury in the canyon liquid wastes went into the high level waste tanks. Mercury in the waste tanks is discussed in the section on releases of mercury to air in Chapter 17. In 1992, a monitoring well with mercury levels exceeding the drinking water standard was reported, suggesting mercury may be leaking from underground waste tanks in the separations areas (Kvartek et al. 1994). However, the groundwater beneath the area has not traveled offsite. There is no evidence that these tanks have contributed to releases to surface water.

Other Surface Water Discharges

Mercury was identified as a contaminant present in three NPDES effluents discharged to streams in 1981, which are identified in Table 18-9 (Lower 1985).

Table 18-9. Mercury Discharged to Streams[a]

Outfall	Mercury concentration (μg L^{-1})	Transport (g d^{-1})	Receiving stream
A-001	0.3	0.34	Tim's Branch to Upper Three Runs Creek
F-008	0.2	0.77	Four Mile Creek
H-012	0.7	1.2	Four Mile Creek

[a] Source: Lower (1985).

Lower (1985) found that mercury concentrations were below the detection limit in stream waters at some undefined point below these three point sources. Sediments below these points were not analyzed for mercury.

M-Area releases of mercury have not been of concern. Measurements of mercury in Tim's Branch sediments in 1984, 1985, and 1986 were reported to be 0.02 mg kg^{-1} in sediments above M-Area and 0.4 mg kg^{-1} in sediments below M-Area. A concentration of 0.001 mg kg^{-1} in sediments at the "creek mouth" was also reported, but it is not clear whether this location was where Tim's Branch joins Upper Three Runs or where Upper Three Runs joins the river. These data were part of a set of presentation overhead transparencies and their source was not referenced. Details about the sample collection, number of samples, the time of year they were taken, and the analysis were not included. It is likely that the concentrations presented were averages (Specht 1991). An explanation for the higher concentration in Tim's Branch below M-Area than above was not given. As far as we know, mercury was not used in M-Area processes and was not discharged to Tim's Branch via the M-Area process sewers. Looney et al. (1987)

estimated that 0.05 kg y^{-1} of mercury was discharged to the settling basin based on effluent monitoring data from the1970s and 1980s.

The 1993 Federal Facility Agreement Progress Report identified mercury as a contaminant for the D-Area oil seepage basin (1952–1975); old TNX seepage basin (1954–1980); Gunsite 218 rubble pit (mid 1950s–mid-1960s); Central Shops sludge lagoon (1950s–1986); TNX groundwater, which does not appear to be migrating offsite; and Par Pond. The document identified seven other units with a potential for mercury contamination but for which no confirmation of contamination had yet been made. These sites were described by Kvartek et al. (1994). From 1974 to 1984, mercury in equipment, lights, etc. was probably buried at the sanitary landfill 740-G. Well samples indicate that mercury was released into groundwater from the sanitary landfill, but this groundwater does not outcrop to surface water and does not flow offsite (Kvartek et al. 1994).

The use and potential release of mercury from the tritium facilities in H-Area are described in the section on air releases of mercury in Chapter 17. Waste mercury from the facilities was buried in the burial grounds. Mercury in liquid effluents from the tritium facilities did not appear to be a concern.

Mercury in the Burial Grounds

Until the end of 1968, all of the mercury drained during pump maintenance and replacement was buried as waste, usually in 1-L polyethylene bottles that were placed in metal cans. The cans were buried in the old burial ground (643-E) of the Solid Waste Disposal Facility (SWDF), formerly called 643-G and also referred to as the burial ground (Orebaugh and Hale 1976). The SWDF is located between F-Area and H-Area. The original burial site was operated from 1953 until 1972 and was called the old burial ground. Another area, called the new burial ground (643-7E), was opened in 1972. The groundwater beneath the old burial ground SWDF flows toward Four Mile Creek. Mercury has been detected in groundwater beneath the SWDF. The maximum concentration detected in 1993 was 0.0046 mg L^{-1}. Soil studies suggest that mercury disposed of at the old burial ground SWDF was adsorbed onto soils as mercury vapor (Kvartek et al. 1994).

Beginning in 1968, metallic impurities and oxidized mercury in the mercury were removed through decanting and the mercury was reused. As a result of this recycling, the amount of mercury buried was reduced and no new mercury was added to the tritium facilities inventory after 1987 (Kvartek et al. 1994). Several reports estimate that between 1956 and 1968, before the Site began recycling mercury drained from the pumps in 1968, the tritium facility disposed of about 10,000 kg of mercury in the old burial ground (643-E) SWDF (Westinghouse 1992b). After 1968, mercury trapped in the gold traps, collected from leaks or spills, and mercury that could not be drained from equipment was buried. The old burial ground was filled in 1972 (Kvartek et al. 1994). In 1973, Horton estimated that 10 tons (20,000 lb or 9126 kg) of mercury had been buried. He considered that of the 1455 kg used from 1968–1972, about 454 kg were recovered from spills and was in storage. The remaining 1000 kg might have existed as unmeasured increased inventory in the process facilities or may have been buried in discarded process equipment (Horton 1973). Horton (1974b) reported that mercury burial was stopped in 1974. Hale (1973) reported that burial began in 1956 and that the records he reviewed did not show any burial of mercury after 1968. After 1968, and certainly after 1974, waste mercury was presumably stored in 234-H or recycled. In early 1968, the mercury drained from the pumps was returned back to

the pumps. Before 1968, mercury was obtained at low cost in a used form from Oak Ridge and purified, and efforts to recycle and conserve mercury were not made (Horton 1973). An example of the kind of disposal occurring after 1968 was found in Murphy (1986). Murphy suggested that two Sprengel pumps were drained and sent to the burial ground; and at least one Sprengel pump, which was replaced in 1985, could not be drained without a significant tritium release, so it was sent to the burial ground undrained and in a welded container. At that time there were 57 mercury pumps operating in the tritium facilities (Murphy 1986).

Most of the estimated 20,000 lb (10 tons) of waste mercury in the burial ground was thought to have been buried in 200–300 5-gal steel cans that contain 2–3 L (about 27–40 kg) of mercury each (Kvartek et al. 1994). Liquid mercury was placed in a 1-L polyethylene bottle, surrounded by two polyethylene bags. Two or three bottles in bags were then placed in a 5-gal steel can. These cans were buried throughout 44 acres of the low-level beta/gamma waste trenches in the 643-G burial ground (Oblath 1985). Burial depth and location were not specified in records or in instructions for burial. The trenches were 20 ft deep with a minimum burial depth specification of 4 ft. The polyethylene packaging was expected to have a lifetime greater than 100 years. A potential source of release to the soil would be rupture of the cans, polyethylene bags, and bottles because of physical contact with earth moving machinery, an activity that has not been reported (Kvartek et al. 1994; Orebaugh and Hale 1976). Reports from the mod-1970s state that the burial containers were expected to remain intact for several decades and routine monitoring to detect transport of mercury from the burial ground was not recommended at the time (Hale 1973; Horton 1973).

Mercury in the groundwater beneath the burial ground (both at 643-G and 643-7G) was monitored annually from 1977 to 1984, except for 1980. The groundwater monitoring data were said to indicate that mercury was not migrating from the burial ground and they suggested that no observable amounts of mercury were being released from the burial ground to Four Mile Creek in the 1980s (Oblath 1985). Because measurements in control wells (adjacent to the burial ground but under an area where no waste was buried) were similar to concentrations from samples taken from wells monitoring the burial ground, the mercury in the burial ground monitoring wells was thought to perhaps be due to sources other than the buried waste (Oblath 1985). This may have been true for the new burial ground, which contained little mercury wastes, but the consistent presence of mercury in some of the wells in the mid to late 1980s may be due to migration from mercury-containing waste in the old burial ground. This groundwater has not migrated offsite. Well samples and soil samples around the SWDF taken in 1992 suggest that mercury has leaked from storage containers into groundwater (Kvartek et al. 1994), but the groundwater has not traveled offsite.

Orebaugh and Hale (1976) mathematically modeled transport using 9000 kg of mercury as the source term, corresponding to the 10 tons of mercury buried in trenches reported in Horton (1973). They examined the soil redox potential, pH characteristics, and inorganic chloride and sulfate concentrations and concluded that mercury in the soil was in a stable complex. The modeling took into account the soil moisture, mercury solubility, soil composition, burial depth, soil porosity, and permeability to water and gases of the trench fill soil and surrounding and underlying soil. This soil had been studied extensively over the years because of a concern about the leaching of buried radioactive materials. Experiments were conducted to measure mercury vaporization from soil and determine the diffusivity of mercury vapor through soil with varying moisture contents and the solubility of mercury was also experimentally determined. In the course

of these experiments, the researchers determined that mercury on colloids could be an important transport mechanism in soil. They found that transport could occur through vaporization into the air or in water. Transport in water could occur by dissolved mercury in water percolating through the soil and soil water containing mercury on oxide colloids of iron and silicon. They concluded that colloidal suspension was the dominant mode of transportation of mercury from the burial grounds. An estimate of the amount of mercury that could travel from the 9000-kg of buried mercury to the water table and travel horizontally to surface water was 219 mg h^{-1}, 206 mg h^{-1} on colloidal suspension of iron and silicon oxides, and 13 mg h^{-1} of dissolved mercury. As a worst case (which assumed that all of the mercury in bottles, bags, and steel cans was released to the soil), they estimated that this flux could contribute 0.2 μg L^{-1} to Four Mile Creek using a flow of 10^6 L h^{-1}. The drinking water standard reported by Orebaugh and Hale for that time was 5.0 μ g L^{-1}. The authors concluded that the contribution from the burial ground would not contribute to an environmental impact (Orebaugh and Hale 1976; Kvartek et al. 1994). Kvartek et al. (1994) calculated that the exposure an adult worker would receive by consuming maximally contaminated water directly from Four Mile Creek, daily for 30 years, corresponded to a risk of 1.9×10^{-6} mg kg-d^{-1}. The risk estimated for an offsite individual consuming the water for 70 years was 5×10^{-6} mg kg-d^{-1}. Neither estimate exceeded the EPA's reference dose for mercury.

We have characterized the burial of mercury to the extent possible. At this time, development of a source term for releases to surface water from the burial grounds is not warranted because groundwater under the burial ground area has not migrated offsite and no problems with surface water runoff or related discharged have been documented.

Mercury Spills

We did not find documentation of mercury spills to surface water. No large spills of mercury were reported in the fault tree databanks or documents reporting spills to the states or EPA. A document about a spill at TNX of an unknown amount of water containing 14 μg L^{-1} mercury reported that concentrations in soil below the spill were measured and ranged from 46 to 545 mg g^{-1}. The researchers estimated that a soil volume of 20 ft^3 might contain mercury in excess of 20 mg g^{-1} (Looney 1994). Monitoring studies have not provided any evidence of large spills of mercury to surface waters onsite.

Summary

Based on available data, it is not apparent that Site activities have contributed to elevated mercury concentration in any offsite environmental media. Chapter 20 provides a detailed discussion of mercury concentrations in environmental media.

NICKEL

M-Area

Nickel salts were released to M-Area liquid effluent from the plating rinse tanks. This was recognized as a problem, and methods to reduce the amounts were being studied in 1970 (Hardt 1970). The *Process Waste Characterization Flow Reduction Study* for M-Area, published in

1983, estimated that the annual use of nickel chloride for Building 313-M, used in a form that was 24.5% nickel, was about 2273 kg (Lockwood Greene 1983).

M-Area effluent discharges to the M-Area settling basin, based on effluent monitoring data from a time period when the average production rate for 313-M was 17,500 cores per month, were estimated by Looney et al. (1987) to have been 1500 kg y^{-1} for nickel. An M-Area settling basin characterization study published in 1985 estimated that the basin then contained 3585 kg of nickel in the sludge, 53 kg in the soil (to a depth of 6 ft), and 3.5 kg in the liquid for a total of 3640 kg. Accumulation of metals was also assessed for sediments of the overflow ditch and Lost Lake. This information was not compiled here because these contaminants did not reach surface water leaving the Site (Colven et al. 1985). In 1984, a partial failure of the wooden dam at Steed's Pond, which received M-Area effluents, occurred and samples were taken to determine if radioactive materials were migrating from the pond. The 1985 Annual Environmental Report reported that the maximum nickel concentration in Steed's Pond sediment was 5300 μg g^{-1}, about 500 times higher than an average background of 10 μg g^{-1}, (ranging from 3-18 μg g^{-1}) in the southeastern U.S. (Pickett 1990). From March through May 1985, the influent to and effluent from the M-Area settling basin was sampled and analyzed weekly for 10 weeks. Based on these sampling data, a removal efficiency was calculated for the basin (the influent concentration minus the effluent concentration divided by the influent concentration). The influent concentration of nickel was 5.95 $\pm$ 5.0 mg L^{-1}, the effluent concentration was 0.573 $\pm$ 0.98 mg L^{-1}, and the removal efficiency was estimated to be about 90% (Colven et al. 1985).

A 1970 memo described releases to Tim's Branch from M-Area processes and estimated that 3544 kg of nickel sulfate and 486 kg of nickel chloride were released each year to the process sewers that went into Tim's Branch (Hardt 1970). A 24-hour composite sample of sewer effluent taken in December 1981 when the plant was operating showed nickel concentrations of 0.89 mg L^{-1} in sewer effluent going to the settling basin and 0.118 mg L^{-1} in sewer effluent going to Tim's Branch. From this data, 13,635 kg were estimated to have accumulated in the M-Area settling basin sludge and sediment, corresponding to an average discharge rate of 1.4 kg d^{-1} (Merz 1982; Bradley 1981). If we assume average discharges to the Tim's Branch were 2.6×10^8 gal y^{-1} or 9.84×10^8 L y^{-1} (Monier 1970; Hardt 1970), then the concentration of 0.118 mg L^{-1} reported by Merz would lead to an estimate of 116 kg discharged each year or 3600 kg of nickel total from 1952 to 1982, a 31-year period, assuming 365 d y^{-1} and 3.78 L gal^{-1}. Measurements of nickel in Tim's Branch sediments in 1984, 1985, and 1986 were reported to be 2.4 mg kg^{-1} in sediments above M-Area and 783 mg kg^{-1} in sediments below M-Area (Specht 1991). Monthly sampling of Tim's Branch and Upper Three Runs Creek sediments in 1985 and 1986 showed that nickel concentrations clearly decreased with distance from M-Area. The furthest sampling point was in Upper Three Runs Creek, just downstream of the confluence with Tim's Branch. Mean concentrations $\pm$ standard error at this point were 13.1 $\pm$ 1.2 mg kg^{-1} for nickel, compared to 783.0 $\pm$ 228.9 mg kg^{-1} just downstream of where the outfall enters Tim's Branch (Pickett 1990).

Releases of nickel to Tim's Branch may have ranged from 116 kg to more than 2000 kg y^{-1}, based on the amount of nickel compounds said to have been used and discharged in the 1970s.

Concentrations of nickel in water and sediments of the Savannah River have been similar upstream and downstream of the SRS, and nickel levels in fish have been less than levels of concern (Westinghouse 1996a).

NITRATES AND NITRIC ACID

The primary human health concern associated with nitric acid discharges is the formation of nitrates that, if they contaminate drinking water, can cause adverse health effects, especially in babies and infants. Nitrates were used and produced by several processes onsite. The H-Area, F-Area, and M-Area seepage basins received process wastewater consisting primarily of nitric acid and sodium nitrate (Peralta and Lewis 1982), and nitrates were released to Tim's Branch and Four Mile Creek.

To allow comparison, amounts in gallons, pounds, and kilograms were converted to tons. In the 1950s, 1960s and much of the 1970s, nitrate measurements for drinking water samples were reported as total nitrate (NO_3). Sometime around 1977, the total nitrogen or NO_3-N was reported. Many of the site documents report 'nitrates', 'as NO_3' or 'NO_3-N, or 'nitrogen' so that it is possible to tell which reporting method was used. In many of the reports the drinking water standard was given as a comparison value. We have assumed that documents reporting a drinking water standard of 45 mg L^{-1} reported concentrations in mg L^{-1} as nitrate, rather than NO_3-N which corresponds to a drinking water standard of 10 mg L^{-1}. The amount of NO_3 is related to the amount of NO_3-N measured by the molecular weight ratio of NO_3/N which is 62 ÷ 14 or 4.43. To facilitate comparisons, and ease in reproducing tables, the old reporting preference was used. Any data reported as NO_3-N was converted to NO_3 by multiplying by 4.43. If the new reporting preference is more familiar to the reader, the NO_3 values could be converted to NO_3-N by dividing these values by 4.43.

Separations Areas

Nitrates were primarily a concern for the releases to surface water or to the seepage basins and underlying groundwater that outcrops to surface streams. Nitrate in Four Mile Creek comes from groundwater beneath the F-Area and H-Area seepage basins outcropping into the creek and from surface water runoff. Information about nitrate concentrations in the seepage basins, groundwater, seepline or outcrop areas, and Four Mile Creek was reviewed and is summarized below.

Seepage Basins

The separations areas have routinely released large volumes of liquids containing nitrates to seepage basins since their startup in 1954. Nitric acid was the primary source of nitrates. Sodium nitrate was produced from neutralization of nitric acid solutions with sodium hydroxide in the uranium recovery process (Monier 1970). For example, plutonium was separated from uranium by adjusting the oxidation state of plutonium to +3 by using ferrous sulfamate and hydroxyamine nitrate as a reductant in the Purex process (Bebbington 1990).

A 1974 memo characterizing nitrate releases to the separations area seepage basins estimated the average annual release based on measurements taken from 1961 through 1970. About 295 tons y^{-1} was discharged to the F-Area seepage basin and 267 tons y^{-1} to the H-Area seepage basin for a total of 562 tons y^{-1}. The total for this 10-year time period was 6182 tons. The releases of nitrate to the seepage basin was determined by considering the total cation content of

the seepage basin wastewater and assuming all of the anions to be nitrate. A study done in 1972 suggested that more than 90% of the anions were nitrate (Horton and Carothers 1974).

Table 18-10 shows an example of the release estimates corresponding to weekly nitrate analysis of liquid released to the seepage basin from 5/25/73 to 7/6/73. These release estimates to the seepage basins were based on nitrate analyses and volume measurements taken by the trebler samplers.

Table 18-10. Estimates of Nitrate Releases to the Seepage Basin Based on Anion Analysis in 1972[a]

	Nitrate release estimates (tons y^{-1})	
Week ending	F-Area	H-Area
5/25/73	177	91
6/01/73	390	57
6/08/73	211	343
6/15	–	211
6/22	257	200
6/29	650	148
7/6	333	268
Average	336	198

[a] Source: Horton and Carothers (1974).

Holcomb and Emslie (1984) reported that in 1975, an estimated 50.5 tons of nitrate was released to F-Area seepage basin and 113.5 tons to the H-Area seepage basin, for a total of 164 tons that year. Hazardous constituent inventories from 1983 reported 3090 tons of nitrate in the F-Area seepage basin and 2900 tons of nitrate in the H-Area Seepage basins (Christensen and Gordon 1983b; DOE 1987). Eleven samples of F-Area and H-Area effluent/ seepage basin influent were taken from September to December 1983. In the F-Area seepage basin the concentration of nitrate averaged 538 mg L^{-1}. A maximum value of 1950 mg L^{-1} and a minimum value of 67 mg L^{-1} was reported (Killian et al. 1987a). In the H-Area seepage basin the concentration of nitrate averaged 1220 mg L^{-1}. A maximum value of 6740 mg L^{-1} and a minimum value of 90 mg L^{-1} was reported (Killian et al. 1987b). Looney et al. (1987) calculated inventories for the H-Area seepage basins from influent data, which included 1 million kilograms, or 1100 tons, of nitrates. The nitrate inventory was based on assuming 115.5 tons y^{-1} was released for the years 1971–1985 (Looney et al. 1987). In the Environmental Information Documents for the H-Area seepage basins and the F-Area seepage basins, Killian et al. (1987a and 1987b) said nitrate releases varied but averaged about 242 tons y^{-1} in H-Area and 257.7 tons y^{-1} in F-Area, based on measurements from 1961–1970, 1975, and 1983. Killian et al. (1987a and 1987b) compiled tables of nitrate releases, based on data from Christensen and Gordon (1983a) and 1983 and 1984 quarterly monitoring data. Their estimates for 1961–1970 agree well with those of Horton and Carothers (1974) on which all of the subsequent estimates were probably based.

We made release estimates for the years in the shaded areas of Table 18-11 by relating the 1961–1970, 1975, and 1983 estimates (Horton and Carothers 1974; Holcomb and Emslie 1984; Killian et al. 1987a and 1987b) to production during these years and attempting to correlate production to release amounts during the years that releases were not reported. The estimates for each year are shown in Table 18-11.

Table 18-11. Estimates of the Amount of Nitrates Released to the F-Area and H-Area Seepage Basins Made by Horton and Carothers (1974) and Later Killian et al. (1987a and 1987b)[a]

Year	Tons to F-Area seepage basin	Tons to H-Area seepage basin	Total tons to both seepage basins	Predicted amount eventually released to Four Mile Creek tons to seepage basins × 0.27
1954	70	76	146	39[b]
1955	245	168	413	111
1956	271	419	690	186
1957	46	431	477	129
1958	0	202	202	54
1959	234	42	276	74
1960	294	110	404	109
1961	91	99	190	51
1962	309	186	496	134
1963	433	310	744	201
1964	820	562	1383	373
1965	175	372	547	148
1966	153	175	328	88
1967	194	204	398	107
1968	244	157	402	108
1969	255	402	658	177
1970	270	204	474	128
Average 1961–1970	295	267	562	152
1971	165	644	809	218
1972	234	250	484	131
1973	247	248	495	134
1974	229	95	324	87
1975	50	113	162	44
1976	262	115	377	102
1977	125	67	192	52
1978	182	84	266	72
1979	172	48	220	59
1980	242	79	321	87
1981	278	126	404	109
1982	193	101	294	79
1983	95	118	212	58
Average 1961–1970, 1975, 1983	258	242	499	135
1984	235	120	355	96
1985	217	147	364	98
1986	194	120	314	85
1987	219	78	297	80
1988	192	33	225	60
1989	59	0	59	16

[a] The shaded values were estimated from reported values based on correlations to F-Canyon and H-Canyon production (mass of U or number of U-235 tubes charged to the dissolvers) for each year or time period.
[b] These amounts are theoretical. Realistically, it may have taken 4 to 9 years for contaminants in the seepage basins to travel to Four Mile Creek.

Groundwater

Studies of nitrate in groundwater in 1968 and 1969 showed nitrate concentrations in the main flow of groundwater from the basins to the creek ranging from 100–300 mg L^{-1} in H-Area and from 100–200 mg L^{-1} in F-Area. The springs along the edge of the swamp bordering the creek contained maximum concentrations of 220 mg L^{-1} in F-Area and 240 mg L^{-1} in H-Area (Horton and Carothers 1974), compared to 3 mg L^{-1} measured in background groundwater (Holcomb and Emslie 1984).

The nitrate concentrations in groundwater were determined in 1973 and 1974 by sampling 65 wells in F-Area and 50 wells in H-Area. The concentration in the main flow path from the basins to Four Mile Creek ranged from 100 to 300 mg L^{-1} in F-Area and from 100–200 mg L^{-1} in H-Area (Horton and Carothers 1974). For comparison, the drinking water standard at that time was 45 mg L^{-1}. The report's authors estimated that it would take more than a decade after seepage basin operation stops for the nitrate in the groundwater to be reduced to acceptable levels based on what was known about biological removal and reduction of nitrate to nitrogen (Horton and Carothers 1974).

Seepline and Four Mile Creek

Nitrates from the seepage basins outcropped to Four Mile Creek. Extensive sampling of the seepline was conducted in 1989 and 1990 and was reported by Haselow et al. (1990). Levels of nitrate were greater than the drinking water standard at both the F-Area and H-Area seepline, but levels in the creek were less than one-half the standard. In 1974, the springs or outcrop areas along the edge of the swamp bordering Four Mile Creek had maximum concentrations of 975 mg L^{-1} in F-Area and 1063 mg L^{-1} in H-Area (Horton and Carothers 1974). In 1982, the average concentration of nitrate discharged to the F-Area and H-Area basins was estimated from measurements to be 5316 mg L^{-1} in F-Area and 2658 mg L^{-1} in H-Area. The nitrate concentration at the 'outcrop location' at Four Mile Creek was reported to be 961 mg L^{-1} for F-Area and 1072 mg L^{-1} (as NO_3-N) for H-Area, suggesting a reduction in the concentrations between the basins and the seepline of 6 fold for F-Area and 2.4 fold for H-Area (Peralta and Lewis 1982).

Dixon and Rogers (1994) collected samples from five locations along the seepline in F-Area, five along the H-Area seepline, and three stream locations on Four Mile Creek. This sampling was the first in a series of three semiannual sampling events aimed at characterizing the shallow groundwater outcropping to Four Mile Creek and associated wetlands. The results showed that groundwater at the seeplines was influenced by contaminates migrating from the seepage basins. Nitrates were found to be greater than the drinking water standard at the H-Area seepline (Dixon and Rogers 1994).

In a short-term study, the flow of the creek and nitrate concentrations were determined using a continuous paddlewheel sampler and U.S. Geological Survey flow gauge at six locations from August 13 to September 24, 1974. From these data, estimates of the amount of nitrate in the creek and flowing past each location were made (see Table 8-12).

Table 18-12. Nitrate Concentrations in Four Mile Creek in 1974

Location	Average nitrate concentration (mg L^{-1})	Maximum nitrate concentration (mg L^{-1})	Quantity of nitrate calculated to flow past each location (tons y^{-1})
H-Area cooling water	0.28	0.58	0.34
H-Area cooling tower effluent	2.3	3.6	2.1
Below H-Area Seepage Basins 1, 2, and 3	2.9	12.8	19
Below H-Area Seepage Basin 4	9.9	18.0	88
F-Area effluents	0.03	0.08	0.08
Below F-Area Seepage Basins 1, 2, and 3	9.6	17.1	153

[a] Source: Horton and Carothers (1974). Reported as NO_3

The authors concluded that the large increases at locations below each of the basins demonstrated that the seepage basins were the major contributor of nitrate. The text explains that based on these measurements, predictions were made about releases to the creek each year. Releases were estimated to be 88 tons y^{-1} for the H-Area basins and 65 tons y^{-1} for the F-Area basins or a total of 153 tons (Horton and Carothers 1974). These studies summarized in this document suggest that 27% (153 tons found in the creek ÷ by 562 tons released to the basins) of the nitrate released to the seepage basins emerges into the creek. They acknowledge that the estimates are based on a limited number of creek measurements taken over a short time and suggest monthly analysis be conducted over several years to determine more accurate values. The results were said to agree with previous findings that the input of nitrate to Seepage Basin 1 of F-Area averaged 1200 mg L^{-1} and the maximum groundwater concentration in the main flow toward Four Mile Creek was reported to be about 300 mg L^{-1}. These values suggest a reduction factor of 0.25 (Horton and Carothers 1974).

Determining the amount of nitrate reaching Four Mile Creek was of interest for understanding dilution of other contaminants as they move from the seepage basin to groundwater and then into Four Mile Creek. As groundwater flowed into Four Mile Creek, the nitrate in it was diluted to about one-fifth of the drinking water standard or about 8.7 mg L^{-1}. Horton and Carothers (1974) said that the concentrations in the creek below the H-Area seepage basins, which averaged about 9.9 mg L^{-1}, were diluted at least 25 times by the C-Area cooling water. Therefore, the concentrations flowing to the river would have averaged less than 0.4 mg L^{-1}. This dilution seems to be based on predicted flow levels rather than concentration measurements. The authors predicted that if all 560 tons y^{-1} of the nitrate discharged to the basins were to enter the creek, the concentration would be 80% of the drinking water standard (presumably about 36 mg L^{-1}). The authors recommended that a monthly nitrate analysis of Four Mile Creek water below the seepage basins be continued to determine the fraction of released nitrate that emerges into the creek and that the rate of chemical reduction of nitrate to nitrogen be determined in the laboratory (Horton and Carothers 1974). Interestingly, the 1974 concentration estimates based on 1973 trebler sample data suggest a reduction factor of 0.32 for H-Area (88 tons/267 tons), 0.22 for F-Area (65 tons/295 tons), and 0.27 for both basins combined (156 tons/540 tons). Based on releases to the seepage basins and calculated dilution and retardation values derived from measurements, we might conclude that as much as 3888 tons (14,402 tons in

the 36 years from 1954 to 1989 × 0.27 y^{-1} = 3888 tons) of nitrates may have reached Four Mile Creek. The estimates are compiled by year in Table 18-11. The creek water was diluted again by at least a factor of 25 as it mixed with C-Area cooling water before entering the river.

In summary, a 6-fold reduction for F-Area and a 2.4-fold reduction for H-Area in the concentrations of nitrates from the basin to the seepline were predicted, based on 1982 monitoring data. About 27% of what was released to the seepage basins was predicted to reach Four Mile Creek based on 1973 monitoring data. Further dilution in Four Mile creek was predicted to be about 25 fold from adding C-Area cooling water to Four Mile Creek. It is likely that further reduction and dilution occurred in the swamp before Four Mile Creek water entered the Savannah River. River water quality monitoring data do not indicate that Four Mile Creek has a significant impact on nitrate concentrations in the river.

M-Area

Large amounts of nitric acid were used in the M-Area processes. Nitric acid was routinely drained from cleaning tanks and discharged to the sewers. It leaked from process equipment and sewer drains. Not surprisingly, there is a nitrate groundwater plume underneath M-Area that has been well characterized in recent years (Gordon 1982; Merz 1982). In a study of waste reduction methods, Hardt (1970) estimated that 135.75 tons of salts (probably primarily nitrates and phosphates), including 83.2 tons of sodium nitrate and 2.35 tons of aluminum nitrate, were being released to Tim's Branch annually (Hardt 1970).

In a file of memos dated from 1968 to 1970, a one-page memo from J.A. Monier estimated volumes and concentrations of chemical wastes discharged to Tim's Branch in 300-Area effluent each year based on production data for 4 months from November 1968 through February 1969 (Monier 1970). References for the estimates, the data from which they were determined, and how they were determined were not included in the report. Monier (1970) estimated that 25,000 gal of 50% nitric acid and 35,500 gal of sodium nitrate solution had been discharged to Tim's Branch in 1 year.

In 1981, Bradley estimated that 0.39 tons of nitrates were discharged each day to the M-Area settling basin. Based on effluent monitoring data from a 13-year time period when the average production rate for 313-M was 17,500 cores per month, M-Area effluent discharges to the M-Area settling basin were estimated by Looney et al. (1987) to have been about 55 tons y^{-1} for nitrate.

Concentrations of nitrates measured in a 24-hour composite sample of sewer effluent sampled in December 1981 when the plant was operating, were 14.6 mg L^{-1} (64.7 mg L^{-1} as NO_3) in effluent going to the settling basin and 10.2 mg L^{-1} (45 mg L^{-1} as NO_3) in effluent going to Tim's Branch. The ratio of inflow and outflow concentrations of nitrate for the M-Area settling basin was calculated to be 13, suggesting a reduction of more than 90% (Merz 1982).

From March through May 1985, the influent to and effluent from the M-Area settling basin was sampled and analyzed weekly for 10 weeks. Based on these sampling data, a removal efficiency was calculated for the basin (the influent concentration minus the effluent concentration divided by the influent concentration). The influent concentration of nitrate was 361 $\pm$ 180 mg L^{-1}, the effluent concentration was 156 $\pm$ 70 mg L^{-1} as NO_3-N, and the removal efficiency was estimated to be about 57% (Colven et al. 1985A *Process Waste Characterization Flow Reduction Study* from 1983 (Lockwood Greene 1983) described process solutions, volumes,

concentrations, and typical flowthroughs for M-Area processes. Consumption data from plant records for 1982 and 1983 were compiled and the content of solvents, nitrates, and metals in effluent from various processes were estimated as a part of the characterization. Based on the concentration of major anions and cations in bulk chemicals, a calculation was made to project the annual use in pounds per year. The study estimated the annual use of 50% nitric acid to be 164.3 tons by 320-M, 260.5 tons by 313-M, and 39.1 tons of 100% nitric acid by 320-M, for a total of 251.6 tons of 100% nitric acid used each year in M-Area (Lockwood Greene 1983). In a study of waste reduction methods, Hardt (1970) estimated that each year about 36 tons of acids, which included about 33.5 tons of nitric acid, were being released to Tim's Branch. These amounts represented 100% nitric acid solutions, calculated from larger amounts of more dilute solutions actually discharged (Hardt 1970).

Nitric acid discharged to the sewer and neutralizer from the 313-M core plating line process in 1983 ranged from 37.35 to 135 tons y^{-1} according to flow process charts compiled by Martin in 1983 and reproduced in a report by Colven et al. (1985). Colven et al. (1985) examined production and throughput data and estimated the amount of chemicals released into effluent in 1982:

- The Cap cleaning process in Building 313-M was said to have released 3.5 tons of sodium nitrate and 16.0 tons of nitric acid to the sewer
- 33.4 tons of nitric acid were discharged to the sewer from the final slug cleaning process
- Building 313-M uranium recovery process discharged an estimated maximum of 53 tons and a minimum of 19.1 tons of sodium nitrate (based on production)
- Core recovery process in Building 313-M released 27.6 tons of sodium nitrate and 0.3 ton of nitric acid
- 321-M component cleaning process discharged 1.5 ton of sodium nitrate and 1.5 ton of nitric acid
- 321-M tube cleaning process discharged 7.85 tons of sodium nitrate and 10.25 tons of nitric acid
- 320-M cleaning room processes discharged 5.52 tons of sodium nitrate and 2.8 tons of nitric acid (Colven et al. 1985). The discharges for all of the processes examined totaled 64.3 tons of nitric acid and 237.3 tons of sodium nitrate.

In 1982, the amount of nitric acid released in 1982 for all of M-Area totaled 302 tons (Colven et al. 1985). If the plant had released 302 tons each year for the entire time period M-Area facilities operated before the effluent treatment plant started up in 1985, a total, very conservative estimate of about 9664 tons results. A more reasonable estimate can be made using estimates of the production during various time periods. The production in 1982 was probably about 90% of the production in 1985, leading us to predict that about 335 tons of nitrates may have been released in 1985. If we presume that production from 1954–1959 was 20–40% of the production in 1985, production from 1960–1981 was 30–80% of that in 1985 and that production in 1983 and 1984 was equal to that in 1982, that production in 1986 equaled 1985 and that production in 1987–1989 was about 40% of that in 1985, then we can estimate the following:

67–135 tons y^{-1} were discharged from 1954–1959
100–268 tons y^{-1} were discharged from 1960–1981
about 302 tons y^{-1} were discharged from 1982–1984
about 335 tons y^{-1} were discharged from 1985–1986

about 135 tons y^{-1} were discharged from 1987–1989.

It is difficult to estimate how much of the nitrate in effluents went to the seepage basins compared to how much went to Tim's Branch. Estimates of the amounts of waste solvents discharged to the seepage basins and Tim's Branch made by Christensen and Brendall (1981) suggest that the volumes discharged were variable but about 15–60% of the total went to the creek in the late 1970s. If we assume 100% of the discharges went to the creek before 1959, 60% between 1959 and 1985, and 20% after 1985 when the treatment plant was operating, the following conservative estimates for the amount of nitrates that may have been discharged to Tim's Branch are the result:

67–135 tons y^{-1} from 1954–1959, or a total of 402–810 tons for those 6 years
60–160 tons y^{-1} from 1960–1981, or a total of 1320–3520 tons for those 22 years
about 181 tons y^{-1} from 1982–1984 or a total of 543 tons for those 3 years
about 200 ton y^{-1} in 1985 and 67 tons in 1986 after the treatment plant was operating
about 27 tons y^{-1} from 1987–1989 or a total of 81 tons for those three years.

These estimates total a range of about 2613–5220 tons for the 36-year time period of M-Area operation, or about 72 to 145 tons y^{-1}. This is a very rough estimate of the discharges to Tim's Branch based on very little information. Estimates for the portion of M-Area effluent discharged to the Tim's Branch from 1981 and production information for 1982–1989 was used to estimate values for the entire 36-year period. Release estimates for 1954–1981 are especially uncertain.

Our estimates compare surprisingly well with the Hardt (1970) estimate of 85.5 tons and the 142 tons estimated by Monier (1970). The 1981 daily discharge estimate of 0.39 tons d^{-1} by Bradley (1981) totals 142 tons at an operating rate of 365 d y^{-1}.

Information on dilution and reduction of nitrate from Tim's Branch to the Savannah River is not sufficient to determine the release of nitrate to the river from the discharge of about 145 tons y^{-1} from M-Area to Tim's Branch. The river water quality monitoring data suggest that the discharge did not cause nitrate levels downstream of the SRS to be significantly different from levels upstream. Sampling data from 1973 found an average of 0.45 mg L^{-1} in Tim's Branch near Upper Three Runs and the same concentration in Tim's Branch at Highway 278. Average nitrate levels in the Savannah River were similar, averaging about 0.30 mg L^{-1} (Du Pont 1967).

Water Quality Monitoring for Nitrates

Nitrate analysis was a part of the Savannah River water quality monitoring program initiated in 1959. In general, nitrate concentrations downstream of the SRS were not different from concentrations upstream. Nitrates in Site streams were less than 9 mg L^{-1} (as NO_3) except for Four Mile Creek at Road A-7, which averaged about 35 mg L^{-1} in the mid-1970s and reached maximum concentrations of about 80 mg L^{-1} in later years. Average nitrate concentrations reported in the health physics section of the June 1973 monthly report for Site streams are shown in Table 18-13.

Table 18-13. Average Nitrate Concentrations Reported in June 1973[a]

Sampling location	mg L^{-1} (NO_3-N)	mg L^{-1} (NO_3)
Upper Three Runs at Highway 278	0.45	1.99
Upper Three Runs at Road C	0.22	0.97
Upper Three Runs at Road A	0.20	0.89
Tim's Branch Near Upper Three Runs	0.45	1.99
Four Mile Creek at Road A-7	7.9	34.99
Steel Creek at Road A	0.27	1.19
Beaver Dam Creek near swamp	1.2	5.32
Savannah River at 681-3G	1.3	5.76
Lower Three Runs at Patterson's Mill	0.16	0.71
Lower Three Runs below Par Pond	0.22	0.97

[a] Source: Du Pont (1973d).

The highest nitrate concentrations were in Four Mile Creek at Road A-7 (Du Pont 1973d) Quarterly monitoring of nonradioactive components of wastewater effluents, including nitrates, began in 1982 in compliance with Resource Conservation and Recovery Act and South Carolina Hazardous Waste Regulations (Holcomb and Emslie 1984).

In 1979, a monthly Works Technical Report gave estimates of the concentration of water quality parameters in the Savannah River, for the month and year-to-date for 1978. The tables were dated December 1978. The average, maximum, and minimum nitrate concentrations for several constituents are summarized in Table 18-14.

Table 18-14. 1978 Nitrate Concentrations in (NO_3-N), Reported in a 1979 Technical Works Monthly Report of Sampling Data[a]

Location	Average concentration (mg L^{-1})	Maximum concentration (mg L^{-1})	Minimum concentration (mg L^{-1})
Savannah River No. 2 upstream	0.85	3.8	<0.02
Savannah River No. 10 downstream	0.63	2.3	<0.02
Upper Three Runs at Highway 278	0.232	0.620	0.12
Upper Three Runs Thermal Effects Lab	0.13	0.18	0.07
Upper Three Runs at Road A	0.13	0.19	0.07
Tim's Branch, Road C	0.11	0.41	<0.02
Four Mile Creek, Road A-7	3.8	6.5	2.0
Steel Creek. Road A	0.49	4.45	0.05
Beaver Dam at Swamp	0.66	3.0	0.23
Savannah River, 3-G	0.28	0.48	<0.02
Par Pond	0.03	0.09	<0.02

Nitrates were also monitored in the swamp receiving effluent from the D-Area ash and coal pile runoff basins. The nitrate concentrations measured in the swamp water averaged about 5.2 mg L^{-1} and ranged from 1 mg L^{-1} to 9.1 mg L^{-1}, below the drinking water standard of 10 mg L^{-1}

(Guthrie and Cherry 1976). These concentrations correspond to 4.43 mg L^{-1} and 40 mg L^{-1} reported as NO_3. Routine releases of nitrates likely overwhelm any accidental spills. In March of 1997, a vendor trailer line broke and spilled 863 kg of 57% nitric acid in F-Area. It was neutralized and washed into the Old F-Area basin. About 38 L reached Three Runs Creek (Durant 1994).

Although elevated nitrate concentrations were measured in site streams, impact beyond the Site boundary is not supported by concentrations measured in Savannah River water. Upriver concentrations were apparently greater than downriver concentrations much of the time.

PESTICIDES

Pesticides were used at the Site for rodent, insect, and vegetation control. Most were applied by outside contractors, and since at least the 1980s, contractors were required to be certified pesticide applicators (DOE 1987). In recent years, the choice and application rate of pesticides are controlled by a guidance committee (Du Pont 1973a). The U.S. Forest Service also uses some herbicides and insecticides in their SRS timber management program (Du Pont 1977b).

There is little documentation of pesticide use before the 1970s. We do not know if DDT and other commonly used insecticides were used to control mosquitoes and other insects during this time period. Concentrations of insecticides and herbicides measured in water, sediments, and soils have been very low or undetectable (Gladden et al. 1985). Very low or trace quantities that were detected were attributed to offsite farm and industrial sources. Pesticides detected in onsite stream sediments were said to have come from upriver sources in river water that was pumped to the SRS facilities and released to streams. None of the pesticides in river water samples have exceeded the drinking water standards.

In the monthly reports from 1970, the Radiological Sciences Division reported using herbicides to destroy contaminated vegetation along seepage basins, creeks near effluent and on top of burial areas. Herbicides were also said to have been used to control *Potamogeton*, a weed in Par Pond. The reports did not specify which compounds were used (Du Pont 1970b).

Beginning in 1967, water and sediment samples were analyzed for pesticides. From 1967 to 1970, the analyses were performed by the Federal Water Pollution Control Administration (known as the Environmental Protection Agency after 1972) under contract with the SRS. Seven stream and two river water samples were analyzed semiannually for pesticides from 1967 to 1971 by the Department of the Interior Laboratory in Athens, Georgia (Arnett et al. 1993), part of the Federal Water Pollution Control Administration. All analysis results showed less than the minimum measurable concentrations except for dieldrin, which was detected in the river water both upstream and downstream of the SRS at a range of 0.03 to 0.04 μg L^{-1} (ppb) (Du Pont 1971c).

From 1971 to 1975, the work was performed by the U.S. Geological Survey. After 1975, the U.S. Department of Interior's Water Quality Laboratory performed the analyses (Du Pont 1978). According to the 1979 Annual Environmental Report, DDD, DDE, DDT, dieldrin, and chlordane were the pesticides most often identified in stream and river sediment samples. These pesticides, with the exception of chlordane, were reportedly not used at the SRS. The annual report states that the "concentrations of pesticides and polychlorinated biphenyls (PCBs) detected in sediment continue to indicate offsite sources" (Du Pont 1980).

In 1971, the river water analysis detected only dieldrin, both above and below the SRS, at concentrations of 0.04 μg L^{-1}. Dieldrin was said to be an agricultural chemical not used at the SRS. Other pesticides, including: aldrin, DDD, DDE, DDT, diazinon, endrin, ethion, heptachlor, lindane, malathione, parathion, methyl-trithion, silvex, trithion, 2,4-D, and 2,4,5-T were not above detection limits (Du Pont 1972c).

In 1972, water samples contained 0.01 to 0.02 μg L^{-1} dieldrin. "Trace" quantities of aldrin, DDT, DDD, DDE, and chlordane, all attributed to agricultural sources, were found in sediments (Du Pont 1973e).

In a Works Technical Monthly Report for 1971, dieldrin in river water up and downstream was reported to be 0.04 μg L^{-1}. Dieldrin in Four Mile Creek and Pen Branch was measured at a concentration of 0.02-0.03 μg L^{-1} and was attributed to the river water taken in for use at the site, then discharged. The herbicide 2,4-D was detected at Four Mile Creek at Road A at very low levels (0.03 μg L^{-1}). This was said to be "the first indication of SRP contribution in our pesticides monitoring program" (Du Pont 1971c).

In 1973, dieldrin was measured in stream and river water at concentrations of 0.01 μgL^{-1}. Trace quantities of aldrin, DDD, DDE, DDT, PCBs, and chlordane were detected in sediments. Of these, only chlordane was said to have been used at the SRS. Although it was not specifically discussed, the data show that chlordane was not detected above the plant but was found below the plant in sediments at a concentration of 1 μg L^{-1}, which is at the limit of detection (Du Pont 1974b).

In 1974, trace quantities of deildrin in river water were again attributed to offsite sources. Chlordane, said to have been used in very small amounts at the SRS, was not above the detection limit in 1974 (Du Pont 1975).

Analyses done in 1975, 1976, and 1977 were similar. The 1977 annual report says that dieldrin, DDT, DDD, and DDE were found in sediments in trace amounts and that "some pesticides and herbicides are used moderately in areas where insect and vegetation control is necessary for security and safety". Information on which pesticides were used in what quantities was not provided (Du Pont 1980).

In 1981, endrin; lindane; methoxychlor; toxaphene; 2,4-D; 2,4,5-TP; and Silvex were added to the list of analytes (Du Pont 1981). Why these chemicals were chosen was not explained and whether these were used or disposed of by the Site is not known. Addition of the chemicals may have been related to the EPA's interest in environmental levels of these chemicals.

In 1980, diazinon was detected in an Upper Three Runs Creek sample, but it was attributed to offsite sources (Du Pont 1981).

Average analytical results and the detection limits for 32 pesticides and 7 PCBs measured in sediment samples were reported in the 1981 annual report (Du Pont 1982). The report presented concentrations of nine compounds in river sediments at sampling location 2 (upstream of the SRS) and location 10 (downstream of the SRS). Most of the values were at the limit of detection.

In 1982, pesticide levels were less than the limit of detection except for heptachlor. Heptachlor concentrations up and downstream of the plant were the same (Du Pont 1983).

Trace quantities of pesticides found from 1983 to 1988 were attributed to forestry and agricultural applications (Zeigler et al. 1985).

Benzene hexachloride, said to be used by farms offsite, was detected in Savannah River sediment in 1985 and in sediments of Upper Three Runs Creek in 1986.

None of the pesticides analyzed for from 1987 to 1990 was above the limit of detection (Cummins et al. 1991).

The Comprehensive Cooling Water study, conducted in 1983 to evaluate environmental effects of intake and discharge of cooling water at the SRS, reviewed the routine annual monitoring program said to have begun in 1976. The program's goal was to determine levels of pesticides, herbicides, and PCBs in stream and river sediments. During 1976 to 1983, water and sediment from seven locations on plant streams and two locations on the Savannah River were analyzed. The number of materials analyzed and the detection limits changed from year to year depending on the offsite vendor who performed the analysis. All of the sediment samples were at or near the limit of detection. Diazinon had been detected in Upper Three Runs since 1981. DDE was detected in Upper Three Runs, but concentrations had decreased since 1979. The highest chlordane concentrations were found in Upper Three Runs at Road F in 1976. This was a control location, upstream of plant effluent, and the contamination was attributed to offsite sources. In 1982, this location again showed the greatest levels of pesticides, attributed again to offsite agricultural uses. In 1982, concentrations for analytes in river sediments were less than the limit of detection except heptachlor. There was no significant difference in heptachlor concentrations above and below the plant, and heptachlor concentrations upstream were greater, leading the authors to suggest that the source was offsite (Gladden et al. 1985).

None of the records we reviewed suggested that any significant spills or leaks of pesticides occurred onsite in the past. No visible evidence of spills or leaks was observed during the inspection of the operations and storage areas conducted in 1987 as a part of the Site Environmental Survey (DOE 1987).

Records do not indicate that pesticide contamination from waste storage areas has been a problem. Documents indicate that the CMP pits previously received pesticides for disposal, but there are no records of the types of materials or quantities received (DOE 1987).

In conclusion, monitoring data indicate that large amounts of pesticides were not released from the Site, and data on the quantities and types of pesticides used before the mid-1970s are lacking. Therefore, source terms for specific pesticides were not determined.

POLYCHLORINATED BIPHENYLS

PCBs, also called arochlors, were used in electrical equipment, such as electrical transformers and capacitors. In the 1970s, it was recognized that PCBs were very persistent in the environment and could bioaccumulate up the food chain. PCBs have also been shown to cause reproductive effects and contribute to cancer in animal studies.

River and stream sediment analyses conducted from 1976 to 1980 showed low concentrations of PCBs at similar concentrations both upstream and downstream of the plant (Du Pont 1977a; Du Pont 1980; Du Pont 1981). The highest level appears to have been 15 μg/kg sediment in 1979 at river sampling location 2, upstream of the SRS.

In most cases of PCB contamination, exposure to fish is one of the pathways of greatest concern. In 1977, fish collected from the River and Site streams had concentrations less than the lower limit of detection, which was 0.5 μg/g (Du Pont 1978).

In 1976 and 1977, it was estimated that the SRS had about 700 PCB-containing transformers, 7 to 12 extrusion presses, 1,500 capacitors, and other hydraulic oils containing PCBs onsite. In 1979, a program was started to identify PCB equipment and PCB-contaminated

items and label them as such. Since early 1985, PCBs have been disposed of at a commercial facility offsite. The SRS has not had any PCB transformers or capacitors in operation onsite since about 1986. The U.S. Environmental Protection Agency (EPA) conducted a multimedia audit at the SRS in July 1986. The Toxic Substances Control Act (TSCA) portion of the audit concentrated on the handling of PCBs. The EPA found one administrative violation involving the lack of an inspection record for one transformer. Other aspects of the PCB program were found to be satisfactory. By 1985, all PCB transformers had been removed from service, and, by 1984, all capacitors containing PCBs had been removed. PCB-contaminated items were stored at one of two places: 740-1A in the salvage yard and in 643-29G at the Radioactive Waste Burial Grounds. Radioactive PCBs that were not accepted by commercial disposal facilities were stored at the Radioactive Waste Burial Grounds. The storage areas were diked and inspected regularly and detailed inventory records were kept (DOE 1987). A letter written in 1986, referencing a request by the EPA for information on hazardous waste treatment at the Site, says about 15 gal of capacitor fluid containing PCBs, weighing about 140 lb was stored inside capacitors packed in 55-gal galvanized drums, stored in a special concrete culvert. This PCB storage began in 1978 (Porter 1986).

Several spills of PCBs have been documented. In 1981, about 100 gal of transformer fluid leaked in the salvage yard. The soil was excavated and disposed of as PCB-contaminated waste. In 1982, 1 L of liquid from a transformer retrofit was spilled inside a truck. In 1985, the extrusion press in 320-M was found to containing about 7000 gal of liquid with about 10,000 ppm PCBs, which apparently spilled and required 103 drums of materials to complete soil and concrete cleanup (DOE 1987).

Nancy J. Lowry (1996) responded to a letter *Radiological Assessments Corporation* sent out to solicit information on inventory amounts from individuals who might have knowledge of chemical usage, purchase, storage, disposal, or other relevant operations onsite. Ms. Lowry said that inventories of PCBs were probably not compiled until the late 1970s when regulations issued under TSCA required PCB users and water generators to develop and maintain records of the PCBs in use and disposed of, beginning July 2, 1978. An annual document log for PCB use and disposal was prepared by July 1979 and each year after that. She believed these records would have been sent to central records storage. She knew of no release of PCBs to the environment.

The information obtained from the records and presented here indicates that large amounts of PCBs were not released from the SRS. Therefore, a source term was not estimated for PCBs.

URANIUM

M-Area

Much of the M-Area process sewer effluent went directly into Tim's Branch until a seepage basin was put into use in 1973. The seepage basin was apparently built to address concerns about uranium discharges to the creek. The releases were not considered to be of environmental health consequence, but the seepage basin was put into use and sampling of the stream for uranium oxide was initiated in the mid-1950s. Releases after 1985 were predicted to have been negligible because of the Liquid Effluent Treatment Facility (Pickett 1990). Estimates of the total uranium discharged to the Tim's Branch were said to have been unavailable until 1974 when flow

instruments were installed. Uranium releases to Tim's Branch and the Savannah River have been characterized and are summarized in Chapter 5.

ZINC

Relatively small amounts of zinc were released from a large number of sources. Lower (1985) estimated that about 1.36 kg d^{-1} of zinc was discharged to onsite streams from numerous outfalls.

Although not used in processes in large amounts, zinc compounds were identified as one of the substances released from the coal and ash piles and basins. Zinc has contaminated groundwater under the coal pile runoff basins in D-Area and A-Area as well as groundwater beneath the Silverton Road Waste Site.

Separations Areas

Zinc was released, in relatively small amounts, to the separations area seepage basins and the SRL basin. About 309 kg of zinc was released to the F-Area seepage basin in 1975 (Lower 1985). About 216 kg of zinc was discharged to the SRL seepage basin during its 28-year operating history (Lower 1985). Eleven samples of H-Area effluent/H-Area seepage basin influent were taken from September to December 1983. The concentration of zinc averaged 3.1 mg L^{-1}. A maximum value of 26.5 mg L^{-1} was reported by Killian et al. (1987b). Zinc concentrations at the seepline and in Four Mile Creek were never large enough to be of concern.

M-Area

In 1981, Bradley estimated that 91 kg of zinc was in sludge and sediment of the M-Area seepage basin, resulting from a discharge of about 0.136 kg d^{-1}. The source of zinc was thought to be galvanized pipe and gratings (Specht et al. 1987). According to the Annual Environmental Reports, zinc concentrations in M-Area effluents in the late 1980s were less than the NPDES permit limit. In March through May 1985, the influent to and effluent from the M-Area settling basin was sampled and analyzed weekly for 10 weeks. Based on these sampling data, a removal efficiency was calculated for the basin (the influent concentration minus the effluent concentration divided by the influent concentration). The influent concentration of zinc was 0.141 $\pm$ 0.065 mg L^{-1}, the effluent concentration was 0.024 $\pm$ 0.03 mg L^{-1}, and the removal efficiency was estimated to be about 83% (Colven et al. 1985). Based on effluent monitoring data from a time period when the average production rate for 313-M was 17,500 cores per month, M-Area effluent discharges to the M-Area settling basin were estimated by Looney et al. (1987) to have been 25 kg y^{-1} for zinc.

In 1985 and 1986, Tim's Branch and Upper Three Runs Creek sediments were sampled monthly at six locations for metals. Eight samples were taken for each location. The most distant sampling point was in Upper Three Runs Creek, just downstream of the confluence with Tim's Branch. Mean concentrations $\pm$ standard error at this location were 8.90 $\pm$ 3.8 mg kg^{-1} for zinc, compared to 128.2 $\pm$ 11.4 mg kg^{-1} just downstream of where the outfall enters Tim's Branch. The control location had levels of 13.9 $\pm$ 2.3 mg kg^{-1} (Pickett 1990). A concentration of 5.2 mg kg^{-1}

in sediments at the creek mouth was also reported (Specht 1991). A typical medium value for zinc in soil samples from the United States is 36 mg/kg amounts (ATSDR 1997).

The drinking water standard for zinc is currently 5 mg L^{-1} for taste. The influent to the seepage basins seems to have been less than this. It averaged 0.14 mg L^{-1} in the M-Area settling basin (Colven et al. 1985) and 0.31 mg L^{-1} in the H-Area seepage basin (Killian et al. 1987b). Sediment concentrations were also below levels of health concern. Zinc is not a carcinogen, but it can cause adverse health effects to the blood, kidneys, and other organs if ingested in relatively large amounts (ATSDR 1997). Zinc is also an essential element and lack of zinc in the diet can cause adverse health effects. Zinc lozenges have recently become a popular cold remedy. Because zinc is relatively nontoxic, very large amounts would have had to be released to Site streams to reach concentrations of concern in the Savannah River. River monitoring does not indicate significant concentrations of zinc in the water, sediments, or fish (Westinghouse 1996a).

SPILLS TO SURFACE WATER

Most of the spills and leaks were routed to seepage, settling, or retention basins but some went into outfalls and Site streams. The following paragraphs summarize some of the more significant spills reported over the years.

Releases to Beaver Dam Creek

Releases of acid, caustic, and other chemicals to Beaver Dam Creek are worthy of special mention. Beaver Dam creek conveyed thermal effluents from the D-Area power plant and, before 1982, effluents from the Heavy Water Facility. It also received process water discharges from the Water Treatment Facility, which has been a source of sulfuric acid spills to the creek. The creek also received coal and ash basin effluent discharges. The swamp area at the mouth of the creek appears to have had stressed vegetation in 1985. The 400-D Area power plant seemed to have caused stressed vegetation in two locations: (1) north of the Ash Basin (488-D) where the vegetation was dead and the surface water (thought to be coal runoff or seepage from the ash basin) was reported to be yellow and (2) a delta affected by coal fly ash basin effluent discharges (DOE 1987). According to a paper published on swamp ecology in 1978, additional compounds routinely added to Beaver Dam Creek from the power plant and the Heavy Water Plant were reported to be 3800 L of 30% silica solution, 1800 kg of various detergents, 134,000 kg of sodium phosphates and phosphoric acid, about 450 kg of concrete cleaner, 675 kg of potassium permanganate and other manganese compounds, and 9000 kg of hydrogen sulfide each year in the mid-1970s. Periodic regeneration of ion exchanger in the power plant every 1 or 2 days involved 25 kg or larger additions of sulfuric acid and sodium hydroxide, which were discharged in alternate pulses to Beaver Dam Creek (Evans and Giesey 1978).

Three relatively large spills of sulfuric acid from the D-Area to Beaver Dam Creek were reported to have occurred in November 1977, December 1981, and May 1982. Additional spills may have occurred in the 1950s and 1960s, but they were not thought worthy of reporting by the SRS. These spills would have caused pH changes in the stream and perhaps contributed to total sulfates but would not have been expected to have caused a health hazard offsite. In May 1982, about 22,000 lb of sulfuric acid were released to Beaver Dam Creek because of a valve failure at the Heavy Water Facilities. The pH in the stream dropped to between 2 and 3 for about 8 hours.

Plant personnel calculated that the Savannah River water pH was lowered about 0.2 pH units or from about 6.6 to 6.4 at the mouth of Beaver Dam Creek. A survey of the river by boat after the spill indicated no dead fish in the river (Du Pont 1983). Sulfate concentrations in river water were recorded as a part of the water quality program started in the 1950s. A source term was not estimated for sulfuric acid releases because of its rapid degradation once released to the environment.

In June 1983, about 1100 gal of concentrated sodium hydroxide were released to Beaver Dam Creek because of a spill at the D-Area water treatment plant. The pH in the creek increased from about 6.5 to a maximum of 10.7 because of this caustic spill. The pH remained above 9 for about 9 hours. A survey team was said to have found "no measurable effect on the environs of the creek or the pH Savannah River on the day after the incident" (Zeigler et al. 1985).

Accidental Spills in Other Areas

On April 27, 1983, a caustic spill occurred in K-Area. About 350 gal of 50% sodium hydroxide overflowed a holding tank and dike because of a faulty automatic shutoff valve. Most of the caustic was contained in the drainage ditch using sandbags; however, the spill occurred during a heavy rain, and it is estimated that about 10 gal were carried to a storm sewer outfall. The contained NAOH was neutralized with sulfuric acid then discharged to the stream. Soil beneath the contained sodium hydroxide was removed to the sanitary landfill (Zeigler et al. 1985).

From 300 to 400 gal of 50% sodium hydroxide was found to have leaked from 211-H caustic storage tanks October 14, 1984. The leaked caustic was contained in a ditch and removed, along with 30 tons of soil, to the sanitary landfill (Zeigler et al. 1986a).

Two hundred gallons of uranyl nitrate were inadvertently released from the uranium recycle lab in 773-A to the process sewer on May 21, 1985. The solution had a pH of 1.5 and contained about 670 mg L^{-1} depleted uranium. The release did not exceed the Comprehensive Environmental Response, Compensation, and Liability Act reporting limit of 5000 lb, but it was of concern because of the low pH (Zeigler et al. 1986a).

Fuel and Oil Spills

Numerous fuel and oil spills have occurred on or near the Site. Investigations and reporting of these spills were required by the EPA in the 1980s and Incident Logs are available after 1980. The spills were generally all contained using booms or other containment, and contaminated soil was removed and put into a landfill. In the case of spills that went into an outfall toward the river, such as the 600-gal spill of fuel oil at TNX in 1984, the outfall was barricaded with booms and absorbent pads were placed into the outfall discharge stream to help keep oil out of the river. A "small amount" of oil did reach the river, and this was reported in the local newspapers and to the EPA. Fish were said not to have been affected.

About 160 gal of diesel fuel were spilled in the separations area December 12, 1983, from an overfilled tank. An estimated 10 gal reached a storm sewer that flowed into Upper Three Runs Creek. Water samples collected from the sewer outfall and creek on December 12, 13, and 14 were analyzed for oil. Maximum oil concentrations were 71,000 mg L^{-1} at the outfall and 11 mg L^{-1} in the Creek at Road A. No oil was detected in the Savannah River. Concentrations in the

creek returned to levels considered normal (less than 5 mg L^{-1}) by December 14. Oil-contaminated dirt and gravel at the spill site was excavated and the oil on the surface was absorbed (Zeigler et al. 1985).

On November 15, 1984, an airplane developed a hydraulic line failure and ejected about 1200 gal of fuel over a 25-mi stretch of the Savannah River between Four Mile Creek and Johnson's landing from an altitude of about 5000 ft. Sampling did not find oil concentrations to be greater than normal (Zeigler et al. 1985).

Approximately 600 gal of number two fuel oil spilled at TNX were reported in the 1984 annual report. Boom equipment was installed and most of the oil was removed with skimmers or with contaminated soil (Zeigler et al. 1985). The spill occurred from a fuel oil truck transferring oil into a tank. According to maps, the spill occurred about 500 yd from the Savannah River. Oil reached two storm drains, about 20 and 40 ft from the spill, that drain into the X- outfall. The X-8 outfall to the river was barricaded with booms to decrease the flow of oil into the river, and absorbent pads were placed into the outfall discharge stream to absorb the oil. According to the Investigation Committee Report, "some amount of oil reached the river." A newspaper article in the file, date and author unknown, states that "A thin sheen of oil was visible on the river early this morning moving downstream to a point where [it] is was breaking up near Plant Vogle." The incident review report says that an oil contaminant boom was acquired when oil was discovered in the river. The SRP-Operations Low Potential Unusual Incident form, filled out by M.L. Todd (1984) (Du Pont 1985) says that Health Protection estimated that about 8 to 10 gal of oil reached the X-8 outfall through the two storm drains. A report filed by R.K. Cauthen, Lower Savannah District Environmental Quality Control, December 5, 1984, states,

> Statements made by personnel in charge of the site indicated that they believed that only 6-8 gallons escaped, based on the flow (120 gpm) at the X-8 outfall [and] no trace of the oil remained in the creek on [or] the river. Their belief was based solely on engineering judgment and not on any physical evidence or investigation. …an inspection of the outfall to the creek was made [by us]. The outfall and creek had a heavy sheen of oil for the first 100 yards that was visible. Officials immediately ordered that absorbent rolls be brought to the site… we decided to take our boat out on the Savannah River to see if any oil remained or was entering from the creek. .. oil was continuing to escape from the creek into the River… indicated a sheen, width approximately one-half of the river, meandering downstream. The sheen was followed downstream approximately 21/2 miles to the intake structure for Georgia's Plant Vogtle Nuclear Station, where the amount of oil became extremely heavy. Further downstream, no evidence of oil was noted (Du Pont 1985).

The annual report for that year described the spill and said that the maximum concentrations of oil detected in the river were 15 mg L^{-1} about 20 yd downriver of the outfall tributary and 1.4 mg L^{-1} at Highway 301. Oil concentrations had decreased to less than 1 mg L^{-1} by November 30 (Zeigler et al. 1985). On March 15, 1984, about 2850 gal of diesel fuel leaked from a corroded underground line to the ground in Central Shops. The soil was removed and was not subjected to runoff (Zeigler et al. 1985).

On record in an administrative consent order were numerous other spills documented by the SCDHEC, including:

- February 12, 1981, spill of approximately 800 lb of hydrogen sulfide from 400-D Area to Beaver Dam Creek
- December 5, 1981, spill of approximately 400 lb of hydrogen sulfide from 400-D Area to Beaver Dam Creek
- May 5, 1982, spill of 9000–10,000 lb of concentrated sulfuric acid from 400-D Area to Beaver Dam Creek
- June 7, 1983, spill of 1000 gal of 50% sodium hydroxide from 400-D Area to Beaver Dam Creek
- October 9, 1983, underground leak of oxalic acid at an unnamed location onsite
- November 8, 1977, spill of 2000 gal of sulfuric acid from 400-D area, presumably to the ground
- November 28, 1984, spill of 600 gal of number two fuel oil to soil that drained to an NPDES outfall in the TNX area.

The consent order concluded that the SRS had violated the South Carolina Hazardous Waste Management Regulations by failing to immediately report these spills and take measures to ensure that releases do not occur or recur at a facility.

The number of spills reported in the 1980s raises concerns about the number of unreported spills that may have taken place in the 1950s, 1960s, and 1970s, when reporting to the State and EPA was not required. Certainly many spills, perhaps all spills, were reported in monthly reports, but we really do not know whether all of the significant spills were reported.

Fish Kills

Several fish kills have been reported. A 1955 Monthly Report notes that from May 26 to June 3, newspapers in the region reprinted a release from the Allendale County Game Warden reporting dead fish in the Savannah River up as far as Steel Creek. SRS personnel conducted a survey of Steel Creek and the river on June 2 and no residual evidence of a fish kill was found (Du Pont 1955).

A one-page memo dated February 15, 1966, reported that a fish kill in the Steel Creek Swamp was noted by biological monitoring technicians who estimated several hundred fish had been killed. Their deaths were attributed to thermal shock from unusually cold weather conditions. A temperature of 3°F had resulted in thick ice the week before the kill was noticed, and the kill occurred when warm temperatures returned. A few dead fish were also observed in the Savannah River during this time (Johnson 1966).

A fish kill at Pond C, involving thousands of small bream, happened 1 week after startup of P-Reactor on April 8, 1979. The reactor had been down for 10 days following a scram and the kill was thought to have been because of rapid water temperature changes that occurred when hot water was put back into the cooled pond. A similar kill occurred in June 1975 (Du Pont 1979).

CONCLUSIONS

In summary, historical releases of chemicals to the Savannah River are difficult to determine and quantify. Release estimates are summarized in Table 18-15. In addition, an undetermined amount of chromium used to treat high level waste tank cooling coil water was released. Coal and

ash pile runoff contributed to elevated arsenic, cadmium, chromium, manganese, mercury, lead, nickel and zinc concentrations in on-site surface streams and the surrounding swamp. Releases of uranium to surface water are described in Chapter 5.

Table 18-15. Summary of the Estimated Releases of Chemicals to Surface Water

Release estimate (maximum or estimated range)	Released to
1 kg y^{-1} of cadmium	To Tim's branch
900 kg y^{-1} of hydrogen sulfide	To Beaver Dam Creek
8–50 kg y^{-1} of lead	To Tim's Branch
15–623 kg y^{-1} of lead	To the Separations Area Seepage Basins
1500–3500 kg y^{-1} of mercury	To the Separations Area Seepage Basins
0.1–8 kg y^{-1} of mercury	Entering Four Mile Creek in Groundwater
116–2000 kg y^{-1} of nickel	To Tim's Branch
0–1383 tons y^{-1} of nitrate	To the Separations Area Seepage Basins
Up to 3890 tons y^{-1} of nitrate	To Four Mile Creek in Groundwater
27–200 tons y^{-1} of nitrate	To Tim's Branch

Considerable amounts of hydrogen sulfide were released to Beaver Dam Creek, but hydrogen sulfide would have been chemically transformed and degraded into less toxic or nontoxic forms within hours. Large amounts of chlorinated solvents were released to the M-Area settling basin and Tim's Branch. These releases are discussed in Chapter 17 because most of the solvent released probably evaporated.

Monitoring data were generally very limited for chemicals, particularly during early years. River water quality monitoring data suggest that the amounts of chemicals (including metals, pesticides, PCBs, nitrates, and solvents) introduced in the Savannah River from SRS streams were not detectable. Chemical concentrations upstream were the same or greater than levels downstream of the Site, or the concentrations were at or below detection limits.

Risk Assessment Corporation researchers have reviewed a large amount of effluent and environmental monitoring data related to measured chemical concentrations. Based on these data, it would be very difficult to conclude that Site activities have resulted in measurable offsite impacts for any of the chemicals discussed in this chapter. Localized areas (such as seepage basin water and sediments; coal and ash pile runoff areas; several seepline areas; and, to some extent, onsite surface water) have been affected by various routine and accidental Site activities. However, the data suggest that impacts to surface water extending beyond the Site boundary are not measurable.

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CHAPTER 19

NONRADIOLOGICAL MONITORING

ABSTRACT

This chapter summarizes available nonradiological monitoring data. Based on these data, there is little evidence to suggest that Savannah River Site (SRS) operations have resulted in elevated concentrations of nonradiological contaminants at locations beyond the Site boundary. This is true for all identified chemicals and heavy metals of concern measured in surface water, groundwater, and ambient air.

INTRODUCTION

We conducted a study to identify available nonradiological monitoring data and evaluate its potential for supporting dose reconstruction. A thorough evaluation of nonradiological contaminant (e.g., chemicals, heavy metals, and pesticides) monitoring is limited by the amount of data available for analysis. More information is available about radionuclides released from the SRS than environmental and effluent monitoring data for nonradiological contaminants. Consequently, conclusions inferred from available nonradiological data are inherently less certain than those based on radiological data.

Environmental information documents developed for environmental impact statements for waste sites are one source of information for characterizing past practices and evaluating their potential impact on current contamination. Other sources of information about the current status of onsite contamination are the Resource Conservation and Recovery Act (RCRA) and Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) projects currently in progress at the SRS. These projects are primarily evaluating present and potential future releases, but the information can also be used to investigate past activities and releases at the Site. Research studies and monitoring conducted to support environmental compliance activities may be especially useful for persistent chemical materials that were not routinely monitored before the compliance regulations or cleanup agreements were established. For the purpose of reconstructing chemical releases, recent monitoring data for RCRA or CERCLA characterization studies may be the best and sometimes only information available.

AVAILABLE MONITORING DATA

Environmental and effluent monitoring for nonradiological materials was not conducted during the early years of operation at the SRS. Most of these monitoring programs were developed in the 1980s. Industrial hygiene measurements for nonradiological compounds in work areas were also very limited before the late 1970s.

Monitoring for nonradiological materials in liquid effluents, streams, groundwater, drinking water, the Savannah River, air, fish, and wildlife was expanded over the years. However, most of the expansion occurred after 1980, and most of this growth was in onsite groundwater sampling. The streams and Savannah River were sampled monthly or quarterly for metals, chlorides, and water quality. Sediments were sampled annually after 1980 (Davis et al. 1989). Beginning in the

late 1980s, ambient air was generally sampled continuously for sulfur dioxide, nitrogen oxides, and ozone. Quarterly monitoring of nonradioactive parameters in wastewater effluent began in 1982 to demonstrate compliance with RCRA and South Carolina Hazardous Waste Regulations. In 1983, many parameters were reduced to an annual monitoring schedule.

Concentrations were often not detectable for nonradiological contaminants, or there was no apparent Site influence on concentrations in the Savannah River, ambient air, or offsite soil. For most of these materials, monitoring data are likely to be of marginal or no use for dose reconstruction. Because existing chemical monitoring data are severely limited and these data were used to estimate source terms for chemicals, a detailed description of the data is not provided here (see Chapters 17 and 18 for a discussion of available chemical monitoring data). We reviewed data for nitrates in surface water and other metals in coal and ash basin and pile runoff. We evaluated these data as supporting evidence for source term estimates and present the data in the source term sections of this report, which detail chemicals released to air and water. Chapter 20 discusses data regarding mercury and chromium concentrations in groundwater, surface water, sediments, and fish. The following sections summarize available surface water quality, groundwater quality, and ambient air monitoring data that have been collected at locations on or in the vicinity of the SRS.

SURFACE WATER QUALITY MONITORING

Chapter 5 describes the surface water affected by the Site, potential contaminant flow to surface waters, and the National Pollutant Discharge Elimination System (NPDES) outfalls. The surface water bodies that flow to the Savannah River are of concern for dose reconstruction. Examples of plant discharges to surface water flowing to the river include Pen Branch, which received heat exchanger cooling water from K-Area and flow from Indian Grave Branch; K-Reactor retention basin, which outcropped into Indian Grave Branch and flows into Pen Branch; Steel Creek, which receives contaminants migrating from the P-Area seepage basins; L-Lake, which enters Steel Creek; and F-Area and H-Area seepage basins, which outcrop groundwater to Four Mile Creek.

The Academy of Natural Sciences of Philadelphia, under contract to Du Pont, has performed independent surveys of water quality since 1951. The program, which included diatom diversity studies and other monitoring, was established at plant startup and was directed for many years by Dr. Ruth Patrick, Curator of Limnology.

The SRS began a routine monitoring program to evaluate the chemical quality of Savannah River water upriver and downriver of the Site in October 1959. Water was collected continuously at an upriver location (R-2) and at the U.S. Highway 301 bridge crossing (R-10) (refer to Chapter 13 for figure showing sampler locations). Samples were analyzed weekly for color, pH, alkalinity, hardness, conductivity, total dissolved solids, dissolved oxygen, sulfide, sulfate, total iron, chloride, nitrate, nitrite, biological oxygen demand, lignin, and surfactant (Zeigler et al. 1986). Following the first year of the program, Van Wyck (1960) concluded that SRS operations did not affect the water quality of the river.

Savannah River water quality monitoring data are reported in monthly and annual reports from 1972 (Du Pont 1973) to the present. The annual reports compare the water quality analysis results to the South Carolina standards for a Class B stream. Class B streams are defined as “freshwaters suitable for secondary contact recreation and as a source for drinking water supply

after conventional treatment in accordance with requirements of the SCDHEC; suitable for fishing, survival and propagation of fish, and other fauna and flora; suitable also for industrial and agricultural uses" (Mikol et al. 1988). For example, the 1985 and 1986 annual reports summarize the water quality program results by stating that, except for temperature in Pen Branch, all stream analyses were within South Carolina standards for a Class B stream, and, except for fecal coliform, there was no difference in water quality above and below the Site (Zeigler et al. 1986, 1987).

By 1988, continuous paddlewheel samplers were used to sample river and stream water for many water quality parameters and chemicals, including heavy metals. Methods of analysis were based on U.S. Environmental Protection Agency (EPA) guidelines and subsequent guidance, which are provided by Cummins et al. (1991).

Table 19-1 summarizes data collected during a 3-month period in 1959. The results indicated that water quality parameters measured downriver were not significantly different from those measured upriver, and that the plant had not adversely affected the water quality as measured by these parameters (Van Wyck 1960).

Table 19-1. Savannah River Water Quality Data Measured in 1959 Above and Below the SRS (mg L^{-1} [ppm])

Water quality parameter	Upstream (mg L^{-1})			Downstream (mg L^{-1})		
	Maximum	Minimum	Average	Maximum	Minimum	Average
Chloride	3.20	0.87	1.72	3.50	0.97	1.68
Nitrate	0.02	0.01	0.01	0.02	0.01	0.01
Sulfate	5.60	0.02	1.21	4.20	0.02	1.13
Nitrite	0.004	0.001	0.002	0.003	0.002	0.002
Surfactants	0.08	0.01	0.03	0.22	0.01	0.03

Although nitrate concentrations averaged 0.01 mg L^{-1} in 1959, Savannah River water quality measurements during January through June from 1960 to 1967 indicated that nitrates upriver and downriver of the plant averaged about 0.30 mg L^{-1}. However, there was generally little difference between upriver and downriver concentrations during this time period. The maximum nitrate value reported was 1.81 mg L^{-1} (Du Pont 1967).

Aperture cards (handwritten compilations of original environmental monitoring results that were photographed in a format similar to microfiche) detailing Savannah River water quality data were located for Savannah River sampling stations R-2 and R-10 for 1960, 1961, 1962, 1963, and 1964. Water quality data for Lower Three Runs Creek for 1960, 1961, 1962, 1963, and January to September of 1964 were also located. These data are generally consistent with the data reported in the various Site reports. For example, the 1961 handwritten data appear to match the more legible summary data in the December monthly report (Du Pont 1961).

Most of the water quality parameters measured before 1973 are not specific for chemicals of concern for this study. We reviewed monitoring results for nitrates and heavy metals, the only analytes selected as chemicals of concern, for the Savannah River and Four Mile Creek. Chapter 18 summarizes nitrate discharges to Four Mile Creek from the separations areas. Chapter 20 evaluates mercury and chromium environmental monitoring results and Chapter 18 characterizes mercury releases.

In 1973, a summary report stated that monitoring for nonradioactive materials in Savannah River water was conducted periodically near Jackson, South Carolina, and monthly grab samples were taken below the plant at the U.S. Highway 301 bridge (R-10). Concentrations at these two sites were compared through the 1960s and 1970s, and there were no significant differences in the concentrations of nitrate, mercury, sulfate, and metals above and below the plant (Reinig et al. 1973). In 1973, concentrations of sulfide and chloride appear to be greater above than below the SRS (Du Pont 1974). The chloride concentrations in Savannah River water had increased both above and below the plant in 1982. The increase was attributed to offsite upriver industrial sources (Du Pont 1983).

In June 1973, routine water quality monitoring was expanded to include carbonate, ammonium, sulfate, phosphorus, calcium, total iron, sodium, mercury, and aluminum. Mercury monitoring in river and stream sediments was also initiated in the early 1970s (Arnett 1993). With the exception of fecal coliform and temperature, water quality standards were not exceeded for any of the onsite streams or the Savannah River.

In 1979, the monthly Works Technical Report gave estimates of the concentration of water quality parameters in the Savannah River. Estimates of contaminant transport (in kilograms) were made for each month and year-to-date based on the concentrations (Du Pont 1979a). The average, maximum, and minimum concentrations; the calculated transport for the month of December; and the totals for 1978 were given for nitrite, nitrate, sulfate, chloride, as well as several other water quality parameters. Values were given for location R-2 above the plant, location R-10 below the plant, and nine onsite stream locations. Lead levels were less than the detection limit, except the maximum concentrations measured in Tim's Branch at Road C (0.5 mg L^{-1}). Iron and aluminum were also included, but no information for the other toxic metals of concern for this study was provided (Du Pont 1979a). Chapter 18 summarizes the 1978 year-to-date transport for nitrates measured in kilograms.

Since 1983, discharges from five ash settling basins in various areas around the SRS and their receiving streams were monitored monthly for pH and five metals and biweekly for suspended solids, oil, and grease. Monthly flow measurements were also made (Zeigler et al. 1985). Suspended solids exceeded the limit once from the H-Area ash basin in 1983 (Cummins et al. 1991).

In 1985, the South Carolina Department of Health and Environmental Control (SCDHEC) required a program to evaluate the quality of stormwater runoff. It required sampling of runoff from 17 industrial outfalls to SRS streams and six waste disposal sites during three separate rain events of 1 inch or more. The data were being evaluated at the time of the annual report, but preliminary data indicated no significant discharges of pollutants from any of the monitored outfalls (Zeigler et al. 1986).

In 1986, the concentrations of chemicals in surface water were regulated by SCDHEC under the NPDES. The NPDES monitoring data were reported in annual reports after 1986 and included outfall concentrations of chlorinated solvents, metals, phosphate, nitrate, sulfide, oil, and grease. Seventy outfalls were monitored in 1986, and 99.4% of all results were reported to have been within permit limits (Zeigler et al. 1987). Sixty-eight NPDES outfalls were monitored in 1987 with similar conclusions reported. In 1989, NPDES noncompliances (exceedance of discharge limits) were found for trichloroethylene from A-Area and for benzene from TNX Area. The Site reported that the source of these contaminants in the outfall effluent was unknown (Cummins et al. 1990). By 1990, surface water was monitored for nonradioactive contaminants at 76 effluent

outfalls from facilities at the SRS. Surface water was also monitored at locations along six onsite streams and at three locations on the Savannah River. After 1992, the annual environmental reports list the NPDES outfall locations and only detail releases that exceed discharge limits. Most often, the exceedance was for oil and grease, which were noted as sheen or another qualitative description (Arnett 1993). Westinghouse (1996) provides a detailed description of recent monitoring in the Savannah River watershed.

More recently, river sediments have been sampled by the Site and/or SCDHEC. Benzene, cadmium, carbon tetrachloride, pesticides, chrysene, manganese, mercury, polychlorinated biphenyls (PCBs), tetrachloroethylene, and toluene were all at levels below detection limits. The mean concentrations for the individual metals detected downstream were 0.016 mg kg^{-1} for chromium, 0.02 mg kg^{-1} for lead, 1.6 mg kg^{-1} for magnesium, 2.6 mg kg^{-1} for manganese, 0.028 mg kg^{-1} for nickel, 10 mg kg^{-1} for uranium, and 0.21 mg kg^{-1} for zinc. These concentrations were less than the screening level, risk-based concentrations derived from EPA values by the Site, including uranium with a screening level of 45 mg kg^{-1} (Westinghouse 1996). Comparisons of concentrations above and below the Site do not suggest that these pollutants originate at the Site. In general, the results of grab sample analysis in the annual reports for chemicals and metals are reported as being within the ranges observed in previous years.

Nonradiological contaminants, including nitrates, mercury, chromium, cadmium, lead, nickel, manganese, zinc, magnesium, arsenic, hydrogen sulfide, uranium, sodium hydroxide, detergents, and oil, have been discharged to surface waters in measurable amounts. Chapter 18 characterizes these releases. However, the transport of nonradiological contaminants offsite in concentrations that would present a hazard has not been demonstrated by environmental monitoring.

GROUNDWATER QUALITY MONITORING

Recent monitoring of onsite groundwater has been extensive; however, it is not relevant to past releases because groundwater exposure pathways to the public are not now, and have not in the past been, complete. Groundwater flowing beneath the waste sites or seepage basins from which chemicals are being leached generally does not flow offsite. Based on evidence from tritium migration to surface water, there are three exceptions: (1) groundwater beneath the H-Area and F-Area Seepage Basins flows into Four Mile Creek, (2) groundwater beneath the K-Reactor retention basin flows into Indian Grave Branch, which flows into Pen Branch, and (3) groundwater beneath the P-Area seepage basins, which flows into Steel Creek.

Extensive groundwater sampling and analysis has been conducted since the mid-1980s. Much of this information has been collected to support characterization and cleanup activities. Improvements were made to groundwater sampling and sampling preservation techniques in 1983, including better flushing of wells before sampling and filtering of samples for metal analysis (Zeigler et al. 1985).

Unfortunately, the majority of the groundwater data are not useful for dose reconstruction because the groundwater contaminants have not traveled offsite. However, understanding which materials were discharged into an area may help evaluate which materials potentially migrated beyond these discharge areas. For example, measurable concentrations of volatile contaminants under waste sites and seepage basins suggest that these compounds might also have evaporated into the air after being discharged. Groundwater monitoring below waste disposal sites and

seepage basins has been used to identify potential contaminants of concern for remediation work. Summaries of contaminants identified in groundwater are provided in monthly, quarterly, and annual reports. Suspected contaminants were defined as materials that may have been introduced into the groundwater by the SRS facility, identified as such by statistical comparison of upgradient and downgradient water quality parameters. Table 19-2 presents examples of this information.

Table 19-2. Chemicals Identified as Potential Contaminants Introduced to Groundwater as a Result of SRS Activities

Location	Chemical Contaminants of Concern	Annual Report (year)
F-Area Seepage Basin	Cadmium, copper, manganese, iron, lead, sodium, nickel, zinc, fluoride, nitrate, foaming agents, and phenol	1983
	Acid, cadmium, copper, manganese, iron, lead, sodium, nickel, zinc, fluoride, nitrate, radium, foaming agents, phenol	1984
	Acid, sodium, nitrate	1985
	Acid, sodium, nitrate, cadmium, chromium, lead	1986
	Nitrates, lead, and sulfate. Also detected arsenic, cadmium, chromium, mercury, selenium and fluoride and chlorinated organics	1989
H-Area Seepage basin	Acid, chloride, mercury, manganese, iron, sodium, nitrate	1983
	Acid, chloride, iron, mercury, manganese, sodium, and nitrate	1984
	Acid, mercury, sodium, nitrate	1985
	Acid, mercury, sodium, nitrate, lead	1986
M-Area Seepage basin	Acid, radium, chloride, iron, mercury, manganese, sodium, and nitrate	1983
	Chloride, organic, cadmium, copper, manganese, nickel, nitrate, and phenol	1984
	Organics, nitrate and sodium	1985
	Nitrate, lead, endrin, trichloroethylene, tetrachloroethylene, carbon tetrachloride, sodium	1986

The M-Area seepage basin was taken out of service in 1985 (Zeigler et al. 1986). Galvanized well casings are thought to have contributed to apparent groundwater contamination by zinc, cadmium, lead, and iron. These casings were replaced with polyvinyl chloride (PVC) in 1985. The Site then reduced the number of contaminants monitored in groundwater in the vicinity of the seepage basins to include only acid, sodium, nitrate, mercury, and organics.

In a 1987 assessment of chemical constituents and estimation of inventories for SRP waste sites, Looney et al. (1987) selected chemicals of concern for each of the waste sites. Cadmium, chromium, lead, mercury, and nitrates were chemicals of concern identified at the Old F-Area Seepage Basin and F-Area and H-Area Seepage Basins. Total organic halogens were also listed for the Old F-Area basin. No contaminants of concern were identified for the Reactor Area seepage basins.

Groundwater near the D-Area had elevated levels of arsenic, cadmium, chromium, fluoride, mercury, lead, and selenium in 1989. Sulfate and chromium were high, exceeding the drinking

water standards by 600 and 55 times, respectively. Trichloroethylene and tetrachloroethylene were also above standards in groundwater below H-Area. The maximum concentration of mercury under the H-Area seepage basins exceeded the drinking water standards by nine times and was the highest level monitored at the SRS. Wells below the K-Area disassembly basin had elevated levels of lead, trichloroethylene, and tetrachloroethylene. No chemical constituents were detected above the drinking water standards at the K-area Reactor Seepage Basin. (Cummins et al. 1990).

Lead, trichloroethylene, and tetrachloroethylene were the most common nonradiological contaminants exceeding standards in monitoring wells onsite during 1992. Cadmium, chromium, lead, mercury, nitrate, and sulfate were greater than the drinking water standards in some wells (Arnett 1993).

AMBIENT AIR MONITORING

Section 110 of the Clean Air Act Amendments of 1970 required each state to establish a network to monitor ambient air quality. In response to a concern about sulfur dioxide releases from the 400-D-Area, three ambient air monitors were located in a straight line from the 400-D-Area to 200-F-Area in 1973. The monitor situated midway between D-Area and F-Area was located on the Cassel's Fire tower (Peterson 1973). By 1976, both South Carolina and Georgia had implemented sampling networks. More extensive monitoring of ambient air quality was initiated at the SRS in 1977 (Zeigler et al. 1987). For example, a program to measure hydrogen sulfide and sulfur dioxide using monitoring equipment in mobile trailers began in 1977. Both were detected in ambient air most frequently at stations near D-Area. The maximum sulfur dioxide concentration was 190 ppb, and the average at all stations was about 4 ppb, well within the air quality standards established by Georgia and South Carolina (Du Pont 1979b). By the late 1980s, ambient air on and around the SRS was sampled continuously for sulfur dioxide, nitrogen oxides, and ozone.

After 1977, air quality data were summarized in tables in the annual environmental reports. Control of total suspended particulates and oxides of nitrogen was assessed by using opacity meters in the powerhouse stacks. Beginning in 1987, compliance with standards was also determined through tests conducted every 2 years by air emissions specialists under contract to the SRS. Although original documentation for most of these tests has not been found, the results of the test were located in Air Emission Inventory and Operating Permit Application supporting documents and files assembled in the early 1990s.

Occasionally, high opacity values in the 291-F stack were measured. The acid absorption column for F-Area separations process stack was renovated in 1986, after which the opacity requirement was met (Zeigler et al. 1987).

Since 1982, Georgia and South Carolina have operated ambient air quality monitoring stations for particulates, lead, ozone, nitrogen oxides, carbon monoxide, and sulfur dioxide near the SRP. Four to six monitoring stations have been operated onsite by private contractors. The air quality measurements onsite have been below the air quality standards, with the exception of ozone, which has been reported at concentrations greater than the standard at several locations.

Zeigler et al. (1986) reported that concentrations of ozone were higher than the annual standard of 120 ppb on 1 day in each of the years 1982, 1983, and 1985. Exceedance of the standard for 1 day each year is allowed by State regulations; consequently, the ambient air

concentrations were all reported as being within the standards. The high concentrations have generally been recorded in the summer months. Ozone concentrations were also reported to be high at offsite monitoring stations at the same time. Some reports attributed the elevated ozone concentrations to photochemical reactions of hydrocarbons and nitrogen oxides. In an environmental survey, DOE (1987) stated that "high concentrations of ozone may be due to volatile hydrocarbons such as those released from the air stripper in M-Area, the laboratories, fuel storage facilities and vehicles." However, elevated ozone concentrations were recorded in 1982 and 1983 before the air stripper was in use. In 1982, ozone concentrations above the standard were measured when the performance of the detection instrument was said to have been erratic, and the annual report states that "it is not known if the ozone level was actually above the 120 ppb standard" (Zeigler et al. 1985).

The annual environmental reports for 1984, 1985, and 1986 state that audits of the ambient air monitoring stations were performed quarterly. The calibration of the equipment was also evaluated quarterly (Zeigler et al. 1985, 1986)

By 1985, the SRS monitored air quality at five or six stations and the states of Georgia and South Carolina monitored air quality at eight stations near the SRS. The stations continuously measured total suspended particulate, sulfur dioxide, oxides of nitrogen, and ozone (Zeigler et al. 1987).

The annual and quarterly or special reports summarizing the data generally maintain that concentrations were within standards. For example, the 1986 annual report listed the state standards compared to the maximum concentration found at any of the SRS monitors for sulfur dioxide (Table 19-3).

In 1986, the annual ambient air concentrations for sulfur dioxide, compiled quarterly at four locations, averaged 2 ppb and ranged from 1 to 4 ppb. Nitrogen dioxide levels, measured at five locations, ranged from 0 to 9.6 ppb and averaged 4.32 ppb (Zeigler et al. 1987). In 1987, annual sulfur dioxide levels ranged from 1 to 6 ppb and averaged 3.2 ppb, while annual nitrogen dioxide levels ranged from 0 to 7 ppb and averaged 2.7 ppb (Mikol et al. 1988).

Table 19-3. Comparison of Maximum Measured Sulfur Dioxide Concentrations by the SRS with Georgia and South Carolina Standards

Measurement Period	Georgia and South Carolina standards ($\mu g\ m^{-3}$ or ppb)	Maximum measured concentration ($\mu g\ m^{-3}$ or ppb)
3 hours	1300	61
24 hours	365	6
Annual	80	3

A series of tests of the powerhouse emissions were carried out in 1988 and showed all were in compliance. The 291-F stack exceeded the limit for oxides of nitrogen. A deteriorating acid absorption column was reworked, which brought the stack emissions into compliance. An absorber column control project was planned for 1989. Emissions estimates and opacity measurement information is compiled in Chapter 17 under Coal Ash in the discussion about releases of coal and oxides of nitrogen to air.

CONCLUSIONS

Based on available monitoring data, there is little evidence to suggest that SRS operations have resulted in elevated concentrations of nonradiological contaminants to locations beyond the Site boundary. Although this assertion is supported by a relatively small amount of data, available environmental monitoring information does not support an appreciable source term from the SRS for any nonradiological contaminant. This holds true for all identified chemicals and heavy metals of concern measured in surface water, groundwater, and ambient air. Refer to Chapter 20 for a more detailed conclusion regarding measured concentrations of mercury and chromium in environmental media.

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CHAPTER 20

METALS IN THE SAVANNAH RIVER SITE ENVIRONMENT

ABSTRACT

This chapter summarizes available monitoring data for mercury and chromium in the Savannah River Site (SRS) environment. Based on a thorough evaluation of these data, including measured concentrations in water, sediment, and fish, there is no evidence to suggest that the SRS has contributed to increased levels of mercury or chromium in the Savannah River. It appears that industries upriver from the SRS are the primary contributors to elevated mercury concentrations in onsite streams and the Savannah River.

INTRODUCTION

Operations at the SRS introduced several heavy metals into the environment. Of the metals that have entered the SRS environs, mercury and chromium appear to be the only metals that have been investigated with sufficient regularity to warrant a thorough analysis of reported environmental monitoring data. This chapter discusses the various sources of available data and attempts to identify the SRS's role as a contributor to the levels of mercury and chromium in the environment surrounding the SRS. See Chapters 15, 17, and 18 for a detailed discussion regarding atmospheric and effluent releases of metals and other chemicals.

Because of the toxic nature of many metals, it is important to evaluate concentrations measured in various environmental media. Water, sediment or soil, and fish are some of the most common media that have been examined. It is particularly important to evaluate fish, not only because of their popularity as a food item, but also because certain metals (including mercury) have the ability to bioaccumulate. Water concentrations of metals that are well below regulatory guidelines can, therefore, cause relatively high concentrations in edible fish tissue that approach and even exceed recommended intake levels.

MERCURY

Mercury in the environment can result from both natural and anthropogenic sources. Natural sources of mercury include various minerals in soil, such as cinnabar (mercury sulfide or HgS), and degassing processes (e.g., the eruption of Mt. St. Helens and other volcanoes). Anthropogenic sources include the burning of fossil fuels, the chlor-alkali industry, the pulp and paper industry, seed fungicide treatment, and mercury catalysts used in industry (EPA 1992; Phillips and Russo 1978; Kvartek et al. 1994; Watras and Huckabee 1994).

Based on groundwater monitoring, the F-Area and H-Area seepage basins appear to be two of the most significant potential sources of mercury to the aquatic environment from SRS operations. Mercury was used at the SRS as a processing aid in the F and H separation areas. As a catalyst, mercury increases the dissolution rate of uranium-aluminum alloy reactor fuel, plutonium-aluminum targets, and scrub-alloy scrap in nitric acid. As a precipitating agent, mercury removes chloride ions, which can corrode stainless steel in processing equipment. It also entered the separation areas as an impurity in NaOH. Effluents associated with these processes

have been discharged to F-Area and H-Area seepage basins since 1959. Between 1959 and 1981, approximately 3600 and 840 lb of mercury were released to F-Area and H-Area seepage basins, respectively (Horton 1974b). Use of the F-Area and H-Area seepage basins was discontinued in November 1988, and the Effluent Treatment Facility became operational to treat wastes that were previously sent to the seepage basins. The SRS continues to burn coal, which can contain 600 ppb mercury (Olotka 1973). Kvartek et al. (1994) provides a detailed summary of the SRS operations that used mercury.

The SRS has historically maintained that the majority of mercury in onsite streams and reservoirs resulted from pumping Savannah River water for use as reactor coolant. Mercury was introduced to the Savannah River by upriver industries, including the Olin Corporation, a mercury-cell-type chlor-alkali plant, near Augusta, Georgia, which began operation in 1965. The plant manufactures chlorine gas and caustic soda through the electrolysis of brine by a mercury electrode. Following notification by the Georgia Water Quality Control Board (GWQCB) in response to a 1970–1971 survey (GWQCB 1971), the Olin Corporation significantly reduced mercury discharges. However, for at least five years it discharged large quantities of mercury directly to the Savannah River.

During September 1970, the release of mercury to the F-Area and H-Area seepage basins came under routine observation, and subsequent attempts were made to determine soil, sediment, and water concentrations to assess the rate of movement into Four Mile Creek. In July 1971, the SRS began monitoring fish collected from onsite ponds and streams and the Savannah River for mercury. This program has consistently monitored both onsite and offsite locations, and large numbers of fish have been analyzed for mercury content. Information regarding water and sediment concentrations, based on the available documents, is more limited. A 1974 document (Horton 1974a) described the mercury environmental monitoring program at that time. Water samples from F and H treblers were composited for weekly analysis; Savannah River water samples from six locations were analyzed monthly; and water samples from four burial ground wells, one 200-H well, and four locations at Four Mile Creek were analyzed semiannually. Sediment samples from two Savannah River locations, four Four Mile Creek locations, and one location each in Upper Three Runs, Steel Creek, Beaver Dam Creek, and Lower Three Runs were analyzed semiannually. Unfortunately, significant gaps appear to exist in actual sampling, but we have attempted to locate all information pertaining to mercury investigations in the Savannah River vicinity by the SRS and other agencies and researchers. Available information is generally restricted to water, soil or surface sediment, and fish concentrations. Consequently, the following discussion focuses on the concentrations measured in these media.

Upriver Industrial Sources of Mercury

The Olin Corporation in Augusta, Georgia, discharged significant amounts of mercury directly to the Savannah River, particularly during the late 1960s. From August 1965 through August 1970, the reported discharge rate from the Olin Corporation was 12 lb d^{-1} (EPASWL 1971, cited in Tilley and Wilhite 1972). Before a study conducted by the GWQCB in 1970, discharge rates of 10 lb d^{-1} were reported (GWQCB 1971). Following the study and notification by the GWQCB, discharge rates were reportedly reduced to less than 0.25 lb d^{-1}. On May 20, 1970, the Olin Corporation had a recorded discharge of 2.65 lb, and by July 14, 1970, the recorded discharge had been reduced to 0.51 lb (D'Itri 1972, cited in Newman and Messier 1994).

Although the reported discharge rates differ slightly, it is clear that significant quantities of mercury were discharged directly to the Savannah River. Assuming a discharge rate of 10 lb d^{-1} for 5 years, more than 18,000 lb of mercury would have been discharged directly to the Savannah River between 1965 and 1970.

SRS Separations Area Seepage Basins

The chemical separations facilities (F-Area and H-Area) and associated seepage basins are located between Upper Three Runs Creek to the north and Four Mile Creek to the south (Fenimore and Horton 1973). Water movement in the area is complex because underlying sediments are not uniformly permeable; however, in general, water moves slowly away from the groundwater divide, which is offset toward Four Mile Creek. Water then moves at an accelerating rate down the gradient to outcrop at the springs, swamps, and beds of the two streams.

Two separate and distinct water tables exist below the F-Area seepage basins. A perched groundwater table is located 10 to 15 ft below the ground surface, and water either seeps through the less permeable underlying strata or flows laterally a maximum of 150 ft to the edge of the supporting strata. Flow is then vertical to the normal water table located 60 to 65 ft below the ground surface. The nearest outcrop area, a line of springs along the edge of the Four Mile Creek swamp, is 1600 ft from the basins. Based on detection of tritium in Four Mile Creek, travel time from the basins to Four Mile Creek was estimated to be between 8 and 12 years with a flow rate of between 0.37 and 0.55 ft d^{-1} (Fenimore and Horton 1973).

Geological characteristics of the H-Area basins are quite different from those of the F-Area basins. The water table is located 15 to 25 ft below the ground surface, and the outcrop area along Four Mile Creek is only 400 to 600 ft from the basins. The soil contains a high percentage of clay and has a relatively low permeability except for narrow zones of extremely high permeability. Based on detection of tritium in Four Mile Creek, travel time from basin 1 was estimated at 3.25 years with a flow rate of about 0.5 ft d^{-1}, and travel time from basin 4 was estimated at 3.75 years with a flow rate of about 1.0 ft d^{-1} (Fenimore and Horton 1973). The flow rate from seepage basins in the H-Area likely is higher than in the F-Area because of the narrow zones of high permeability. See Chapter 5 for a more detailed description of surface water flow, groundwater outcropping, and seepage basin geography.

Between 1959 and 1982, more than 3600 and 840 lb of mercury were released to H-Area and F-Area seepage basins, respectively (Figure 20-1) (Horton 1974b). Use of these basins continued through 1988, after which they were capped and did not receive additional effluent. Information regarding annual releases of mercury from F-Area and H-Area from 1971 through 1982 was provided in a 1985 memorandum (Lower 1985). No information was available regarding annual releases between 1959 and 1970, but cumulative (1959 through 1981) releases of 3600 and 840 lb from the H-Area and F-Area, respectively, have been reported. Figure 20-1 shows the reported releases from 1971 through 1982 and average annual releases from 1959 through 1970 based on cumulative releases between 1959 and 1981.

During 1972, several studies were conducted to identify mercury concentrations in the environs of H-Area seepage basins (Horton 1974b). Soil samples were collected from seepage basins 1, 2, and 4. Basins 1, 2, and 3 were full of water, and basin 4 was dry at the time. Seepage basin 3 was not sampled because it had not been used since 1964.

Two locations each in basins 1 and 2 were sampled above the water level. Basin 1 had concentrations of 135 and 6500 ppm, and basin 2 had concentrations of 970 and 5040 ppm. Four soil samples each in basins 1 and 2 were collected from the top 6 in. of soil in the bottom of the basins. Basin 1 had concentrations of 87, 1640, 1730, and 780 ppm, and basin 2 had concentrations of 150, 850, 830, and 410 ppm. The higher concentrations in soil in one location above the water level in both basins (6500 and 5040 ppm) were attributed to splashing and evaporation. The total volume of soil containing these higher concentrations was estimated to be very small. Based on these concentrations, it was estimated that 99% of the mercury releases to these basins could be accounted for in the soil. However, the large variation between samples and the limited number of samples makes this estimate questionable.

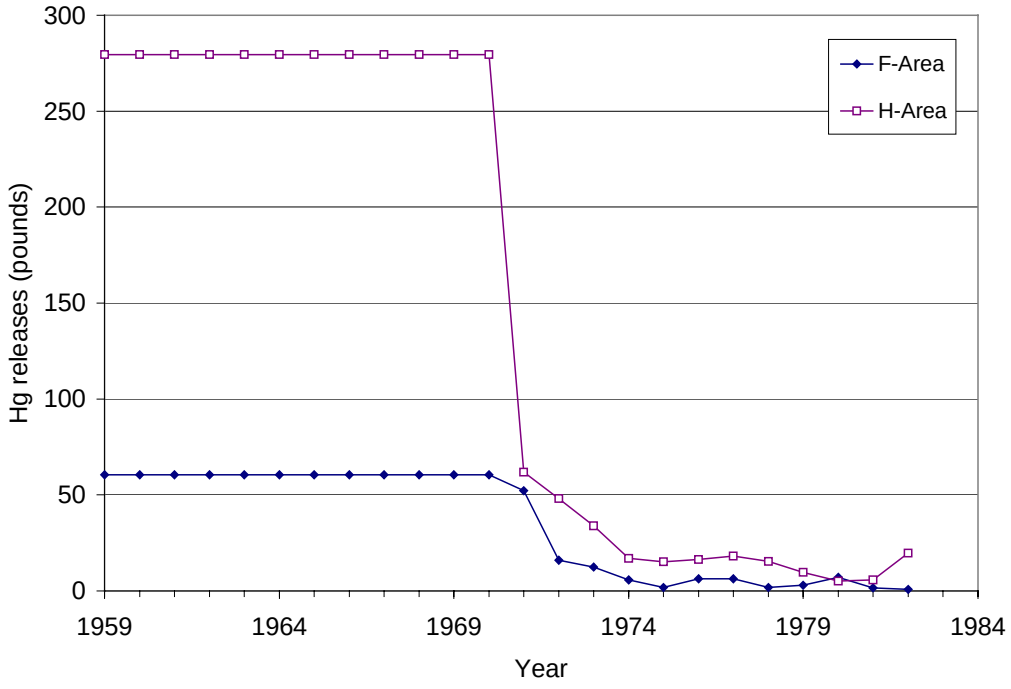

Figure 20-1. Annual releases to F-Area and H-Area seepage basins from 1959 through 1982. Link to tabulated data.

The higher concentrations in the bottom of basins 1 and 2 compared to those in basin 4 indicated that much of the mercury was precipitated in basins 1 and 2. Leaching studies were conducted on soil from basins 1 and 2 to assess the mobility of the mercury in these basins. The first test involved leaching a soil sample with a mercury concentration of 5000 ppm with groundwater collected above the H basins. Only 0.08% was removed after 350 pore volumes. The second test involved leaching a soil sample with a mercury concentration of 630 ppm with 1360 pore volumes of distilled water to remove soluble mercury and 199 pore volumes of 1N $NH_4C_2H_3O_2$ (ammonium carbonate) to remove exchangeable mercury. These leaching processes removed 0.15% of the mercury. These experiments showed that nearly all the mercury in basins 1 and 2 was both insoluble in water and nonexchangeable with a salt solution, and that the mercury in these basins was virtually immobile.

Since basin 4 was dry, soil samples were collected in two locations from the basin bottom to the water table, 14 ft below the basin bottom. Seven depth increments were analyzed, and elevated mercury concentrations were evident at all depths (Figure 20-2). For reference, typical background mercury concentrations in soil and sediment range from 0.02 to 0.05 ppm (Horton 1974b), and concentrations at all depths were at least an order of magnitude greater than 0.03 ppm.

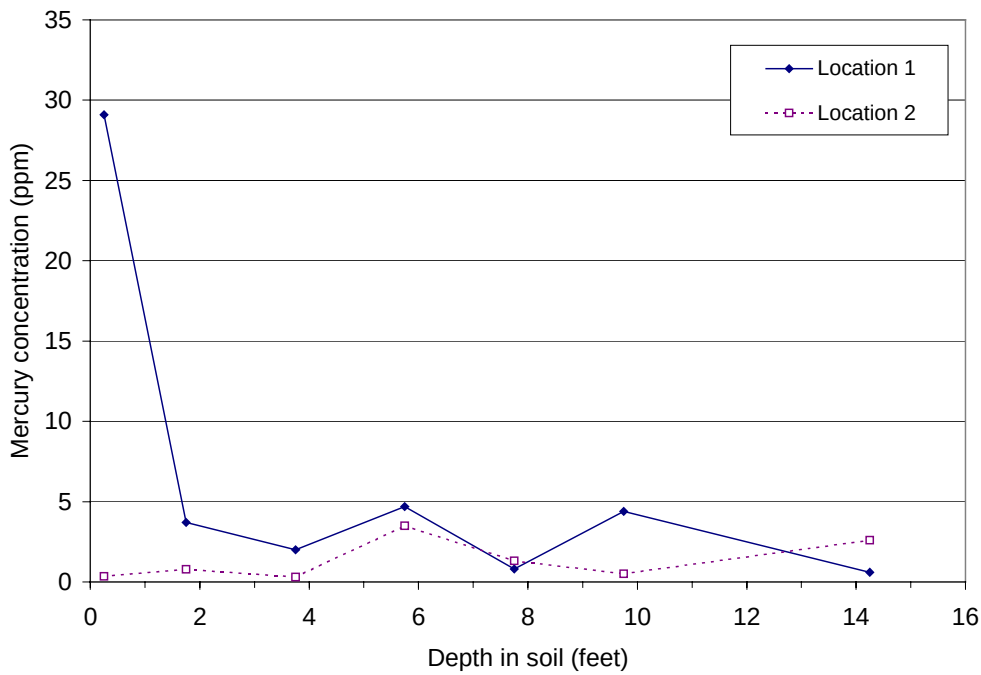

Figure 20-2. Mercury concentrations as a function of soil depth in H-Area seepage basin 4. Link to tabulated data.

An attempt was made to calculate the H-Area seepage basin contribution of mercury to Four Mile Creek based on analysis of suspended sediments in Four Mile Creek water above and below the H-Area seepage basins. However, the estimate of mercury transport, 0.36 g d^{-1}, was based on only two samples. In addition, the concentration above the seepage basins (0.023 ppb) was higher than the concentration below the seepage basins (0.020 ppb). A positive mercury transport rate was calculated only because the water flow rate above the basins (15,900 L min^{-1}) was significantly lower than the water flow rate below the basins (30,850 L min^{-1}). A lower concentration below the seepage basins seems contrary to a measurable mercury transport rate from the H-Area seepage basins.

Based on the difference in flow rate above and below the H-Area seepage basins, an alternative method for calculating mercury transport is possible. Assuming a uniform mercury concentration in seepline water of 1 ppb (which is a conservative assumption) based on reported seepline water concentrations, an estimated mercury transport rate can be calculated from the H-Area seepage basins. If the increase in water flow rate below the basins were attributed only to seepline water contributions, the mercury transport rate would be about 22 g d^{-1}. This is a conservative estimate because seepline vicinity measurements indicate that water concentrations

are generally less than 1 ppb, and the highest concentrations in both sediment and water appear to be in localized areas, likely along a few channels of high permeability. It also assumes no background contribution, and background mercury concentrations of 0.5 ppb have been measured in Four Mile Creek water. It does not seem likely that mercury transport rates would have exceeded this value, and actual transport rates were likely much lower. At any rate, the transport rate of mercury to Four Mile Creek (and certainly to the Savannah River) from the F-Area and H-Area seepage basins appears negligible relative to the discharge rate of mercury by the Olin Corporation directly to the Savannah River during the late 1960s.

Water in H-Area seepage basin 1 was sampled in 1970, and concentrations ranged from 100–150 ppb with a mean of 114 ppb (n [number of samples] = 5). Basin 4 was also sampled, and concentrations ranged from 3–8 ppb with a mean of 6.3 (n = 4). One sample was collected from basin 2 and it had a concentration of 15 ppb (Bebbington 1971).

Data from trebler samplers, which were used to collect liquid samples in F-Area and H-Area effluent streams from October 1980 through October 1981, provided average effluent concentrations of 2.9 for the F-Area and 25.6 ppb for the H-Area (attachment to Horton 1974b). These data appear to have been composited weekly and apparently were used to estimate annual releases of mercury to the basins.

In 1981, two National Pollutant Discharge Elimination Systems (NPDES) effluent outfalls from F-Area and H-Area facilities that flow directly into Four Mile Creek (F-008 and H-012) contained measurable mercury concentrations of 0.2 and 0.7 ppb, respectively. Dixon and Rogers (1994) concluded that a water concentration of 0.5 ppb was in the background range based on samples taken upstream from the separations areas, so it would be difficult to conclude that these effluent concentrations were elevated above background.

Well Water

Concentrations measured in two water samples taken from 200-H well #4 (just east of seepage basin 1) in 1970 were 5 and 25 ppb (Bebbington 1971). The 25-ppb sample was analyzed with solids removed (the report does not specify a definition for solids) and a concentration of 2 ppb was recorded, indicating most of the mercury was associated with suspended sediment. A sample collected from this well on May 26, 1973, had a concentration less than 2 ppb (Du Pont 1973a). Water samples from seven H-Area seepage basin wells were collected on November 11, 1981, and five contained concentrations less than 0.05 ppb. The other two samples had concentrations of 0.5 and 3.4 ppb (attachment to Horton 1974b).

In general, maximum reported mercury concentrations in SRS groundwater have occurred in H-Area seepage basin wells (Mikol et al. 1988a, 1988b; Davis et al. 1989a, 1989b; Cummins et al. 1990, 1991; Arnett et al. 1992). Figure 20-3 shows mercury concentrations for H-Area seepage basin wells that have historically shown some of the highest concentrations. The highest concentrations in these wells have generally occurred in wells situated south and east (downgradient) of the seepage basins. Concentrations measured between 1984 and 1996 are also generally consistent with concentrations reported for wells sampled in 1970 and 1981 (discussed above) although data are very limited before 1984.

Seepage basin operations halted in November 1988, and rainfall infiltration between that time and completion of the cap raised the pH significantly in the immediate vicinity of the basins. Reduced hydraulic load in the basins combined with a higher pH would be expected to slow the

migration of mercury. Dissolved mercury concentrations were reported through the third quarter of 1992, after which total mercury concentrations have been reported. This makes interpreting the data somewhat difficult, but it appears that concentrations in the wells depicted in Figure 20-3 have decreased since closure of the basins.

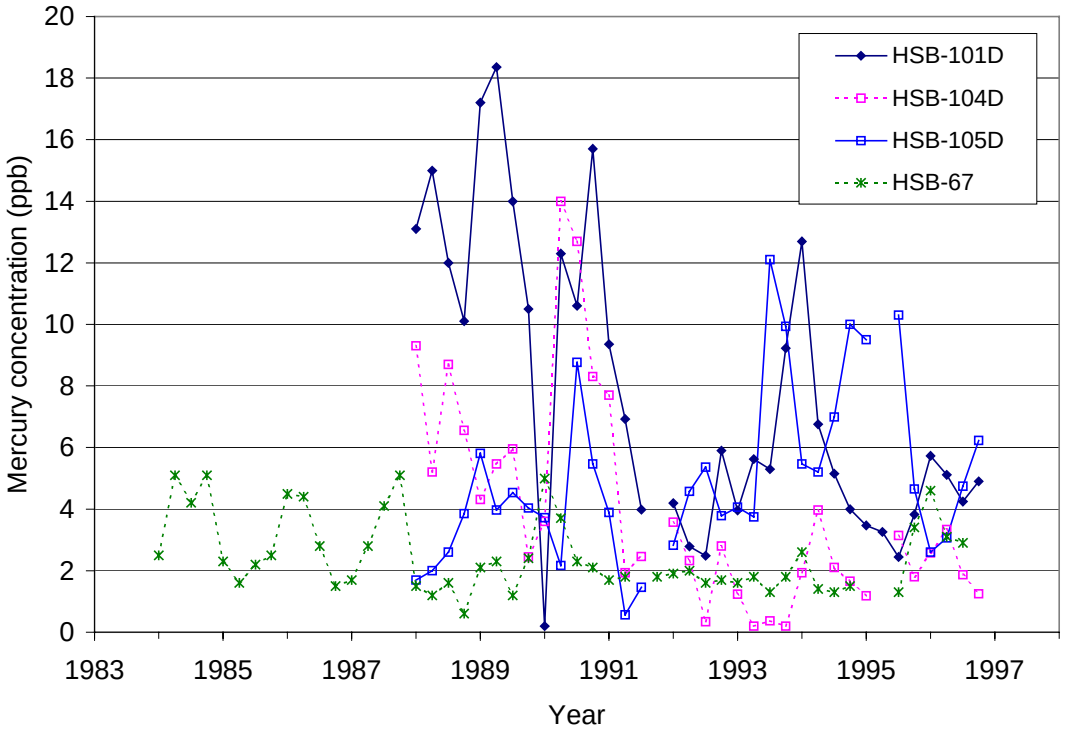

Figure 20-3. Mercury concentrations at four H-Area wells (HSB-101D, HSB-104D, HSB-105D, and HSB-67) that have historically had some of the highest measured concentrations. Link to tabulated data.

In 1991 and 1992, 65 and 67 F-Area seepage basin wells were sampled, respectively. Six seepage basin wells had water concentrations greater than the U.S. Environmental Protection Agency's (EPA's) Primary Drinking Water Standard (PDWS) of 2 ppb (Arnett et al. 1992; Arnett 1993). The maximum concentrations measured in these wells were 7.4 and 12 ppb, respectively. In 1991 and 1992, 108 H-Area seepage basin wells were sampled and 11 and 15, respectively, had concentrations greater than the PDWS. The maximum concentrations measured in these wells were 9.4 and 7.9 ppb, respectively.

Based on these data, it is clear that measurable but relatively small amounts of mercury from the F-Area and H-Area seepage basins are being leached into the groundwater. Temporal trends in groundwater concentrations for individual wells vary considerably, and it is difficult to draw many conclusions regarding the stability of mercury in groundwater below the seepage basins. However, elevated concentrations (above the PDWS) have only been measured in a small fraction (about 10%) of the total number of F-Area and H-Area seepage basin wells. The wells with water concentrations consistently above the PDWS are situated downgradient of both F-Area and H-Area seepage basins. Based on 1989 data, concentrations above the PDWS have only been measured in the upper saturated zone of the water table. Concentrations above the PDWS were

not reported for any well in lower geologic layers, including the Dry Branch, McBean, Santee, and Congaree Formations.

Seepline and Swampy Outcrops of Four Mile Creek

Surface Sediment Monitoring

In 1972, 19 sediment samples were collected from the swampy outcrop along Four Mile Creek (Horton 1974b). Two samples collected in the vicinity of basins 1, 2, and 3 had concentrations of 30 and 20 ppb. Nine samples collected from the swampy outcrop south of basin 4 had an average concentration of 60 ppb, with concentrations ranging from 30 to 80 ppb. These concentrations are near background concentrations of 20 to 50 ppb reported by Horton (1974b). Similar values have been reported for uncontaminated lakes in South Carolina (Abernathy and Cumbie 1977). Similar background values have also been reported for river sediment in the Mt. Amiata region of Tuscany and sediment in Lake Michigan, with elevated values (in the vicinity of cinnabar mineralization) approaching or exceeding 1000 ppm (Dall'Aglio 1970, cited in Jonasson and Timperley 1973; Kennedy et al. 1971, cited in Reimers et al. 1973). Eight samples collected from the same swampy outcrop in the vicinity of the first side stream[1] into Four Mile Creek had an average concentration of 4100 ppb, with concentrations ranging from 160 to 19,400 ppb. The two highest concentrations by far were 10,100 and 19,400 ppb. These data appear to confirm migration of mercury to these outcrop areas, likely along a channel of high permeability leading to the first side stream.

Sediment samples were collected in 1988 and 1989 from the Four Mile Creek seepline in the vicinity of the F-Area and H-Area seepage basins (Haselow et al. 1990). In 1988, concentrations in the vicinity of the F-Area and H-Area seepage basins ranged from 40 to 310 ppb (mean = 166 ppb, n = 9) and from 70 to 220 ppb (mean = 131 ppb, n = 7), respectively. In 1989, concentrations at all F-Area (n = 6) and H-Area (n = 4) seepage basin vicinity seepline locations were less than 100 ppb (lower limit of detection [LLD]).

Water Monitoring

Three studies in recent years have analyzed for mercury in the groundwater intersecting the surface at the seepline along Four Mile Creek, downgradient from F and H-Areas. Water samples collected in 1987 from the seepline downgradient of F and H-Area seepage basins had maximum unfiltered mercury concentrations of less than 0.1 (LLD) and 0.14 ppb, respectively (Looney et al. 1988). The authors concluded that the majority of mercury was sorbed to subsurface materials or neutralized and that transport from both F-Area and H-Area seepage basins was very slow.

Water samples collected in 1988 from similar locations had maximum mercury concentrations of less than 0.2 (LLD) and 0.3 ppb, respectively (Haselow et al. 1990). Water samples collected in 1989 from similar locations had maximum mercury concentrations of less than 0.2 ppb (LLD).

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[1] The frequently mentioned side streams in this chapter refer to the many small drainages or seeps that outflow into Four Mile Creek. The first side stream refers to the first of these encountered downstream of the Separations Area seepage basins.

Water samples collected in July 1992 from a "background" location upstream from the General Separations Areas had mercury concentrations ranging from less than 0.2 (LLD) to 0.47 ppb (Dixon and Rogers 1994). Water samples collected from the F-Area seepline had concentrations ranging from less than 0.2 (LLD) to 0.54 ppb (average of two measurements). The authors concluded that this was within the range of "background" concentrations (<0.2 to 0.47 ppb). Water samples collected from the H-Area seepline had concentrations ranging from less than 0.2 (LLD) to 1.7 ppb (average of three measurements). The 1992 data appear to confirm some measurable migration of mercury in the groundwater at localized areas of the H-Area seepline, but concentrations at the F-Area seepline do not appear elevated above background.

Four Mile Creek

Surface Sediment Monitoring

In 1972, sediment samples were collected from nine locations in Four Mile Creek from Road 4 to Road C (Horton 1974b). These samples had an average concentration of 240 ppb. The two highest concentrations, 620 and 590 ppb, were in sediment collected from locations at the first side stream and at Road C, just below F-Area effluent, respectively. This suggested maximum introduction of mercury from H-Area seepage basin 4 and F-Area cooling water. There is little question that mercury has been introduced to the Four Mile Creek swampy outcrop along the first side stream based on the seepline sediment concentrations discussed earlier. However, subsequent analyses of sediment collected at Road C indicated concentrations of 100, 190, <30, and <1 ppb, which are not elevated above average concentrations measured in 1972 (Horton 1974a; Du Pont 1972b; Du Pont 1973a; Mikol et al. 1988a). Sediment concentrations appear quite variable, and it would be difficult to conclude with confidence that discharge of F-Area cooling water has resulted in increased sediment concentrations at Road C.

The four above reports also provided sediment concentrations measured at other locations along Four Mile Creek and at other SRS locations (Table 20-1). Du Pont (1973a) provided concentrations at 12 locations in Four Mile Creek. The average concentration was 310 ppb, with the two highest concentrations (800 and 1060 ppb) occurring at Cassel's Pond above the weir and in sediment from C-Area cooling tower effluent at Road 3, respectively. Du Pont (1971a) provided concentrations at three locations in Four Mile Creek. One sample collected downstream from Road 4 had a concentration of 50 ppb, and the other two samples had concentrations less than 30 ppb. A 1974 memorandum (Horton 1974a) provided concentrations at 11 locations. The average concentration was 500 ppb, with the two highest concentrations (1860 and 810 ppb) occurring at Cassel's Pond above the weir and in sediment associate with H-Area cooling tower effluent.

Stream sediment concentrations measured in the vicinity of the F-Area and H-Area seepage basins were generally lower than concentrations measured at other locations, and the data for various stream locations are variable. Elevated concentrations measured at locations upstream from Road 3 can possibly be attributed to migration from the F-Area and H-Area seepage basins because Savannah River water was pumped to C Reactor, which only discharged water into Four Mile Creek below Road 3. The H-Area cooling tower received water pumped from groundwater wells, which could have contained elevated mercury concentrations based on groundwater concentrations in the H-Area water table. However, well water used at the Site comes from fairly

deep aquifers (in H-Area as well as other areas). None of the water supply wells is located between the basins and Four Mile Creek, and, in H-Area, the aquifer that supplies the production wells has higher pressure than its overlying aquifer (Heffner 1997). In general, sediment concentrations in Four Mile Creek appear higher below Road 3, indicating Savannah River contributions through C-Area cooling water.

The high concentrations reported for Cassel's Pond above the weir are likely the result of increased deposition of suspended sediments above the dam. Sediments at the discharge points for H-Area cooling tower effluent and C-Area cooling water and from cooling water settling basins (C-Area, K-Area, and L-Area) also appear to have elevated concentrations. Aside from these areas, onsite sediment concentrations are similar to the concentrations reported for Savannah River locations (see Table 20-2).

Table 20-1. Mercury Concentrations (ppb) in Sediment Collected at Various SRS Locations

Location	1972[a]	February 1972[b]	May 1973[c]	June 1974[d]	1987[e]
Four Mile Creek					
Effluent at Road E		130	<30	210	
Cooling tower effluent		540		810	
Downstream from Road 4	70	175	50	250	
Above entry of F-Area effluent	190	70		100	
F-Area effluent at Road E		150		210	
Below F-Area effluent at Road C	590	100	<30	190	<1
Downstream at Road A7		80		120	<1
Downstream at Road 3		240		600	
C-Area cooling water at Road 3		1060			
Downstream at Road A		250		430	<1
Cassel's Pond, above weir		800		1860	
Cassel's Pond, below weir		220		690	
Steel Creek					
At Road B				20	
At Road A-14				210	
At Road A		160			
At Road A-17		6			
Par Pond					
At pump house				60	
At hot dam				170	
At cold dam				210	
Cooling water settling basins					
C-Area		320		700	
K-Area		400		850	
L-Area				1410	
R-Area				200	

[a] Source: Horton (1974b).
[b] Source: Du Pont (1972b).
[c] Source: Du Pont (1973a).
[d] Source: Horton (1974a).
[e] Source: Mikol et al. (1988a).

In 1987, sediment concentrations in Four Mile Creek at the locations shown in Table 20-1 and at two additional locations (where the stream enters the river swamp and at the mouth) were all less than 1 ppb (Mikol et al. 1988a). This makes it difficult to conclude with any confidence that migration of mercury from seepage basins has resulted in contamination of Four Mile Creek sediments except in areas associated with the swampy outcrop soil samples collected in the vicinity of the first side stream (previously discussed). The fact that sediment concentrations appear to have decreased by over 2 orders of magnitude since the early 1970s suggests that much of the sediment-bound mercury in Four Mile Creek has been buried or eroded into the Savannah River. The decrease also illustrates the decreased mercury discharges from the Olin Corporation. However, similar decreases were apparent at locations both upstream and downstream from Road 3. This is not consistent with the fact that Savannah River water has never been introduced to locations above Road 3. These stream surface sediment concentrations are also significantly lower than concentrations measured in F-Area and H-Area seepline sediment in 1988 (previously discussed). This suggests that mercury migration from the F-Area and H-Area seepage basins may be resulting in measurable contamination of the local seepline sediment but not widespread contamination of Four Mile Creek. It is also possible that seepline sediments are not as susceptible to erosion as stream sediments, and the mercury in these sediments is less mobile.

The data for sediment concentrations in Four Mile Creek are quite variable, and sediment samples collected from the Savannah River indicate that mercury concentrations may vary by more than a factor of 2 for sediment collected in similar locations (GWQCB 1971). These differences are likely due to natural heterogeneity in sediment composition. In addition, based on samples collected in 1972 and 1974, stream sediment concentrations in the vicinity of the F-Area and H-Area seepage basins appear lower than sediment concentrations below Road 3, which were impacted by C-Area cooling effluent. Based on 1972 measurements, there is little question that small amounts of mercury have migrated from the seepage basins, resulting in significantly elevated seepline sediment concentrations in the vicinity of the first sidestream (Horton 1974b). However, given the general variability in the data and the lack of an apparent disparity between Savannah River and Four Mile Creek sediment, it would be difficult to conclude with very much confidence that the F-Area and H-Area seepage basins have introduced sufficient amounts of mercury to measurably impact the Savannah River.

Water Monitoring

A water sample collected from the Four Mile Creek river swamp area (confluence with the Savannah River) in September 1970 had a mercury concentration less than 0.1 ppb. Water samples collected from Four Mile Creek at Roads E, 4, and C in May 1973 all had mercury concentrations less than 2 ppb, the apparent LLD (Du Pont 1973a). In 1973 and 1974, maximum concentrations of mercury were less than 1 ppb. Between 1984 and 1990, maximum concentrations of mercury in Four Mile Creek at Roads A7 and A were below the LLD of 0.2 ppb, except in 1988 (Road A7) and 1989 (Road A) when maximum concentrations were 0.22 and 0.4 ppb, respectively. In 1991, maximum mercury concentrations were below the LLD of 0.0001 ppb at these two locations.

Savannah River

Surface Sediment Monitoring

Sediment samples collected from the Savannah River have had similar mercury concentrations to those collected from many of the onsite streams (Table 20-2). However, the data that have been available for review are somewhat limited, and a significant gap exists between 1974 and 1992–1994.

Table 20-2. Mercury Concentrations (ppb dry weight) in Sediment Collected at Various Savannah River Locations Shown with the Approximate River Mile Designation

Location	1970—1971[a]	February 1972[b]	May 1973[c]	June 1974[d]	1992–1994[e]
River mile (RM) 187	272				<0.13
RM 183	85				
Jackson's Landing (RM 165)		60	<30	90	
Shell Bluff Landing RM 162	346				
Upper Pumphouse (RM 158)		290			
Lower Pumphouse (RM 155)		320			
Griffin's Landing (RM 147)	875				
Steel Creek Landing (RM 142)		60	<30	210	
Highway 301 bridge (RM 119)	667	80	<30	210	<0.13
RM 78	811				
RM 61	143				
RM 45	277				
RM 22	175				

[a] Source: GWQCB (1971).
[b] Source: Du Pont (1972b).
[c] Source: Du Pont (1973a).
[d] Source: Horton (1974b).
[e] Source: Westinghouse (1996).

In 1970–1971, the GWQCB, in cooperation with the Georgia Game and Fish Commission and the EPA, surveyed several media (including sediment) collected at various locations along the Savannah River (GWQCB 1971). Sediment concentrations ranged from 85 ppb, at a location just downriver from the Olin Corporation's discharge points (between river miles [RMs] 187 and 183), to 875 ppb, at a location near Griffin's Landing (RM 147). Given the general variability in sediment concentrations, it is difficult to make many conclusions other than that mercury concentrations appear elevated at distances greater than 100 mi downriver from the Olin Corporation's points of discharge. In addition, concentrations are similar to those reported for Four Mile Creek and other onsite locations in 1972–1974 (Table 20-1).

Westinghouse (1996) provided data for three- and four-sediment samples collected from the Savannah River between 1992 and 1994 at RMs 120 and 187, respectively. All seven samples had concentrations below the detection limit of 0.13 ppb. These concentrations are significantly lower (about 3 orders of magnitude) than the concentrations that were measured in the 1970s.

This suggests that much of the sediment-bound mercury in the Savannah River has been buried or eroded downriver; however, additional information would be helpful to more conclusively assess this situation. A similar decrease in sediment concentrations was noted for Four Mile Creek. It is interesting to note that the mercury concentrations measured in Four Mile Creek sediment in 1987 (<1 ppb) and Savannah River sediment in 1992–1994 (<0.13 ppb) are significantly less than background concentrations (20–60 ppb) reported for sediment in the SRS vicinity and in other areas. Further analysis of these data is hindered by the general spatial variability of mercury deposition in the river and the fact that precise sampling locations for different years are not known.

Water Monitoring

On July 9, 1970, mercury concentrations were measured in 25 water samples collected from the Savannah River between Augusta to Savannah, GA. Analyses indicated concentrations less than 0.2 ppb (the apparent LLD) for all of the samples. Westinghouse (1996) provided data for 16 water samples collected from RM 120 of the Savannah River. These samples all had concentrations below the detection limit of 0.2 ppb.

In 1973 and 1974, maximum concentrations of mercury in Savannah River water collected from locations above and below the SRS were less than 5 and 2 ppb (the apparent LLDs), respectively. Between 1984 and 1990, maximum concentrations of mercury in Savannah River water collected from locations above the SRS, below plant Vogtle, and below the SRS were below the apparent LLD of 0.2 ppb; however, in 1988 maximum concentrations at these three locations were 0.7, 0.39, and 0.26, respectively. In 1991, maximum concentrations at all three locations were less than 0.05 ppb. For comparison, wastewater from the Olin Corporation contained 50 ppb of mercury on July 17, 1970 (St. John 1970).

Fish

We examined three sets of routine SRS annual environmental monitoring reports spanning the years 1971 through 1991 to summarize mercury concentrations in fish collected from locations on or in the vicinity of the SRS:

- Environmental Monitoring at the Savannah River Plant, 1971–1984 (Ashley 1972; Ashley and Zeigler 1973–1984; Ashley et al. 1984a, 1984b; Zielger et al. 1985)
- Environmental Monitoring in the Vicinity of the Savannah River Plant, 1971–1983 (Du Pont 1972c, 1973b, 1974–1984)
- Savannah River Plant Environmental Report, 1985–1991 (Zeigler et al. 1986a, 1986b, 1987a, 1987b; Mikol et al. 1988a, 1988b; Davis et al. 1989a, 1989b; Cummins et al. 1990, 1991; Arnett et al. 1992).

Monitoring for mercury in fish by the SRS began in 1971, and, in general, values reported in the three sets of monitoring reports were consistent during this time period.

Sample Collection and Preparation

In July 1971, the SRS began monitoring fish for mercury. Collected fish were composited quarterly for analysis through 1972 and semiannually from 1973 through 1975. In 1976, analysis of individual fish began and has continued through 1991. Du Pont (1972a) described the method of analysis for fish and indicated that 1-g samples of flesh were used for analysis. A LLD of 20 ppb was also reported. A 1975 memorandum indicated that 2-g portions of flesh taken from behind the dorsal fin were used for analysis Cumbie 1975). A 1990 document (Loehle and Paller 1990) indicated that 0.3-g plugs of fish flesh were routinely collected for analysis. Concentrations were consistently reported for Savannah River locations, but the Clark Hill control location and onsite location concentrations were reported more sporadically. Concentrations have generally been reported as micrograms mercury per gram of flesh wet weight (ppm).

Basis for Analysis

For brevity and clarity, only average concentrations for all species are graphically depicted in this report. However, based on the reported concentrations for various species of fish, there is evidence to suggest that bass (and other predatory fish as well as bottom-dwelling fish) typically accumulate higher concentrations of mercury than other species. Concentrations for bream, catfish, and other species relative to those for bass at all locations between 1971 and 1991 (1981 through 1991 for other species) were 0.49, 0.50, and 0.70, respectively. These data suggest that bass may exhibit concentrations by at least a factor of 2 greater than bream or catfish. In 1973, bass from Four Mile Creek and Steel Creek had concentrations more than a factor of 5 greater than bream collected at the same locations (Du Pont 1974; Ashley and Zeilger 1974). In 1976, bass from Four Mile Creek had concentrations more than a factor of 10 greater than bream or catfish collected at the same location (Du Pont 1977; Ashley and Zeilger 1978a).

Because average concentrations and concentrations for individual species are discussed, it is helpful to look at the relative numbers of each species of fish collected at the various sampling locations (Table 20-3). In general, a larger percentage of bass has been collected from onsite locations, particularly Par Pond and Pond B. This is considered when evaluating concentrations measured in fish from the Savannah River and various onsite locations.

The total number of fish collected yearly from the Savannah River and from onsite locations has varied significantly since 1971 (Figure 20-4). Onsite and Savannah River locations are the same as described in Table 20-3 (see Chapter 14 for a map illustrating specific locations). Very few fish were collected from the Savannah River and onsite locations from 1976 through 1980. Additionally, concentrations were reported for onsite locations in 1974 and 1975, but sample numbers were not provided. No fish were analyzed in 1987 because of the construction of a new laboratory.

Table 20-3. Percentage of Bream, Bass, Catfish, and Other Fish and the Total Number of Fish Collected for Mercury Analysis at Various Sampling Locations on or in the Vicinity of the SRS Between 1971 and 1991

Location	Bream	Catfish	Bass	Other	Total #
Clark Hill/Thurmond Lake	45.7	26.7	15.7	11.9	210
Savannah River Locations					
Above SRS (R-2)	43.4	45.0	5.2	6.3	553
Adjacent to SRS (R-8)	40.8	43.3	5.9	9.9	573
Below SRS (R-10)	44.7	44.6	5.3	5.4	704
River total	**43.1**	**44.3**	**5.5**	**7.1**	**1830**
Onsite Locations					
Upper Three Runs	37.8	34.6	16.2	11.4	185
Four Mile Creek at Cassel's Pond	76.3	3.6	17.7	2.4	413
Steel Creek at Road A	44.0	29.5	23.5	3.0	366
LTR at Patterson Mill Road	54.3	29.6	13.5	2.7	223
Par Pond	34.5	8.1	38.6	18.8	197
Pond B	33.1	8.8	52.9	5.1	136
Onsite total	**51.3**	**18.5**	**24.1**	**6.1**	**1520**
Grand Total[a]	**46.8**	**32.2**	**14.0**	**6.9**	**3560**

[a] This total does not include fish collected from Four Mile Creek at Road 3 or the mouth of Steel Creek because these locations were sampled very infrequently.

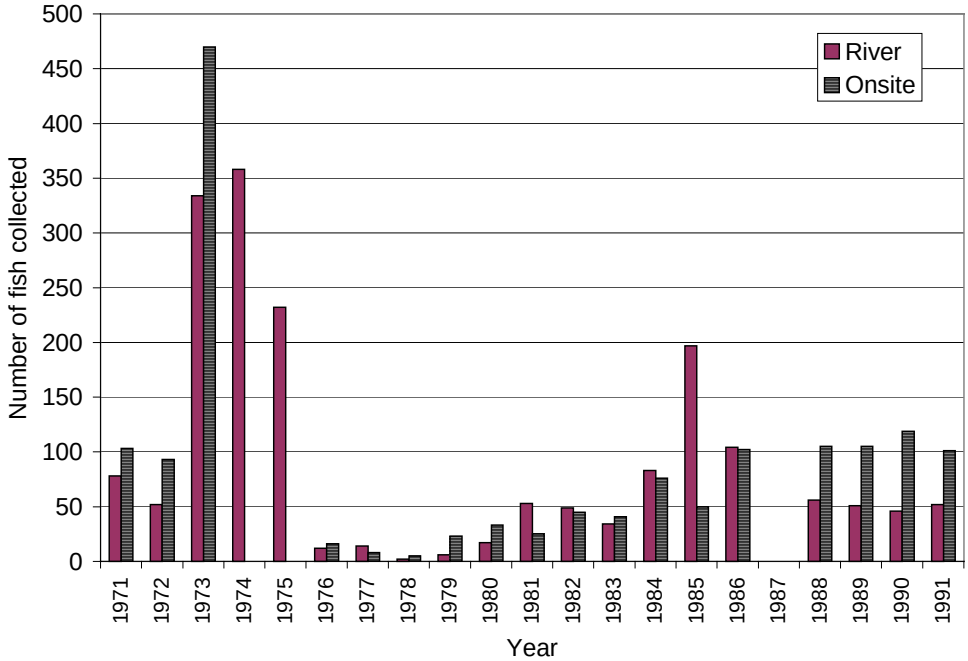

Figure 20-4. Total number of fish collected annually for mercury analysis from three Savannah River locations and five onsite locations. Link to tabulated data.

Mercury Concentrations Reported in SRS Annual Monitoring Reports

To establish whether the SRS has been responsible for introducing mercury to Savannah River fish, it is instructive to examine concentrations measured in fish from Clark Hill (a control location upriver from the SRS) and three Savannah River locations (above, adjacent to, and below the SRS) (Figure 20-5). Figure 20-5 also shows the Food and Drug Administration (FDA) guide for daily intake of mercury in edible fish. Concentrations have fluctuated somewhat since inception of the monitoring program, but they have fluctuated rather consistently at all locations. This is indicative of similar mercury levels in fish from various locations along the Savannah River. In addition, average concentrations in Savannah River fish (at all three locations) between 1971 and 1991 (see Table 20-3) are not statistically different from the average fish concentrations measured at Clark Hill. This is not consistent with the fact that Olin Corporation discharges to the Savannah River are well downriver from the Clark Hill Reservoir, and concentrations of mercury in fish from Clark Hill Reservoir would not be expected to be the same as for fish from the Savannah River. An explanation for the lack of a difference is not apparent, but differences in fish size and age as well as differences in mercury methylation rates for different locations could be factors.

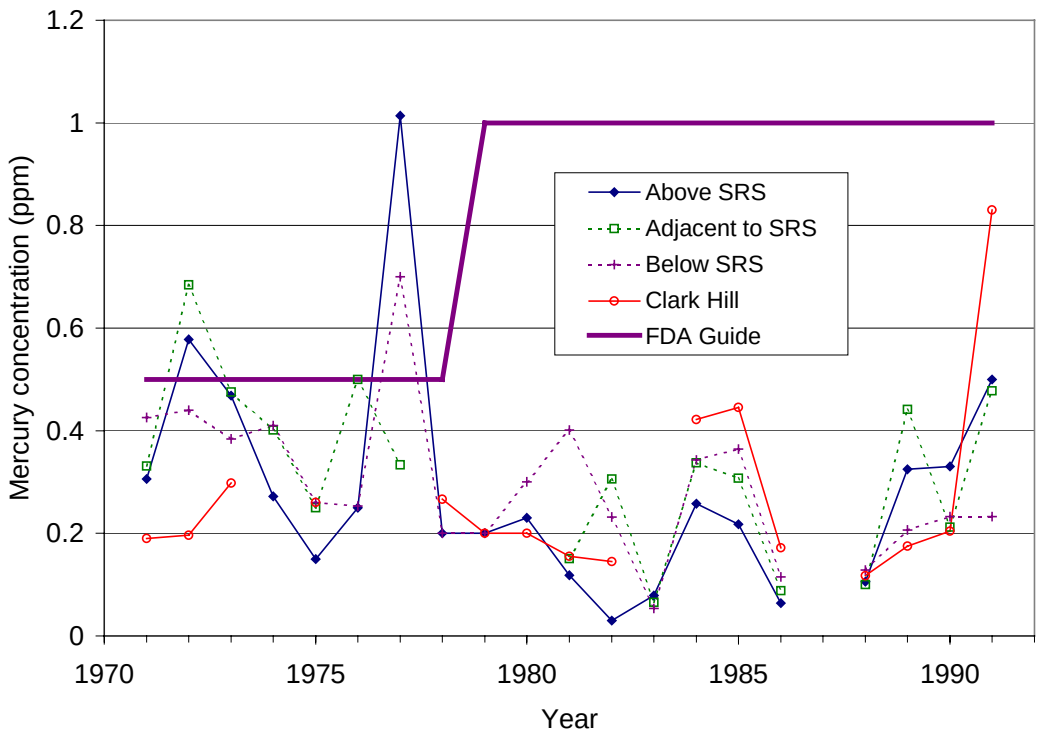

Figure 20-5. Average mercury concentrations measured in fish collected from Clark Hill and the Savannah River at locations above, adjacent to, and below the SRS. Link to tabulated data.

It is also instructive to examine average onsite concentrations and average Savannah River concentrations relative to those reported for Clark Hill (Figure 20-6). Very few fish were

collected at any location from 1976 through 1980, but mercury concentrations in fish collected from onsite locations are clearly elevated relative to concentrations in fish collected from the Savannah River and Clark Hill locations. In general, onsite concentrations are about a factor of 2 greater than Savannah River concentrations. It should be noted that a greater percentage of bass has typically been collected from onsite locations. This may contribute somewhat to the higher onsite concentrations, but there was likely some other mechanism that resulted in higher concentrations in onsite fish.

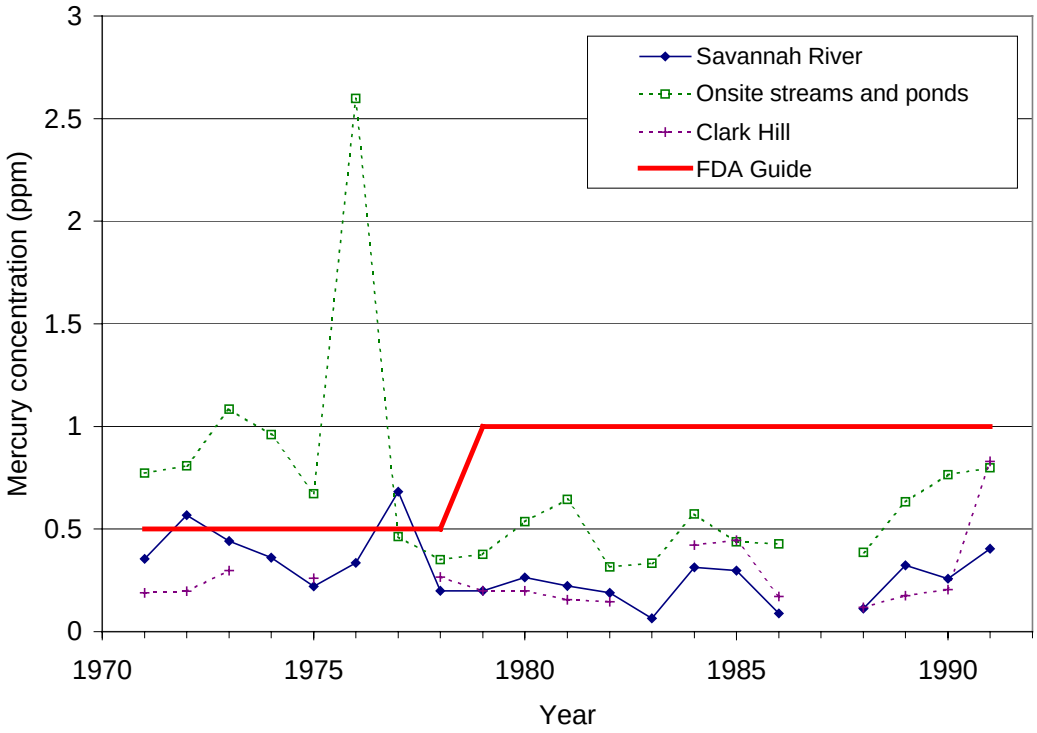

Figure 20-6. Average mercury concentrations measured in fish collected from Clark Hill, Savannah River, and onsite locations. Link to tabulated data.

Onsite concentrations appear significantly elevated in 1976. However, few fish were collected and the majority of fish collected onsite were bass, which had concentrations roughly an order of magnitude greater than bream and catfish collected at the same locations. No bass were collected from the Savannah River in 1976.

To assess the credibility of a significant F-Area and H-Area seepage basin source term for mercury, it is instructive to look at mercury concentrations measured in fish from various onsite locations (Figure 20-7). Concentrations appear similar at all locations, which suggests that F-Area and H-Area seepage basins have not contributed significantly to elevated mercury concentrations in Four Mile Creek. In fact, with the exception of 1971 and 1973, concentrations in fish from Four Mile Creek are lower than concentrations in fish from other onsite streams. Concentrations measured in onsite fish appear significantly elevated in 1976 at all locations. As discussed above, measured mercury concentrations in 1976 were heavily influenced by the fact that the majority of

fish sampled were bass, which had significantly higher mercury concentrations that other species collected at the same locations.

Unfortunately, the routine sampling location in Four Mile Creek was at Cassel's Pond, which is below the C-Area cooling tower effluent discharge point. Concentrations were reported from 1971 through 1973 for fish collected from Four Mile Creek at Road 3, which is above the C-Area cooling tower effluent discharge point. During this time period, concentrations in fish at Road 3 averaged 0.66 ppm, and concentrations in fish at Cassel's Pond averaged 1.15 ppm. This suggests that C-Area cooling water discharges (water originating from the Savannah River) had a larger impact on mercury concentrations in Four Mile Creek fish than did F-Area and H-Area seepage basins. The fact that fish from other onsite streams have historically had mercury concentrations similar to those measured in Four Mile Creek also suggests limited impact from F-Area and H-Area seepage basins.

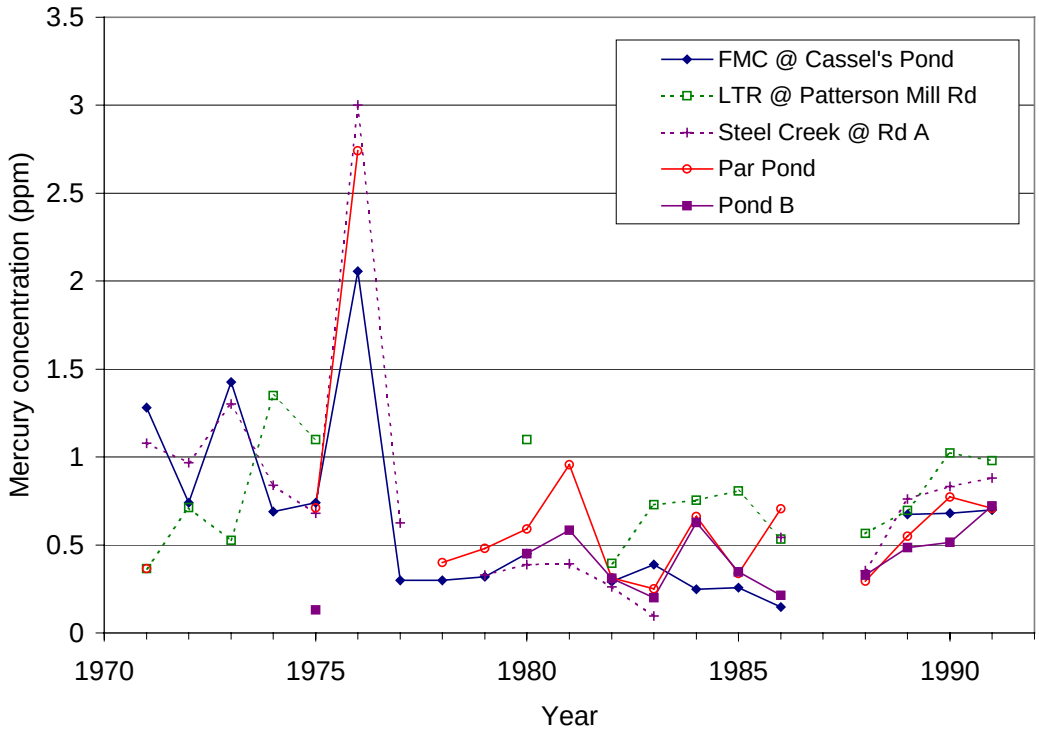

Figure 20-7. Average mercury concentrations in fish collected from five onsite locations. Link to tabulated data.

Table 20-4 shows average mercury concentrations for Savannah River and onsite locations from 1971 through 1991. Average concentrations for each individual species (i.e., bass, bream, catfish, and other) are shown as well as mean concentrations for all species and normalized mean concentrations for all species after normalization to bass concentrations. Normalized mean concentrations were calculated by dividing the bream, catfish, and other species concentrations by an adjustment factor (0.49, 0.50, 0.70, respectively). These adjustment factors were calculated by dividing the concentrations measured in bream, catfish, and other species by the concentrations measured in bass. There is little question that average mercury concentrations in all species of fish collected from onsite locations appear higher than the concentrations in fish collected from

the Savannah River. The concentrations appear, on average, to be about a factor of 2 greater in onsite fish. The mean concentration for all species from onsite locations is a factor of 2.0 greater than for Savannah River locations. The mean concentration for all species after normalization to bass concentrations is a factor of 1.6 greater than for Savannah River locations. These mean concentrations were calculated considering the number of each species. Therefore, the difference in relative numbers of each species collected from the Savannah River and onsite locations (Table 20-3) likely contributed to the disparity in mercury concentrations. However, it does not appear that this is the only factor that contributed to higher onsite fish concentrations. Concentrations measured in fish collected from Four Mile Creek and from other onsite locations that would have been exposed to water pumped from the Savannah River (Steel Creek, Lower Three Runs, and Par Pond) do not appear to be significantly different.

Table 20-4. Average Mercury Concentrations (μg mercury g^{-1} flesh wet weight) Reported for Clark Hill, Savannah River, and Onsite Locations from 1971 through 1991

Location	Bass	Bream	Catfish	Other	Mean	Mean[a]
Clark Hill	0.35	0.20	0.16	0.39	0.27	0.40
River above SRS	0.51	0.26	0.30	0.26	0.28	0.53
River adjacent to SRS	0.52	0.28	0.30	0.27	0.32	0.58
River below SRS	0.60	0.21	0.32	0.32	0.29	0.54
Savannah River average	*0.54*	*0.25*	*0.30*	*0.28*	*0.30*	*0.55*
Upper Three Runs at Road F and Highway 278	0.98	0.38	0.45	0.51	0.44	0.78
Upper Three Runs at Road A	0.58	0.33	0.30	0.39	0.35	0.55
FMC at Cassel's Pond	1.22	0.48	0.44	0.18	0.65	0.94
Steel Creek at Road A	1.43	0.52	0.54	0.40	0.78	1.09
Lower Three Runs at Patterson Mill Road	1.31	0.51	0.83	0.60	0.78	1.12
Par Pond	0.94	0.52	0.42	0.49	0.68	0.88
Pond B	0.59	0.23	0.28	0.45	0.41	0.48
Onsite average	*1.02*	*0.45*	*0.46*	*0.43*	*0.59*	*0.89*

[a] Mean concentrations for all species normalized to bass concentrations.

Examining the maximum reported concentrations from 1981 through 1991 (the reports did not provide maximum concentrations before 1981) reveals similar maximum reported concentrations for Clark Hill, Savannah River, and onsite locations (Figure 20-8). However, maximum concentrations appear consistently higher for the Savannah River and SRS streams and ponds than for Clark Hill. The highest reported concentration during this time period was at Clark Hill.

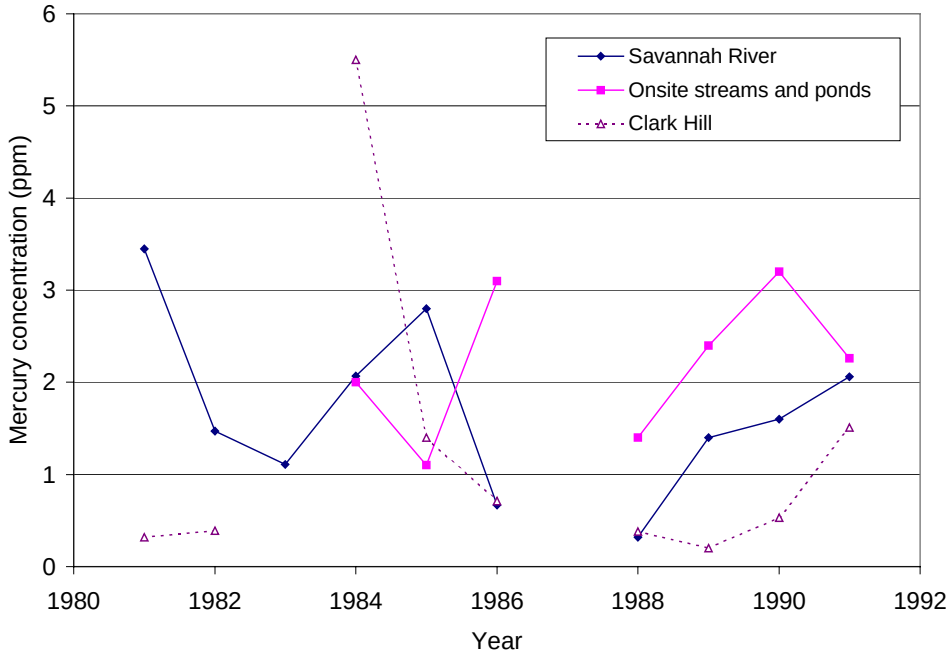

Figure 20-8. Maximum reported mercury concentrations for Clark Hill, the Savannah River, and onsite locations from 1981 through 1991. Link to tabulated data.

There appears to be little evidence to suggest that mercury introduced to Four Mile Creek from the F-Area and H-Area seepage basins has resulted in the elevated concentrations measured in Four Mile Creek based on concentrations measured by the SRS. It is not clear why onsite fish have consistently had higher concentrations than fish from the Savannah River, but potential explanations will be explored later in this section.

Other SRS and Savannah River Studies

In 1970–1971, the GWQCB, in cooperation with the Georgia Game and Fish Commission and the EPA, conducted an extensive survey of several media, including fish. Concentrations for bass, bream, suckers, and shad collected at various locations along the Savannah River and from seven other major river basins in Georgia are reported in Table 20-5. Concentrations in fish collected from various locations along the Savannah River below the Olin Corporation's discharge points (between RMs 183 and 187) appear very similar, and they are significantly elevated relative to concentrations in fish collected below Clark Hill dam. This suggests that mercury contamination extends at least 150 mi below the Olin Corporation's discharge points. A study on mercury accumulation in fish of the North Fork Holston River in Virginia and Tennessee reported elevated mercury concentrations in fish more than 80 mi below an inactive chlor-alkali plant (Hildebrand et al. 1980). Similar concentrations (between 1 and 2 ppm) were reported for fish collected in this study compared to fish collected by the GWQCB. Concentrations for fish collected upriver of the Olin Corporation's discharge points (below the Clark Hill dam) were significantly less than for fish collected below the discharge points and similar to average mercury concentrations measured for fish collected from the seven other major

river basins in Georgia. The average concentration across all species (not just those listed in Table 20-5) was 0.34 ppm for the seven major river basins exclusive of the Savannah River and 0.95 ppm for the Savannah River from Augusta to Savannah.

Table 20-5. Average Mercury Concentrations (ppm mercury wet weight) in Fish Collected in 1970–1971 by the GWQCB[a]

Location	Bass	Bream	Suckers	Shad
Savannah River				
RM 238[b, c]	0.47	0.06		0.04
RM 162	1.53	0.70	1.30	0.52
RM 119	1.72	0.70	1.41	0.50
RM 78	1.63	1.06	1.36	0.52
RM 22	1.80	0.80	1.80	0.36
Other river basins[d]	0.37	0.37	0.44	0.38

[a] Source: GWQCB (1971).
[b] RM = river mile.
[c] Below Clark Hill dam.
[d] Seven major river basins in Georgia exclusive of the Savannah River.

Concentrations measured in bass and suckers were significantly higher than for bream and shad, and the data indicated that piscivorous fish at the top of the food chain (e.g., largemouth bass) had the highest mercury concentrations. However, suckers were found to contain mercury concentrations comparable to concentrations in the piscivorous species. Because suckers are bottom feeders, they may ingest greater amounts of sediment than other species of fish and, therefore, may have accumulated higher mercury concentrations. In addition, large specimens of a species were generally found to contain higher concentrations than smaller specimens of the same species from the same area.

Newman and Messier (1994) conducted an extensive study to address the consistently higher reported mercury concentrations for mosquitofish collected from SRS streams relative to fish collected from the Savannah River. The results indicated no significant differences in mercury concentrations between onsite and offsite fish. There were, however, significant differences in mercury concentrations between fish from locations associated with Savannah River water input and fish from locations isolated from river water input. Additionally, mosquitofish collected from the Olin Plant Canal had concentrations roughly an order of magnitude greater than fish from other locations. Furthermore, the authors examined Westinghouse Savannah River Company data (which is reported in the annual environmental monitoring reports) and concluded that fish size and age were confounding factors in interpreting the data. This is the only study that *RAC* has been able to locate that was designed specifically to address the question of higher concentrations reported for onsite locations.

O'Connell (1971) compared mercury concentrations for several species of fish collected from Four Mile Creek to concentrations in fish from other "control" locations, including Upper Three Runs Creek, Tinker's Creek, Dick's Pond, Skinface Pond, and an unnamed creek. There were no statistically significant differences for any species except largemouth bass. Concentrations in largemouth bass from Four Mile Creek were approximately an order of magnitude greater than in bass from the "control" locations. Only three bass were collected from Four Mile Creek, and detailed information regarding fish size and age was not provided, although

the authors reported that the sizes of fish varied greatly. Based on the results of this study, concentrations in fish from Four Mile Creek do not appear higher than in fish from "control" locations, and the difference noted for largemouth bass may have resulted from collecting larger or older fish from Four Mile Creek. Samples of homogenized whole fish and, when possible, fish muscle were analyzed, and the limit of detection was reported as 0.01 ppm.

In July 1990, fish were collected from six locations in Four Mile Creek and one location in Pen Branch (Loehle and Paller 1990). Fish were gutted, and the entire fish was homogenized before analysis. The detection limits were reported as 0.004 to 0.019 mg g^{-1}, depending on the fish weight. It is assumed that the detection limit units were erroneously reported as milligrams per gram and should have been reported as micrograms per gram (ppm) because concentrations well below the reported limit were measured.

Concentrations of mercury in fish collected from Four Mile Creek at Road F, which is upstream from the seepage basin outcrops, and at Road 4 and Road C, which are in the vicinity of H-Area and F-Area seepage basins, were below the detection limits. Concentrations in all fish collected at Road 3 were also below the detection limits. Four out of six fish collected from Four Mile Creek at the west end of Banana Road had concentrations greater than the detection limit and a mean of 0.017 ppm. Seven out of ten fish collected from Four Mile Creek at Cassel's Pond had concentrations greater than the detection limit and a mean of 0.008 ppm. Two out of eight fish collected from Pen Branch at Road B had concentrations above the detection limit (0.24 and 0.038 ppm). All fish with concentrations greater than the detection limit were sunfish.

One fish from Pen Branch (0.24 ppm) had a concentration similar to concentrations reported in the annual environmental monitoring reports in 1989 and 1990. However, the concentrations reported for Four Mile Creek location are nearly 2 orders of magnitude lower than concentrations reported in the annual monitoring reports and significantly lower than concentrations reported in other studies. For comparison, mean concentrations reported for bream collected from Four Mile Creek at Cassel's Pond in 1989 and 1990 were 0.48 and 0.52 ppm, respectively. An explanation for the large discrepancy between concentrations for fish collected by Loehle and Paller and fish collected routinely by the Westinghouse Environmental Protection Department is not apparent. The method of analysis may account for some of the difference because Loehle and Paller analyzed individual homogenized fish, and the SRS reportedly analyzed 0.3 g plugs of flesh.

As part of the National Pesticide Monitoring Program, the Bureau of Sport Fisheries and Wildlife collected fish from the Savannah River near Savannah, Georgia, in 1969 and 1970 (Henderson et al. 1972). In 1969, mercury concentrations ranged from 0.36 to 1.00 ppm wet weight; in 1970, concentrations ranged from 0.17 to 1.80 ppm. Composite samples of homogenized tissue from two to four fish were analyzed, and the LLD was reported as 0.05 ppm. Species of fish included carp, bluegill, and largemouth bass.

As part of the same program, the U.S. Fish and Wildlife Service collected fish from the same location in 1971 through 1973 (Walsh et al. 1977). In 1971, mercury concentrations ranged from <0.05 to 0.72 ppm wet weight; in 1972, concentrations ranged from 0.14 to 1.25 ppm; and in 1973, concentrations ranged from 0.08 to 0.73 ppm. Composite samples of homogenized tissue from two to five fish were analyzed, and the LLD was reported as 0.05 ppm. Species of fish included carp, bluegill, catfish, sunfish, and largemouth bass.

As part of the National Contaminant Biomonitoring Program (formerly the National Pesticide Monitoring Program), the U.S. Fish and Wildlife Service collected fish from the Savannah River near Savannah, Georgia, in 1978 and 1980 (Lowe et al. 1985). Mercury

concentrations ranged from 0.08 to 0.19 ppm wet weight in 1978 and from 0.06 to 0.57 ppm wet weight in 1980. Three composite samples were analyzed in both years of collection, and each composite contained homogenized tissue from three to five fish. The LLD was reported as 0.01 ppm. Species of fish included catfish, bowfin, and largemouth bass.

Concentrations reported through the National Pesticide Monitoring Program and the National Contaminant Biomonitoring Program indicate significantly elevated mercury concentrations in the Savannah River near Savannah, Georgia. These data also suggest that mercury discharges from upriver industries may have resulted in widespread contamination extending more than 150 mi downriver from the source.

In February 1984, spotted gar and largemouth bass collected from the Savannah National Wildlife Refuge near Savannah, Georgia, had mercury concentrations ranging from 0.17 to 0.41 ppm wet weight (Winger et al. 1990). A follow-up study in August 1985 collected and analyzed fish and other organisms from the Savannah National Wildlife Refuge and the lower Savannah River. Mercury concentrations in these specimens ranged from 0.02 (fiddler crab) to 0.78 (bowfin) ppm wet weight. Aliquots of homogenized whole organisms were analyzed, and the limit of detection was reported as 0.01 ppm.

Information obtained from the Georgia Department of Natural Resources (GDNR 1994) included data regarding a summary of toxic substances found in fish fillet composite samples collected in the fall of 1993. However, a very limited number of samples were collected. Fish collected from the Savannah River in Richmond County (above the SRS) included one largemouth bass and three suckers with mean concentrations of 0.20 and 0.18 mg kg^{-1} (ppm) wet weight, respectively. Concentrations ranged from 0.10 to 0.25 ppm. Fish collected from the Savannah River in Chatham County (near Savannah, Georgia) included three channel catfish and three largemouth bass with mean concentrations of 0.06 and 0.04 ppm, respectively. Concentrations ranged from 0.02 to 0.08 ppm. Samples of fish muscle tissue were analyzed, and the detection limit was reported as 0.01 ppm.

Other South Carolina and Georgia Studies

Various species of fish were collected in 1974 from several South Carolina lakes and rivers (Koli et al. 1977). These locations included the Savannah River and Clark Hill, but relatively few fish were sampled. Because of the small number of fish collected and the fact that different species were collected from the various lakes and rivers, it is difficult to make many conclusive comparisons to SRS data. Mercury concentrations in fish from the Savannah River were similar to concentrations in fish from Clark Hill and generally lower than concentrations in fish from other locations. However, the collection location along the Savannah River was not specified and may have been upriver from Augusta, Georgia. Only two fish (both catfish) were collected from the Savannah River, and concentrations were about a factor of 5 less than concentrations measured by the SRS in catfish from the Savannah River in 1974. Concentrations generally increased with fish size, and liver, kidney, and muscle had the highest concentrations among various tissues.

Largemouth bass were collected from three unpolluted South Carolina reservoirs between March 1973 and November 1975 (Abernathie and Cumbie 1977). Fish muscle tissue was analyzed and the reported detection limit was 0.05 ppm. Fish were grouped according to length, and mean concentrations ranged from 0.34 to 4.49 ppm. Variations in concentrations between the

three reservoirs (differences were statistically significant) were attributed to the ages, trophic states, and water quality characteristics of the reservoirs.

Various species of fish were collected from the Suwannee River below Okefenokee Swamp in Georgia in 1973 (Cumbie 1975). This river was not associated with a recognized point source of mercury contamination. Fish axial muscle was analyzed, and the LLD was reported as 0.02 μg total mercury per specimen. Average mercury concentrations ranged from 0.14 (chubsucker) to 0.81 ppm (chain pickerel), with a maximum measured concentration of 1.40 ppm in a chain pickerel.

Factors Influencing Accumulation of Mercury in Fish

A number of water quality and biological factors can influence the degree to which mercury bioaccumulates in fish. Eisler (1987) presents a very detailed discussion of mercury in the environment. The majority (80 to 99%) of mercury found in fish tissue is organic in the form of methylmercury (Phillips and Russo 1978; Hildebrand et al. 1980; Grieb et al. 1990; Winfrey and Rudd 1990). Accumulation of mercury is generally greatest in liver, kidney, and muscle tissue (Wojtalik 1971; Koli et al. 1977; EPA 1992). A study by the EPA (EPA 1992) generally found higher mercury concentrations in fillet samples than in whole body samples, but the opposite was also true for several locations. This disparity may have resulted from a number of factors, including species variability and stomach content, which can include significant quantities of contaminated sediment.

It is widely accepted that mercury can be bioconcentrated in organisms and biomagnified through food chains. Estimated biomagnification factors (water concentration divided by fish concentration) range from 10^3 to $10^{6.5}$ (Wojtalik 1971; Watras and Huckabee 1994). Consequently, fish living in waters with mercury concentrations well below the PDWS (2 ppb) can accumulate mercury levels in muscle tissue exceeding the FDA “Action Level” of 1.0 μg g^{-1} (ppb) mercury.

In general, mercury concentrations tend to increase with fish age, length, and weight (Grieb et al. 1990; GWQCB 1971; Doi and Ui 1973; Hildebrand et al. 1980; Abernathy and Cumbie 1977; Koli et al. 1977; and Stiefel 1976). Other factors that may influence the formation of methylmercury and subsequent accumulation by fish include pH, dissolved organic carbon, temperature, microbial activity, sulfate, chloride, calcium, suspended sediment load, and sedimentation rates (Wren and MacCrimmon 1983; Bisogni and Lawrenece 1975; Hildebrand et al. 1980; Grieb et al. 1990; Watras and Huckabee 1994; Tsai et al. 1975; Rodgers and Beamish 1983; and Eisler 1987). In addition, piscivorous (e.g., largemouth bass) and bottom dwelling fish have been shown to contain higher mercury concentrations than other species of fish; however, top predatory species generally accumulate mercury to the highest concentrations (GWQCB 1971; Grieb et al. 1990; Koli et al. 1977). The construction of artificial reservoirs has also contributed to elevated mercury concentrations resulting from mercury releases from flooded soils when the reservoirs are first filled (Eisler 1987). Concentrations in fish collected from various reservoirs have been shown to decrease with increasing reservoir age (Abernathy and Cumbie 1977). It is also likely that the rate of mercury methylation is different for reservoirs and streams, resulting in different rates of bioaccumulation.

Miettinen (1973) described a biexponential equation for mercury excretion in aquatic animals. The biological half-life. of the fast component usually varies from 1 to 10 days, while

the slow component ranges from 200 to 1200 days. Tollefson and Cordle (1986) described accumulation of mercury in fish muscle tissue over a period of a few weeks, and a biological half-life of 2 years. In addition, methylmercury has been shown to have the slowest rate of elimination among different mercury species. Methylmercury production is greatest at the sediment-water interface. Methylation generally takes place only in the top few centimeters of sediment (Jernelöv and Åséll 1973), and methylation of mercury by microorganisms can occur under both anaerobic and aerobic conditions (Bisogni and Lawrence 1975; Eisler 1987).

The factors that appear to consistently be correlated with mercury accumulation in fish include species, age, length, weight (positively correlated), and pH (negatively correlated). Other water quality characteristics have been shown to have varying effects in different aquatic systems. In addition, much of the research on factors affecting mercury accumulation in fish has been conducted in lakes and not streams or rivers.

Potential Explanations for Higher Mercury Concentrations Measured by the SRS in Onsite Fish

A definitive explanation for higher mercury concentrations measured by the SRS in onsite fish is not readily apparent. However, based on the vast amount of information in the open literature regarding mercury in aquatic ecosystems, there appears to be a number of potential explanations.

A study conducted by the GWQCB (GWQCB 1971) indicated significantly higher mercury concentrations in Savannah River fish below the Olin Corporation's discharge points than above the discharge points. This appears consistent with the fact that the Olin Corporation has historically discharged significant amounts of mercury to the Savannah River. Information reported in the SRS annual environmental monitoring reports indicates very similar average concentrations in fish from the Clark Hill reservoir and the Savannah River (Table 20-4). This is not consistent with the fact that the reservoir is located a significant distance upriver from the Olin Corporation's discharge points or with the results presented by the GWQCB. Very similar concentrations were measured by the GWQCB in Savannah River fish and by the SRS in onsite fish from areas that received pumped Savannah River water (Table 20-6). In addition, concentrations in fish from onsite locations that did not receive Savannah River water appear similar to concentrations measured in fish from Clark Hill. Concentrations measured in Savannah River fish by the SRS in 1971 are significantly lower than concentrations measured by the GWQCB in 1970–1971.

Average concentrations measured by the SRS in fish from Upper Three Runs at Highway 278 (which would not have been affected by SRS activities or received any pumped Savannah River water) are actually higher than concentrations measured in Savannah River fish (Table 20-4). The concentrations measured in Upper Three Runs at Highway 278 are similar to concentrations measured in other river basins in Georgia (GWQCB 1971) and in "background" locations throughout the United States (EPA 1992). Newman and Messier (1994) reported significant differences between mercury concentrations in fish from locations associated with Savannah River water input and locations isolated from Savannah River water input.

Table 20-6. Mean Mercury Concentrations (ppm) Reported for Bass and Bream Collected by the GWQCB and SRS in 1971

Species of fish	GWQCB (1970–19-71)		SRS (1971)			
	River mile 162	Clark Hill	River	Clark Hill	Onsite[b]	Onsite[c]
Bass	1.53	0.47	ns[a]	0.25	1.74	ns
Bream	0.7	0.06	0.34	ns	0.72	0.30

[a] ns = no sample collected.
[b] Onsite streams that received pumped Savannah River water (FMC, Steel Creek, and Lower Three Runs).
[c] Onsite streams that did not receive pumped Savannah River water (Upper Three Runs).

Savannah River and onsite stream water quality data were examined for the years 1984 through 1991. The pH appears lower for onsite streams (this is based on minimum and maximum yearly values that were provided in the annual monitoring reports), and sulfate and chloride concentrations were consistently lower for onsite streams. Calcium concentrations were significantly higher in Lower Three Runs Creek than in other onsite streams, but mercury concentrations do not appear significantly different in fish from Lower Three Runs than in fish from other onsite streams that received pumped Savannah River water. More acidic conditions in onsite streams may result in increased accumulation of mercury by fish.

It appears likely that the lower concentrations measured in Savannah River fish by the SRS may, in part, be the result of a combination of factors. However, the fact that average concentrations in Upper Three Runs fish have been consistently higher than Savannah River fish and that average concentrations in Savannah River fish have been similar to Clark Hill fish suggests that variations in fish size or age may account for much of the discrepancy. Concentrations have been shown to vary by more than a factor of 3 between fish ages 2 and 9 (Grieb et al. 1990). Concentrations in largemouth bass collected from a South Carolina reservoir have also been shown to vary by more than an order of magnitude between fish less than 230 mm and fish greater than 380 mm (Abernathy and Cumbie 1977). If smaller and younger fish were consistently collected from the Savannah River relative to fish collected from onsite streams and the Clark Hill reservoir, differences in mercury concentrations would result. This certainly seems possible since the Savannah River is commercially fished, while onsite streams and ponds are restricted from public access. Unfortunately, information regarding the size and age of fish collected by the SRS is not available for examination. However, Newman and Messier (1994) concluded that fish size and age were confounding factors in interpretation of SRS fish data. A larger percentage of bass has also consistently been collected from onsite streams and likely accounts for some part of the discrepancy between onsite and Savannah River fish. It is also possible that differences in analytical techniques (e.g., analysis of muscle tissue or homogenated whole fish) have resulted in different concentrations.

It is not clear why fish concentrations measured by the SRS do not appear to have mimicked the decreases noted for sediment concentrations based on data reported by the SRS. Hildebrand et al. (1980) reported a linear relationship between fish and sediment mercury concentrations, with a two-fold reduction in fish mercury concentrations roughly corresponding to a three-fold reduction in sediment mercury concentrations. However, a study of the distribution and accumulation of mercury in the Ottawa River (Miller 1977, cited in Hildebrand et al. 1980) indicated that fish concentrations did not decrease as rapidly as sediment concentrations. This is apparently related to the slower turnover rate of mercury in fish relative to sediment. Eisler (1987) stated that

elevated levels of mercury in aquatic biota can persist for up to 100 years after the pollution source is eliminated. Concentrations reported for mosquitofish collected from an Olin Corporation canal in 1993 suggest that significant mercury contamination remains in this area and may contribute to continued elevated mercury concentrations in Savannah River fish (Newman and Messier 1994). Inconsistencies in fish size and age from year to year may also hinder definitive interpretation of the SRS data.

Conclusions Regarding Mercury

In an attempt to determine whether SRS operations have resulted in elevated mercury concentrations, reported concentrations for several environmental media have been examined. In general, water concentrations measured in onsite streams and in the Savannah River have consistently been near or below the detection limits and well below the PDWS of 2 ppb. There have been no apparent differences between concentrations in the various onsite streams and the Savannah River. Based on water concentrations, there is little evidence to suggest that mercury from F-Area and H-Area seepage basins has resulted in elevated mercury concentrations in any onsite (except at the Four Mile Creek groundwater seepline near the H-Area seepage basin) or Savannah River surface water.

There is little question that small amounts of mercury have migrated from F-Area and H-Area seepage basins into the groundwater. Monitoring of seepage basin wells has confirmed this, but the majority of seepage basin wells (~90%) have consistently had concentrations below the PDWS (2 ppb). In general, the highest groundwater mercury concentrations have been measured in H-Area seepage basin wells. This suggests that migration of mercury through the groundwater to any onsite stream has likely been greatest to Four Mile Creek in the vicinity of the H-Area seepage basins. Seepage basin closure in 1988 and the reporting of total mercury beginning in the third quarter of 1993 (instead of dissolved mercury) hinders complete interpretation of temporal trends.

It appears likely that much of the F-Area and H-Area seepage basin mercury inventory (about 4500 lb) has remained in the environs of the basins, and that the rate of mercury migration into Four Mile Creek (and certainly into the Savannah River) has been small relative to the rate of mercury discharge from the Olin Corporation. At one time, the rate of discharge was reported to be 12 lb d^{-1}. At this rate, Olin plant operations would result in an annual discharge of about 4400 lb of mercury directly to the Savannah River. For comparison, approximately the same amount was discharged by the SRS to F-Area and H-Area seepage basins between 1959 and 1982.

Sediment samples collected from the swampy outcrop along the first sidestream into Four Mile Creek have indicated migration of mercury to this area. However, sediment samples taken from various locations in Four Mile Creek imply that the mercury has remained in rather localized areas. In general, the highest sediment concentrations have been measured in ponds, reservoirs, and settling basins. This suggests accumulation of mercury from increased deposition of suspended sediments in these areas. Sediment concentrations measured in the Savannah River were similar to concentrations measured in sediment collected from many onsite locations, including Four Mile Creek, in the early 1970s. In addition, mercury concentrations in sediment samples collected from Four Mile Creek in 1987 and from the Savannah River between 1992 and 1994 indicate significant decreases since the early 1970s. While this is based on limited data, it

suggests that much of the sediment-bound mercury has been buried or eroded. It also reflects the significant decreases in mercury discharge to the Savannah River from upriver industries.

Sediment concentrations in Four Mile Creek appear to have decreased significantly since the early 1970s. However, concentrations reported for sediment samples collected from the Four Mile Creek seepline near the F-Area and H-Area seepage basins in 1988 and 1989 were more than 2 orders of magnitude greater than concentrations reported for Four Mile Creek stream sediment in 1987. This may be the result of more stable seepline sediments (i.e., not as exposed to erosive forces) as well as localized accumulation of mercury leached from the seepage basins. Based on mercury concentrations measured in Savannah River and Four Mile Creek sediment, however, SRS activities have not resulted in an appreciable mercury releases to the Savannah River.

Based on concentrations of mercury measured in fish collected from the Savannah River at locations above, adjacent to, and below the SRS, which were very similar, SRS activities have not resulted in measurable mercury releases to the Savannah River. Mercury concentrations measured by the SRS in fish collected from the Savannah River have also been very similar to concentrations measured in fish from Clark Hill, which would not have been impacted by Olin Corporation discharges. This is inconsistent with other studies and suggests that collecting smaller or younger fish from the Savannah River may have resulted in lower measured concentrations.

Mercury concentrations measured in fish collected from onsite ponds and streams have consistently been elevated (by about a factor of 2) relative to Savannah River fish concentrations. However, concentrations in fish from Four Mile Creek are similar to or lower than concentrations in fish from other onsite locations. There is little evidence to suggest that mercury from F-Area and H-Area seepage basins has resulted in elevated fish concentrations in Four Mile Creek or any other onsite stream. The highest concentrations appear to be in streams and reservoirs that have received reactor cooling effluent. It is unlikely that cooling effluent would contain mercury resulting from reactor operations, and it appears that the primary source of mercury at the SRS has been the continuous pumping of Savannah River water for use as reactor coolant. In addition, several studies have reported elevated mercury concentrations in fish (approaching and exceeding concentrations measured in onsite fish) collected from South Carolina and Georgia reservoirs lacking a known mercury point source of contamination. A number of factors may be related to the higher measured onsite concentrations. Given the general variability that occurs between fish of various sizes and ages and the variability that appears to occur between fish from various water systems in South Carolina and Georgia, it would be almost impossible to develop a credible SRS mercury source term based on fish concentrations.

It is difficult to determine the exact source of the onsite mercury, but it is certainly possible (and seems likely) that suspended sediment in water pumped from the Savannah River for use as cooling water has accumulated in onsite streams, reservoirs, and settling basins. However, concentrations measured in fish collected from Upper Three Runs Creek at Road F and Highway 278 were also elevated above concentrations in fish from the Savannah River. These locations are upstream from SRS operations and were not impacted by continuous circulation of Savannah River water for use as reactor coolant. Clearly, the Savannah River is not the sole source of onsite mercury contamination, and other anthropogenic (coal burning) and natural (volcanoes and igneous rock) mercury sources contribute to accumulation in onsite fish. It is not clear why fish from Upper Three Runs Creek would have higher concentrations than fish from the Savannah

River, which have been exposed to industrial mercury sources. Differences in fish size and age, however, are likely factors.

In addition, mercury concentrations in fish measured by the SRS have been relatively constant between 1971 and 1991. This is inconsistent with the significant (2 or 3 orders of magnitude) decreases apparent in Four Mile Creek and Savannah River sediment concentrations. If sediment-bound mercury has been buried or eroded, as recent analyses of FMC and Savannah River sediment suggest, it is possible that a consequent reduction in fish concentrations would not be evident for several years. It is also possible that seepage from waste disposal areas at the Olin Corporation continues to release mercury to the Savannah River.

It is clear that some mercury has been leached from the H-Area seepage basins and has migrated at least to the seepline area at Four Mile Creek. However, even if SRS activities have resulted in some mercury releases to onsite streams, Four Mile Creek in particular, they do not appear to have impacted concentrations in Savannah River media, including water, sediment, and fish. Regardless, continued monitoring of environmental media for elevated mercury concentrations is warranted because of the small difference between tolerable natural background levels of mercury and possible harmful effects in the environment.

CHROMIUM

SRS Seepage Basins

In 1975, nonradioactive releases included about 270 and 5100 lb of chromium to F-Area and H-Area seepage basins, respectively (Holcomb and Emslie 1984). Data from trebler samplers in the H-Area effluent stream from October 1980 through October 1981 provide an average effluent concentration of 2200 ppb (addition to Horton 1974b).

SRS Well Water

Water samples from seven H-Area seepage basin wells were collected on November 10, 1981, and four contained concentrations less than 5 ppb. The other three samples had concentrations of 7, 16, and 16 ppb (addition to Horton 1974b).

Data reported in SRS environmental monitoring reports from 1984 through 1991 indicate F-Area and H-Area well water concentrations to be near or below the PDWS of 50 ppb (the PDWS was raised to 100 ppb in 1991) (Figure 20-9). An abnormally high concentration was measured in an H-Area well (HSB 68) in 1984 (1188 ppb), but subsequent analyses of water from this well have indicated concentrations less than 4 ppb. However, considerably higher concentrations have consistently been measured in other areas, particularly in the C-Area burning/rubble pit and D/TNX coal pile runoff basin areas (D-Area). Concentrations in D-Area groundwater have been as high as 2760 ppb (Cummins et al. 1990).

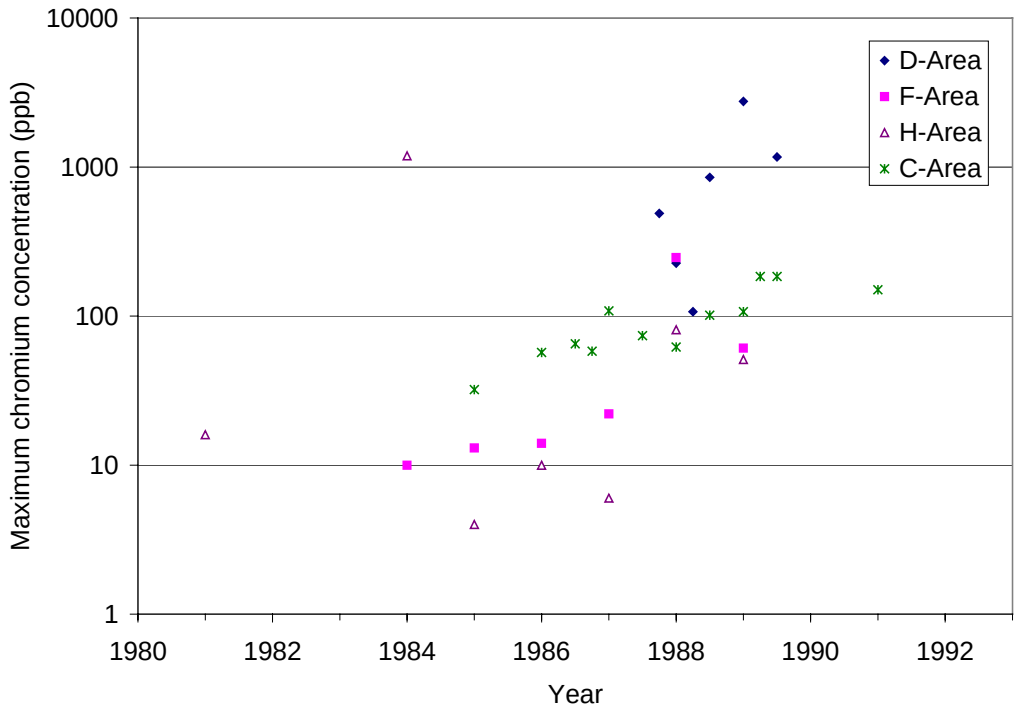

Figure 20-9. Maximum chromium concentrations measured in F-Area, H-Area, C-Area, and D-Area groundwater wells. Link to tabulated data.

Onsite Streams and Ponds and the Savannah River

Water

Geisy and Weiner (1977) measured chromium concentrations in Par Pond water collected between December 1974 and March 1976. The mean concentration for all samples was reported as 0.35 ppb and was described as characteristic of relatively uncontaminated lentic systems in the southeastern United States. Concentrations were determined by flameless atomization.

From 1984 through 1986, maximum measured chromium concentrations in Upper Three Runs, Four Mile Creek, Steel Creek, and Lower Three Runs Creek were reported as less than 10 ppb. In 1987, maximum measured concentrations in the same onsite streams were reported as less than 50 ppb. From 1988 through 1991, maximum measured concentrations in these streams were reported as less than 20 ppb and were generally indistinguishable from maximum concentrations for Savannah River water.

An April 1985 report (Lower 1985) provided mean chromium concentrations of <6 ppb in Savannah River water collected at RMs 155.5 and 157.5. In 1980, Savannah River water samples collected at six stations along the SRS boundary all had concentrations less than the 1 ppb detection limit, except one sample that had a concentration of 2 ppb (ANSP 1980). A September 1996 report (Westinghouse 1996) provided data for 36 samples (12 at each location) collected from the Savannah River between 1992 and 1994 at RMs 120, 150, and 160, which all had concentrations less than 7.5 ppb, the apparent LLD.

Water samples collected in 1987 from Four Mile Creek upstream and downstream of F-Area and H-Area seepage basins and from the seep area downgradient of F and H-Area seepage basins all had chromium concentrations less than 1 ppb (Looney et al. 1988). The authors concluded that the majority of chromium was sorbed to subsurface materials and that transport from both F and H-Area seepage basins was very slow.

Water samples were collected from the Four Mile Creek seepline in the vicinity of the F-Area and H-Area seepage basins in 1988 and 1989 (Haselow et al. 1990). Concentrations in water samples collected from the seepline in the vicinity of the H-Area seepage basins were less than 10 ppb (LLD) in 1988 and ranged from 2.1 to 3.4 ppb in 1989. Concentrations in water samples collected from seepline areas between the F-Area and H-Area were less than 10 ppb (LLD) in 1988 and ranged from less than 3 (LLD) to 4.3 ppb in 1989. Concentrations in water samples collected from the seepline in the vicinity of the F-Area seepage basins ranged from less than 10 (LLD) to 27 ppb in 1988 and from less than 3 (LLD) to 4.8 ppb in 1989.

Water samples were also collected from the Four Mile Creek seepline in the vicinity of the F-Area and H-Area seepage basins in July 1992. Concentrations were reported to be less than the LLD of 4 ppb at the F-Area seepline, Four Mile Creek stream water in the vicinity of the seepage basins, and in seepline and stream water collected upstream from the general separations areas. Water samples collected from the seepline in the vicinity of the H-Area seepage basins had concentrations below the 4 ppb LLD, except one that had a concentration of 13 ppb (an average of three measurements) and one that had a concentration of 4.4 ppb (an average of two measurements).

Sediment

Surface sediment samples were collected and analyzed for chromium from five locations in Four Mile Creek during 1987 (Mikol et al. 1988a, 1988b). Sample locations were Road C, Road A7, the Road A7 flood plain, the place where FMC enters the river swamp, and the mouth of FMC. The respective concentrations were 6, 5, 4, 3, and 7 ppm.

Sediment samples were collected from locations along the Four Mile Creek seepline in the vicinity of the F-Area and H-Area seepage basins and analyzed for chromium in 1988 and 1989 (Haselow et al. 1990). Concentrations in sediment collected from the seepline in the vicinity of the F-Area seepage basins ranged from less than 1 to 11 ppm (mean = 5 ppm, n = 9) in 1988 and from less than 0.6 to 26.2 ppm (mean = 12, n = 6) in 1989. Concentrations in sediment collected from the seepline in the vicinity of the H-Area seepage basins ranged from less than 2 to 15 ppm (mean = 8 ppm, n = 7) in 1988 and from less than 0.6 to 34.4 ppm (mean = 14, n = 4) in 1989.

In October 1976, sediment box core samples were collected from the Savannah River at a location 18 km above the mouth (Goldberg et al. 1979). Concentrations ranged from 150 ppm in the 0–1-cm depth interval to 110 ppm in the 44–48-cm depth interval. The maximum concentration in the core was 260 ppm in the 16–17-cm depth interval.

Winger and Lasier (1995) collected sediment samples from 26 sites along the Savannah Harbor between RMs 9.5 and 23.0. Acid-digested samples were analyzed by inductively coupled plasma emission spectrophotometry. Chromium concentrations ranged from about 10–80 ppm (values were estimated from a figure).

As part of the Savannah River Integrator Operable Unit Study (Westinghouse 1996), several data sets were compiled to provide chromium concentrations measured in Savannah River

sediment from 1992 through 1994. One data set provided concentrations of 0.0193 and 0.0158 ppm for RMs 120 and 184, respectively. Another data set provided concentrations of less than 0.01 ppm for RMs 120, 150.7, and 160. These concentrations are more than 2 orders of magnitude less than concentrations measured in Four Mile Creek in 1987, 1988, and 1989 and four orders of magnitude less than concentrations measured in the Savannah River in 1976.

Chromium concentrations in both water and sediment samples collected from the SRS vicinity are certainly within the ranges of "background" concentrations and well below concentrations measured in contaminated areas compiled for these media by Eisler (1986). Lower (1985) reported a typical soil and crustal rock chromium concentration of 100 ppm. Several of the above studies reported sediment concentrations well below expected background concentrations, which calls into question the accuracy of the detection techniques for chromium.

Fish

Limited data are available for chromium concentrations measured in fish. Ten catfish and two suckers collected from the Savannah River along the SRS boundary in 1980 had mean concentrations of 1.4 and 0.7 ppm dry weight, respectively (ANSP 1980). Five-gram samples of dried muscle tissue were analyzed, and the apparent detection limit was 0.05 ppm. Samples were analyzed by atomic absorption spectrophotometry. For comparison, Mathis and Cummings (1973) reported chromium concentrations in fish from industrialized areas of the Illinois River ranging from 0.02 to 1.06 ppm dry weight.

In August 1985, fish were collected from the Savannah National Wildlife Refuge near Savannah, Georgia, and from the lower Savannah River and analyzed for chromium content (Winger et al. 1990). Concentrations ranged from 0.50 to 10.2 ppm dry weight (converted from wet weight, assuming 80% moisture). Aliquots of homogenized whole fish were analyzed, and the limit of detection was reported as 0.1 ppm wet weight or 0.5 ppm dry weight.

Ten predatory fish, 19 bream, and 12 catfish collected from the Savannah River in the vicinity of RM 150 from 1992 through 1994 had mean concentrations of 3.0, 0.86, and 2.35 ppm dry weight, respectively (Westinghouse 1996). Based on these limited data, concentrations of chromium in fish from the Savannah River in the vicinity of the SRS appear to have remained relatively stable from 1980 through 1992–1994.

Between December 1974 and March 1976, 35 bluegill, 35 blueback herring, 29 brook silverside, 50 golden shiners, and 40 chain pickerel were collected from Par Pond located on the SRS (Geisy and Wiener 1977). Mean chromium concentrations were 0.16, 0.09, 0.28, 0.19, and 0.15 ppm dry weight, respectively. Aliquots of acid-digested, dried whole fish were analyzed by flameless atomization, and the LLD was not specified.

In July 1990, fish were collected from six locations in Four Mile Creek and one location in Pen Branch (Loehle and Paller 1990). Fish were gutted, and the remainder of the entire fish was homogenized before analysis by EPA standard methods (i.e., atomic absorption spectroscopy). The detection limits were reported as 0.1 to 0.2 $\mu g\ g^{-1}$ wet weight (ppm), depending on the fish weight. Assuming 80% moisture, this corresponds to a detection limit of 0.5 to 1.0 ppm dry weight. Mean concentrations ranged from 0.33 to 0.49 ppm wet weight (1.65 to 2.45 ppm dry weight). There were no apparent spatial trends, and the lowest mean concentrations were recorded for fish collected from Four Mile Creek in the vicinity of the F-Area and H-Area seepage basins.

Differences in chromium concentrations in fish collected from the SRS and the Savannah River are not evident. Based on the data that have been examined, there is little evidence to suggest that SRS activities have resulted in elevated chromium concentrations in fish.

Eisler (1986) reported that tissue levels in excess of 4.0 ppm dry weight should be regarded as presumptive evidence of chromium contamination. Winger et al. (1990) reported a maximum chromium concentration in fish collected from the Savannah River Wildlife Refuge, a few miles upriver from the mouth of the Savannah River, of 10.2 ppm. This provides some evidence of chromium contamination in the Savannah River, but there is no indication that this contamination has resulted from SRS activities. In addition, there is some concern about the uncertainties involved with the analysis of some types of biological and environmental samples. Collaborating laboratories have reported order of magnitude differences in measured chromium concentrations in standard bovine liver.

Ecological Aspects of Chromium

In general, fish accumulate chromium to the largest extent in organs such as the spleen, kidney, gall bladder, and gill tissue, while relatively little chromium accumulates in muscle tissue (Knoll and Fromm 1960; Phillips and Russo 1978). No biomagnification of chromium in food chains has been observed, and concentrations are generally highest at the lowest trophic levels (Eisler 1986). In general, chromium compounds have a low fractional absorption from the gut (EPA 1980). Eisler (1986) provides a very thorough review of the ecological and toxicological aspects of chromium in the environment.

In aquatic environments, chromium is virtually always found in the trivalent (+3) or hexavalent (+6) states (EPA 1980). Hexavalent chromium is more toxic than trivalent chromium because of its oxidizing potential and biologic membrane permeability. In addition, biological interactions with chromium involve reduction to the trivalent form and eventual coordination to organic molecules (NAS 1974). Consequently, consumption of fish will involve exposure to primarily trivalent chromium in the form that functions as an essential element in mammals. Most investigators agree that chromium is almost always in the trivalent state in biological materials, and that no adverse effects have been reported for human populations from exposure to chromium in this state through the diet (Eisler 1986). Chromium is also widely accepted as an essential trace element, and a “safe” acceptable daily intake of 0.175 mg d^{-1} was reported in EPA (1980).

Conclusions Regarding Chromium

It is clear that some groundwater wells have contained water with chromium concentrations far exceeding the PDWS (50 ppb through 1990 and 100 ppb since 1991), particularly in the vicinity of the D-Area coal pile runoff basin. However, based on data reported from 1984 through 1991, there do not appear to be significant differences in chromium concentrations measured in onsite stream and Savannah River water, which have both been well below the PDWS and near or below detection limits. In addition, maximum chromium concentrations measured in water collected from the Four Mile Creek seepline downgradient from the F-Area and H-Area seepage basins have also been well below the PDWS.

Chromium concentrations measured in sediment collected from the Savannah River in 1976 were about 4 orders of magnitude greater than concentrations measured in the Savannah River in

1992–1994. Chromium concentrations measured in sediment collected from Four Mile Creek in 1987 were more than an order of magnitude less than in sediment collected from the Savannah River in 1976. Maximum concentrations measured in sediment collected from the seepline in the vicinity of F-Area and H-Area seepage basins in 1988 and 1989 were nearly an order of magnitude less than concentrations measured in the Savannah River in 1976. These data suggest a significant decrease in chromium concentrations in the Savannah River between 1976 and 1992–1994, but without additional data, it is difficult to ascertain the contribution of SRS activities based on sediment concentrations. Additionally, several studies have reported sediment concentrations significantly lower than typical "background" concentrations.

Fish concentrations appear to have remained relatively stable in the Savannah River at locations in the vicinity of the SRS between 1980 and 1992–1994, and concentrations measured in 1985 in the Savannah River near the mouth appear similar to or greater than concentrations measured in the vicinity of the SRS. In addition, concentrations measured in fish collected from Four Mile Creek in 1990 are similar to or less than concentrations measured in fish collected from the Savannah River.

Based on limited data, SRS activities do not appear to have affected chromium concentrations in the waters of onsite streams or the Savannah River. Chromium concentrations measured in fish collected from onsite streams and the Savannah River also provide no evidence for a chromium source term from the SRS. Given the chromium concentrations measured in the various SRS vicinity media and the potential uncertainties associated with these measurements, there is little evidence to suggest elevated levels of chromium in any media resulting from SRS activities. Because chromium does not bioaccumulate significantly in edible portions of fish and trivalent chromium (the form found in fish tissues) is essentially nontoxic to humans, it does not appear to present a health concern to people who may regularly consume fish from the Savannah River.

TABULATED DATA USED TO PRODUCE THE FIGURES IN THIS CHAPTER

The various data summarized in this chapter are electronically compiled in one workbook that contains the figures depicted in this chapter as well as the tabulated data that were used to produce the figures. In this workbook, there is a separate worksheet for each figure and one worksheet that contains the tabulated data for all of the figures. These data can be accessed by clicking on the following hyperlink: Ch20-Figure_data.xls.

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APPENDIX A

SAVANNAH RIVER SITE ANALYTICAL AND COUNTING PROCEDURES FOR RADIONUCLIDES

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SAVANNAH RIVER SITE ANALYTICAL AND COUNTING PROCEDURES FOR RADIONUCLIDES

This appendix presents the sample preparation and analysis methods used to measured radionuclides in environmental samples collected at or in the vicinity of the SRS by the contractor. We focused on those procedures used to measure radionuclides that were determined in this report to be potentially quite useful to the dose reconstruction. The researchers who are involved in the next phase of the dose reconstruction will most likely concentrate their efforts on these radionuclides in their calculations.

The following discussion addresses methods used to measure key radionuclides in ambient air, vegetation and agricultural products, milk, wild game, fish, sediments and soil, and water.

AIR SAMPLE PREPARATION AND ANALYSIS

Based on the evaluation of environmental monitoring in Chapter 8, the key contaminants in air that appear to be most useful to a dose reconstruction are radioiodine and tritium. The following discussion thus focuses on the analyses of air samples for these two radionuclides. Except for details concerning specific analytical procedures, information on sample preparation and analyses were obtained primarily from routine monitoring reports. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series.

Analysis of Air Filters for Iodine

The Health Physics Control Laboratory was responsible for sample preparation and analysis of environmental and regional survey samples beginning with the preoperational survey. Preoperational monitoring of the Savannah River Site (SRS) was conducted in 1951 and 1952 and results are reported in Reinig (1952) and Reining et al. (1953). Du Pont (1953a) contains details of the sampling procedures followed during this period. All samples collected in the field were taken to the Sample Receiving Room in Building 735A and placed in the proper samples storage shelf section. Procedures for laboratory analyses are described below as they are found in Du Pont (1953b).

Air sampling equipment was protected from the elements in housing called 614 buildings. Initially, airborne radioactivity analysis consisted of continuous beta counting of filter paper to determine radon and thoron concentrations. The period from January 1953 to July 1953 was a time of transition to a routine site monitoring program, coinciding with the startup of operations. Airborne particulates were collected by continuous sampling of air, at 2.5 cubic feet per minute (cfm), through a CWS #6 filter. The filters were collected weekly. Two sizes of air filters were used: 2 × 2-in. and 8 × 10-in. filters. (The smaller filters were changed to 2-in. diameter filters and the flow rate was decreased to 2 cfm in mid-1955.) After collection, 3 days were allowed for decay of radon and thoron daughters. The smaller filters were counted for gross alpha activity, using a scintillation counter, and for gross beta activity, using an end-window Geiger-Mueller (GM) tube. The larger filters were analyzed for the number of particles from global particulate

fallout using radioautograph techniques. These filters were assessed for the number of particles per area.

Monitoring for radioiodine in air began in mid-1955. Silver nitrate impregnated HV70 filters were used to collect radioiodine. The flow of air through these filters was 2 cfm, and the filters were changed weekly. The filters were analyzed in the laboratory using a technique that involved dissolution of the filter and extraction with chloroform. Details of this procedure can be found in Du Pont (1959a) and are summarized here. The filters were first cut into pieces and dissolved in nitric and hydrochloric acids. Later in the procedure $CHCL_3$ was added to the solution. The iodine was later recovered by silver iodide precipitation. The precipitate produced was collected on a millipore filter. The dried filter, with precipitate, was then counted on a Geiger-Mueller counter calibrated with ^{131}I.

In early 1962, the silver nitrate-impregnated HV70 filters were replaced with charcoal filters. These filters were counted directly using gamma pulse height analysis. Gamma-emitting radionuclides (including ^{7}Be, $^{141,144}Ce$, $^{134,137}Cs$, ^{40}K, ^{54}Mn, $^{103,106}Ru$, $^{95}Zr/Nb$, and ^{131}I) were determined by counting each particulate or charcoal air filter in a 9 × 9-in. sodium iodide [NaI(Tl)] well detector with a 400-channel gamma spectrometer through 1985.

Beginning in 1986, all samples were analyzed for gamma-emitting radionuclides using a hyper-pure germanium (HPGe) detector. A detailed description can be found in Du Pont (1989).

Detection Limits

Detection limits were reported in semiannual and annual reports for radioiodine from 1957 onward and are presented in Table A-1. In addition, the original data sets typically reported a detection limit when an individual measurement was below detection. The exception was during 1961, 1963, and 1964, when values of “0” were used to represent measurements below the detection limit.

The sensitivity or limit of detection for radioiodine in air reported after 1960 (0.018 pCi m^{-3}) is consistent with detection limits reported elsewhere. Sill and Flygare (1960) reported a detection limit of 13 pCi m^{-3} at a flow rate of 1 ft^3 min^{-1} for a collection time of 1 hour. This correlates to a detection limit of approximately 0.015 pCi m^{-3} for a 2-week collection time and a flow rate of 2.5 ft^3 min^{-1}. A detection limit of 0.1 pCi m^{-3} at a flow rate of 0.35 ft^3 min^{-1} and a collection time of 7 days was reported in APHA (1972). This correlates to a detection limit of approximately 0.007 pCi m^{-3} for a 2-week collection time and a flow rate of 2.5 ft^3 min^{-1}.

In principle, the upper limit of detection is dependent on the state of the iodine and the adsorption capacity of the charcoal. In practice, however, it is limited by the dead time of the detector. This would become appreciable for samples containing more than 1 mCi (1×10^{12} pCi) of activity, which exceeds the amount likely to be collected in any environmental sample (APHA 1989). For comparison, sampling of the maximum measured air concentration at F-Area in 1964 (0.37 pCi m^{-3}) at a flow rate of 2.5 ft^3 min^{-1} for 2 weeks would result in the collection of about 500 pCi total activity.

Table A-1. Detection Limits Reported in Semiannual and Annual Reports for Radioiodine in Air Samples

Time Period	Reported detection limit (10^{-14}μCi mL^{-1})
Jan – June 1957	3.7 ± 0.15
July-Dec 1957	2.3 ± 0.09
Jan – June 1958	2.3 ± 0.09
July-Dec 1958	2.3 ± 0.1
Jan – June 1959	3.3 ± 0.2
July-Dec 1959	3.6 ± 0.1
Jan – June 1960	3.7 ± 0.1
July – Dec 1960	1.8 ± 1.1
Jan – June 1961	1.8 ± 1.2
July – Dec 1961	1.8 ± 1.2
June 1962	1.8 ± 1.2
July 1963	1.8 ± 1.2
1964 through 1965	1.8 ± 0.2
1966 through 1970	1.8 ± 0.2
1971 through 1982	1.8 ± 0.2
1983 through 1985	1.8 ± 1.2
1983 through 1990	1.8 ± 1.2

Analysis of Water Vapor and Rainwater for Tritium

Tritium monitoring in air was initiated in 1956. Tritiated water was captured using silica gel samplers. The water was recovered from the gel in the laboratory via distillation Du Pont (1959a). The distillate was then analyzed using scintillation counting methods described later in this appendix on water sample preparation and analysis.

Rainwater was also analyzed for tritium and other radionuclides. Tritium data were determined to be the most useful for a dose reconstruction, as discussed in Chapter 8. Rainwater samples were analyzed using scintillation counting methods described later in this appendix on water sample preparation and analysis.

Detection Limits

Detection limits were reported in semiannual and annual reports for airborne tritium samples from 1956 onward and are presented in Table A-2 for semiannual and annual averages. In addition, the original data sets typically reported a detection limit when an individual measurement was below the detection limit.

Table A-2. Detection Limits Reported in Semiannual and Annual Reports for Tritium in Air Samples (1956–1980)

Time period	Reported detection limit (pCi L^{-1} water vapor)	Reported detection limit (pCi m^{-3} air)[a]
Jan 1956–June 1958	1000	Not reported per volume of air
July–Dec 1958	6300 ± 900	Not reported per volume of air
Jan–June 1959	4500 ± 600	Not reported per volume of air
July—Dec 1959	3900 ± 200	Not reported per volume of air
Jan–June 1960	3800 ± 300	Not reported per volume of air
July–Dec 1960	4000 ± 400	Not reported per volume of air
Jan–June 1961	4000	40
July–Dec 1961	4000 ± 300	60
Jan–June 1962	3000 ± 200	60
1963–1968	3000 ± 200	60
1969–1970	1000 ± 100	60
1971 – 1973	300 ± 20	10
1974–1980	300 ± 10	Not reported per volume of air

[a] Approximate, varied with absolute humidity.

Table A-3 summarizes the reported lower limits of detection (LLDs) for tritium in rainwater since 1962. The reported LLDs have been relatively stable since 1971.

Table A-3. Lower Limits of Detection for Rainwater Tritium Reported in Semiannual and Annual Reports from 1962 through 1991

Year	LLD (pCi mL^{-1})[a]	Standard deviation [b]
1962–1968	3 ±.2	.003
1969–1970	1 ±.1	.003
1971–1973	0.3 ±.02	.003
1974–1982	0.3 ±.01	NR[c]
1983	0.35 ±.05	NR
1984–1986	0.3 ±.05	NR
1987	0.54	NR
1988—1991	0.4	NR

[a] Based on statistical counting error (90 or 95% confidence levels).
[b] Calculated from spike recovery values.
[c] NR = not reported.

Uncertainties Associated with Air Sampling Procedures

Iodine

The apparatus for sampling iodine in ambient air consists of a cylinder of flexible acetate plastic filled with activated charcoal held in place by a screen. A high-efficiency filter is also used for removal of particulate matter. Air is drawn through the apparatus, and iodine is adsorbed by the charcoal, which can be directly counted for activity determination. Before May 1962, a silver nitrate filter was used to collect radioiodine.

The apparatus used at the SRS initially consisted of silver nitrate-impregnated filters located downline from a 2-in. diameter high-efficiency asbestos paper filter. Silver nitrate filters were replaced by charcoal cartridges in March and April 1962. A continuous flow rate of 2.5 $ft^3\ min^{-1}$ was reported. The filters were collected weekly or biweekly.

Asbestos paper filters were reported to be used in 1971. Mixed fiber papers, including the cellulose-asbestos type, are highly efficient, exhibit low pressure drop, and have relatively low flow reduction because of loading APHA (1977). It is extremely difficult to remove dust and other particulate matter from these types of filters, but they are commonly used to collect radioactive contaminants whose activity can be determined by direct counting of the filters. The filters effectively collect any iodine adsorbed to particulates and a small amount of vaporous iodine. The remainder of vaporous iodine is adsorbed by the charcoal cartridge.

Activated coconut charcoal was reported to be used after April 1962. Silver nitrate was used before that time, until in-plant testing showed that collection efficiencies were far less than the 99% initially reported. Ordinary charcoal is activated by heating it with steam to 800–900°C. This results in the formation of a porous submicroscopic internal structure, which has a very high surface area that can approach 10,000 $ft^2\ g^{-1}$ of charcoal (ACGIH 1989).

Coconut charcoal has a naturally high K^+ content and high pH, both of which enhance adsorption of iodine. This material has been preferred as the natural base material for nuclear air cleaning applications (Holladay 1979).

Iodine is a fairly reactive element and can be found in the air in several chemical and physical forms including gaseous vapor, vapor adsorbed on particulates, or as a gaseous or solid iodine compound. Activated charcoal may not be as effective for removal of iodine compounds. For example, methyl iodide (CH_3I) is quite poorly adsorbed under humid conditions (APHA 1972; ACGIH 1978). However, the capacity of activated charcoal to adsorb organic iodides can be enhanced by impregnation with chemicals such as I_2, KI, triethylenediamine (TEDA), and hexamethylenetetramine (HMTA).

Although activated charcoal is an excellent adsorbent material, there are a few problems associated with its use: precisely, its high adsorbent capability. The process of adsorption is nonspecific and can result in collection of other gases, such as radon, xenon, and krypton. In addition, the majority of adsorbed iodine is usually concentrated on the initial portion of the charcoal bed, which can result in counter geometry problems. The use of silver zeolite cartridges drastically reduces the retention of noble gases. Cline (1981) reported ^{133}Xe retention on silver zeolite to be 15,000 times less than on charcoal. We found no evidence of silver zeolite filters being used within the time period of interest.

For elemental iodine vapor, collection efficiencies for activated charcoal can approach 100%. This is apparently true for all iodine isotopes produced in major quantities in fission processes, but only ^{131}I has a half-life long enough to be of major importance. Collection efficiencies for other forms of iodine, as previously discussed, may be much less. However, with the employment of charcoal impregnated with KI or TEDA, the collection of all forms of nonparticulate iodine will approach 100% (APHA 1989).

Holladay (1979) reported that any good grade of activated carbon, with or without impregnant, will deliver a decontamination factor of 100 (i.e., 99% efficient) for elemental iodine with any combination of temperature and humidity encountered in a nuclear air-cleaning system.

The second and subsequent editions (the first edition was unavailable) of ACGIH (1962, 1967, 1972, 1978) have reported collection efficiencies of elemental iodine greater than 99% for activated charcoal.

Sill and Flygare (1960) conducted a series of experiments that demonstrated the efficiency of charcoal as an adsorbent for iodine. In a laboratory run, which used a series of five 0.25-in. segments (5/8-in. diameter) of activated charcoal and a flow rate of 1–1.5 ft^3 min^{-1}, the activity found in the segments was 58 900, 4 600, 490, 80, and 0 c/m, respectively. The efficiency was excellent (all of the activity was collected in the first four segments and none was collected in the fifth segment) considering the small amount of carbon and the relatively high flow rate. For field samples, most of the activity (about 80%) was also collected in the first 0.25 in. of the cartridge. When two cartridges were placed in series, the first cartridge retained more than 99% of the total activity collected by both cartridges.

Flow measurement accuracy of about 3% is obtainable with most commonly employed properly calibrated devices. A calibration accuracy of 5–10% is achievable depending on how well the distribution of iodine in the calibration standard conforms to the actual distribution of iodine in the sample cartridge (ACGIH 1962).

Various factors can potentially affect the collection efficiency of activated charcoal. In general, the collection efficiencies for elemental iodine are stable with regard to several operating parameters. Efficiency increases with pH and bed depth; decreases with high temperature, highly intense radiation fields, aging, and weathering; and is not affected by charcoal mesh size or relative humidity under 100% (Holladay 1979). Holladay (1979) suggests that ignition caused by radioiodine decay heat can be avoided by maintaining air flow in excess of 5 ft min^{-1}. The SRS flow rate of 2.5 ft^3 min^{-1} through a 2-in. diameter filter correlates to a flow rate of approximately 115 ft min^{-1}. The factors that result in decreased efficiency for collection of elemental iodine are not likely to have been encountered during routine environmental sampling by the SRS.

Particulate Sampling

Biases associated with particulate sampling, using an air filter, would affect gross beta, ^{90}Sr and ^{137}Cs results. The two most important contributors to particulate sampling uncertainty are (1) the volume of air sampled and (2) the total efficiency factor, which incorporates collection efficiency of samplers and filters. The collection efficiency of the ambient samplers is one significant source of negative bias (measurement will be lower than true value).

The volume of air sampled at each SRS monitoring location was calculated by multiplying the flow rate by the time period of collection. This assumes that the flow rate is constant, which is not always the case. Electrical surges and filter loading can affect the flow rate. These variables are situation-specific and would require a study of the monitors used at the Site. Unfortunately, no studies of flow rates could be found at the SRS.

The total collection efficiency of an air sampler can be viewed as having two components:

1. The *inlet collection efficiency* of the sampling device (how accurately the device draws the ambient aerosol into the filter)
2. The *filter collection efficiency* (the amount of the material drawn into the filter that is retained by the filter, i.e., does not pass through it).

For monitoring of SRS releases and global fallout, inlet collection efficiency is not likely to be an important factor. The particle sizes that are subject to large-scale transport are small enough that inlet collection efficiency is expected to be high.

From a review of the technical literature, filter collection efficiency is a much less important source of bias than the inlet collection efficiency, which was discussed in the previous section. A similar conclusion is reached by Eisenbud (1973), which states:

> The uncertainties in estimating the human hazard from inhaling radioactive dust are so great that small differences of the order of 10 to 20% owing to imperfection in filter performance are relatively unimportant and would not affect one's evaluation of a given set of data. All the commercial filter media, when used properly, have efficiencies that are more than adequate to serve the purpose.

Tritium Sampling

Tritium in Air. Tritium in air was sampled by pumping air through a silica gel column, which collected the water vapor. Columns were changed every 2 weeks. The water vapor was separated from the silica gel by distillation. The radioactivity of tritium in the vapor was then determined by liquid scintillation counting. The air concentration was calculated by taking into account the volume of the air sampled and the absolute humidity during the sampling period. Because recovery of water is very near 100% (Murphy et al. 1991), the sources of uncertainty in the sampling procedure would be inaccuracies in the volume and absolute humidity. We could find no uncertainty values associated with these data reported in SRS documents.

Tritium in Rainwater. Rainwater was collected in pans and analyzed in the laboratory for tritium. The major source of uncertainty in the sampling procedure would be inaccuracy in the volume collected. We could find no uncertainty values associated with these data reported in SRS documents.

Uncertainties Associated with Laboratory Analyses

Beginning in 1958, uncertainties associated with laboratory analyses, based on the analyses of spiked samples, were reported in semiannual and annual reports. Unfortunately, standard deviations were not reported for iodine in air. Table A-4 presents standard deviations for tritium in low-level samples for the years 1958 through 1980.

Table A-4. Standard Deviations Reported in Semiannual and Annual Reports for Tritium in Water Vapor Samples

Time period	Reported standard deviation (%)[a]
Jan–June 1958	5
July–Dec 1958	6
Jan–June 1959	3
July–Dec 1959	6
Jan–June 1960	6
July–Dec 1960	6
Jan–June 1961	5
1962–1973	3
1974–1980	Not reported

[a] Calculated using spike recovery values.

VEGETATION AND AGRICULTURAL PRODUCT SAMPLE PREPARATION AND ANALYSIS

Based on the evaluation of environmental monitoring in Chapter 9, the key contaminants in air that appear to be most useful to a dose reconstruction are ^{131}I and tritium. The following discussion thus focuses on the analyses of vegetation and agricultural product samples for these two radionuclides. Except for details concerning specific analytical procedures, information on sample preparation and analyses were obtained primarily from routine monitoring reports. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series.

Vegetation and Agricultural Product Sample Preparation

Following collection, plant samples were oven dried at 80–90°C for 24 hours. A portion of each sample was retained before drying for tritium analysis of free water obtained through freeze-drying. Concentrations were reported as activity per gram (dry weight), except tritium, which was reported as activity per milliliter, and radioiodine, which was reported on a wet weight basis through June 1961.

Analysis of Vegetation and Agricultural Products Samples for Iodine and Gamma-Emitting Radionuclides

Concentrations of ^{131}I were reported from 1955 through 1991. Before 1961, radioiodine concentrations were determined by counting on a GM counter calibrated with ^{131}I (Du Pont 1959b). Undried vegetation and agricultural product samples were first digested with chromic and sulfuric acids, and the radioiodine was reduced with a 30% solution of phosphoric acid and removed by distillation. A chemical separation technique involving chloroform extraction followed by iodine recovery using silver iodide precipitation was then used to isolate the

radioiodine. In 1961, the chloroform extraction procedure was replaced by gamma spectrometry (described below).

Gamma-emitting radionuclides (including ^{7}Be, ^{141,144}Ce, ^{134,137}Cs, ^{40}K, ^{54}Mn, ^{103,106}Ru, ^{95}Zr/Nb, and ^{131}I) in vegetation were determined by counting dried, briquetted samples in a 9 × 9-in. NaI(Tl) well detector with a 400-channel gamma spectrometer from July 1961 through 1985. The plant material was dried at temperatures less than 110°C, briquetted in a hydraulic press, and canned with a commercially available canner. Beginning in 1986, all samples were analyzed using a HPGe detector.

Gamma-emitting radionuclide concentrations in agricultural products were measured by counting a bottled sample in hydrochloric acid solution. The methods of detection were not always explicitly stated, but it is assumed that samples were counted similarly to vegetation using a 9 × 9-in. NaI(Tl) well detector through 1985 and a HPGe detector thereafter

Analysis of Vegetation and Agricultural Product Samples for Tritium

Tritium concentrations in vegetation and agricultural products were reported from 1974 through 1991. Free water was obtained by freeze-drying and counted using a liquid scintillation detector. The LLD for tritium from 1974 through 1985 was 1 pCi mL^{-1} and for 1986 through 1991 ranged from 1.09 to 1.85 pCi mL^{-1} for a short count (20 minutes) and from 0.28 to 0.38 pCi mL^{-1} for a long count (300 minutes).

Agricultural products were prepared in a manner similar to that used for eating. Peelings, seeds, and other nonedible portions were removed. Wheat, containing the whole grains only, and oats, containing both grains and husks, were processed unwashed. In general, the majority of samples have been collected during the harvest season.

Analysis procedures for tritium and strontium were the same as those described for vegetation. Concentrations have been reported as picocuries per gram wet weight, except for tritium, which was reported as picocuries per milliliter free water. The analysis procedures for meat samples were not explicitly stated, but it is assumed that analysis procedures were similar to those described for wild game.

MILK SAMPLE PREPARATION AND ANALYSIS

Based on the evaluation of environmental monitoring in Chapter 10, the key contaminants in air which appear to be most useful to a dose reconstruction are ^{131}I, ^{137}Cs and ^{89,90}Sr. The following discussion thus focuses on the analyses of milk samples for these radionuclides. Except for details concerning specific analytical procedures, information on sample preparation and analyses were obtained primarily from routine monitoring reports. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series.

As of 1961, it is stated that controls were established in the laboratory groups when the "rerun" system was started. Spiked samples were analyzed with sets of routine samples and at intervals established by the laboratory supervision. In addition to this program, routine samples were selected randomly and analyzed again a week or more later. Although it was reported (Marter and Boulogne 1961) that the results of the spike samples and the laboratory "reruns" were published monthly, such results have not been found when reviewing the documents. Results of the SRS's participation in the interlaboratory cross-check program are provided in

annual reports starting in 1977. An appendix detailing the SRS's quality control program can be found in the annual reports starting in 1980.

The minimum detection limit with 90% confidence interval was reported for milk analyses based on statistical counting error. No self-absorption corrections were applied, but probably contribute minimally to the total uncertainty. Percentage standard deviations were also reported based on the spike recovery values. It was stated that these values were applicable to the reported 6-month average data. These data are summarized for ^{131}I and ^{90}Sr in Tables A-5 and A-6, respectively.

Analysis of Milk Samples for Iodine

The earliest analyses (1956–1960) used a radiochemical separation technique and the sample was counted with a calibrated GM counter. An ion exchange - gamma spectrometric technique was introduced in 1960 that greatly improved the sensitivity of the analysis. This general technique has remained essentially the same ever since. Subsequent improvements resulted from using deep well detectors, larger crystals, and more sophisticated multichannel analyzers. The sample size and counting time, which also affect the sensitivity of the analyses, have also changed over time. Table A-5 summarizes the detection limit that was achieved for milk sample analysis. A description of the techniques used is provided in the following paragraphs.

Before the third quarter of 1960, milk samples were analyzed for radioiodine using a chemical separation technique that involved a chloroform extraction procedure to extract the iodine from other elements, followed by iodine recovery using silver iodide precipitation. To prepare the milk for chloroform extraction, most of the water was removed by alkaline distillation. The organic fraction of the milk was digested with chromic and sulfuric acids. The iodine was reduced with phosphoric acid and removed by distillation (Du Pont 1959a). The final sample (filter paper on a planchet) was counted on a GM counter calibrated with ^{131}I. For procedure control, a spike and a blank were included with each group of samples. According to Du Pont (1959a), the manual for standard operating procedures at the time, the results were corrected if the recovery efficiency of the procedure was less than 95%. All results were corrected for decay. It would appear that a 100 mL sample of milk was used for the analysis. Sample counting time is not reported.

An ion exchange - gamma spectrometric technique was introduced in 1960 that greatly improved the sensitivity of the analysis. The new technique was first described in the March 21–25, 1960, Control and Methods Weekly Report (Du Pont 1960b). It was introduced in the second half of 1960 for routine analysis of ^{131}I in milk (Boni 1960, 1962; Du Pont 1960b). The technique involved the direct absorption of ^{131}I on anion exchange resin, transfer of the resin to a 500-mL polyethylene bottle, and the subsequent analysis on top of a 3 × 3-in. sodium iodide, thallium activated crystal (NaI[Tl]), gamma detector. A 1-gal sample of milk was used during a 10-minute counting time with a 200 RIDL multichannel analyzer. Although the third quarterly report for 1960 (Du Pont 1960c) gives a detection limit of 9 pCi L^{-1}, the minimum detection limit was reported as 11.0 pCi L^{-1} in subsequent tabulations of the results and a subsequent memo (Boni 1960). This suggests that for routine analyses, it was not possible to achieve the sensitivity initially anticipated. The reports state that a correction for background was made in the analysis (Boni 1960). The procedure was also reported at the annual Health Physics Society Meeting in

1962 (Boni 1962) with a minimum detection limit of 3.5×10^{-15} Ci mL^{-1}. Again, this suggests that for routine analysis this sensitivity was not achieved.

Table A-5. Changes in Detection Limit for Radioiodine in Milk Samples

Time period	Reported detection limit (pCi L^{-1})	Standard deviation (%)
January–June 1957	120 ± 5	22
July–December 1957	Not reported	Not reported
January–June 1958	92 ± 5.3	18
July–December 1958	100 ± 6	23
January–June 1959	150 ± 10	26
July-Dec, 1959	160 ± 10	15
January–June 1960	140 ± 7	10
July–December 1960	10 ± 1.5	2.5
January–June 1961	9.8 ± 1.5	11
July–December 1961	5.4 ± 1.0	15
June 1962	2.2 ± 0.2	10
July 1963	11.3	Not reported
1964 through 1965	2.2 ± 0.5	10
1966 through 1970	5[a]	Not reported
1971 through 1982	1 ± 0.5	Not reported
1983 through 1985	1.2 ± 0.5	Not reported
1983 through 1990	3[b]	Not reported

[a] Detection limit reported as 2.2 ± 0.5 pCi L^{-1} (10% Std. Dev.) in the annual reports, but 5 pCi L^{-1} appears to be the norm based on the aperture cards. It is not clear what (sample size, counting time) accounts for the difference.

[b] Detection limit is reported as 1.2 ± 0.5 pCi L^{-1} in the minimum detectable concentration table but as 3 pCi L^{-1} in text of the annual reports. It is possible that the detection limit for ^{131}I in milk was erroneously reported in the text and that the error was propagated in subsequent reports.

The detection limit was reduced from 11.0 to 6 pCi L^{-1} starting in July 1961. This improvement appears to have resulted from counting the sample directly in a well NaI detector and no longer transferring the resin to a 500-mL polyethylene bottle; however, no explicit documentation for this has be found to date.

A further improvement on the ion exchange technique was introduced for the routine analysis of milk during the last week of June 1962 (Du Pont 1960b). The milk sample was counted directly with a large (9 × 9-in.) NaI(Tl) crystal with a deep (3.25 × 6-in.) well (Boni 1962). The sample size and sample counting time remained unchanged. The minimum detection limit was reported as 2.2 ± 0.2 pCi L^{-1}. The uncertainty was based on statistical counting error (90% confidence interval) (Du Pont 1963). The sample size was reduced to one-half gallon in July 1963 and the minimum detection limit was reported as increasing to 11.3 pCi L^{-1}.

In 1980, the milk sample size was 3.8 L (1 gal). The sample was counted for 200 minutes on a (9 × 9-in.) NaI(Tl) well detector. The detection limit was reported as 1 ± 0.5 pCi L^{-1}. Based on the reported detection limit it would appear that this procedure was used from 1971 through 1982.

In 1983, a 2-L (slightly in excess of one-half gallon) sample of whole milk was passed through an anion exchange resin ("Dowex L-X8") column. The column was counted in a (9 × 9-in.) NaI(Tl) well detector.

The 1989 and 1990 annual monitoring reports (Cummins et al. 1990, 1991) state that one-half gallon of fresh raw milk was obtained from each sample location, but just 1-L aliquots (0.2642 gal) were determined by direct count on a high purity germanium (HPGe) detector. The count time is not specified.

Problems associated with the analytical procedures are occasionally noted in the reports. However, it is difficult to relate this information to the reported measurements. For example, in the September 26–30, 1960, weekly report (Du Pont 1960d) it is stated that the ^{131}I recoveries in spiked milk were suddenly low and variable. Close control of the pH of the spike solution was found to be very important. The presence of sodium hydroxide in the milk inhibited the concentration of ionic iodine by the anion resin. It was speculated that high basicity probably caused saponification of the butterfat that absorbs iodine and inhibits complex formation with the resin.

Analysis of Milk Samples for Cesium

In 1963, milk samples were counted directly in 500-mL volumes in 16-oz narrow-mouth polyethylene bottles for ^{137}Cs (Boni 1963). If no other isotopes were present, a sensitivity of 23 pCi L^{-1} was reported for a 10-minute count time. By 1965, one-half gallon milk samples were passed through an ion column. The ^{137}Cs in the column effluent was concentrated on an ion exchange column for gamma spectrometric analysis. A minimum sensitivity of 11 pCi L^{-1} was reported (Du Pont 1965). A two-part ion exchange column was used to sequentially separate and concentrate radioiodine and cesium from the same sample of milk (1 gal). The second part of the column contained granular potassium cobalt ferrocyanide (KCFC), a highly selective absorbent for cesium. A minimum sensitivity of 3 pCi L^{-1} with a 10-minute count time and a 9 × 9-in. well crystal was reported (Marter 1965).

By 1972, ^{137}Cs was removed from the sample by passing approximately 2 L of milk through a KCFC resin column. The column was counted directly using a 9 × 9-in. NaI(Tl) well detector with a 400-channel gamma spectrometer (Du Pont 1973). This procedure continued through 1986. From 1987 through 1990, a HPGe detector was used to count a 1-L aliquot of milk directly.

Analysis of Milk Samples for Strontium

The method used to analyze ^{90}Sr in milk samples appears to have changed very little over the entire time period of the monitoring program. The milk was first passed through an ion exchange resin to chemically extract the strontium, followed by acid leaching of the resin. The short-lived ^{90}Y daughter was removed from the eluate before ingrowth was allowed to begin. After equilibrium of ^{90}Y with ^{90}Sr was established, the ^{90}Y was again stripped from the solution and counted with a low background beta counter. The minimum detection limit for the analysis has remained very similar throughout the entire time period as a consequence. Table A-6 summarizes the detection limit achieved for milk sample analysis. In April 1961, ^{90}Sr determinations in milk were made using a di-2-ethylhexyl phosphoric acid (HDEHP) extraction

procedure that used NH_4OH instead of NaOH in the neutralization step. Total radiostrontium ($^{89,90}Sr$) was determined using a rhodizonic acid procedure.

In 1962, milk samples were prepared using a chemical extraction process that involved ion exchange, acid leaching, dissolving in a dilute solution of HCl, and extraction ^{90}Y into HDEHP in toluene solvent. The ^{90}Y was finally counted on a planchet in a low background beta counter. A 500-mL sample of milk and a counting time of 60 minutes were (Butler 1962). The minimum detection limit was stated as 1.0 ± 0.1 pCi L^{-1} based on statistical counting error (90% confidence interval). In March 1964 (Du Pont 1964), the analytical procedures for determination of ^{90}Sr and $^{89,90}Sr$ were combined to reduce analysis time and to improve precision. Recovery of ^{90}Sr was 82% ± 5%, and total radiostrontium was 68% ± 5%. In 1965 a rhodizonic acid separations procedure was used. It had a minimum detection limit of 1.0 ± 0.1 pCi L^{-1}.

The same procedure appears to have been used from 1972 through 1990 for the analysis of ^{90}Sr in milk. One-half-gallon samples were collected and 0.5-L aliquots were analyzed. The strontium and yttrium were removed as chlorides by a slurry of the whole milk with a cation resin. The resin was leached with 8N nitric acid. The acid was evaporated to dryness and dissolved in 0.08N hydrochloric acid. To begin the ^{90}Y ingrowth, ^{90}Y is removed from the ^{90}Sr by liquid ion-exchange using HDEHP in toluene. Equilibration of ^{90}Y with ^{90}Sr is approached over a 15-day period, after which the short-lived ^{90}Y daughter is stripped and counted in a low-level gas flow beta proportional counter (Du Pont 1973). The amount of ^{90}Sr is calculated by relating the ^{90}Y buildup to the original ^{90}Sr concentration. The sample count times are known to have changed—a 50-minute count was used from 1974 through 1982 and a 20-minute count time was used from 1983 through 1985. These differences probably account for the small changes in the minimum detection limit for the analysis (Table A-6).

Table A-6. Changes in Detection Limit for ^{90}Sr in Milk Samples

Time period	Reported detection limit (pCi L^{-1})	Standard deviation (%)
1959	Not reported	Not reported
1960	1	Not reported
January–June 1961	1.6 ± 0.3	11
July–December 1961	1.0 ± 0.1	7
1962 through 1973	1.0 ± 0.1	7
1974 through 1982	1.1 ± 0.12	Not reported
1983 through 1985	2.4 ± 0.75	Not reported
1986 through 1988	1.62	Not reported
1988 through 1991	1.9	Not reported

WILD GAME SAMPLE PREPARATION AND ANALYSIS

Based on the evaluation of environmental monitoring in Chapter 11, the key contaminants in wild game that appear to be most useful to a dose reconstruction are ^{137}Cs and $^{89,90}Sr$. The following discussion thus focuses on the analyses of wild game samples for these two radionuclides. Except for details concerning specific analytical procedures, information on sample preparation and analyses were obtained primarily from routine monitoring reports. See

Chapter 7, Table 7-1 for a complete description of the various monitoring report series. Methods of analysis and lower limits of detection have not always been explicitly stated in the environmental monitoring reports. The following paragraphs summarize the information given in these reports regarding sample preparation and analysis (primarily for deer). It is assumed that similar methods were used for other wild game samples. All concentrations have been reported as picocuries per gram wet weight.

Analysis of Wild Game for Cesium

Between 1965 and 1969, approximately 20% of the total number of deer harvested were monitored for ^{137}Cs and nonvolatile beta activity by random sampling of foreleg flesh and bone for laboratory analysis. Additionally, deer were monitored for external contamination with a Thyac GM survey meter. Beginning in 1970, all individual deer were monitored in the field by 2-minute duplicate counts using two different portable NaI 2 × 2-in. detectors before their release to the hunter. Details regarding the operation of this instrument can be found in Rabon and Johnson (1973) and McMahan and Wright (1973). Nonvolatile beta concentrations were reported for deer muscle and bone tissue before 1969, but the methods of sample preparation and analysis were not clearly specified. However, a gas-flow proportional detector was likely used, following wet-ashing of collected tissue with nitric acid and hydrogen peroxide

Gamma spectrometry was first used in 1956 to identify gamma-emitting radionuclides in environmental samples (Harvey and Rabon 1965). While this process identified specific radionuclides, average nonvolatile beta concentrations were predominantly reported through the early 1960s. In 1962, concentrations of specific radionuclides in wild game samples began to be routinely reported.

Cesium-137 concentrations have been consistently reported for deer muscle tissue samples since 1965. Tissue samples were collected for laboratory analysis from approximately 20% of the total number of deer harvested before 1970. Since 1970, a fraction (ranging from 2 to 20%) of the total number of deer harvested have been analyzed in the laboratory to verify the field measurements. The SRS environmental monitoring reports did not specify methods of sample preparation or analysis before 1987. However, Rabon (1968) and McMahan and Wright (1973) detailed a laboratory method of analysis for deer collected from the SRS. A 500-g (wet weight) sample of muscle was wet-ashed in nitric acid and hydrogen peroxide and subsequently placed in a 3 × 6-in. well of a 9 × 9-in. NaI(Tl) crystal connected to a 400-channel pulse height analyzer for counting.

Table A-7 complies data comparing field and laboratory measurements of ^{137}Cs in deer from aperture card printouts for several years between 1972 and 1981 and the Savannah River Plant Site Environmental Reports for 1985 through 1991. In general, the field and laboratory measurements have been in good agreement, and neither method appears to have provided consistently higher values. Figure A-1 shows concentrations measured in the field as a function of concentrations measured in the laboratory for the same years described in Table A-7. The relationship appears very linear with a slope of 1 through about 35 pCi g^{-1}. Beyond this concentration, the field measurements appear to slightly underestimate the concentrations measured in the laboratory. However, this should not have presented a problem because all animals with field estimates of 25 pCi g^{-1} or greater were sampled for laboratory analysis. McMahan and Wright (1973) also reported a plot of field estimates versus laboratory estimates,

and the data appeared linear with a slope of 1 to about 30 pCi g^{-1}. Beyond 30 pCi g^{-1}, the field estimates again appeared to slightly underestimate the laboratory estimates although there were few data points.

Table A-7. Comparison of ^{137}Cs Concentrations Measured in the Field and Laboratory

Year	Ratio of field to laboratory measurements	Number of samples
1972[a]	0.93	20
1974[a]	1.01	8
1975[a]	1.21	56
1977[a]	0.96	82
1978[a]	0.98	79
1979[a]	1.01	52
1980[a]	0.71	58
1981[a]	1.13	53
1985	1.21	42
1986	1.37	41
1987	0.90	35
1988	1.20	67
1989	0.96	96
1990	0.69	93
1991	0.69	124[b]
Overall average	0.96	Total number = 906

[a] Data are from aperture card printouts.
[b] Includes measurements for both deer and hogs.

Methods of sample preparation and analysis for feral hogs and other fur-bearing animals have generally not been explicitly described in the environmental monitoring reports, but it is assumed that analysis techniques similar to those described for deer were used.

Methods of sample preparation and analysis for waterfowl have also not been explicitly described in the environmental monitoring reports. However, Straney et al. (1975), Fendley et al. (1977), and Domby et al. (1977) described a method that was used for counting 500-g samples as well as individual whole birds collected from the SRS. A Packard Model 446 Armac liquid scintillation detector equipped with a Packard tri-carb scintillation gamma spectrometer was used for radiocesium counting in these studies.

Thyroid samples were monitored for ^{131}I by the SRS using the same detection methods as described for ^{137}Cs. The LLD reported for ^{131}I in 1965 was 6 pCi g^{-1} and has been 1 pCi g^{-1} since 1976. Beginning in 1984, thyroid samples were analyzed for ^{129}I by the University of Tennessee, Memphis. Samples were analyzed by the Center for Health Sciences (1984–1986) and the Department of Physiology and Biophysics (1987–1991). The detection method or LLD for these samples were not specified in the SRS environmental reports.

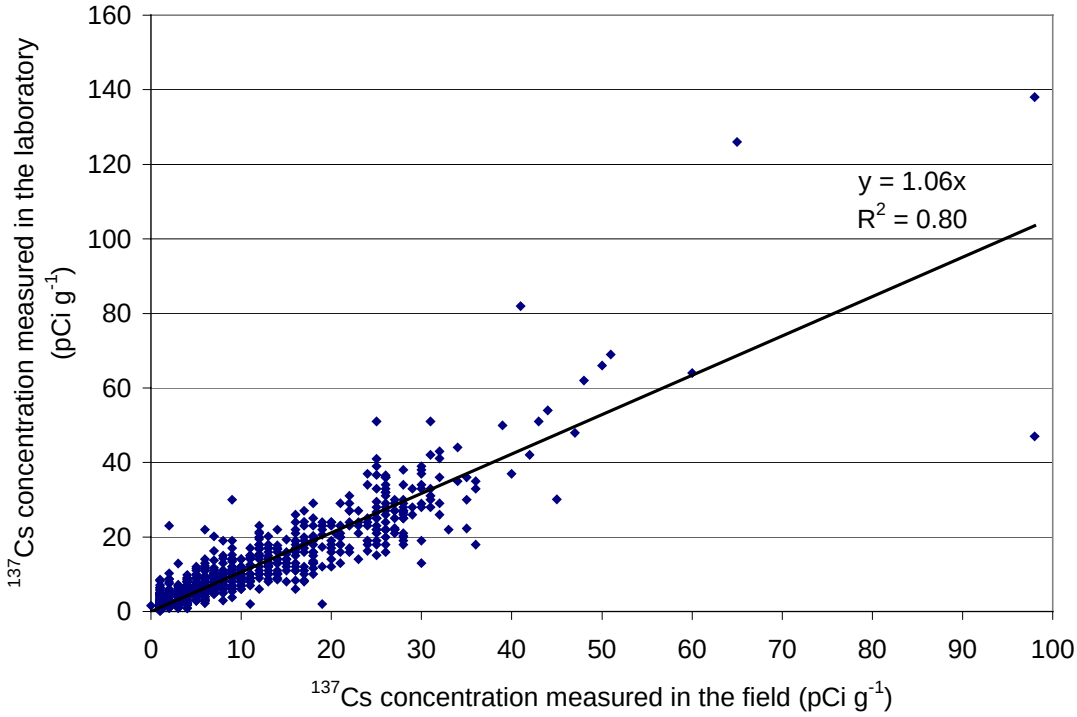

Figure A-1. Relationship between ^{137}Cs concentrations measured in the field and laboratory.

Analysis of Wild Game for Strontium

Bone samples were analyzed for radiostrontium by the SRS using a low-background gas-flow proportional counter (Du Pont 1977). Samples were ashed in a furnace at 700–900°C, leached with 6N hydrochloric acid, evaporated to dryness, and dissolved in 0.08N hydrochloric acid. Yttrium-90 was then stripped from the strontium by liquid ion exchange using di-2-ethylhexyl phosphoric acid. Equilibrium was then established with the short-lived ^{90}Y daughter over a 15-day period, which was then once again stripped. The remaining radiostronium was then transferred to a stainless steel planchet for counting. Rabon (1968) described a method for separating radiostrontium from large amounts of calcium by using potassium rhodizonate as a selective precipitating agent.

FISH SAMPLE PREPARATION AND ANALYSIS

Based on the evaluation of environmental monitoring in Chapter 14, the key contaminants in fish, which appear to be most useful to a dose reconstruction, are ^{65}Zn, ^{137}Cs and ^{89,90}Sr. The following discussion thus focuses on the analyses of fish samples for these radionuclides. Except for details concerning specific analytical procedures, information on sample preparation and analyses were obtained primarily from routine monitoring reports. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series.

Analysis of Fish Samples for Cesium and Zinc

Composite flesh samples were analyzed for ^{137}Cs from 1961 through 1970; beginning in 1971, homogenized whole individual fish were analyzed. In 1990 and 1991, samples were blended and decanted and placed in sample bottles for analysis. Sample preparation methods were not clearly specified between 1971 and 1990, but samples were most likely prepared in a similar fashion. Through at least 1965, samples of muscle or homogenized fish were wet-ashed in nitric acid and hydrogen peroxide and subsequently placed in a bottle for analysis (Harvey and Rabon 1965). Methods of sample preparation for counting were not clearly specified until 1990, when blended samples were analyzed with no further preparation.

Before July 1961, only nonvolatile beta concentrations in fish muscle and bone tissue were routinely reported. Nonvolatile beta concentrations were also reported for fish through June 1969, but the methods of sample preparation and analysis were not clearly specified. However, following wet-ashing of collected tissue with nitric acid and hydrogen peroxide, a gas-flow proportional detector was likely used.

Gamma spectrometry was first used in 1956 to identify gamma-emitting radionuclides, such as ^{65}Zn and ^{137}Cs, in environmental samples (Harvey and Rabon 1965). While this process served to identify specific radionuclides, average nonvolatile beta concentrations were predominantly reported through the early 1960s. In July 1961, concentrations of specific radionuclides in fish samples began to be routinely reported.

Since July 1961, samples have been analyzed in a 3 × 6-in. well of a 9 × 9-in. NaI(Tl) crystal connected to a 400-channel pulse height analyzer. Beginning in 1986, both NaI and HPGe detectors were used, and HPGe detectors have been used exclusively since 1989.

Analysis of Fish Bone Samples for Strontium

Strontium-89,90 concentrations were reported for fish bone samples from 1961 through 1980. Bone samples for all species were composited for monthly analysis. Samples were analyzed using a low-background gas-flow proportional counter. Further details regarding the counting procedures for radiostrontium are discussed in the wild game sample preparation and analysis section.

SEDIMENT/SOIL SAMPLE PREPARATION AND ANALYSIS

Based on the evaluation of environmental monitoring in Chapters 12.1 and 12.2, the key contaminants in sediments and soil which appear to be most useful to a dose reconstruction are the long-lived alpha-emitting radionuclides, uranium and plutonium. The following discussion thus focuses on the analyses of sediment or soil samples for these two radionuclides. Except for details concerning specific analytical procedures, information on sample preparation and analyses were obtained primarily from routine monitoring reports. See Chapter 7, Table 7-1 for a complete description of the various monitoring report series.

According to in Du Pont (1953b), sample preparation in the early years of the program consisted first of drying and sifting the sample, using a No. 8 sieve. Approximately 5 grams of this sample was ground, using an electric mortar and pestle, until the entire sample could pass through a 200-mesh sieve. One gram of this material was placed in a crucible and ashed in a muffle furnace. The ash was dissolved using nitric and hydrofluoric acids. Dried residue was then analyzed for gross alpha concentrations using a ZnS scintillation detector through 1982 and was

analyzed using a gas flow proportional counter from 1983 through 1991. For specific determination of uranium and plutonium, the uranium and plutonium were extracted using tri-n-butyl phosphate (TPB) and then back-extracted into distilled water. The sample is dried on planchet. Alpha pulse height analysis was then used to determine the ratio of uranium activity to plutonium activity. Du Pont (1989) does not contain the procedures used to extract of uranium and plutonium from soil samples. However, a procedure for triisooctylamine (TIOA) extraction for vegetation samples. Since the TBP extraction was used for both types of media earlier, it could be assumed that the TIOA extraction was also used for sediment and soil samples, beginning in the early 1980s.

WATER SAMPLE PREPARATION AND ANALYSIS

The Health Physics Control Laboratory was responsible for sample preparation and analysis of environmental and regional survey samples beginning with the preoperational survey, and are quite well documented. Preoperational monitoring of the SRS was conducted in 1951 and 1952 and is reported in Reinig et al. (1953). Water samples from about 15 locations along the river were analyzed for gross alpha, gross beta, uranium/plutonium alpha, radon, radium, and ^{40}K content. The earliest sampling and procedures guide for SRS environmental samples was dated May 1953 and provided concise steps in obtaining and handling weekly water samples (Du Pont 1953a).

A jug sinker was used to obtain water samples when there was sufficient water depth to do so. A jug sinker was a device designed to hold either a 1-gal or a 1-L water jug. It was constructed of iron with the bottom and side runners of wood. The bottles were held in place by an iron hasp and a strap with a center hole that enclosed the jug neck. A rope was attached to a pail-type handle for lowering into the water. Random samples were obtained by lowering the jug sinker to the bottom of the river, stream, pond, or well to be sampled and raising the bottle slowly to allow filling. When water depth was not sufficient, bottles were held on their sides with the mouth pointing upstream, about 1 ft below the surface of the water. All samples collected in the field were returned to the Sample Receiving Room in Building 735A (Du Pont 1953a).

At the time of plant startup, analysis of water samples was limited primarily to gross alpha and nonvolatile beta measurements. The original procedures, outlined in 1953, gave “an approximate value for the level of alpha and beta contamination in water samples. Self-absorption of radiation in the sample and loss of volatile emitters lead to variation in results. Consequently, the method serves only as a rough check and must be supplemented and supported by specific analyses.” Uranium nitrate-spiked samples were used as a control, with a reported average alpha recovery of 35% and a standard deviation of 19%. Self-absorption was determined by an analysis of 11 spiked samples of tap water with a recovery of 55% (standard deviation of 12%). Samples were evaporated to a small volume (100 mL) and transferred by pipette to a stainless steel planchet. After evaporation, the planchet was flamed, mounted on a card, and sent to the counting room for gross activity counting.

Special analyses for uranium by fluorophotometry, potassium by spectophotometry, thorium by spectrophotometry, and plutonium by tributyl phosphate (TBP) extraction were described in the initial procedures (Du Pont 1953a). For tritium analysis in the 1950s, the tricarb scintillation method was described and the calculations for their spike method and method for converting

counts per 30 minutes of an unspiked sample to microcuries per liter of tritium were laid out (Du Pont 1959a).

As 1960 approached, there were continuing efforts to review and modify procedures, especially for sampling and analysis of onsite samples (Johnson 1960). To inventory radioactive waste released to effluent streams, technical standards were requested to separately address methods for the release for short-lived versus long-lived radioactive materials (isotopes with half-lives less than or greater than 15 days, respectively). For beta activity analysis, a 100-mL sample was obtained from each flowing weir from the disassembly basin effluent once per shift. The samples were mixed in proportion to the flow from each weir. For alpha activity measurements, one additional sample was submitted to the laboratory "on each Wednesday 12–4 shift" (Johnson 1960). A method and step-by-step worksheet guided the analyst to estimate the ^{90}Sr content of the total release.

Routine tritium monitoring of the river began as early as 1958, and as early as 1960, samples from key locations were routinely analyzed for radioiodine, radiostrontium, and radiocesium. By late 1960, ion exchange media were used to concentrate radiostrontium and radiocesium in water samples, resulting in a roughly four-fold decrease in the detection limits from approximately 4 to 0.8 pCi L^{-1} (Du Pont 1960a). Analysis for ^{90}Sr in the river samples began in September 1959 (Van Wyck 1960). Routine analysis of river samples for specific radionuclides by gamma spectroscopy did not begin until 1963. Some of the monthly and semiannual reports for the early years of operation do provide data from decay studies that were periodically performed on river water samples. Radioactivity release limits in 1960 specified "no limit" for tritium released to plant effluent streams, seepage basins, or the disposal pits (Johnson 1960). By 1964, the annual operating release guide was 100,000 Ci of tritium to the streams (Ashley 1965).

As 1960 approached, there was a general expansion of the quantitative analytical techniques for a variety of environmental samples. With the automation of gamma spectrometry, isotopic analysis was possible for routine sampling of effluent streams, river water, reactor area effluent, biological specimens, and other samples not previously analyzed quantitatively (Johnson 1960). The Site made progress on increasing the sensitivity of the tritium analysis, especially for large volume samples. Working with personnel at the University of California and at the University of Chicago, the analytical laboratory made a number of recommendations to improve the sensitivity for tritium analysis (Butler 1960). A solvent was developed and modified for use in the liquid scintillation process that permitted use of relatively large samples without serious loss of efficiency (Albenesius and Meyer 1962).

By September 1962, the analytical procedure used for tritium analysis was fairly well established at SRS. Low energy of the tritium beta, an average of 6 keV, required internal counting systems. At this time, the liquid scintillation provided a rapid and sensitive technique to determine tritium concentration in streams and river water samples. (In liquid scintillation counting, the tritiated water is dissolved in a solvent that contains an organic scintillator. Particles of radioactive decay [beta] excite the organic molecules, which then emit light pulses. Light pulses detected by a photomultiplier tube, amplified and electronically counted.)

For the basic tritium analytical procedure, the tri-carb liquid scintillation counter was placed in operation by June 1958 (Du Pont 1958). The optimum conditions for counting with the organic scintillator solution were described. The efficiency was reported as 5.3% for a 3-mL water sample dissolved in the scintillator solution. The background was approximately 50 cpm using "Wheaton Co., low K-40 content, vials." The tritium analytical procedure, described in 1953 and 1959 (Du

Pont 1953a, 1959a) was modified for more sensitive analysis of water samples (Butler 1960). Two methods were compared for the accurate determination of tritium in urine and water: liquid scintillation counting and vibrating reed electrometer rate-of-drift determination (Butler 1961).

A new type of water sampler was used beginning in the spring of 1964. This sampler used an ion column to collect radioactive materials from a large volume of water and was designed to operate continuously without electrical power. A water wheel drove two piston pumps. One of these pumps passed water through a prefilter and the ion column. The other pump, which was geared much slower, delivered water directly into a 2-gal container. Because each pump had a fixed gear ratio, the volume of water in the container could be used to determine the volume of water that had passed through the ion column. For the first few months of operation (April through October 1964), the volumes of water sampled ranged from 40 to 96 L depending on the river flow rate. A comparison of the results obtained at the downstream Savannah River sampling location during this period, using both the ion exchange sampler and the standard paddle wheel sampler, found that the average concentrations for all radionuclides, except ^{131}I, were consistently higher for the ion exchange sampler. This difference was attributed to possible settling of radionuclides associated with suspended solids within the container of the paddle wheel sampler over the sampling period. Because the volume of water sampled with the ion exchange sampler was two to four times greater than the volume collected by the paddle wheel sampler, the number of positive analyses was also consistently higher for this new sampling technique (Du Pont 1964).

It is not clear from the available information how long SRS used the ion-column equipped samplers that were originally tested in 1964. For the first few months of 1965, aperture card data are reported as prefilter, ion column, and total, indicating that these samplers were put into routine use. However, by the early 1970s when sampling methods were first described in the annual, offsite monitoring reports, ion-exchange results were no longer used as part of the sampling collection process. The 1972 annual monitoring report describes continuous sampling of Savannah River water as follows:

> . . . a sampler consisting of a ‘Plexiglass’ water wheel suspended on two pontoons. As the water wheel is turned by flowing water, a small cup (or cups) on one paddle picks up a sample of water and deposits it into a trough. The sampled water flows by gravity from the trough through connecting tubing into a large polyethylene jug which trails the sampler. The sampled water (up to six gallons) is collected weekly at river locations above and below SRP. Increased analytical sensitivity for water samples (containing insufficient radioactivity for direct processing) is achieved through concentration of radionuclides by ion exchange. The ion exchange column is counted directly for gamma emitting radionuclides (Du Pont 1973).

The sampling equipment used by the SRS does not appear to have been modified from that time up through the early 1990s when the ISCO samplers were installed.

By the early 1970s, the procedures for water collection and counting were standard for the time. Tritium was measured in distilled water samples with a liquid scintillation spectrometer. Alpha and beta-emitting radionuclides were measured by passing 25 L of water through a cation-anion resin column and direct counting the column using a NaI(Tl) well detector with a 400-channel gamma spectrometer. The resin column was then eluted with nitric acid for subsequent strontium analysis. Strontium-90 was recovered from a potion of the sample by liquid

ion exchange. Equilibrium of ^{90}Y was allowed over a 15-day period and then the short-lived ^{90}Y daughter was counted in a low-level gas flow proportional counter (Du Pont 1975).

The analytical procedure for the determination of ^{35}S was adapted for use with stream and river water and used routinely, as reported in biweekly Environmental Monitoring and Allied Studies (Du Pont 1959c). The procedure was extended in early 1962 to determine ^{32}P levels in reactor moderator and disassembly basin weir water. The wastewater sample was passed through a cation column to remove ^{239}Np, then a step removed iodine. Following a second cation column treatment, an ammonia phosphate precipitation was done. This precipitate contained the ^{32}P, which was then plancheted and counted.

By 1966, a technique based on liquid ion exchange was developed to determine ^{35}S in water because the previous method was lengthy and produced low, variable recoveries. Overall, the recovery of ^{35}S in this procedure was 85%, and the method was routinely used during this time (Du Pont 1966).

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APPENDIX B

ANALYTICAL PROCEDURES FOR CHEMICALS

APPENDIX B

ANALYTICAL PROCEDURES FOR CHEMICALS

ABSTRACT

This appendix summarizes the various methods that were used by the Savannah River Site (SRS) laboratories to analyze samples for nonradiological or chemical contaminant concentrations. Methods of analysis used by organizations other than the SRS are not provided.

MERCURY

A variety of units was used for reporting mercury concentrations in environmental samples. In general, sediment and soil concentrations were reported as parts per million (ppm), parts per billion (ppb), or μg Mercury g^{-1} soil dry weight. Water concentrations were reported as ppm, ppb, or μg Mercury l^{-1} water. Fish concentrations were reported as ppm, ppb, or μg Mercury g^{-1} flesh wet weight. A May 1971 report (Du Pont 1971a) described a procedure for analysis of mercury in environmental samples by the SRS. Water samples were buffered at pH 6.0 ± 0.5, 200 mg of mercury-free cadmium sulfide were added, and the slurry was mixed for 15 minutes. The sulfide and sorbed mercury were filtered on an asbestos pad, which was washed with methyl alcohol to remove organic material and dried overnight at 100°C. The mercury was then distilled at 550°C, and vapor was collected in an absorption cell for atomic absorption spectrophotometry. The lower limit of detection (LLD) in water samples was 0.5 ppb, and analytical precision was about 10 to 15% for 1 to 5 ppb.

A June 24, 1971 letter from W.P. Bebbington to N. Stetson (Bebbington 1971) indicated the LLD in water samples analyzed in 1970 to be 2 ppb for the routine method, which was not described. However, a special and tedious method not practical for routine monitoring, which also was not described, appeared to have a LLD of 0.1 ppb, based on tabulated data.

A new method for mercury analysis of environmental samples (flameless atomic absorption spectrophotometry) was described in a Savannah River Laboratory September 1971 report (Du Pont 1971c). Sediment samples were prepared by oxidation with $KMnO_4$ followed by leaching with concentrated HCl or H_2SO_4. The mercury was then reduced to elemental Mercury with $SnCl_2$. Air was bubbled through the solution, which carried Mercury vapor through the absorption cell of a Perkin-Elmer Model 403 atomic absorption spectrophotometer. Quantitative assessment of mercury concentrations were then made based on the measured absorption of the 2537 Å line. Tracer studies showed that $H_2SO_4/KMnO_4$ digestion recovers 96% of the mercury. The LLD for sediment samples was not provided, but a 1974 memorandum (Horton 1974) indicated a LLD of 2 ppb for the same method of analysis. This was considered sufficient since background levels in soil and streambed sediments vary from 20 to 50 ppb. A LLD of 30 ppb was reported for a study conducted on stream and river sediment in 1973 (Gladden et al. 1985).

Water samples were analyzed following overnight digestion with H_2SO_4 and $KMnO_4$ at room temperature. Fish samples were dissolved in concentrated H_2SO_4 at 55° C, and organic material was oxidized by $KMnO_4$ at room temperature. Mercury concentrations in the resulting solution were determined as described for sediment. The LLD for water samples was reported as 0.05 ppb with an analytical precision of about 10% at 0.1 ppb. The LLD for fish samples was not

given but analytical precision was reported as 12% for 500 ppb. An August 1971 document (Du Pont 1971b) reported a LLD of 10 ppb using flameless atomic absorption spectrophotometry with a Coleman mercury analyzer system. Mercury measurements for all environmental media appear to have been made using standard flameless atomic absorption spectrophotometry as described by the Environmental Protection Agency (EPA) (EPA 1983) since 1971.

CHROMIUM

Methods of analysis for chromium and other metals were not explicitly described in the environmental monitoring reports that have been reviewed. The annual report for 1983 (Ashley et al. 1984) indicated that a large part of the nonradioactive analyses was performed by an offsite vendor (Envirodyne Engineers, Inc. was the vendor in 1983). Standard EPA methods of analysis (EPA 1983), such as atomic absorption spectroscopy for chromium, were likely used for these analyses.

OTHER CHEMICALS

Other chemical analyses were carried out using standard EPA methods of analysis, many of which are described in EPA (1983).

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APPENDIX C1

TRADE NAME MATERIAL COMPONENTS LISTINGS AND TOTALS

APPENDIX TO CHAPTER 16

APPENDIX C1

TRADE NAME MATERIAL COMPONENTS LISTINGS AND TOTALS

Information on chemicals used, stored and disposed of at the SRS was compiled for Chapter 16. Trade name materials (commercial products) accounted for a significant amount of the inventory for some materials. The information in the Chemical Information and Inventory System (CIIS) database suggested that 5.2% of the trichloroethane, all of the dimethyl phalate, 4.2% of the nickel, 6.8% of the ethylene oxide, 74.6% of the trichloroethylene, and 19% of the phosphoric acid onsite was contained in trade name compounds.

Numerous entries for ethyl alcohol (ethanol, solvent alcohol, dehydrating alcohol, etc.) totaling 6930 kg were found in the CIIS database. Ingestion of alcohol is believed to increase the risk for various human cancers and alcohol is considered to be a reproductive and developmental hazard. Alcohol can be inhaled as a result of its use as a cleaner, solvent, and disinfectant. Methanol is more toxic than ethanol and was not combined under the entry alcohol.

References to chrysotile, crocidolite, anthophyllite, cristobalite, filterbestos, asbestos siding, and asbestos tile were combined under asbestos.

On the basis of the toxicity values for the most toxic components and inventory amounts listed, ABA Plus bacteriacide and biocide, Garlon 3A Herbicide, Biosperse 261T Microbiocide, and Kathon FB 1.5 biocide and algaecide were deleted from the list of chemicals to be ranked. These compounds are severe irritants but ranking ratios, determined using acceptable exposure limits proposed in the material safety data sheets or threshold limit values for the most toxic components, would be well below 0.001.

Table C1-1 summarizes the trade materials accounted for in the summed inventory totals used for chemicals. The chemical component and the percentage is given if known.

Table C1-1. Summary of Trade Materials

Material	Component and estimated percentage if known	CIIS inventory amount (kg)
ALCOHOL - ETHANOL		
Copalite		408.6
Reducer No 54	23% ethanol	1.8
Tuff-bond	?% ethanol	0.5
Gram safranin	20% ethanol	4.6
Safranin solution	9% ethanol	0.9
Nycote 7-11	50% ethanol	0.4
Dodecanol, dodecyl alcohol		5.5
ISOPROPANOL		
Isopropanol		7,480.1
IPA anhydrous		90.7

Table C1-1. (Continued)

Material	Component and estimated percentage if known	CIIS inventory amount (kg)
ASBESTOS		
Kaocrete HS	15% cristobalite	498.9
Dicalite 21	55–15% cristobalite	43.5
Hyflo Super Gel	<60% cristobalite	9.9
Celite	20% cristobalite	1,533.1
CL7824	?% chrysotile	0.9
CHLOROFLUORCARBONS (CFC, FREONS)		
Forane 11	Trichlorofluoromethane	340.2
E Series UltraJet	Chlorodifluormethane	326.6
R 12	Dichlorofluromethane	0
R 22	Chlorodifluromethane	0
CDC 3-36 Aerosol		425.9
Forane 113		453.6
CHROMIUM		
Colloidion	98% chromium trioxide	1.4
Chronamig 333M	3–60%	45.3
Knight/Laggi System	Zinc chromate	238.1
Smootharc 307R 309 MC-150	1–10%	77.1
Potassium zinc chromate		14.9
Sodium chromate		17.3
Ammonium chromate		0
Ammonium dichromate		1.36
Potassium chromate		17.7
Sodium dichromate		442.7
DIOXANE		
Remac 71	?%	97.9
ETHYLENE OXIDE		
Rhodameen VP-532	<01 %	29.9
Iguafen VP-532 SPB	<0.1%	Listed under rhodameen
Katapol VP 532 SPB	<0.1%	Listed under rhodameen

Table C1-1. (Continued)

Material	Component and estimated percentage if known	CIIS inventory amount (kg)
METHYLENE CHLORIDE		
SCI-125	Up to 100%	4.08
Virginia No 10 degreasing solvent	10–15%	14.51
TFE Dry Spray	34%	0.4
Strip Solve	98%	0.9
PHOSPHORIC ACID		
Naval jelly rust dissolver	25–30%	5,062.1
TOLUENE/XYLENE		
Byco 300 Hb catalyst		299.3
Bycothane 900		1,428.8
CP 30 Chilperm	Xylene 50%	0
HYDROCARBONS/PETROLEUM DISTILLATES		
Bioact AE-P	Terpene hydrocarbons	381.0
Novoid A	Asphalt	34.0
MP-429	Hydrocarbons > 6%	0
Ardox 906/p303A	Petroleum, naptha	117.9
Blue ribbon prime neatsfoot	Petroleum distillate	54.9
Diesel fuel antigel		23.6
Diesel tone fuel conditioner		16.3
NICKEL		
Active raney catalyst	88–92%	1.8
Savin developer	Nickel oxide <99%	278.0
MetCo		68.0
Green Nickolous	98% nickel oxide	4.5
FEL-PRO N-5000	Nickel flake 16%	0.7
Raney 200, active raney catalyst	88–92% nickel	0.4
Raney 2924, active raney catalyst	88–92% nickel	0.4
Chronamig 333M	1–15%	45.3
Smootharc 307R 309 MC 150	1–3%	77.1
INCO Srounds Electrolytic	99% nickel metal	3,749.0
Nickelous oxide green		5.0

Table C1-1. (Continued)

Material	Component and estimated percentage if known	CIIS inventory amount (kg)
STYRENE		
Muffle lag	14%	163.2
Ceilline Saurant Part A	5–15%	35.3
T-431 solvent		64.4
P380 primer liquid	54%	4,440.0
Hetron 435P	33% styrene	347.0
Muffle Lag Part 1	14%	163.2
Cielcrete 695	36%	34.4
TRIBUTYL PHOSPHATE		
Opti-fluor		2,363.6
TETRACHLOROETHYLENE		
Virginia No 10 degreasing solvent	10%	14.5
Tremply solvent		16.1
TRICHLOROETHANE		
BCR 10288-22	92–94%	6.8
SP-400 solvent	30%	904.9
Tremply solvent		16.1
TRICHLOROETHYLENE		
Aerothane ST solvent typethinner	<10%	297.6
Deglazing solvent	96%	180.9
Vucanizing fluid 201-207	50%	8.6
ZINC		
Metalhydride primer		0
HG absorber	94–96%	49.8
Texamatic 9226 tranmission fluid	Up to 11% zinc compounds	571.5
Knight/Laggi System	Zinc chromate	238.1
Potassium zinc chromate		14.9
Metalhide 1001 green zinc		34.0
DIMETHYLPHTHLATE		
NoRox MEKP-9	52%	408.2

Table C1-1. (Continued)

Material	Component and estimated percentage if known	CIIS inventory amount (kg)
PESTICIDES		
ABA Plus	Biocide bacteriacide	512.4
Pronone 10G granular herbicide	61% ammonium hydroxide	136.1
Garlon 3A herbicide	Petrolleum distillates	1,088.1
Amdro fire ant poison	1% hydramethylnon	56.7
Accord	41% glycophosphate isopropylamine	90.72
Biospherse 261T microbiocide	Bromochlorodimethylimidazoidinedione. Algacide/biocide, no ingredients listed	333.7
Kathon FB 1.5 biocide/algacide	Dipropylene glycol	1,587.6
Garlon 4 herbicide		90.7

Although silica was not considered a hazardous environmental pollutant and was not included in the ranking, trade name materials containing silica are listed below in Table C1-2.

Table C1-2. Summary of Trade Name Materials Containing Silica

Material	Component and estimated percentage if known
QUARTZ/SILICA	
IMSILA 108	Silicon dioxide
HIT-C100 dowelling	Quartz
Frit 202	Glass frit
Ionsiv zeolite cation exchange	Silicon oxide
L&M Cure	Sodium silicate
CP 10 Vi Cryl	Quartz 18–25%
Kaocrete HS	Quartz 15%
CP11-Vi Cryl	18–23% quartz
Dicalite 215	2–9%
Bento Seal	1% quartz
Type S-15 aggregates	90% quartz
Waterplug	25–30%

Table C1-2. (Continued)

Material	Component and estimated percentage if known
Hyflo Super Gel	
Celite	3% quartz
Carboline Surfacer	70% quartz
Bisco SE Form Part A	17% quartz
Bisco Se Form B	16% quartz
S1 Powder	90% quartz
CP-10 Vi-Cryl	18–23%
Sikadur 31 hi od gel	17% silica flour
Imasil A 108	Silicon dioxide
Carboline	65% quartz
Nibo Chemtro Heavy Duty Gray	Cement
Smooth on MT 13	Quartz
Amorphous silica	
Crystal lime silica	
Portland cement	
Talc	
Tricalcium silicate	
Polydimethylsiloxane	

APPENDIX C2

USAGE AND HAZARD RANKING INFORMATION FOR CHEMICALS

APPENDIX TO CHAPTER 16

APPENDIX C2

USAGE AND HAZARD RANKING INFORMATION FOR CHEMICALS

The following information about the hazard associated with many of the chemicals considered in Chapter 16 was obtained from Sax, N.I. 1975. *Dangerous Properties of Industrial Materials, 4th ed*. New York, New York: Van Nostrand Reinhold Company; Sax, N.I. and R.J. Lewis. 1992. *Dangerous Properties of Industrial Materials.* New York, New York: Van Nostrand Reinhold; or from ATSDR's (Agency for Toxic Substances and Disease Registry's). 1992–1997. Toxicological Profiles.

The material name and chemical abstract services (CAS) number is given in the first column. The toxicity ranking in the middle column is relative and is defined in *Dangerous Properties of Industrial Materials* as:

0 No toxicity, materials that cause no harm under any conditions of normal use, or materials that produce toxic effects in huamsn only under the most unusal conditions or by overwhelming dosage.

1 Slight Toxicity, in general, these are materials that produce changes in the human body which are relatively reversible and that will dissappear following termination of exposure, either with or without medical treatment.

2 Moderate Toxicity, materials that may produce irreversible as well as reversible changes in the human body. The changes are not of such severity as to threaten life or produce serious physical impairment.

3 Severe Toxicity, materials that can cause injury of sufficent severity to threaten life.

The information in the third column addresses the common uses of the chemical and other information which may be of interest for dose reconstruction. Mention is also made of materials listed for the EPA Toxic Substances Control Act (TSCA) and compounds that may cause reproductive effects or that may have mutagenic or carcinogenic activity. Many of the chemicals used at the SRS and in industry world-wide have not been subjected to extensive toxicity testing and a hazard ranking could not be provided for all of the chemicals listed.

Usage and Hazard Ranking Information for Chemicals

Material (CAS number)	Toxicity ranking	Uses and Other Information
Acetone (64-1)	3	Paint, paint remover and thinner, used to clean precision parts of equipment, airplane glue solvent
Acetonitrile (75-05-8)	3 EPA TSCA, suspected mutagen	Pesticides, solvent, floor polish, water proofing, antistatic, detergent, water softeners brighteners for metals

Acrolein (107-02-8)	3 Possible carcinogen, experimental reproductive effects	Making plastics, artificial resins, and synthetic fibers, herbicide to control aquatic weeds in drainage and irrigation channels, fungicide for slime, tear gas
Acrylonitrile (107-13-1)	3 Confirmed human carcinogen, mutagen	Pesticide, to make synthetic fibers, plastics, surface coatings, adhesives, cotton pesticide, grain fumigant. Breaks down quickly in air, 1–2 weeks In water, metabolites include cyanide
Aldrin (309-00-2)	3 Suspected carcinogen, suspected mutagen, suspected reproductive hazard	Insecticide, no longer used, used in the 1950s–1970s. on crops such as corn and cotton. Its use was stopped in 1970 but it was approved for use on termites from 1972–1987. Aldrin readily changes into dieldrin once it enters the environment or a person. Dieldrin sticks to the soil strongly and may remain in soil, unchanged for many years. Not very water soluble. Attaches to sediments, bioconcentrates in fish. Causes liver tumors in rodents. There are a number of negative or inconclusive epidemiological studies.
Aluminum (III) nitrate (13473-90-0)	3 Also called aluminum slat, Norway saltpeter	Making fertilizers explosives, insecticides, and other chemicals; tanning leather, textiles, anti-perspirants, anti-corrosives, rocket propellant
Ammonia (7664-41-7)	3 Suspected mutagen, corrosive	Making fertilizers and other chemicals; in synthetic fibers, dyestuffs, explosives, cosmetics, hair dyes; electroplating, blueprint and film developing, processing leather, pulp and paper, textiles, refrigerant, flame retardant, cleaning solutions. Rapidly degraded.
Ammonia hydroxide (1336-21-6)	3 Also called ammonia water. EPA TSCA suspected mutagen	Textiles, making of rayon, rubber, fertilizers, metallic hair dyes, inks, refrigeration, photography, soaps, lubricants, protective skin creams, fire proofing of wood, explosives, ceramics, detergents, food additives, household cleaners
Aniline (62-53-3)	3 Also called phenylamine EPA TSCA, EPA Ext Haz Sub carcinogen, mutagen, a NIOSH study links to cancer	Making dyes, hair dyes, colored pencils, crayons, lithographic and other printing inks, perfumes, pharmaceuticals, nylon fibers, resins, varnishes, industrial solvent, rubber processing, photographic chemical, fungicide, artificial sweetener, and corrosion agent
Anthracene (120-12-7)	3 Suspected carcinogen, mutagen, allergen	Dyes, wood preservative, semi-conductor research, smoke screens

Antimony trioxide (1327-33-9)	Also called antimony oxide, carcinogen	Flame proofing textiles, paper, plastics, paint pigment, staining iron and copper, glass decolorizer
Arsenic (7440-38-2)	3 Confirmed human carcinogen, mutagen, suspected reproductive hazard	Making alloys (especially lead and copper), semiconductors, germanium and silicon, solid state products, types of glass, harden alloys, radioactive tracer for spirochetal infections, blood disorders and skin disease, pesticides, insecticide, rodenticide, solders
Asbestos (1332-21-4)	3 Carcinogen	Fireproofing, brake linings, roofing, electrical and heat resistant insulation, paint filler, chemical filters, reinforcing agent in plastic, and rubber
Benzene (71-43-2)	3 Also called carbon oil, coal naptha, mineral naptha; confirmed human carcinogen, mutagen, possible reproductive hazard	Perfumes, paints and coatings, processing nylon and photographic materials, making gasoline, styrene, pesticides, plastics, resins, synthetic rubber, aviation fuel, dyes, explosives and other chemical using phenols
Benzidine (92-87-5)	3 Confirmed human carcinogen, mutagen	Making dyes, rubber, detection of bloodstains, stain in microscopy, laboratory chemical
Benz (a) anthracene (56-55-3)	3 Confirmed carcinogen; it is isolated from products such as soot, coal, tar, tobacco smoke, petroleum and cutting oils	
Benzo (a) pyrenene (50-32-8)	3 Confirmed human carcinogen, possible reproductive hazard. common air contaminant found in food, water, and smoke; by-product of burning various carbon products (e.g., tar, soot, carbon black, coal, and coke)	Laboratory chemical
Benzo perylene	Questionable carcinogen	
Benzo fluoranthene (207-08-9)	3 Confirmed carcinogen	
3,4 Benzofluoranthene (205-99-2)	Confirmed carcinogen	

Beryllium (7440-41-7)	3 Carcinogen	Making electrical components, ceramics and chemicals, in computer parts, solid propellant and rocket fuel
Bis (2-chloroethoxy) methane (542-88-1)	3 Confirmed human carcinogen	laboratory chemical; used to make ion exchange resins
Bromacil (314-40-9)	Very low toxicity, 8 mg/kg mild AchE inhibition, carcinogenic report in USSR literature; possible reproductive hazard	Herbicide, weed killer
Bromine (7726-95-6)	3 Poison, corrosive	Making other chemicals, anti-knock gas, bleach, water purification, solvent, fumigant, fire retardant for plastics, dyes, pharmaceuticals, photography
1-bromo-3-chloro-5,5,-dimethylhydantoin	Relatively low toxicity, biodegradable	Bromocide® a disinfectant used at wastewater treatment plants at the SRS
Bromoform (75-25-2)	3 Questionable carcinogen, narcotic	Solvent for fats, waxes and oils, making other chemicals and drug
Bromophenyl phenyl ether (1985-12-2)	3	
Butyl benzyl phthlate (85-68-7)	3 Possible carcinogen	
Cadmium compounds	3 Confirmed human carcinogen, mutagen, reproductive hazard	Electroplating, making batteries, plastics, TV tubes, fumigant, lithography, ceramic glazes, nuclear reactor rods, electronic devices, photo-electric cells
Calcium fluoride	2 Also called fluorite, fluorspar	Steelmaking, smelting, arc welding, glass and ceramics, fluoridation of drinking water
Carbon tetrachloride (56-23-5)	3 Confirmed carcinogen, mutagen	Spot remover, refrigerant, making fire extinguishers, metal degreaser, aerosols, pesticide, agricultural fumigant, production of semi-conductors, solvent
Chlordane (57-74-9)	3 Carcinogen, mutagen	Insecticide, fumigant
Chlorine gas (7782-50-5)	3 Mutagen	Chlorine bleach cleaners, making solvents and other chemicals, water purification, disinfectant, flame retardant, processing food, lithium and zinc batteries
Chlorobenzene (108-90-7)	Mutagen, poisonous, causes narcosis	Making other chemicals, dyes, pesticides, rubber dyes, solvent, heat transfer

Chlorodibromomethane (124-48-1)	2	
Chloroethane (38915-59-2)	3 Possible carcinogen	
Chloroform (67-66-3)	3 Confirmed carcinogen, mutagen, reproductive hazard	Making other chemicals including pesticides, drugs, and dyes
Chloronapthalene 1, chloronapthalene (90-13-1) 2, chloronapthalene (91-58-7)	2	
Chlorophenol	Questionable carcinogen	
2, chlorophenol (95-57-8)		Making dyes
3, chlorophenol (108-43-0)		Making other chemicals
4, chlorophenol (106-48-9)		Making other chemicals, topical antiseptic, corrosive
Chromium compounds chromium (744-47-3)	3 Chromium salts are human carcinogens,	In plating, metals and plastics, increasing corrosion resistance, nuclear and high temperature research, inorganic pigment
Chrysene (218-01-9)	3 Confirmed carcinogen	Never found alone. Found in coal tar, found in distillation of coal, tar, asphalt, coal tar pitch.
Coal fly ash (68131-74-8)		By-product of burning coal
Coal tar (80007-45-2)	3 Confirmed human carcinogen	Raw material for plastics, solvents, dyes, drugs; the crude or refined products or fractions are used for waterproofing, paints, pipe coating, roads, roofing, insulation and as pesticides and sealants; direct burning as fuel; component of products for treatment of psoriasis; alcohol denaturant; binder and filler in surface coatings; modifier in epoxy resin surface coating
Coal tar pitch volatiles (65996-93-2)	3	

Copper compounds (7440-50-8)	3 Questionable carcinogen, possible reproductive hazard	Making brass; in industry and construction
Cristobalite (crystalline silica) (14464-46-1)	3 Carcinogen	Making fiber glass, ceramics, foundry molds, iron and steel castings
Cyanide (57-12-5)	3 Very poisonous	Found in rat poison and pest poison, silver and metal polishes, photographic solutions, fumigating products
DDD or 1,1-bis(4-chlorophenyl) -2,2-dichloroethane (72-54-8)	3 Carcinogen	Insecticide
DDE or 2,2-Bis (p-chlorophenyl) -1,1-dichloro-ethylene (72-55-9)	3 Suspected carcinogen, mutagen	Insecticide
DDT or 1,1,1 tri-chloro-2,2-di (4-chorophenyl)-ethane	Carcinogen, mutagen, reproductive hazard	Insecticide
Diazinon (333-4-5)	3 EPA TSCA, EPA Gen Tox poisonous	Insecticide
Di (2 ethylhexyl) phthalate or di-sec-octyl phthalate (1117-81-7)	3 Carcinogen, mutagen, possible reproductive hazard	Plasticizer
Dibenzanthracene or dibenz (a,c) anthracene 1,2:3-4 - dibenzanthracene 1,2:3-4 - dibenzoanthracene benzo(b) triphenylene (215-58-7)	3 Suspect carcinogen, mutagen	
Dibutyl phthlate (84-74-2)	3 Mutagen, reproductive hazard	Insect repellent, lacquer solvent, used to make flexible plastics

p-dichlorobenzene (106-46-7)	3 Carcinogen, mutagen, possible reproductive hazards	Fumigant, to control mold and mildew, also as a deodorant
3,3 dicholorbenzidine (91-94-1)	3 Carcinogen, mutagen	
Dichlorobromomethane or bromodichloromethane (75-27-4)	3 Suspected carcinogen	
1,1 dichloroethane (75-34-3)	3 Carcinogen	Solvent, fumigant
1,2 dichloroethylene (540-59-0)		Solvent for organic materials, dye extraction, perfumes, lacquers, thermoplastics, making other chemicals
dichloromethyl ether (4885-02-3)	3 Confirmed human carcinogen, mutagen	Lab chemical for making ion exchange columns
Dichlorophenol		
2,4 dichlorophenol (120-83-2)	3 Suspected carcinogen, poisonous	
2,6 dichlorophenol (87-65-0)	3 Poisonous depending on exposure route	
Dieldrin (60-57-1)	3 Probable carcinogen, reproductive hazard	Insecticide, wood preservative
Di (2-ethylhexyl) phthalate (117-81-7)	3 Carcinogen, mutagen, possible reproductive effects	Plasticizer
Diethyl phthlate (84-66-2)	3 Possible reproductive effects, poisonous	
Dimethylformamide (68-12-2)	3 Suspected carcinogen, mutagen, possible reproductive hazard	Solvent, making other chemicals
Dimethyl aniline or xylidine (1300-73-8)	3 EPA TSCA suspected human carcinogen	Making dyes, pharmaceuticals, and other chemicals

Dinitrotoluene (25321-14-6)	3 Confirmed carcinogen, mutagen	2,4 dinitrotoluene 121-44-2 is used for making plastics, dyes, other chemicals and explosives
Dioxane (123-91-1)	3 Carcinogen	Solvent for many products, lacquers and paints, varnishes, paint and varnish remover, in textile processing ,dye baths, stain and printing, cleaning and detergent preparations, cements, cosmetics, deodorants, fumigants, polishing compositions
Diphenylhydrazine (122-66-7)	3 diphenylhydrazine 1,2 – diphenylhydrazine carcinogen	
Ethylbenzene (100-41-4)	3 possible reproductive hazard	Making styrene and synthetic polymers, solvents, in automotive and aviation fuels
Endosulfan (115-29-7)	3	
Ethlyene dichloride (107-06-2)	Confirmed carcinogen, reproductive hazard, narcotic	Making vinyl chloride, in gasoline, paint varnish and finish removers, metal degreasing, soaps, to make other chemicals
Endrin (72-20-8)	3 also called Hexadrin, Medrin mutagen, possible reproductive hazard	Insecticide, rodenticide
Ethoxyethanol (110-80-5)	2 Mutagen	Widely used as a solvent, in brake fluid and aviation /automotive fuels
Ethyl benzene (100-41-4)	3 Moderately toxic	Making styrene and synthetic polymers, solvent, automotive and aviation fuels
Ferrous sulfamate	Lethal dose is probably related to the iron content	
Fluoranthene (206-44-0)	3 Carcinogen	
Fluoride (16984-48-8)	3	Widely used in chemical, steel and aluminum industry, toothpaste, other dentifrices
Formaldehyde (50-00-0)	3 Confirmed carcinogen	Germicide, embalming fluid, it is in home insulation and wood products, resins, making other chemicals, fertilizer, dyes, preservatives, treatment of textiles, treatment of grain
Gadolinium (III) nitrate (1:3) (10168-81-7)	Questionable carcinogen	

Gasoline (8006-61-9)	3 Mutagen	Solvent, fuel for internal combustion engines
Heptachlor (76-44-8)	Suspected carcinogen, mutagen	Pesticide (especially termites)
Heptachlor epoxide or epoxyheptachlor (1024-57-3)	3 Carcinogen, mutagen, suspected reproductive hazard	Pesticide
Hexachlorobenzene (118-74-1)	3 Carcinogen, mutagen, suspected reproductive hazard	Fungicide for treating seeds, wood preservative, making other chemicals
Hexachlorobutadiene (87-68-3)	3 Suspected carcinogen, reproductive effects	Solvent, heat transfer fluid, transformer hydraulic fluid and wash liquor, used in making other chemicals
Hexachlorobutadiene (lindane) benzene hexachloride (608-73-1)	3 Carcinogen, mutagen	Pesticide
Hexachloroethane (67-72-1)	3 Suspected carcinogen, mutagen	Insecticide, animal medicines, solvent, explosives, camphor substitute in smoke making devices
Hydrochloric acid (7647-01-0)	3 Mutagen, corrosive	Used for making many other chemicals, to adjust pH in swimming pools, for metal processing, food processing, general cleaning, common laboratory chemical
Hydrofluoric acid (7664-39-3)	3 Mutagen corrosive	Making other chemicals, fluorocarbons, etching glass
Hydrofluosilicic acid (2589-15-3)	2	
Hydrogen sulfide (7783-06-4)	3	To test and make other chemicals, by-product of sewage treatment and other chemical reactions
Hydroxylamine nitrate hydroxylamine (7803-49-8)	3	

Hydroxylamine sulfate or oxammonium sulfate (10034-54-0)	3 Mutagen, corrosive	
8, hydroxyquinoline or 8, quinolinol	3	
Indeno pyrene (193-39-5)	3 Mutagen	
Isophorone (78-59-1)	3 Possible carcinogen, mutagen	Solvent for resins, making pesticides and lacquers
Kerosene (8008-20-6)	3 Suspected carcinogen, mutagen	Fuel, domestic heating, solvent, insecticide spray
Lead (7439-92-1)	3 Suspected carcinogen, mutagen	Resistant to corrosion, used in pigments and paints, solder, fusible alloys, storage batteries, gasoline additive, radiation shielding, cable covering, ammunition foil, bearing alloys
Lithium compounds	D Toxicity of lithium is a function of solubility in water, except strong bases such as lithium oxide; most hazardous is lithium hydride	
Manganous nitrate o (10377-66-9)	Mutagen	As a color agent in porcelain and ceramics, to make other chemicals
Mercury (7439-97-6)	3 Poison, corrosive, experimental reproductive effects	In dental fillings; making other chemicals; electrical equipment and instruments; mercury vapor lamps; mirror coating; as a coolant and neutron absorber in nuclear power plant
Methoxychlor (72-43-5)	3 Suspected carcinogen	Insecticide
Methyl bromide (74-83-9)	3 Suspected carcinogen, corrosive	Pesticide (use has been suspended), fumigant gas
Methyl chloride (74-87-3)	3 Chloromethane suspected carcinogen, mutagen, fire hazard	Making other chemicals, refrigerant, solvent, herbicide, topical anesthetic
Methyl ethyl ketone (MEK) (78-93-3)	3	Powder, cleaning fluids, printing, to produce “drugs of abuse,” solvent, paint remover, cements and adhesives, making other chemicals, plastics, smokeless

Methyl isobutyl ketone (108-10-1)	3	Solvent
Methylene chloride (75-09-2)	3 Carcinogen,	Paint remover (stripping) in foods, furniture and plastics processing
Naptha (8030-30-6)	3 (and petroleum distillates) also called Benzin, coal tar naptha, varsol	Solvent, paint thinner, making other chemicals, in dynamite
Napthalene (91-20-3)	3 Also called camphor tar, white tar Possible carcinogen	Moth repellent, making other chemicals, dyes, plastics, resins, explosives and lubricants; fungicide; smokeless powder; cutting fluid; lubricant; synthetic tanning; preservative; antiseptic
Nickel compounds	3 Carcinogen, mutagen, human carcinogen if inhaled	Making metal alloys, cars, stainless steel, magnets; electroplating; alkaline storage batteries
Nitric acid (7697-37-2)	3 Corrosive	Making chemicals, dyes, fertilizers, drugs and explosives; used in metallurgy, photo-engraving, urethanes, rubber chemicals, and reprocessing nuclear fuel
Nitrilotriacetic acid or aminotriacetic acid (139-13-9)	3 Possible carcinogen	
Nitrobenzene (98-95-3)	3 Also called oil of mirbane poison, reproductive effects	Making shoe polish, dyes, explosives, floor and metal polish, other chemicals and paints
Nitrogen dioxide (10102-44-0)	3 reproductive effects	in production of chemicals – particularly nitric acid, in rocket fuels, polymerization, inhibitor of acrylates
Nitrophenol		
2, nitrophenol (88-75-5)	3	Making pesticides, dyes and other chemicals
3, nitrophenol (554-84-7)	3	Making pesticides, dyes, indicator solutions and other chemicals
4, nitrophenol (100-02-7)	3	Fungicide, making other chemicals

N-nitrosodimethylamine (62-75-9)	3 Confirmed carcinogen	Antioxidant, additive for lubricants, and softener of copolymers, solvent in fiber and plastics industry, used in condensers to increase dielectric constant, formally used as a nematocide, formerly used in production of rocket fuels, laboratory chemical
N-nitrosodiphenylamine (156-10-5)	3 Carcinogen	
N-nitrosodi-N-propylamine (621-64-7)	3 Confirmed carcinogen	
Nitrous oxide (10024-97-2)	2 Mutagen	Anesthetic in dentistry and surgery, propellant gas in food aerosols, leak detectors, foaming agent in whipped cream
Oxalic acid (144-62-7)	3	Wide-range of uses, automobile radiator cleaner, general metal and equipment cleaning, leather tanning, laboratory chemical, bleaching textiles and woods, permanent press resins
Ozone (10028-15-6)	3 Questionable carcinogen powerful oxidizing agent	Purifying air and water; industrial water treatment; bleaching waxes, oils, paper, textiles; making other chemicals, bactericide, steroid hormones
Pentachlorophenol (87-86-15)	3 Also called Pentacon, Permacide, Permite, Weedone, Thompsons Wood Fix; suspected human carcinogen	Making fungicides, bactericides, algaecides, herbicides; wood preservative; insecticide for termite control; general herbicide
Phenol (108-95-2)	3 Questionable carcinogen	Making pharmaceuticals, chemicals, plastics, resins, plywood, rubber, refining oils, fertilizer, coke, points, paint removers, asbestos goods, perfumes; general disinfectant in solution, bactericide, fungicide, veterinary antiseptic and anesthetic
Phenyl phenol		
o-phenyl phenol or 2-biphenylol (90-43-7) 2-phenyl phenol (90-43-7)	3 mutagen	Making fungicides, rubber, chemicals and dyestuffs

p-phenyl phenolor 4, biphenylol 4- phenyl phenol (92-69-3)	3 Acute poison, questionable carcinogen	
Phosphoric acid (7664-38-2)	3 Corrosive	Making metal products, gelatin , fertilizers; soaps and detergents, rust-proofing, pharmaceuticals, sugar refining, water treatment, animal feeds, eletro-polishing, gasoline additive, coating for metals, making other chemicals, fabric dyeing, yeasts, waxes and polishes, in ceramics, in foods and carbonated beverages, laboratory chemical
Polychlorinated biphenyl (PCB) (1336-36-3)	3 Suspected carcinogen	
Potassium fluoride (7789-23-3)	3 Highly corrosive	Etching glass, preservative, insecticide, in organic synthesis, solder flux
Potassium cyanide (151-50-8)	3 Deadly poison, mutagen, corrosive, reproductive hazard	To extract gold and silver from ore, electroplating, laboratory chemical, insecticide, to harden steel, fumigant
Potassium ferrocyanide two possibilities were suggested in SAX:		
Potassium hexacyanoferrate (II) (1445-95-1)	2	
Tetrapotassium hexacyanoferrate (forgot CAS)	1	
Potassium permanganate	3 strong irritant	In production of “drugs of abuse”, as a topical antibacterial agent, chemical reagent
Pyrene (8003-34-7)		Insecticide for store foods, household use, dairies
Ruthenium compounds (7440-18-8)	3	Hard metal crystals, toxic fumes when burned

Sodium arsenate or arsenic acid, sodium salt (7631-89-2)	Confirmed human carcinogen, reproductive hazard	Dyeing and printing, making other chemicals, germicide
Sodium hydroxide (1310-73-2)	3 Also called caustic soda, lye, strong base, corrosive; among the highest volume chemicals produced in the U.S.	Making other chemicals, mercerized cotton, processing vegetables and fruits, laboratory chemical, electro-plating, extraction of zinc, tin plating, oxide coating, etching, food additive, metal cleaner, laundering, bleaching, in making plastics
Stoddard solvent (8052-41-3)	3 Also called white spirits, dry cleaning solvent, varsol, veroline	Dry cleaning, spot and stain remover, degreasing, cleaning in mechanical shops, herbicide
Styrene (100-42-5)	3 Suspected carcinogen	Solvent, making plastics, synthetic rubber, protection coatings, resins, polyesters; making other chemicals
Sulfamic acid (5329-14-6)	3 Corrosive	Stabilizer or swimming pool chemicals, metal cleaning, making paper
Sulfuric acid (7664-93-9)	3 Powerful acid oxidizer, corrosive, combustible	Fertilizers, dyes, pigments, petroleum refining, etching, electro-plating, rayon, film, explosives, laboratory chemical, making iron and steel, making other chemicals
Tetrachloroethane (25322-20-7)	Suspected carcinogen, considered one of the most toxic of the chlorinated hydrocarbons	Making other chemicals, insecticides, paints, rust removers and varnishes
Tetrachloroethylene or percholorethylene (127-18-4)	3 Confirmed carcinogen	Solvent and dry cleaning agent, degreasing , heat exchange fluid, insulating fluid and cooling gas for transformers, drying agent for metals, grain fumigant
Trimethylbenzene (25551-13-7)	1	Making other chemicals
Titanium compounds	Considered to be physiologically inert; dusts are considered in the nuisance category	Metal used in structural material in jet engines, marine equipment, chemical and surgical equipment
Toluene (108-88-3)	3 Derived from coal tar, commercial grades usually contain small amounts of benzene poison	Paint solvent, in aviation gasoline and high octane blending stock, making other chemicals, TNT, detergents, saccharine, medicines, dyes, perfumes, adhesive solvent
Toxaphene or chlorinated camphene	3 Confirmed carcinogen, reproductive hazard	Insecticide that resembles chlordane, primary for foliage of cotton, small grass and vegetables, livestock pest control, maggot killer

(8001-35-2)		
Tributyl phosphate (126-73-8)	3	Solvent, plasticizer, making pigments, anti-foam agent
Trichloroacetic acid (76-03-9)	3 Also called AMCHEM grass killer, Vantox questionable carcinogen, corrosive	Herbicide: to make medicines, drugs, herbicides, pesticides and other chemicals; laboratory chemical
1,2,4 trichlorobenzene (120-82-1)	3 Poison, possible reproductive hazard	Making insecticides, fungicides and other chemicals; heat transfer fluids
Trichloroethane or methyl chloroform (71-55-6)	3 Suspected carcinogen, poisonous	Cleaning solvent, metal degreasing, pesticide, making and finishing textiles, making other chemicals, aerosol propellant
Trichloroethylene (79-01-6)	3 Also called Vestral, Fleck-flip, Trimar, Perm-a-clean suspected carcinogen, mutagen	Solvent for oils, fats, waxes; degreasing and dry cleaning; solvent; dyeing; refrigerant; heat exchange fluid; fumigant; in paints and adhesives; textile processing; making other chemicals; a common air contaminant
2,4,6 trichlorophenol (88-06-2)	3 Confirmed carcinogen, some mutagen data	Preservative, in insecticides, treating mildew, germicide
2,3,6, trichlorophenol	3	
2,4,5 trichlorophenol (95-95-4)	3 Suspected carcinogen	
Uranium (7440-61-1)		Production of nuclear fuel
Uranyl nitrate (36478-76-9)	3	
Velpar or hexazinone (51235-04-2)	2	Herbicide
Vinyl chloride (75-01-4)	3 Confirmed human carcinogen, mutagen	Making other chemicals, refrigerant, adhesive for plastics
Xylene (1330-20-7)	3 May contain benzene, a carcinogen	Aviation gasoline, protective coatings, solvent for alkyd resins, lacquers, enamels, rubber cements, synthesis of organic chemicals

Xylidine (1300-73-8)	3 Also called acid leather brown 3G, or acid orange 24, resource brown J, dimethylaniline suspected human carcinogen	Making dyes, pharmaceuticals, and other chemicals
Zinc compounds (7440-66-6)	Variable toxicity, carcinogens	Galvanized coatings in iron and steel sheet metal production, making paint, dyes, brass metal alloys, batteries, laboratory chemicals

APPENDIX C3

NONCARCINOGENS AND CARCINOGENS

APPENDIX TO CHAPTER 16

Table C3-1. Noncarcinogens

Chemical name	Inventory amount (kg)	Develop-mental	Repro-ductive	RfD (mg/kg/d)	RfD converted to water conc (mg/L)	RfC (mg/m^3)	Drinking Water Standard (mg/L)	TLV/10, PEL/10 or (NAAQS) mg/m^3	RF air	Ratio air	RF water	Ratio water
Acetone	3,872.6		R	0.1	3.5			178.0	1	3.69×10^{-8}	0.02	2.74×10^{-6}
Acetonitrile	214.3			0.006	0.21				1	5.44×10^{-8}	0.02	2.53×10^{-6}
Acrolein				0.02	0.7	0.00002		0.23	1		0.02	
Aluminum nitrate	952,560							0.2	0.005	4.05×10^{-5}	0.1	
Ammonia	1,113.3					0.1	34	1.7	0.005	9.46×10^{-8}	0.1	4.06×10^{-7}
Antimony trioxide	5.4		RR	0.0004	0.014		0.006	0.05	1	1.84×10^{-7}	0.02	2.23×10^{-6}
Boron	78.8			0.09	3.15				0.005	1.34×10^{-9}	0.1	3.10×10^{-7}
Bromacil	275.6		R	0.13	4.55				0.005	2.34×10^{-9}	0.1	7.51×10^{-7}
Bromine	0.3							0.066	1	7.72×10^{-9}	0.02	
Bromofluorobenzene	204.1							0.52	1	6.67×10^{-7}	0.02	
Calcium fluoride	824.0							0.25	0.005	2.80×10^{-8}	0.1	
Ceric ammonium nitrate	1,050.8							5.1[b]	0.005	1.72×10^{-9}	0.1	
Cerium (III) oxalate hydrate	35.0							0.1[c]	0.005	2.98×10^{-9}	0.1	
Chlorine gas	348,364.8			0.1	3.5				1	3.95×10^{-3}	0.02	2.47×10^{-4}
Chloroethane						10			1		0.02	
Chloroethylvinyl ether	17.7							1000 ppm	1	6.68×10^{-12}	0.02	
Chloronapthalene	0			0.08	2.8				1		0.02	4.43×10^{-11}
Chlorophenol	0.09		RRR	0.005	0.175				1		0.02	1.27×10^{-9}
Coal	41,010,886						0.002 (BaP)	0.2	0.005	1.74×10^{-3}	0.1	2.54×10^{2}
Coal fly ash	488.1						0.005	0.2	0.005	2.07×10^{-8}	0.1	1.22×10^{-4}
Coal slag	567						0.005	0.2	0.005	2.41×10^{-8}	0.1	1.40×10^{-4}
Copper compounds	142,111.5						1.3	0.02	0.005	6.04×10^{-5}	0.1	1.35×10^{-3}
Cyanide (KCN,NaCN, etc.)	100.7	D		0.005	0.175	0.003	0.2	0.5	0.005	2.85×10^{-7}	0.1	7.14×10^{-6}
Diazinon	10.5	DDD		0.00009	0.00315				0.005	8.92×10^{-9}	0.1	4.13×10^{-5}
Dibutyl phthalate	3,910.9	DD		0.1	3.5				1	1.33×10^{-5}	0.02	2.77×10^{-6}
Dichlorodifluoromethane (F21)	3,041.2							495.0	1	1.04×10^{-8}	0.02	
Dichlorophenol	0.02			0.003	0.105				1		0.02	4.73×10^{-10}
Dimethylformamide	9.6	DDD	R	0.1	3.5			3.0	1	5.44×10^{-9}	0.02	6.80×10^{-9}

Endosulfan	0			0.00005	0.00175		0.074	0.01	1	1.70×10^{-10}	0.02	1.42×10^{-9}

Table C3-1. (Continued)

Chemical name	Inventory amount (kg)	Develop-mental	Repro-ductive	RfD (mg/kg/d)	RfD converted to water conc (mg/L)	RfC (mg/m$^{3)}$	Drinking Water Standard (mg/L)	TLV/10, PEL/10 or (NAAQS) mg/m^3	RF air	Ratio air	RF water	Ratio water
Endosulfan sulfate	0			0.00005	0.00175		0.074	0.01	1	1.70×10^{-10}	0.02	1.42×10^{-9}
Endrin	0		R	0.0003	0.0105		0.002	0.01	0.005	8.50×10^{-13}	0.1	1.18×10^{-9}
Endrin aldehyde	0		R	0.0003	0.0105		0.002	0.01	0.005	8.50×10^{-13}	0.1	1.18×10^{-9}
Ethoxyethanol	920.8							1.8	1	8.69×10^{-7}	0.02	
Ethylbenzene	12,260.4			0.1	3.5	1.0	0.7	43.4	1	2.08×10^{-5}	0.02	4.34×10^{-5}
Fluoranthene	0			0.4	14				1	1.70×10^{-11}	0.02	1.77×10^{-13}
Fluoride	0.18				0.12		4.0	0.25	0.005	6.12×10^{-12}	0.1	1.86×10^{-8}
Gadolinium compounds	408.2			0.06	2.1				0.005	1.51×10^{-6}	0.1	2.41×10^{-6}
Gadolinium nitrate	5,774.3				2.1				0.005	2.13×10^{-5}	0.1	3.41×10^{-5}
Hexanol	14,600				0.0072[a]				1	3.44×10^{-3}	0.02	6.82×10^{-6}
Hydrochloric acid	1,483.5							0.75	1	3.36×10^{-6}	0.02	5.02×10^{-3}
Hydrofluoric acid	45,400			0.06	2.1		4.0	0.26	1	2.97×10^{-4}	0.02	5.36×10^{-5}
Hydrofluosilicic acid	46.6							0.25	1	1.98×10^{-8}	0.02	
Hydrogen sulfide	454,000							1.4	1	5.50×10^{-4}	0.02	
Hydroxylamine nitrate	454,000							5.2[b]	1	1.48×10^{-4}	0.02	
Hydroxylamine sulfate	45,400				0.00102 [a]				1	7.56×10^{-2}	0.02	1.10×10^{-1}
Iron (ferrous) sulfamate	353,808							0.1 (Fe)	0.005	3.00×10^{-5}	0.1	
Kerosene (n-parrafin, ultasene)	89,812.8							0.2 (fume)	1	7.63×10^{-4}	0.02	
Lithium compounds	36,160.1							0.0025	0.005	1.22×10^{-4}	0.1	
Manganous nitrate	45,400		RR	0.005	0.175	0.0003		0.3 (5)	0.005	1.28×10^{-3}	0.1	3.22×10^{-3}
Mercuric nitrate	5,659.2		R				0.002 (Hg)	0.01	0.005	4.81×10^{-6}	0.1	3.53×10^{-2}
Mercury	9,797.6	DDD	RR	0.0003	0.0105		0.002	0.005	1	3.33×10^{-3}	0.02	1.21×10^{-2}
Methanol	8,648.9							26.2	1	5.61×10^{-7}	0.02	
Methoxychlor	0		RR	0.005	0.175		0.04	1.0	0.005	8.50×10^{-15}	0.1	3.10×10^{-10}
Methyl bromide				0.1	3.5			1.9	1		0.02	
Methyl chloride	0							10.3	1		0.02	

Methyl ethyl ketone	9,656.0			2	70	1.0		59	1	1.64×10^{-5}	0.02	3.42×10^{-7}
Nitric acid	4,545,072.0							5.2	1	1.48×10^{-3}	0.02	
Nitrogen dioxide	13.7		R					0.56 (100)	1	4.15×10^{-8}	0.02	

Table C3-1. (Continued)

Chemical name	Inventory amount (kg)	Develop-mental	Repro-ductive	RfD (mg/kg/d)	RfD converted to water conc (mg/L)	RfC (mg/m^3)	Drinking Water Standard (mg/L)	TLV/10, PEL/10 or (NAAQS) mg/m^3	RF air	Ratio air	RF water	Ratio water
Nitrophenol	2.2						65		0.005		0.1	4.19×10^{-10}
Nitrosobenzene								0.05	1	3.40×10^{-11}	0.02	
Oxalic acid	45,400							0.1	1	7.72×10^{-4}	0.02	
Ozone	0							(235)	1		0.02	
Phenol	814.4			0.6	21				1	7.29×10^{-7}	0.02	9.62×10^{-8}
Phosphoric acid	8,041.0							0.1	1	1.36×10^{-4}	0.02	
Potassium cyanide	5.1	D		0.05	1.75				0.005	8.67×10^{-11}	0.1	3.61×10^{-8}
Potassium ferrocyanide	124.1	D						0.5	0.005	2.11×10^{-9}	0.1	
Potassium fluoride	827.0							0.25	0.005	5.62×10^{-6}	0.1	
Potassium permanganate	32,659.2							0.5	0.005	1.11×10^{-4}	0.1	
Pyrene	0.05			0.03	1.05				1	1.7×10^{-10}	0.02	1.18×10^{-10}
Rhodium nitrate	50.4							0.001	0.005	4.28×10^{-7}	0.1	
Ruthenium compounds	3.4				0.0036[a]				0.005	8.02×10^{-9}	0.1	1.17×10^{-5}
Sodium hydroxide	4,540,000							0.2	0.005	1.93×10^{-4}	0.1	
Stoddard solvent	63,798.6							51.5	1	2.11×10^{-6}	0.02	
Styrene	5,125.7			0.2	7	1.0		21.3	1	8.71×10^{-6}	0.02	1.82×10^{-10}
Sulfamic acid	5,740.4							0.1	1	9.75×10^{-5}	0.02	
Sulfur oxides	172.7							(80)	1	3.67×10^{-9}	0.02	
Sulfuric acid	544,320							(0.07)	1	1.32×10^{-2}	0.02	
Titanium compounds (dioxide)	358,781.7							1.0	0.005	3.05×10^{-6}	0.1	
Titanium tetrachloride	13.9							1.0	0.005	1.18×10^{-10}	0.1	
Toluene	40,493.2	DD		0.2	7	0.4	1.0	18.8	1	1.72×10^{-4}	0.02	1.00×10^{-4}
Tributyl phosphate (TBP)	62,087.2							0.22	0.005	2.39×10^{-6}	0.1	
Trichloroacetic acid	1.99						0.06	0.67	0.005	2.52×10^{-11}	0.1	4.11×10^{-7}
Trichlorobenzene	5.5		RR	0.01	0.35	0.2	0.07	3.7	1	4.68×10^{-8}	0.02	1.95×10^{-7}
Trichlorophenol (245-T)	0.6			0.01	0.35				1		0.02	4.25×10^{-9}

Trimethylbenzene	134,996.2							12.3	1	1.86×10^{-5}	0.02	

Table C3-1. (Continued)

Chemical name	Inventory amount (kg)	Develop-mental	Repro-ductive	RfD (mg/kg/d)	RfD converted to water conc (mg/L)	RfC (mg/m^3)	Drinking Water Standard (mg/L)	TLV/10, PEL/10 or (NAAQS) mg/m^3	RF air	Ratio air	RF water	Ratio water
Uranium	15,880,776.8	DDD	R	0.003	0.105		0.1	0.005	0.005	2.69×10^{-2}	0.1	1.87
Uranyl nitrate	0.09							0.005	0.005	1.53×10^{-10}	0.1	
Velpar L herbicide	0			0.033	1.15				1		0.02	
Xylene	100171.6			2	70		10.0	43.4	1	3.92×10^{-6}	0.02	2.48×10^{-5}
Xylidine	1.3							0.25	1	8.84×10^{-9}	0.02	
Zinc compounds	416663.4		R	0	10.5		5.0	0.1	0.005	3.54×10^{-5}	0.1	1.03×10^{-3}

[a] The lowest LD50/10,000.
[b] Toxicity value for nitrates or nitric acid.
[c] Toxicity value for oxalate.

Table C3-2. Carcinogens (Known, Suspect, Possible, Probable, and Mutagens)

Chemical name	Inventory amount (kg)	C	D	R	SF oral (mg/kg/d)	10^{-5} water conc (mg/L)	INHUR risk per Tg/m^3	10^{-5} air conc (mg/m^3)	TLV, PEL/10 or (NAAQS) mg/m^3	RfD	RfD converted to water conc	RfC
Acrylonitrile	1.7	B1	D	R	0.54	6×10^{-4}	0.000068	1×10^{-4}	0.43	0.001	0.035	0.002
Aldrin	2.0	B2			17	2×10^{-5}	0.0049	2×10^{-6}	0.025	3×10^{-5}	0.00105	0.005
Aniline	11.9	B2			0.0057	0.06			0.76			0.001
Anthracene	55.4	D							0.02	0.3	10.5	
Arsenic	0.3	A		RR	1.5	2.3×10^{-4}	0.0043	2×10^{-6}	0.02	0.003	0.0105	
Asbestos	442.62	A					0.23	4×10^{-8}	0.05			0.23
Benzene	7,350.1	A	DD	RR	0.029	0.012	8.30×10^{-6}	1.2×10^{-3}	3.2			
Benzidine	0	A			230	1.5×10^{-6}				0.003	0.105	
Benzo(a) anthracene	0	B2										
Benzo (a) pyrene	0	B2			7.3	5×10^{-5}						
Benzofluoranthene	0	D										
Benzoperylene	0	D										
Beryllium	1.4	B2			4.3	8.3×10^{-5}	0.024	4.2×10^{-7}	0.0002	0.005	0.175	
Bromoform	3.3	B2			0.0079	0.044	1.12×10^{-6}	9.0×10^{-3}	0.52	0.02	0.7	
Butyl benzyl phthalate	103.1	C		RR						0.2	7.0	
Cadmium compounds	1,781.2	B1		RR			0.0018	6.0×10^{-6}	0.0002			
Carbon tetrachloride		B2		R	0.13	0.0027	1.50×10^{-5}	7.2×10^{-4}	3.1	0.0007	0.0245	
Clordane		B2			1.3	2.7×10^{-4}	0.00037	3.0×10^{-5}	0.05	6×10^{-5}	0.0021	
Chlorobenzene	2.4	D							4.6	0.02	0.7	0.02
Chlorodibromomethane	10.3	C			0.084	4.21×10^{-3}				0.02	0.7	
Chloroethene (ethylchloride)		A3							26.4			
Chloroform	18.57	B2			0.0061	0.0573	2.3×10^{-5}	4.0×10^{-4}	4.9	0.01	0.35	
Chloromethane (methylchloride)		C		RR	0.0126	0.027				0.004		
Chromium compounds	187,046.6	C,A							0.001			
Chrysene	0	B2							0.02			
Coal tar (coke oven emissions)	3,589.5	A							0.02			
Cristobalite	10,296.7								0.05			
Dibenzanthracene	0.009											
DDT dichloro diphenyl trichloroethane	0	B2	DD	R	0.34	1.03×10^{-3}			0.1	0.0005	0.0175	

Table C3-2. (Continued)

	MCL				

Chemical name	(mg/L)	RF air	Ratio air	RF water	Ratio water
Acrylonitrile		1	2.89×10^{-5}	0.02	7.03×10^{-6}
Aldrin		1	1.70×10^{-3}	0.02	2.48×10^{-3}
Aniline		1	2.66×10^{-8}	0.02	4.92×10^{-7}
Anthracene		1	4.71×10^{-6}	0.02	2.81×10^{-9}
Arsenic	0.05	0.005	1.11×10^{-8}	0.1	1.62×10^{-5}
Asbestos	7 f	0.005	9.40×10^{-2}	0.1	
Benzene	0.005	1	1.04×10^{-2}	0.02	3.64×10^{-3}
Benzidine		1		0.02	1.65×10^{-6}
Benzo(a) anthracene	0.002	1		0.02	8.50×10^{-10}
Benzo (a) pyrene	0.002	1		0.02	3.43×10^{-8}
Benzofluoranthene	0.002	1		0.02	8.50×10^{-10}
Benzoperylene	0.002	1		0.02	8.50×10^{-10}
Beryllium	0.004	0.005	2.83×10^{-5}	0.1	2.09×10^{-4}
Bromoform	0.08	1	6.23×10^{-7}	0.02	1.86×10^{-7}
Butyl benzyl phthalate	0.1	1		0.02	2.56×10^{-6}
Cadmium compounds	0.005	0.005	2.52×10^{-3}	0.1	4.42×10^{-3}
Carbon tetrachloride	0.005	1		0.02	
Clordane	0.002	1		0.02	
Chlorobenzene		1	2.04×10^{-7}	0.02	8.50×10^{-9}
Chlorodibromomethane	0.08	1	1.65×10^{-10}	0.02	6.07×10^{-6}
Chloroethene (ethylchloride)		1		0.02	
Chloroform	0.08	1	7.89×10^{-5}	0.02	8.04×10^{-7}
Chloromethane (methylchloride)		1		0.02	
Chromium compounds	0.1	0.005	1.59×10^{-3}	0.1	2.32×10^{-2}
Chrysene	0.002	1	8.50×10^{-11}	0.02	1.24×10^{-9}
Coal tar (coke oven emissions)	BaP	1	3.05×10^{-4}	0.02	4.45×10^{-3}
Cristobalite		0.005	1.75×10^{-6}	0.1	
Dibenzanthracene	BaP	1		0.02	1.12×10^{-8}
DDT dichloro diphenyl trichloroethane		1		0.02	

Table C3-2. (Continued)

Chemical name	Inventory amount (kg)	C	D	R	SF oral (mg/kg/d)	10^{-5} water conc (mg/L)	INHUR risk per Tg/m^3	10^{-5} air conc (mg/m^3)	TLV, PEL/10 or (NAAQS) mg/m^3	RfD	RfD converted to water conc	RfC
Dichlorobenzene	0.1	C		R	0.024	1.45×10^{-2}			6.0	0.09	3.15	
Dichlorobenzidine	0	B2			0.45	7.7×10^{-4}						
Dichloroethane	3.2	B2			0.091	3.8×10^{-3}			40.5	0.1	3.5	0.5
Dichloroethylene	0.6	B2 to C			0.6	5.8×10^{-4}	0.00005	2×10^{-4}	79.3	0.009	0.315	
Dichloropropylene		B2								0.0003	0.0105	0.02
Dieldrin	0.004	B2	DD	R	16	2.2×10^{-5}	0.0046	2×10^{-6}	0.025	0.00005	0.00175	
Diethylhexyl phthalate	13.1	B2	D	RR					0.5			
Diethyl phthalate	212.3	D	DD	R					0.5	0.8	28	
Diisodecyl phthalate	51,327.1			R					0.64[a]		0.64[a]	
Dimethylaniline	104.5		D		0.75	4.7×10^{-4}			2.5	0.002	0.07	
Dimethyl phthalate	175.0	D	D						0.5			
Dioctyl phthalate	450.8	B2	DD						0.5	0.02	0.7	
Dioxane	18.35	B2			0.011	0.031			9.0			
Diphenylhydrazine	0.09	B2			0.8	4.4×10^{-4}	0.00022	5×10^{-5}				
Ethanol	6,930.0								188.0			
Ethylene oxide	42.1	B1			1.02	3.4×10^{-4}			0.18			
Formaldehyde	80.9	B1					0.000013	8.0×10^{-4}	0.037	0.15	5.25	
Gasoline, diesal fuels, fuel oil	3,583,569.7								89			
Heptachlor	0	B2			4.5	8.0×10^{-5}	0.0013	8×10^{-6}	0.05			
Heptachlor epoxide	0	B2			9.1	4.0×10^{-5}	0.0026	4×10^{-6}				
Hexachlorobenzene	0.004	B2			1.6	2.0×10^{-4}	0.00046	2×10^{-5}	0.0025			
Hexachlorobutadiene	1.0				0.078	6.0×10^{-5}	0.000022	5×10^{-4}	0.021	0.002	0.07	
Hexachloroethane	0.04	C			0.014	2.5×10^{-2}	0.000004	3.0×10^{-3}	0.97	0.001	0.035	
Hexachlorocyclohexane (BHC) (lindane)	0				6.3	5.5×10^{-4}	0.0018	6.0×10^{-6}	0.05			
Hydrazine	45,400	B2			3.0	1.16×10^{-4}	0.0048	2.1×10^{-6}	0.01			
Hydrazine mononitrate	1,483.4	B2			3	1.16×10^{-4}	0.0049	2.0×10^{-6}	0.1			
Isophorone	110.1	C			0.00095	0.368			2.8	0.15	5.25	
Lead compounds	392,850.7	B2	DD						0.015			
Methylene chloride	525.2	B2							17.4	0.06	2.1	

Table C3-2. (Continued)

Chemical name	MCL (mg/L)	RF air	Ratio air	RF water	Ratio water
Dichlorobenzene	0.075	1		0.02	3.37×10^{-9}
Dichlorobenzidine		1		0.02	3.22×10^{-9}
Dichloroethane		1	1.08×10^{-8}	0.02	2.08×10^{-6}
Dichloroethylene		1	5.10×10^{-6}	0.02	2.56×10^{-6}
Dichloropropylene		1	3.40×10^{-10}	0.02	9.45×10^{-10}
Dieldrin		1	8.53×10^{-7}	0.02	1.13×10^{-7}
Diethylhexyl phthalate		1	4.45×10^{-8}	0.02	
Diethyl phthalate		1	7.22×10^{-7}	0.02	1.18×10^{-8}
Diisodecyl phthalate		1	1.36×10^{-4}	0.02	1.98×10^{-4}
Dimethylaniline		1	7.12×10^{-8}	0.02	5.51×10^{-4}
Dimethyl phthalate		1	5.95×10^{-7}	0.02	
Dioctyl phthalate		1	1.53×10^{-6}	0.02	1.59×10^{-6}
Dioxane		1	3.46×10^{-9}	0.02	1.47×10^{-6}
Diphenylhydrazine		1	3.06×10^{-6}	0.02	5.07×10^{-7}
Ethanol		1	6.26×10^{-8}	0.02	
Ethylene oxide		1	3.97×10^{-7}	0.02	3.07×10^{-4}
Formaldehyde		1	1.72×10^{-4}	0.02	3.82×10^{-8}
Gasoline, diesal fuels, fuel oil	benzene	1	6.85×10^{-5}	0.02	1.77
Heptachlor		1	2.12×10^{-7}	0.02	3.10×10^{-8}
Heptachlor epoxide		1	4.25×10^{-7}	0.02	6.23×10^{-8}
Hexachlorobenzene		0.005	1.70×10^{-9}	0.1	2.48×10^{-7}
Hexachlorobutadiene		0.005	1.71×10^{-8}	0.1	2.06×10^{-4}
Hexachloroethane		1	2.26×10^{-8}	0.02	3.97×10^{-9}
Hexachlorocyclohexane (BHC) (lindane)	0.0002	1	2.83×10^{-7}	0.02	4.51×10^{-9}
Hydrazine		1	3.67×10^{1}	0.02	9.71×10^{-1}
Hydrazine mononitrate		1	1.26×10^{0}	0.02	$3.17_{x}\ 10^{-2}$
Isophorone		1	6.68×10^{-8}	0.02	7.42×10^{-7}
Lead compounds	0.15	0.005	2.23×10^{-4}	0.1	3.25×10^{-2}
Methylene chloride	0.005	1	5.13×10^{-8}	0.02	2.60×10^{-4}

Table C3-2. (Continued)

Chemical name	Inventory amount (kg)	C	D	R	SF oral (mg/kg/d)	10^{-5} water conc (mg/L)	INHUR risk per Tg/m^3	10^{-5} air conc (mg/m^3)	TLV, PEL/10 or (NAAQS) mg/ m^3	RfD	RfD converted to water conc	RfC
Napthalene	4,540	C to D	D						5.2	0.004	0.14	
Nickel compounds (refining dust)	96,921.6	A					0.00048	2.1×10^{-5}	0.005	0.02	0.7	
Nitrilotriacetic acid	4.2	2B				0.011[a]			0.011[a]			
Nitrobenzene	65.3	B2 to D	D						0.5	0.0005	0.0175	0.002
Nitrosodimethylamine	0	B2		RR	51	7×10^{-6}	0.014	7.0×10^{-7}				
Nitrosodiphenylamine	0	B2				7×10^{-6}		7.0×10^{-7}				
Pentachlorophenol	0.2	B2	DD		0.12	3.3×10^{-3}			0.05	0.03	1.05	
Phenyl phenol	178.9	B2 to C	D	R	0.00194	0.181				0.00002	0.0007	
Polychlorinated biphenyls (PCBs, arochlors)	0	B2		R	7.7	0.0454			0.05	0.00002	0.0007	
Samarium nitrate	1.5								10.0			
Sodium arsenate	0.22	A	DD						0.001			
Tetrachloroethane	0	C			0.2	1.7×10^{-3}	0.000058	1.7×10^{-4}	0.69	0.03	1.05	
Tetrachloroethylene (PCE) (perc) withdrawn from IRIS	462,672.0	B2 to C	D						17	0.01	0.35	
Toxaphene	0	B2,2B	D		1.1	3.2×10^{-4}	0.00032	3.0×10^{-5}	0.05	0.1		0.005
Trichloroethane	5,610.4	C			0.057	6.2×10^{-3}	0.000016	6.2×10^{-4}	1.9	0.004	0.14	
Trichloroethylene withdrawn from IRIS	631.1	C							26.9	0.002		0.005
Vinyl chloride	9.9	A	DD		1.9	1.8×10^{-4}	0.000084	1.2×10^{-4}	1.3			0.002

Table C3-2. (Continued)

Chemical name	MCL (mg/L)	RF air	Ratio air	RF water	Ratio water
Napthalene		1	1.48×10^{-6}	0.02	8.04×10^{-5}
Nickel compounds (refining dust)	0.1	0.005	3.96×10^{-2}	0.1	1.20×10^{-2}
Nitrilotriacetic acid		0.005	7.72×10^{-10}	0.1	1.13×10^{-6}
Nitrobenzene		1	5.55×10^{-5}	0.02	9.25×10^{-6}
Nitrosodimethylamine		1	2.43×10^{-6}	0.02	3.54×10^{-7}
Nitrosodiphenylamine		1	2.43×10^{-6}	0.02	3.54×10^{-7}
Pentachlorophenol	0.001	1	6.8×10^{-9}	0.02	1.50×10^{-7}
Phenyl phenol		1		0.02	6.34×10^{-4}
Polychlorinated biphenyls (PCBs, arochlors)	0.0005	0.005		0.1	
Samarium nitrate	10 (NO_3)	0.005	1.27×10^{-12}	0.1	1.86×10^{-9}
Sodium arsenate	0.05 (As)	0.005	1.87×10^{-9}	0.1	5.46×10^{-8}
Tetrachloroethane	1.4	1	1.0×10^{-8}	0.02	1.46×10^{-9}
Tetrachloroethylene (PCE) (perc) withdrawn from IRIS	0.005	1	4.63×10^{-5}	0.02	2.29×10^{-1}
Toxaphene	0.003	1	5.66×10^{-8}	0.02	7.75×10^{-9}
Trichloroethane	0.005	1	1.54×10^{-2}	0.02	2.24×10^{-3}
Trichloroethylene withdrawn from IRIS	0.005	1	2.15×10^{-4}	0.02	3.13×10^{-4}
Vinyl chloride	0.002	1	1.40×10^{-4}	0.02	1.36×10^{-4}

APPENDIX D

PUBLIC INVOLVEMENT

APPENDIX D

PUBLIC INVOLVEMENT

INTRODUCTION

Public involvement is critical to the success of the Savannah River Site (SRS) Environmental Dose Reconstruction Project. Dose reconstruction requires a level of commitment to public interaction not encountered in most scientific studies. Scientists performing the research combine technical rigor and credibility with open and responsive communication with interested individuals and groups.

Radiological Assessments Corporation (RAC) scientists have been dedicated to encouraging the involvement of key stakeholders in the SRS Dose Reconstruction Phase II study. Researchers have spent many hours interacting with concerned citizens and workers via workshops, meetings, roundtable discussions, and other opportunities. It is our belief that early and effective stakeholder involvement is crucial to the success of the study.

Public Involvement Plan Summary

The project has been open to the public and conducted in a manner that maximizes effective public interaction. *RAC* has taken responsibility for many aspects of public communication, education, and outreach. Key points of the plan include:

- Proactive interactions with the media (print, radio, television); community organizations; citizens of the surrounding communities; and the SRS Health Effects Subcommittee.
- Conduct of public meetings and workshops in the communities surrounding SRS.
- Public outreach through newsletters, fact sheet, and project reports.

Public Involvement Deliverables

- A Public Involvement Plan.
- Toll-free telephone number, computer bulletin board, and a published mailing address to receive public input.
- Public meetings, workshops, and press briefings.
- Meetings with Health Effects Subcommittee
- Newsletters.
- Fact Sheets.
- Technical support for the SRS Health Effects Subcommittee.

PUBLIC INVOLVEMENT STAFF	
TECHNICAL STAFF	**SUPPORT STAFF**
John E. Till (lead)	Phoebe Boelter
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Marilyn Case	Marilyn Langan
Mona Dreicer	
Helen Grogan	
Kathleen Meyer	
Patricia McGavran	
Justin Mohler	
Ron Rope	
Sue Rope	
Paul Voillequé	

Philosophy of Public Involvement

The public involvement task is critical to the success of the project as a whole. In dose reconstruction, we cannot afford to achieve scientific credibility and not achieve public credibility. The project was designated a "public study" during Phase I and this approach continued throughout the course of the work. Scientists on the *RAC* research team have made the presentations, worked with the press, and written the fact sheets and newsletters.

Public Involvement Plan

The Public and Scientific Community: Providing Information and Soliciting Input

Radiological Assessments Corporation has ensured that all phases of the dose reconstruction has been open to the public, and have considered all public concerns while planning and conducting the Phase II research. Key individuals and organizations with whom RAC has worked with throughout the Project include:

- Brian Costner (Energy Research Foundation)
- Mildred McClain (Citizens for Environmental Justice)
- Carolyn Cain (Georgians Against Nuclear Energy)
- Mary Crum (South Carolina Wildlife Federation)
- Debra Hasan (Southern Association of Black Educators)
- Theloneous Jones (The Frontier)
- King Singleton (The Augustan's)
- Veronica Thomas (Taxpayers Advocacy Support).

RAC scientists recognize that a successful public involvement program is the result of careful planning. To build public credibility, a public involvement program must:

- Be initiated early;
- Be constant and consistent;
- Make members of the public a part of the process, involved in decision-making;
- Be forthcoming, responsive and open;

- Develop relationships between project participants and interested parties; and
- Speak to individual and community concerns.

Public Meetings, Workshops, and Press Briefings. Phase II public meetings, workshops, and press briefings have been designed both to inform the public and to solicit feedback and suggestions for the work. These meetings were publicized through mailings to individuals on the project mailing list; press releases distributed to newspapers, television and radio stations; announcements in *Dose Reconstruction News*, the quarterly project newsletter; and through published notices in the *Federal Register*. Meeting locations were varied among the communities surrounding SRS in South Carolina and Georgia. Every effort was made to hold these meetings in familiar locations with complimentary parking for the convenience of the public and the press.

Meetings consisted of a project overview for new attendees, a status update on specific aspects of Phase II, and a period for public questions, comments, and suggestions. Visual aids, such as overheads and computer demonstrations, were used during the meetings to enhance the technical presentations. A mailing list sign-up sheet, past issues of project newsletters, and fact sheets were made available to attendees at each of the meetings.

Fact Sheets. Fact Sheets were prepared when new information became available or in response to public concerns. They were designed to provide the public with a better understanding of the dose reconstruction process through definitions of commonly used scientific terms and descriptions of key processes or concepts. Electronic copies of fact sheets have been provided to CDC for use on their web site.

Fact Sheet Topics	Volume
Source Term Estimates: Types of releases; and Source term estimates (2/96)	No. 1
Radiation in our Environment: Natural sources; Manmade sources; Current breakdown of average doses received in 1 year; and Typical radiation exposures (3/96)	No. 2
Environmental Exposure Pathways: Environmental pathways; Food chain; Exposure assessment; and Food pathway factors (5/96)	No. 3
Glossary of Dose Reconstruction Terms (10/96)	No. 4
Accessing and Declassifying Historical Records: Access to historical records; Classification of records; Declassification of historical records; Public access to project records; Categories of classified information; Levels of classified information; Why some secrets remain; and Public participation (12/96)	No. 5
Tritium & SRS Offsite Releases - An Update: Tritium production and releases at SRS; The relative hazard of tritium; Tritium releases seen via monitoring records; and Monitoring tritium releases to water (2/97)	No. 6
Plutonium at the Savannah River Site: History of the Savannah River Site; Studying SRS operations; Characteristics of plutonium; Health studies; and Dose and risk from plutonium (3/97)	No. 7

Newsletters. The purpose of the project newsletter, *Dose Reconstruction News*, has been to address questions and concerns and to summarize discussions from public meetings. Newsletters were distributed via the SRS project mailing list. Electronic copies of the Newsletters have been provided to CDC for use on the web site.

Newsletter Topics	Volume
Phase II of SRS Dose Reconstruction Projects Begins: Estimating past releases; Identifying release pathways; How will so much information be stored; Evaluating environmental information; Clarifying ranges for uncertainty; and So many chemicals - A method to evaluate SRS chemical releases (2/96)	Vol. I, No. 1
A Piece of the Grand Design - SRS Demographic Database and Geographic Information System: Demographic database; Categories of information; Geographic information system; Collecting data; and Release estimates expected to differ (3/96)	Vol. I, No. 2
Evaluating SRS Environmental Monitoring, Data: Where and when are effluent and environmental monitoring data collected?; How will the environmental monitoring data be gathered in Phase II of the Project?; What types of data support the dose reconstruction project?; Why are environmental monitoring data so important?; What are some of the difficulties of using environmental monitoring data?; and How will the data be evaluated? (7/96)	Vol. I, No. 3
Estimates of Releases from the Savannah River Site: Release categories; Examining releases that are most important; Best estimates sought; Time histories will be prepared; Uncertainties to be estimated; Methods of estimating releases; Searches for original records; and Comparisons with environmental data (10/96)	Vol. I, No. 4
Selecting the Savannah River Site Study Area: Available information and accuracy of estimates; Measurements in the body; Measurements in the environment; Measurements at the points of release; Doses decrease with distance; and Recommended study area (2/97)	Vol. II, No. 1
Public Involvement at the Savannah River Site: Public Meetings, Workshops, and Press Briefings; Newsletters; Fact Sheets; Working With the Savannah River Site Health Effects Subcommittee; Education and Outreach Activities; Project Mailing List; Citizen Input and Evaluation (5/97)	Vol. II, No. 2
Estimating the Atmospheric Tritium Source Term at SRS: A Progress Report: Equipment; Diagram of Stack Tritium Monitor; Preliminary Atmospheric Tritium Source Term for 1955-1964; Atmospheric Tritium Releases from SRS in the Early Years (in curies); Measurement Uncertainties (7/97)	Vol. II, No. 3
Using Information About Savannah River Site Power and Production Levels To Fill Data Gaps: Introduction; Searching for Production Data; A Brief History of Operations; R Reactor Power Levels; P Reactor Power Levels; L Reactor Power Levels; K Reactor Power Levels; C Reactor Power Levels; The Canyons – Fuel Reprocessing; F Canyon U to Dissolver; H Canyon U to Dissolver; U235 Tubes to H Dissolver; SRS Tritium Estimated Production; Conclusions (12/97)	Vol. II, No. 4

Project Mailing List. The Project mailing list was used for the distribution of meeting announcements, newsletters, and notice of the issuance of Phase II reports. It includes names collected from sign-up sheets used at public meetings held during Phases I and II of the SRS project, and of individuals and organizations requesting information on the study. Those wishing to be added to the newsletter distribution list or to request past newsletter issues were able to contact *Radiological Assessments Corporation*, 417 Till Road, Neeses, SC 29107 (800-637-4766).

Education and Outreach Activities. During Phase I and Phase II of the Project, RAC scientists have established working relationships with instructors at several colleges and universities in the SRS region, including the University of South Carolina in Aiken, Savannah State College in Savannah, The University of South Carolina in Columbia, the Medical University of South Carolina in Charleston, South Carolina State University in Orangeburg, and Emory University in Atlanta. Scientists have supported university requests for project information by: 1) public or university presentations at or near these institutions; 2) establishing the Phase I database at the DOE reading room at USC Aiken and at Savannah State College; 3) mailing diskette copies of the database to a number of schools, organizations, and individuals; and 4) supporting a student tour of research facilities at SRS.

Researchers use the Phase I database to support education and outreach activities because it is a unique research tool for college staff and students. It represents the first comprehensive historical records review of a major weapons facility, with all documents declassified and available for offsite use. Data from the Phase I database have been input into an expanded, Phase II document database, containing additional information on records reviewed since the completion of Phase I. The Phase II database is discussed in Appendix H of this report, and will be available to interested parties upon request to the CDC.

Citizen Input and Evaluation. Following public workshops or meetings, RAC evaluated results, summarized the material presented, and considered comments and criticisms offered by attendees. The information obtained via these meetings allowed scientists to rate the effectiveness of the meetings and make adjustments as Phase II progressed. To request information on the project or talk with one of the scientists, citizens were able to contact *Radiological Assessments Corporation*, 417 Till Road, Neeses, SC 29107 (800-637-4766. (Appendix Reference)

Supporting CDC Project Staff at Public Meetings

One or more members of the *RAC* research staff attended each public meeting. *RAC* supported CDC Staff by being prepared to knowledgeably deal with the study's status, progress, and problems. *RAC* scheduled meetings associated with each draft or final report related to either radiological or chemical dose reconstruction at SRS during Phase II.

Working With the Savannah River Site Health Effects Subcommittee (SRSHES)

RAC has kept the Savannah River Site Health Effects Subcommittee informed as to the study's status, progress, and problems. The Subcommittee has been given the opportunity to review and comment on draft report materials produced during Phase II. When feasible, meetings with these members were combined with the public meetings or workshops discussed above. If not, separate meetings were held to ensure that the Subcommittee members were kept informed

and able to contribute to the study. *RAC* has provided copies of draft and final plans, reports, newsletters, and fact sheets to the CDC. In conjunction with many presentations at the HES meetings, *RAC* has provided handout copies, and copies of key research materials during those meetings. *RAC* has also provided assistance to HES members making presentations of their own at related meetings. *RAC* also set aside a section of the FTP server for use by the Subcommittee.

Preparing For Public Meetings, Workshops, And Press Briefings

RAC realizes the importance of proper preparation for all parties involved in the meetings. Leadership, both before and during the meeting, is essential. *RAC* staff was comfortable in roles as presenters and facilitators, and professional support staff. *RAC* was responsible for or assisted in planning and executing public meetings and workshops, with full participation of CDC staff. Meetings were held in communities in the vicinity of the Savannah River Site (Aiken, Barnwell, Beaufort, Columbia, Savannah, Hilton Head, Augusta) to provide convenient opportunities for interactions with all interested parties. *RAC* made arrangements as required by the CDC for public meetings.

Site selection and contract negotiation. When requested by the CDC, *RAC* selected meeting locations which best meet the goals and objectives of the overall program.

Meeting room setup and audiovisual support. When requested by the CDC, *RAC* arranged for all necessary meeting room setups and audiovisual support. A table was placed at the entrance of the room for displaying reports, fact sheets, and newsletters. Overhead projectors, slide projectors, LCD panels, electric pointers, screens, flip charts, and microphones were ordered as needed.

Travel assistance. *RAC* was available to assist with travel arrangements as needed for invited speakers.

Meeting announcements and media interviews. Meeting announcements were prepared for timely distribution to the project mailing list, media list (newspapers, TV, radio, regional newsletters), and *Federal Register*. *RAC* worked with CDC to respond to requests for media interviews resulting from public meeting announcements.

Receiving Public Comments

RAC established several systems for receiving public comments as part of the citizen input and involvement effort.

- A toll-free telephone number (800-637-4766)
- Computer bulletin board
- FTP server
- Mailing address (*Radiological Assessments Corporation*, 417 Till Rd., Neeses, SC 29107)
- Opportunities for verbal and written comment at public workshops/meetings

RAC has provided copies of draft and final plans, reports, newsletters, and fact sheets to members of the public upon request, as well as presentation handouts at all public and HES meetings. Newsletters and fact sheets containing summaries of plans and reports were sent to those on the project mailing list.

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APPENDIX E

SAMPLING PROCEDURES USED AT SRS FOR MONITORING ATMOSPHERIC TRITIUM RELEASES

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SAMPLING PROCEDURES USED AT SRS FOR ATMOSPHERIC TRITIUM RELEASES

ABSTRACT

This appendix describes the monitoring instrumentation, sampling procedures, and data analyses used to measure or estimate the reported releases of tritium. The accuracy of reconstructing the atmospheric tritium source term is largely dependent on the type of monitors used to measure the releases. Atmospheric tritium releases have been monitored using dehumidifiers, silica gel monitors, Kanne ionization chambers, the stack tritium monitor (STM), the Berthold tritium monitor (BTM), the stack monitor integrator (SMI), and the FORMS in the reactor and tritium facilities on the Savannah River Site (SRS). This appendix describes the monitoring equipment and how it was used. We have performed an extensive review of documentation, but it has been difficult to determine the exact times that each type of monitor operated at each of the facilities.

Radiological Assessments Corporation (RAC) researchers identified no clear deficiency in the methodologies used at SRS. Therefore, we initially believed that no correction factors should be applied to the SRS data in producing the final source term estimates presented in Chapter 4.1. To verify that this was an acceptable conclusion, we used an uncertainty assessment to evaluate the effectiveness of the monitors. This appendix presents the estimated uncertainty of the tritium measurements by type of monitor.

Using the results of the uncertainty assessment, we concluded that the "best estimate" values should be used as the atmospheric tritium source term without correction, and the range of uncertainty should be applied to release data by facility and year. If more information is found about the monitors or when they were used in each facility, we can apply these results more specifically.

INTRODUCTION

Our annual or monthly release estimates of tritium to the atmosphere are based on Site documents, memos, and logbooks. To develop the tritium facilities and the reactor areas atmospheric source term and its associated uncertainty, it was necessary to determine the methods by which the reported releases were measured or estimated. This appendix describes the monitoring instrumentation, sampling procedures, and data analyses used to measure or estimate the reported releases of tritium.

The equipment used at the SRS to monitor tritium, in most cases, was developed at the Site using existing components. Usually, off-the-shelf equipment that could fulfill the monitoring requirements was not available. The work being done by the scientists and engineers at the SRS was at the forefront of tritium technology, so new approaches and instrumentation were developed as the need arose. Letters that were found from other facilities requesting advice from the SRS staff confirm this fact. It appears from old documentation that the adequacy of the monitoring capability was evaluated regularly. The development of new technology can be

tracked in the memos that propose, justify, and evaluate the new monitors as they were implemented onsite.

It has been difficult to determine the exact times that certain equipment was installed and operated at each of the facilities. In reviewing the documentation, it is possible to find dates on equipment operation procedures or reports of plans or prototype tests. However, it has not been possible to reconstruct an exact chronological history of which monitoring equipment was used in each facility to measure and report the atmospheric releases. Unless more specific information is found for each SRS facility, Table E-1 provides as a general guide to tritium monitoring systems that were the primary source of the reported tritium releases since the 1950s.

Table E-1. Guide for Use of Tritium Monitors at SRS

Monitor	Facility	Time period in use
Dehumidifier	Reactors	1954–shutdown
Silica gel	Reactors and disassembly areas	during 1954–1958 or until shutdown in some cases
Kanne ionization chambers	Tritium facilities	1954–present
Stack tritium monitor (STM)	Reactors	1970–1988
Berthold tritium monitor (BTM)	Reactors	1988–present
Stack monitor integrator (SMI)	Tritium facilities	1974–present
FORMS	Tritium facilities and reactors	1985–present

We made assumptions about which monitors were used in specific facilities based on the reactors' years of operations. For example, BTMs only came into use after the R-Reactor was shut down, so we concluded that BTM monitors were never used in the R-Reactor facility.

The documentation we found that describes testing or operation of prototypes showed that monitoring equipment was tested in parallel to provide some redundancy and check results. Therefore, we could not identify exact dates, by month, when specific monitoring equipment was used. In the cases where data sheets reported the results of multiple monitoring devices, it is possible to check the measurement results of one system against the reported monthly value and determine which monitor results were used. However, we could not determine the exact date that the primary monitoring device replaced an older system, or when it was used during unplanned malfunctions of equipment.

The overall uncertainty includes the representativeness of the sample, the frequency of sampling, efficiency of sampling, the effects of environmental conditions, the accuracy of the measurements, and errors in reporting results. This appendix describes each monitoring system or component of the system. The latter part of this appendix addresses the uncertainty associated with the results from each method of monitoring.

In many cases, existing systems were coupled to create "new" monitoring systems. For example, silica gels or dehumidifiers were combined with the Kanne chambers to create the STM. The SMI is a Kanne chamber coupled with an electronic method of integrating the continuous monitoring data. The FORMS monitor installed in the Tritium Facilities appears to be similar to

the STM because the Kanne chambers that are part of the FORMS monitor measure the stack releases with and without water vapor being removed.

EXHAUST AIRFLOW AND AIR STREAM SAMPLING

Estimates of exhaust airflow rates and additional errors introduced because of sampling could possibly be a major influence on the final estimated stack releases. Detailed descriptions of methods of exhaust airflow measurements taken at the SRS facility stacks have not been found; however, it is clear that there was some capability to measure these flows. A historical copy of Health Physics Procedures (Du Pont 1965) presented average exhaust air stream values to be used for various stacks. These changed slightly through time, indicating that some measurements were actually made. Measurements were definitely made in later years.

In health physics work area sampling documents, we found a description of a system designed to individually sample widely separated work areas or simultaneously average the air activity from any work areas. By using electrically or manually operated valves, samples could be selected from one or more points at a work site or the size of the area being monitoring by one system could be changed. The piping was sized to correct for the pressure drop in long piping runs or restricting orifices (Morris 1967). Although the sampling documents described how these types of measurements were made, in general, we found no specific evidence on how stack measurements were actually made.

There were, however, airflow diagrams of the reactors and Tritium Facilities Buildings 232-H, 234-H, and 244-H were found in a Radioactive Air Emissions Monitoring document (Westinghouse 1991e). Chapter 4.1 presents these diagrams, which also include the exhaust monitoring sampling locations. For different areas, it seems that airflow in the stack was either measured directly or estimated based on building and exhaust system designs; the method of determination is not clear. For example, Table E-2 presents the ventilation airflow rates for reactor stacks provided in a written procedure for silica gel monitoring written in the 1950s.

Table E-2. Ventilation Airflow Rates for Reactor Stacks[a]

Location	Pounds of air per day
Purification exhaust	4,220,000
148-ft stack	35,300,000
Process room near exhaust	8,780,000
Process room for exhaust	7,660,000
-40 pump rooms	12,800,000

[a] Source: Du Pont (1954).

We found some discussion indicating that in 1989, the airflow in the stack was determined by measuring the pressure drop across the carbon filters. This was measured in inches of water and was converted to airflow using a vendor quality assurance test of the filters. The error in the stack flow estimates was estimated to be about 10% based on experience and review of stack flows. The pressure drops are only accurate to the nearest 0.05 in., which introduces a potential additional error of ±5%. Therefore, the conservative estimate of airflow measurement accuracy is ±15% (Smith 1989). However, exactly how knowledge of this accuracy was used in day-to-day operations is not clear.

A data sheet for August 17, 1990, shows that data were collected daily on reactor ventilation systems. Airflow was checked five times a day and showed no variation for the main exhaust stack (Westinghouse 1990a), but methods used were not outlined.

In *SRS Alternative Methods Information* (Westinghouse 1991b), rotameters were the principal method of regulating and measuring sample flow rates for the Kanne chambers in the reactors. It was indicated that the rotameters were calibrated anually and at any other time that maintenance or changes occurred. Unfortunately, no information was found about how often rotameters were calibrated or the range of variability.

In 1991, a report indicated that for K-Reactors and L-Reactor ventilation systems, five hot wire anemometers were used to monitor the total effluent air discharge, and the flow was indicated in the central control room (Suttinger and Burgo 1991). Data sheets found for reactors in the 1970s and late 1980s list airflow values that vary, indicating measured values were being used. From this information, we concluded that when possible (and in later years) actual measurements were used daily.

To sample the air leaving the stack, pumps were installed to withdraw effluent and route it to the samplers. In the earlier years, the samples were not necessarily continuous, which affected the overall accuracy of the release estimates. We describe the sampling regimes in the following sections for each type of monitor used. For tritium gas and vapor, we assumed that sampling air directly from the outflow stack did not introduce measurement errors.

TRITIUM LOSSES

Moderator Loss

The earliest monitoring operations were mostly concerned with measuring loss of moderator. To keep the reactors running, there needed to be sufficient moderator in the system to ensure the core of the reactor was cooled. The moderator is comprised of heavy water and was produced at SRS or elsewhere (Dana Plant). Because of the bombardment of the moderator by fission neutrons, tritium was created in the moderator. Because tritium has a relatively long half-life, its concentration in the moderator built up over years of continuous reactor operation. Moderator could be lost to the atmosphere or by direct spills because of evaporation from leaks in the system, spills during maintenance, or planned liquid discharges. These losses were economically important because moderator was costly to replace and exposures to the environmentally mobile tritium oxide were a potential cause of human health and environmental impacts. For these reasons, the loss of moderator was routinely monitored. By measuring moderator losses and the concentration of tritium, total tritium losses were measured. Daily releases to the stack would vary depending on the type of operations and maintenance carried out in the reactor.

Initially, “releases of tritium to the atmosphere from the 105 (reactor) building are computed from the per cent of D_20 contained in water samples condensed from the stack stream and from moderator losses as determined by inventory” (Caldwell 1958). The method of tracking the moderator inventory reflected all phases of loss; therefore, stack discharges could not be differentiated from losses through sump collection, losses to the disassembly basin water during reactor discharge, or other inventory unaccountability. We did not find an indication of the exact time period that this method was used.

Chapter 5 of this report, which deals with aquatic releases, addresses moderator losses as direct liquid releases. Caldwell (1958) indicates that individual inventory loss measurements were considered accurate to within a factor of 2.

It appears that the first direct method used to determine moderator loss was measuring the inventory of moderator in the tank. When the volume was measured visually on a sight glass attached to the tank, it was possible to discriminate by 1/8 in. or ±149 lb in the reactor tank. The amount that evaporated was dependent on the bulk moderator temperatures. It was reported that the effect of a fluctuation of the moderator temperature of ±1°C would lead to a change of ±300 lb in apparent inventory (Longtin et al. 1973; Jacober et al. 1973). The reports states, without more explanation, that the inventory precision was about ±550 lb. Assuming a density of 1.105 g cm^{-3} for heavy water moderator (Shleien 1992), 1 lb of moderator is approximately 410 mL.

The moderator samples were analyzed for tritium content using a liquid scintillation counter. Using the estimated volume of moderator lost (pounds or liters) and the concentration of the tritium in the moderator (curies per pound or curies per liter), the total loss of tritium (curies) could be estimated. The efficiency of counting (measuring) the concentration of tritium in the moderator sample was quoted as 57%, and it was calibrated over a range of 5×10^{-5} to 0.4 µCi mL^{-1}. The cited variation at 0.038 µCi mL^{-1} was determined to be 0.8% relative at the 95% confidence level (Longtin et al. 1973; Jacober et al. 1973). This information is included in this report for possible use in understanding the uncertainty associated with the early values reported for atmospheric tritium releases.

Limited data found for the early years showed that in 1955, the average total loss of moderator per reactor month was 1453 lb. If the liquid discharge is not taken into account, the loss was 740 lb. The 1956 losses were estimated at 1197 lb mo^{-1}, with no discharge (Rubin 1958). The average concentration of tritium in the moderator was about 1–3 Ci L^{-1}, which was relatively low because the reactors had just begun operation (1955–1957). By the early 1980s, the concentration was about 9 Ci L^{-1}. Kiger (1955) indicates that the losses from R-Reactor ranged from 3 to 80 lb d^{-1} in January and February 1955.

Because of the limitations of this method, other methods of determining moderator loss were developed using silica gel, cold traps, dehumidifiers, and the STM. The monitors, described below, were used to determine the tritium losses as well as total moderator loss.

Silica Gel

Silica gel was used, since start up, to determine the amount of moderator that was lost from the reactor stacks (Miller 1956). The description of the procedure for measuring moderator loss to the stack was given in Du Pont (1954), dated July 23, 1954, and is presented below.

Moderator loss was to be measured three times a week during the day shift, with other samples taken as required if high moderator loss was indicated. Moisture from the exhaust stack was absorbed on the silica gel desiccant during a sampling period. The gel was desorbed, the water collected, and tritium content determined (counted). Environmental releases of tritium were monitored at the +148-ft stack sampling point. The silica gel sampling line was left in place for 3 hours to collect the required 25 g of water. On dry winter days, more time was needed because of the relatively low humidity in the stack exhaust. This confirms that the level of humidity in the

outside environment did have an impact on the humidity levels in the stack at the sampling locations.

The temperature and humidity that was needed for this calculation was read three times during the sampling period using a hydrothermograph in the air supply duct at the +33-ft level. The average of these values was used. The health physics laboratory extracted water from the silica gel and submitted the samples to the 100-R laboratory of D_2O (moderator) and P-10 analysis. Although P-10 was not defined in Du Pont (1954), *RAC* believes that "P-10" in this document refers to tritium, based on the procedure outlined in this and other documents from the 1950s (Anonymous 1950). The procedure called for the laboratory to determine the difference between the sample and natural abundance of D_20 (Du Pont 1954). The airflow rate values presented in Table E-2 were used to calculate tritium released depending on sampling location.

As part of the procedure given in Du Pont (1954), a calculation sheet (Table E-3) was provided to calculate the loss of moderator to the stack. Using the temperature and humidity measurements (three per sampling period), the absolute humidity was determined using a chart provided with the procedure. If the chart was in grains water per pound air, the value was to be divided by 7000 to convert to pounds of water to pounds of air. If the chart was in units of moles water per mole of dry air, then the value was to be multiplied by 0.62 to obtain pounds of water per pounds of air (Du Pont 1954). The sample analysis (in mole percent) minus the natural abundance of D_2O (0.0147) was multiplied by 1.11 to provide weight % D_2O in the exhaust air. This value times the absolute humidity (pounds H_2O per pound air) by ventilation airflow (pounds air per day) divided by 100 gave the moderator loss per day (pounds per day) (Du Pont 1954). Table E-3 summarizes these steps.

Table E-3. Area Procedure P–Moderator Loss to Stack Data and Calculations[a]

Column	Data
1	Date and time sampling of air sample point
2	Temperature (°F)
3	Relative humidity (%)
4	Absolute humidity (lb water lb^{-1} air) A[b]
5	Ventilation airflow (lb air d^{-1}) B[b]
6	Sample analysis—lab result (mol %) C[b]
7	Sample analysis—wt % above natural 1.11 (C–0.0147) = D[b]
8	Moderator loss (lb d^{-1}) $E = A \times B \times D/100$[b]

[a] Source: Du Pont (1954).
[b] Letters correspond to columns on data sheets.

The amount of moisture a silica gel monitor could absorbed was limited, and the monitors could become saturated. When the monitors were saturated, moisture (and therefore tritium) would be lost if the sample was not changed when needed.

Kiger (1955), in presenting the use of a better method (the dehumidifier), states that the silica gel monitor has been "very erratic and non-reproducible with little correlation to system losses as determined by process water inventories." The dehumidifier set-up did not have the limitation of becoming saturated. Kiger (1955) presents 6 days of data from R-Reactor using the dehumidifier method and P-Reactor using the silica gel method. Although the data presented show the silica gel to have produced a much wider range of daily data, they cannot be used to

make a quantitative judgment of the accuracy of the dehumidifier method because of cross-contamination of samples results from using the sample bottles to sample high purity moderator. It is not clear whether the reported tritium losses from the reactors were derived from the dehumidifier or the inventory method.

Dehumidifier

In 1955, dehumidifiers were first used in the R-Reactor to measure tritiated water vapor (HTO) releases from the reactor facilities (Kiger 1955). No specific date of the transfer from the silica gel method to stack dehumidifiers was found, but it appears that they were used after the beginning of 1955. Reports indicate that dehumidifier use continued until the shutdown of the reactors, although other systems were installed over time. Dehumidifiers appear to have been the primary monitors for tritium releases (Murphy et al. 1991). Air effluent monitoring by the dehumidifier method also was used to monitor areas other than the reactors, such as daily stack dehumidifier sampling of the 772-D, 420-D, and 421-2D stacks in the 400-D area, (Westinghouse 1991a).

The methodology for the dehumidifier was essentially the same as for the silica gel sampler, except that the moisture (and, therefore, HTO) was condensed out of the air on a set of cold coils rather than being absorbed by desiccant. The exhaust air was passed over the cold coils and water in the exhaust stream was condensed and frozen. The collection was interrupted daily (usually at night) to thaw for about 30 minutes to collect the 60 mL of sample needed for tritium analysis by liquid scintillation counting (Westinghouse 1979). Other documents (Anonymous 1950) indicate that another method collected the condensate immediately as it dripped into a collection pan. The unit was then dried by infrared heat lamps.

In 1958, Caldwell (1958) indicated that the dehumidifier was run three times a week for 24 hours, but it was not operated during reactor outages. Therefore, it was reliable for computing releases during normal operations of the reactor (Caldwell 1958) but not necessarily during outages. Westinghouse (1982) indicates the dehumidifiers were sampled daily in 1982.

The amount of tritium released from the stack was calculated based on the relative humidity of the air in the exhaust stream, the exhaust flow rate, and the concentration of tritium in the water sample collected from the exhaust (Longtin et al. 1973; Jacober et al. 1973) (Equation E-1). Tritium concentrations in air were measurable over the range of 10^{-5} to 5 µCi cm^{-3} (Albenesius and Meyer 1962). The assumption was made that no significant incidents occurred while the dehumidifier was not running.

Health Physics Standard Operating Procedure (DPSOP) 193-HPP 241 (3/16/65 and 7/14/65) (Du Pont 1965) describes the method used to calculate the tritium activity loss in stacked air from the 100 Areas using the dehumidifier samples. The stack losses were determined using Equation (E-1):

$$H_3\ (\text{Ci}) = (H)\ (A)\ (\text{days/month})\ (K)\ (D) \qquad \text{(E-1)}$$

where

H_3 = tritium discharged from stack (Ci)

H = average absolute humidity of stack air (grains lb^{-1})

A = tritium in dehumidifier sample (average daily) (µCi HTO L^{-1})

K = conversion factor; obtained from a calibration curve of K factor related to the air flow in cfm (converts μCi to Ci; cfm [from the x-axis of the curve] to cfd [unit retained in conversion factor]; and grains lb^{-1} to l ft^{-3}) (Du Pont 1965)

D = air density correction factor (this factor was not introduced until the 3/16/65 version of the DPSOP): below 58.5°F (14.5°C) = 1.035; 58.5–77°F (14.5 to 25°C) = 1.000; above 77°F (25°C) = 0.967.

Stack airflow for normal operating conditions in the reactors was indicated to be 110,000 cfm ±20% and for shutdown conditions usually 350,000 cfm ±20% (Du Pont 1965). It appears that the stack flow was not directly measured because the same exhaust air flow chart was used for many years.

In later published procedures, these same methods seem to have been adapted (Westinghouse 1979). However, the average airflow for the exhaust stream appears to be determined based on measurements at the time the air sample was obtained (this was usually the average flow for the day). The grams of moisture per unit of dry air was determined using a computer program.

In Westinghouse (1979), we see the dual use of the dehumidifier results to determine both moderator loss (pounds per day) and tritium released up the stack. The moderator loss was determined using the tritium data for the vapor in the air sample and in the moderator using Equation (E-2). This was independent of the type of tritium monitoring method used.

$$\text{Tritium loss per day } (D_2O\ d^{-1}) = F \times W \times 0.0168 \times \text{T-ratio} \qquad \text{(E-2)}$$

where

F = exhaust stream ($ft^3\ min^{-1}$)

W = moisture content (grains lb^{-1})

T-ratio = tritium concentration in the water extracted from the air sample by the dehumidifier, divided by the tritium concentration in the moderator

0.0168 = conversion of $ft^3\ min^{-1}$ × grain lb^{-1} to lb d^{-1}.

The "T-ratio," which is the ratio of the tritium content of the water condensed from the air sample to the tritium content in the reactor moderator (the most recent value known), was introduced. The tritium values are obtained from laboratory analysis of the water collected (A in Equation E-1). Using these data, the loss of D_2O (in pounds per day) was calculated for the +148-ft elevation stack exhaust, 0-ft level, –40-ft level, and the purification exhausts (see Figure 4.1-1 for air flow schematic of a reactor). The results of the +148-ft elevation were used to determine the total releases of tritium from the stack. The next step was to use the same laboratory analysis of tritium in the dehumidifier sample to calculate the tritium released to the stack. DPSOP-105-1886 PLCK (Westinghouse 1984a) outlines the procedures for analyzing the amount of tritium in the moderator. The water samples were sent to 735-A for tritium analysis (Suttinger and Burgo 1991). The conservative estimate of the error in determining tritium in the water sample because of equipment counting statistics is 5%. The precision of the tritium level in the sample at 0.038 μCi mL^{-1} was 0.8% relative at the 95% confidence level (Smith 1989).

Although the dehumidifier monitoring method was used until 1986, other methods eventually were used simultaneously, and the newer methods were probably the primary sources

of atmospheric tritium release data after they were installed. DPSOL 105-1878-PLK (Westinghouse 1979) determined the loss of D_2O from the STM (which was a more modern monitor that was put into use in 1970 and is described in a later section) rather than laboratory analysis of the dehumidifier sample, even though the dehumidifier sample results were still reported on the data sheets. Later, the data sheets showed that the STM was used to determine the amount of tritiated water vapor that was released.

The final step of this procedure was to calculate the tritium loss in curies per day for the +148-ft elevation stack using the tritium value from the water condensed from the exhaust stream or the STM measurement. This same procedure was followed whether the tritium release data were from the dehumidifier; the STM in DPSOL 105-1886 PLCK (Westinghouse 1984a); or the latest monitor type, the BTM (which was put into use in 1988 and is described in a later section) in DPSOL 105-1887-PLK (Westinghouse 1988). Data sheets from 3 weeks in 1990 (Westinghouse 1979, 1990b) for one of the reactors (P, L, or K) include BTM sample results as well.

We have found no indication of how often the tritium concentration in the moderator was measured. Intermittent monthly data was found over the years (Lee 1998 - these data are included in Chapter 4.1). During the late 1950s, at the beginning of reactor operations, the tritium concentration gradually more than doubled over the course of 15 months. In the 1980s and 1990s, the monthly values stayed virtually the same. Additional evidence found on data sheets from the 1990s (Westinghouse 1990b) shows the value did not vary during the month.

Kanne Chambers

A method to compute releases of radioactive gases from the 105 building (reactor) stacks was required to compensate for the drawbacks of the dehumidifier (e.g., inactive during reactor outages) and the moderator inventory methods (e.g., differentiating between atmospheric releases and other types of moderator losses) (Caldwell 1958). The Kanne chamber (ionization chamber) was considered a good monitor for use following reactor shutdowns or when activity increases in the stack air were known to result from tritium releases rather than other noble gases. Also, a method was needed that could measure tritium gas and vapor online in the Tritium Facilities where the releases were not predominantly tritiated water vapor.

Ellett and Bulter (1963) indicated that the only practical method of monitoring stack exhausts for tritium was an ion chamber called the Kanne chamber. Kanne air monitors were used as permanently mounted instruments for air monitoring. Two sizes of ionization chambers were used (51.5 and 18.5-L), and the current was amplified and measured with a Beckman micromicroammeter or a vibrating reed electrometer. The measurements were continuously recorded. Longtin et al. (1973) indicated, without providing reasons, that use of the smaller 18.5-L stainless steel chamber was preferable to the older 51.5-L aluminum chamber. A schematic of the Kanne chambers can be seen in Figure E-1.

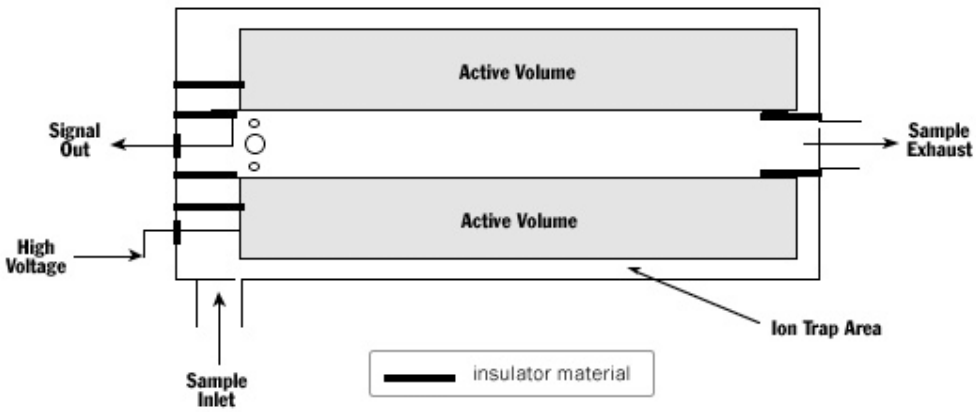

Figure E-1. Functional layout of a 18.5 and 51.5-L Kanne chamber (Litrell 1989).

The ionization chambers that were used consisted of a high-voltage electrode and a collector electrode, each separated from ground by a high-resistance insulator (Teflon). The current was generated as a tritium atom emitted a beta particle in the chamber. As the beta moved through the surrounding gas molecules, electrons were knocked off to form positive ions, and the electrons quickly formed negative ions. The positive and negative ions migrated toward the opposite charge elements in the chamber, creating a flow of current that could be measured by an electrometer or a picoammeter (Litrell 1989).

A linear relationship exists between the concentration of tritium in air and the current from the Kanne chamber if enough potential is present to collect all of the ions formed by radioactive decay of tritium in the chamber. Therefore, it is possible to estimate the active volume of an ion chamber to calculate the conversion factor of microcuries per cubic centimeter per ampere. By introducing a known amount of tritium, this factor can also be determined. Calibration curves were provided for each chamber size, and once determined for a type of ion chamber, this value will not change. It will be the same for identical chambers. The response of the 51.5-L chamber was 2×10^7 μCi/mL/ampere and the response in the 18.5-L chamber was 6.85×10^7 μ Ci/mL/ampere. The measured accuracy was quoted as ±10% for low energy beta-emitting radioisotopes such as tritium. An external gamma source can also be used to check the ion chamber, so it is not necessary to use known concentrations of tritium in air to calibrate the chambers (Longtin et al. 1973; Litrell 1989).

Reactor process stack releases were monitored by Kanne chambers at an elevation of +148 ft (Hoy 1961). Here, the Kanne chamber detected releases to the atmosphere while a dehumidifier method was used to measure the releases quantitatively. An alarm was set to signal at a

predetermined release level. Because of noble gas activities present in the stack effluent, the Kanne does not give a quantitative measure of tritium released.

The Kanne was installed at the +50-ft level in buildings 232-H and 234-H. The basic configuration of the Kanne chamber in Buildings 232-H and 234-H can be seen in Figure E-2. An air sample was drawn continuously through the ionization chamber. Hoy (1961) indicates that at SRS, the flow rates of the Kanne systems were set between 2 and 4 ft^3 min^{-1}.

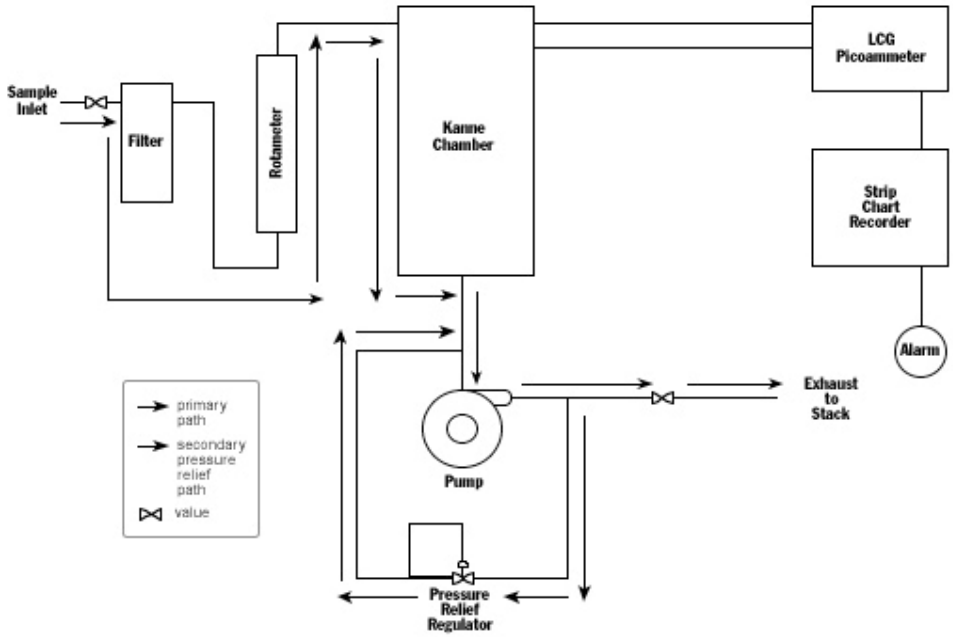

Figure E-2. Typical Configuration of the Kanne chamber monitor (Litrell 1989).

Difficulties in operating Kanne chambers included high gamma background fields and high noble gas activity in the air sample. Kanne chambers are affected by background radiation and cannot differentiate between radioisotopes (noble gases versus tritium releases in the reactor stacks). This caused problems with using them in the reactor but not for the Tritium Facilities stacks. Another drawback was that their sampling capacity was quite small. However, the great advantage of the Kanne ionization chamber was that it provided continuous sampling at a sufficient flow rate to provide real-time information. It was also found that the internal surfaces of the chamber could experience tritium buildup that required decontamination (Longtin et al. 1973; Jacober et al. 1973). Consistency of the results using this method could be affected by system pressure (Litrell 1989).

Stack calibration, rather than only checking the monitor itself, was carried out in the Receiving Basin for Offsite Fuel (RBOF) and the four stacks in the Tritium Facilities. This was done by releasing spikes of about 100 Ci (in Building 238-H only 20 Ci was released because of the much lower height of the building stack) through process hoods into the system (Davis 1977). The results, presented in Table E-4, show that the Kanne did an adequate job detecting the tritium released.

Table E-4. Calibration Test on the 200-H Tritium Facilities Exhaust Stacks Performed during Late February and Early March 1977

Date	Stack	Tritium released (Ci)	Release detected by stack Kanne (estimated) (Ci)
2/28/77	234-H	110	105[a]
3/3/77	232-H Lines I & II	104	104
3/15/77	232-H Line III	100	97
3/17/77	238-H	20	22

[a] 234-H stack integrator was online during the test to compare results. It registered 100 Ci.

In 1989, the calibration methods for the ionization chambers that were used at SRS for measuring tritium in air were reevaluated. This is important because it may indicate that the calibration factors used during the earlier years were incorrect. Table E-5 lists the documented calibration values that have been used since the late 1950s. The 1989 study confirmed that the factors being used at that time were adequate. Reviewing the factors used through time showed that there was a small decrease in the values. This change does not appear to be significant (Litrell 1989). If corrections were to be made to the tritium release estimates based on this information, it would be to decrease the earlier estimates slightly. The propagation of errors in the calibration factors used for the ion chambers was discussed in Metlin (1989). Metlin's calculations show that for the 51.5-L ionization chamber, the measurement uncertainty results in a calibration uncertainty of 2.3% of the reading; for the 18.5-L ionization chamber, it is 2.0% of the reading; and for the high volume chamber, it is 2.6% of the reading.

Table E-5. Documented Calibration Factors for Three Types of Kanne Chambers

Year	High range (μCi cm^{-3} A^{-1})	18.5-L Kanne (μCi cm^{-3} A^{-1})	51.5-L Kanne (μCi cm^{-3} A^{-1})
1959[a]		6.85×10^7	2×10^7
1960s[b]		6.25×10^7	2×10^7
1971[a]	3.3×10^9		
1974[c]		6.25×10^7	2×10^7
1988[a]	3.2×10^9		
1989[a]	3.152×10^9 ±4.3% at 3σ	6.319×10^7 ±4.0% at 3σ	1.866×10^7 ±4.1% at 3σ
1993[d]	3.15×10^9 ±11.5%	6.32×10^7 ±11.5%	

[a] Source: Litrell (1989).
[b] Source: Du Pont (1965).
[c] Source: Westinghouse (1974).
[d] Source: Westinghouse (1993).

The discussion of Hoy (1961) on the calibration of ionization chambers used at the SRS for measuring tritium in air is useful for evaluating the uncertainty of the Kanne monitoring results. The following numbered list summarizes the main points:

1. During **calibration**, tritium absorbs on to the walls of the calibration system and there is a loss of tritium in the chamber. The calibration factors are not corrected for this, so the curves used provide pessimistic or higher tritium values. The article indicates that for the larger volume Kanne, there could be up to an 11.5% loss to the walls, and in the smaller chambers, a 28% loss compared to calculated responses.
2. The mixture of other beta-emitting gases in the sample can also cause an error in the measured values. The results are based on the average beta energy versus response of the system, which would be increased by the presence of increased beta emissions.
3. **Pressure and temperature** variations (flow rate) can cause errors (in 1961, the sample flow rate was set at 2-4 ft^3 min^{-1}). *Stack Monitor Integrator System Pressure Factor Calculation* (Westinghouse 1989a) indicates that the Kanne chamber data are based on the chamber operating at atmospheric pressure. This procedure was developed in the late 1980s to calculate a pressure factor. Unfortunately, it does not explain how it was applied or how much difference it makes.
4. **Humidity** up to 96% seem to be OK. If condensation occurs in the instrument, there is a problem and it must be dried out. The determination of absolute humidity and temperature might also be important factors in determining the uncertainty of these measurements
5. If the air is filtered, there seem to be few problems with contamination in the sample; however, residual activity may build up. This may require air purging.

We considered theses influences when conducting the uncertainty assessment (discussed later in this chapter).

Manual Integration of Stack Recorder Charts

Initially, the Kanne chamber results were recorded on a paper strip chart that was manually integrated by the health physics staff. The height of the trace on the strip chart was proportional to the radioactivity in the airflow. The area under the trace, minus the background, was related to the radioactivity release. The Kanne would measure tritium and ^{41}Ar, so this method was best used when the source of the activity was known. The tritium releases from the stack were computed using the Kanne recorded chart as follows in Equation E-3 (Caldwell 1958):

$$Q = c\,[\,\Sigma\,(k_s A_s) - a_{bg} T] * 1.7 \times 10^6 \qquad \text{(E-3)}$$

where

Q	=	amount of tritium (Ci)
T	=	the release time period (taken from chart) (hours)
c	=	the air exhaust rate during time, T. This may be computed from the rated capacities of the exhaust fans (ft^3 m^{-1})
a_{bg}	=	background tritium air activity (obtained by purging the Kanne with clean air) (Ci cm^{-3})

A_s = area between trace and zero axis on recorder chart generated during time period, T, during which its amplifier was set at a particular setting, s (sensitivity setting of the amplifier) (in^2 or % of scale x in)

k_s = proportionality constant depending on s, the chart speed, and chamber calibration (see Table E-6)

$\Sigma (k_s A_s)$ = sum of all products of k_s and A_s for each amplifier setting, s, that occurred during time period, T.

1.7×10^6 = conversion of time and area units.

Caldwell's (1958) procedures for the reactors indicated that the area measurements A_s could be carried out using one of the three methods:

1. Straight-line trace method, where the chart is sectioned into rectangles or trapezoids and the area of each section is added to get the total.
2. Approximate integration method, where the sections are created at equal lengths along the time scale (base section); the heights of the right side of the sections are added and multiplied by the length of the time section.
3. Equal area method, where within a section, a line is drawn through the trace so that about the equal amounts of area are enclosed below and above the rule. The area between the zero axis and the drawn line is determined by the height of the line at the midpoint of the base (time) section; these sections are summed.

Table E-6. Summary of Proportionality Constant, k_x, Chart Speed, and Chamber Calibration[a]

s		k_S	
Beckman settings (A)		% of scale measurements	inch measurements
I A	3×10^{-13}	3×10^{-14}	2.7×10^{-13}
II A	10×10^{-13}	10×10^{-14}	9.0×10^{-13}
II B	3×10^{-12}	3×10^{-13}	2.7×10^{-12}
II C	10×10^{-12}	10×10^{-13}	9.0×10^{-12}
II D	3×10^{-11}	3×10^{-12}	2.7×10^{-11}
III A	10×10^{-11}	10×10^{-12}	9.0×10^{-11}
III B	3×10^{-10}	3×10^{-11}	2.7×10^{-10}
III C	10×10^{-10}	10×10^{-11}	9.0×10^{-10}
III D	3×10^{-9}	3×10^{-10}	2.7×10^{-9}
IV A	10×10^{-9}	10×10^{-10}	9.0×10^{-9}
IV B	3×10^{-8}	3×10^{-9}	2.7×10^{-8}
IV C	10×10^{-8}	10×10^{-9}	9.0×10^{-8}
IV D	3×10^{-7}	3×10^{-8}	2.7×10^{-7}

[a] Source: Caldwell (1958).

If a logarithmic amplification system was used the following relationship (Equation E-4) was used (Caldwell 1958):

$$Q = c\,[\,k_e \Sigma A_e - a_{bg}T] * 1.7 \times 10^6 \qquad \text{(E-4)}$$

where, in addition to the parameters defined above:

A_e = effective area below the trace in a section of the chart. The height of the area is taken as the ampere reading of the trace (A) and the base is measured inches (in)

k_e = chamber calibration $2.0 \times 10^7 \mu Ci\ cm^{-3}\ A^{-1}$

ΣA_e = sum of all A_es during the tritium release of interest.

The area measurements could be made either as a constant activity trace (so it was a rectangle) or using the approximate integration method described above (referred to as the variable activity trace) (Caldwell 1958). Stack recorder charts for the Tritium Facilities (B232-H, 234-H, and 238-H) Kanne chambers were manually integrated to determine the tritium releases from the stacks. In later years, when the electronic SMI was being used, it was used to verify that the SMI and FORMS monitors (monitor used to determine the ratio of tritium vapor to tritium gas) were working correctly. The stack recorder charts were integrated for each shift at the beginning of the next shift. Special integration rulers were used for each stack to take into account the different airflow characteristics (Westinghouse 1994, 1991c).

The integration rules that were specific to the Tritium Facilities are summarized as follows (Westinghouse 1994, 1991c):

1. If the release rate is <100 Ci min^{-1}, use one of the following:
   - use the constant rate
   - average the rates measured at the beginning of minute and the rate at the end of the minute
   - if rate increased and decreased, take into account the portion of the minute involved in the increase and decrease to use 1(b) and then add the portions together for the total 1-min release.
2. If the release rate is >100 Ci min^{-1}, break the minute into three parts, determine the amount released during each third, and then take the average to determine the release for the 1-min period.
3. The actual activity release in all peaks must be corrected for stack background. This is done by multiplying the background by the time in minutes for each operation or peak shown on the chart and subtracting the background reading for the period from the uncorrected release totals determined in Steps 1 and 2 above.
4. All peaks must be identified and attributed to the process that would have caused it.

Stack Tritium Monitor

In the early 1960s, a STM system was developed by Du Pont engineers at the SRS (Ray 1991). The systems used up until this point were neither adequate for measuring rate and release or time of release (the dehumidifier) nor could they differentiate between tritium and other radioactive gases (the Kanne chamber). This new method was designed to allow for measuring tritium online in the presence of noble gases by combining the dehumidifier with a compensated ionization chamber (Kanne chamber). A prototype of the system was described in 1961 using a vibrating reed electrometer, an Esterline-Angus recorder, a control circuit, and a mechanical

counter to record the amount of tritium released (Kincaid 1961). The initial version is shown in Figure E-3, and an updated version, put to use in later years, is shown in Figure E-4.

In August 1965, the STM description was slightly different than the prototype developed in 1961. The stack gas sample was split in two: one-half going directly to a 4-L ion chamber measuring elemental tritium, tritium oxide, and noble gases, and the other half passing through a cold trap first for the removal the tritium vapor (HTO) and tritiated water (T_2O) before flowing through another identical ion chamber measuring elemental tritium and noble gases. The resulting current was proportional to the difference in activity between the two chambers, which is equal to the tritium activity removed by the cold trap, thus, allowing for an estimate of the amount of tritium oxide released (Rampey 1965). This method eliminated background and noble gas interference, but it measured only tritium in the form of HTO. This could be corrected if a catalytic recombiner cartridge was inserted ahead of the moisture trap to convert the T_2 to HTO and, therefore, include it in the measurement (Longtin et al. 1973; Jacober et al. 1973).

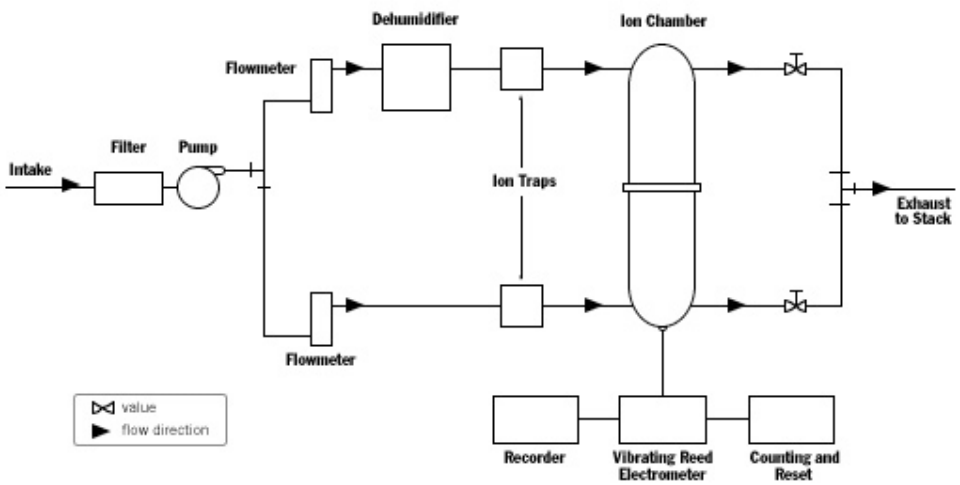

Figure E-3. Configuration of the early stack tritium monitor (Rampey 1965).

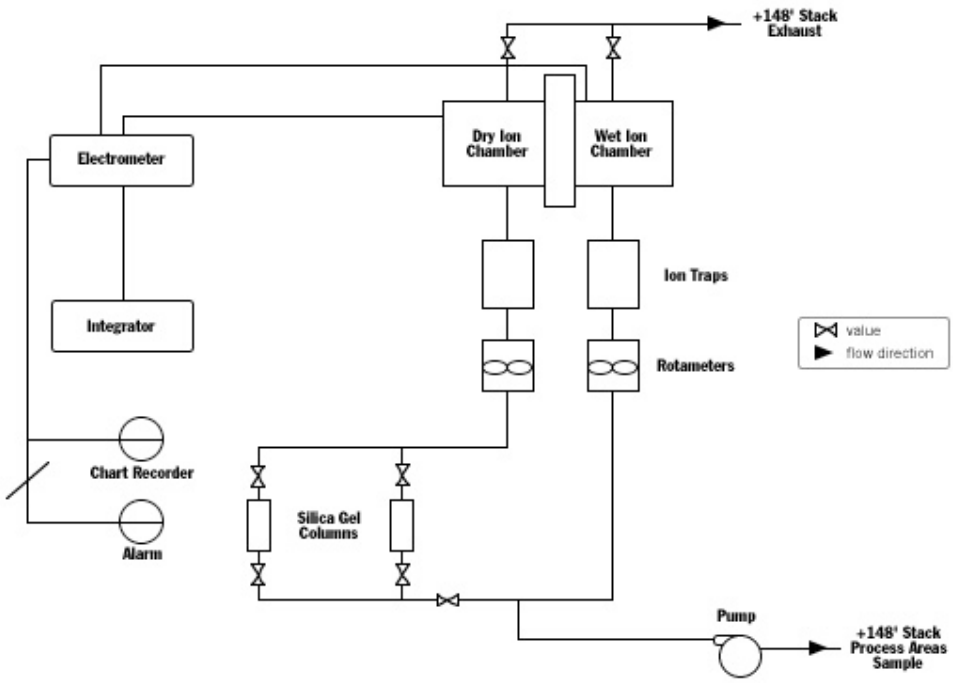

Figure E-4. Configuration of the later stack tritium monitor (Westinghouse 1990c).

Two Kanne ionization chambers were used, and the sample of exhaust gas flowed continuously through the chambers. A Beckman logarithmic micromicroammeter measured the current that was produced. A digital computer was used to integrate the ion chamber current data. The output signal from the micromicroammeter was converted to pulses, with the frequency of the pulses proportional to the rate of discharge of tritium. The computer automatically integrated this data and allowed for integration of tritium releases for up to 1 month. The Kanne chamber and integrator system covered a range of tritium concentrations from 6×10^{-6} to 60 µCi cm^{-3}. Tests showed that at the levels of 25 to 130 Ci of tritium, the accuracy for the monitoring system was better than 10% (Albenesius and Meyer 1962).

The minimum detectable tritium level in air when there were no other radionuclides present was 0.5×10^{-5} µCi cm^{-3}. Therefore, the minimum detectable level in 105 buildings stack gas, when other noble gases were present, was reduced to 1.0×10^{-5} µCi cm^{-3} at full reactor power (Rampey 1965). The STM could be used to monitor stack exhaust during normal operations and shutdown. One disadvantage was that if there were a partial core melt accident, the higher levels of releases would render the system useless.

The stack monitor operated under slight positive pressure so that if there was a leak, air would not dilute the gas sample. Direct readings of gas stream activity could also be monitored. There might have been an increase in response time, but pulse indications caused by changes in noble gas activity were prevented (Rampey 1965). All components (except for the dehumidifier) were mounted in a 4-ft rack cabinet. The prototype of this instrument was demonstrated in 105-K (Rampey 1965).

The amount of tritium oxide released from the exhaust stream was calculated using Equation (E-5) (Rampey 1965):

$$C = K_1 N \tag{E-5}$$

where
C = curies tritium oxide released (Ci)
K_1 = constant determined from the stack monitor calibration and exhaust stream flow rate (Ci/counts)
N = number of counts recorded.

On the daily data sheets, K_1 was determined from the use of a Keithly Amplifier Factor based on the multiplier range, and instantaneous calibration factor based on field calibration, K_2, and the air exhaust rate (Westinghouse 1984a, 1990c).

The rate of tritium released was determined by Equation E-6:

$$R = K_2 / t \tag{E-6}$$

where
R = rate of release (Ci h^{-1})
K_2 = constant determined from the stack monitor calibration and the exhaust stream flow rate (Ci)
t = time required for count (h).

Although the STM was able to discriminate tritium from other radioactive noble gases, it was only effective for monitoring tritium oxide. It was insensitive to elemental tritium, which was why it was used in the reactors and not in the Tritium Facilities. Another drawback of the STM was that it was more difficult to calibrate because tritiated water had to be handled.

In later years, the cold trap that was used to remove the tritiated water vapor was replaced by a 60-in.^{3} silica gel drying column. The silica gel provided a more efficient method for removing water than the cold trap, which fluctuated with changes in relative humidity and temperature (Smith 1989). An ion trap was also added and set at the inlet of each of the ion chambers to remove ions produced in the gas outside the chambers that might disrupt the proper tritium measurement. These traps consisted of 14 parallel plates with a potential difference of 70 V between adjacent plates (Smith 1989).

Stack tritium monitoring DPSOLs (Westinghouse 1984a, 1984b) indicate the monitors were calibrated annually, when required by supervision, or when there was a question about the accuracy of the existing calibration factor. The calibration documentation provides equations to calculate the rate of drift of current measurements; a calibrations procedure print list; and the recommended initial installation procedure for prototype design, including routine operation and maintenance (Westinghouse 1984a, 1984b). It was not easy to calibrate the equipment because tritiated water had to handled by the technicians (Merz 1988).

A strip chart was used as an integrator/recorder of the average daily moderator loss rate. With the strip chart, it was possible to determine the instantaneous loss rate at anytime (Smith 1989). The total releases were integrated manually.

In 1965, releases from the STM and the existing dehumidifier in the reactor stacks were compared during normal operations (Rampey 1965). Table E-7 provides the data. The releases are for a 24-hour sampling period ending at 2 p.m.

Table E-7. Stack Monitor and Existing Dehumidifier System Estimated Releases during Normal Operations of a Reactor in 1965[a]

Day	Estimated release ($Ci\ d^{-1}$)	
	Stack monitor	Dehumidifier
Wednesday	218	336
Thursday	221	372
Friday	211	413
Saturday	162	341
Sunday	156	346
Monday	152	321
Tuesday	240	390
Wednesday	240	320

[a] Source: Rampey (1965).

Although the trends seem to be the same for this reported comparison, the STM release estimates are, on the average, about 40% lower than the dehumidifier estimates. An illustration (Rampey 1965) of the hourly rate of loss over 5 days shows a variation of up to a factor of 2 (i.e., 6 to 12 $Ci\ h^{-1}$) over the course of 1 day. However on some days the release rates were more uniform (Rampey 1965). If the STM releases are about 40% lower than the dehumidifier estimates, the tritium release estimates based on the dehumidifier might have overestimated the actual tritium releases from the reactors. Lee (1998) reports data that indicate that the STM may have overestimated releases compared to the newer BTM; this would support the view that the STM overestimated the results rather than the dehumidifier. Insufficient evidence exists to draw a conclusion.

In July 1972, a memo was written to recommend a few changes to the newly installed STMs in each of the three reactor buildings (Ray 1991). The instruments were designed based on the prototype that was operating in K-Area, and there had been some problems in installation. The recommended changes were mainly to find better locations for equipment and improvements on instrument piping. We did not find documented evidence about how these changes might have improved measurement efficiency.

By 1991, it was recommended that the three remaining STM systems be replaced by a new BTM system for the following reasons:

- The STM chambers required extreme care and cleanliness in assembly to ensure that leakage current was not greater than the signal current.
- Condensation in the “wet” chamber could cause slight negative readings; the amplifier must operate in “fast” mode and is noisy; therefore, there was a slower response time.
- Releases to the environment would be underestimated if the recorder went offscale during a major reactor incident because of inaccessibility of the instrument.
- The amplifier was sensitive to temperature and vibrations that caused significant error.

- STMs were difficult to keep calibrated. The STM did not require mechanical calibration, but it required a "calculational" calibration using two factors: instantaneous (K_2) and daily loss rates (K_3) of process water, where $K_3 = K_2/60$. The K_2 was dependent on the response of the specific STM and the tritium concentration in the moderator; therefore, known quantities of tritium needed to be introduced into the chamber. This factor should be fairly constant, but historical data from the reactor areas do not support this (Table E-8). The error in the estimates of tritium releases would vary with the error of these factors (Ray 1991).
- These factors should be fairly constant over time for a particular STM in one of the areas (Ray 1991). If the calibration factors were off by 100%, the moderator loss rates and tritium releases rates would be incorrect by the same factor.
- The STM and BTM loss rate calculations were required to yield values within 25%. However, for low release days (during shutdown periods), the losses were below the STM sensitivity; therefore, it was not possible to test the BTM.
- There was a greater uncertainty of the STM-calculated moderator loss rates because of poorer counting statistics under conditions of high noble gas concentrations.

Table E-8. Calibration factors (K_2) for Stack Tritium Monitors in P-Area from 7/87 to 3/90

Date	Stack tritium monitor A	Stack tritium monitor B	Stack tritium monitor C
3/8/90	0.00452		0.00286
2/26/90		0.00156	
2/23/90			0.00179
2/20/90	0.00167		
2/11/90		0.00266	
2/9/90	0.00417		0.0125
1/22/90	0.00146		
1/10/90			0.00175
1/8/90		0.00172	
1/4/90	0.00106		
12/22/89	0.00490	0.00325	
11/15/89	0.00321		
11/6/89		0.00240	
8/3/89	0.00356		0.00128
7/3/89		0.00428	0.00179
6/18/89	0.00277		
5/18/89			0.00696
5/17/89	0.00229	0.00368	
5/12/89		0.00506	
5/11/89	0.00126		0.00940
4/15/89	0.00185	0.00600	0.00200
3/8/89		0.00183	
10/28/88	0.00229		0.00032
7/30/87	0.00078	0.00151	0.00087

Berthold Tritium Monitor

During the late 1980s, BTMs were added to monitor the moderator and tritium releases from the reactor areas. The first BTM was installed in L-Area in 1985 (Ray 1991). The BTM was developed because the threshold sensitivity of detection for the STMs was too low to detect the small stack moderator losses expected from the restarted L-Reactor, and the BTM could discriminate between the tritium and the radioactive noble gases released from the stack. The tritium activity in the moderator of L-Reactor was 175 times less than the activity in the P-Reactor and K-Reactor. As mentioned earlier, the tritium concentration in the moderator increased with reactor operation time, so a newly started reactor had much lower tritium releases. With such low activity, the existing STM was unable to detect a moderator loss of less than 2000 lb d^{-1}. The newly developed BTM was able to detect losses of tens of pounds per day. It was first installed in 105-L (Merz 1988). The BTM was going to be the main source of monitoring stack moderator loss rates in the +148-ft stack in L-Area and the dehumidifier would provide a redundant (or backup) measurement.

The BTM system was comprised of a tritium monitor and a radiation monitoring system, along with associated flow monitoring and control devices (Figure E-5). The gas flow proportional counter could discriminate between tritium and noble gases using two single channel analyzers employing rise time discrimination methods. The sample air was mixed with counting gas before it passed through a 1.3-L gas flow-through proportional counter. (In this chapter, the counting gas is called P-10. It is composed of 10% methane and 90% argon. This appears to be different from the P-10 referred to in documents from the 1950s, which appear to be referring to tritium gas).

The lower energy tritium beta particles, with a short range, interacted with the counting gas in a limited volume in the counter tube. The noble gases in the exhaust air reacted in a larger volume of the tube because of higher beta particle energies. These higher energies could be discriminated by using two single channel analyzers because of the difference in the pulse rise times (Merz 1988). Pulse rise time discrimination depended only on the energy of the emitted beta particle, not the chemical composition in which the isotope of interest was found. The BTM was equally useful in monitoring elemental tritium as tritium oxide.

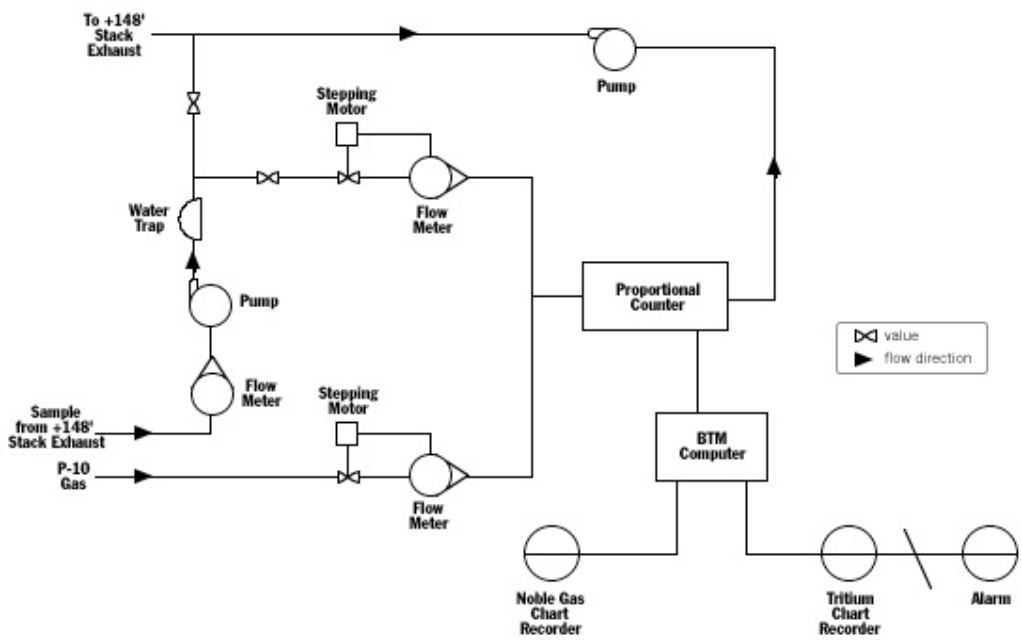

Figure E-5. Schematic of the Berthold tritium monitor (Merz 1988).

The accuracy of the tritium measurements from the BTM depended on the ratio of exhaust air to counting gas mixture. A control unit regulated this ratio with a light barrier control. The measurement was accomplished using a floating body flow-through meter using photoelectronic position scanning. The detector automatically shut down if the counting gas flow was lost (Smith 1989). After counting, the gas and sample were pumped out to the exhaust air system. A water

trap was added to the exhaust air system to alleviate the buildup of moisture in the sample lines and the counter tube.

The Berthold Analytical Instruments Company quoted a sensitivity that was not achieved in the field (0.01 μCi tritium per cubic meter air or between 0.01–0.04 lb moderator per day depending on the reactor). In fact, a sensitivity of <0.5 lb d^{-1} was found with operating experience. This was attributed in part to the uncertainties in ventilation airflow rates and moderator tritium content. In calibration exercises, the BTM was able to measure tritium in the presence of a simulated noble gas background to an accuracy of <5 ppm (Merz 1988). The threshold of detection for stack releases was improved by a factor of about 200 to 10^8 μCi of tritium per cubic centimeter of air with the shift from the STM to the BTM (Westinghouse 1985; Merz 1988).

In 1988, a BTM was also installed in K-Reactor (Merz 1988), and there were recommendations to replace the existing BTMs with newer systems. It was also recommended that the STMs currently installed in each reactor building continue to be used to monitor tritium concentration of below grade, purification, and process room air. It was suggested that the use of the dehumidifiers could be discontinued because the STMs could be used as a redundant measure for the BTMs, but the data sheets we found indicate that all three systems continued to operate.

The procedure for BTM operation (DPSOL 105-1887-PKL) (Westinghouse 1988) describes how the equipment worked, and the Special Procedure SP-2381 (Westinghouse 1985) provides a Daily BTM Stack Loss Data Sheet requiring measurements at 4-hour intervals starting at 2:00 a.m. The daily stack loss (in pounds per day) was calculated by the product of the concentration of tritium in the moderator (microcuries per milliliter; weekly lab results), a conversion factor, the integrated counts in Channel B, and the average duct flow rate (cfm). Data sheets were used to calculate the instantaneous loss rate (Rp) using Equation (E-7):

$$Rp = k \times F \times Bc/T \qquad \text{(E-7)}$$

where

k = conversion factor from DPSOL 105-1885A-PLK, *BTM/STM Calibration* (Westinghouse 1989b)

F = flow in duct being sampled (cfm)

T = tritium content in moderator, μCi mL^{-1} from latest laboratory result

Bc = [Ba-Bo] – S[Ag-Ao]

where

Ba = count rate from recorder or count rate meter Channel B

Bo = Channel B background, Data Sheet 1 DPSOL 105-1885A

Ag = count rate from recorder or count rate meter Channel A

Ao = Channel A background, Data Sheet 1 DPSOL 105-1885A

S = spillover factor Data Sheet 1 DPSOL 105-1885A.

As was described in the earlier section on dehumidifiers, the calculated loss of moderator in pounds per day was used to calculate the amount of tritium oxide released to the atmosphere. However, in this case, both elemental and oxide forms were included in the release estimate.

Some BTM data were available for the newly installed equipment in P-Area from May 1988. The BTM was reported to show an accuracy of measurement of ±3%. It was suggested that a conservative estimate of ±5% be assumed to take into account 2% drift in the calibration measurement. At the time of the report, it indicated that an accuracy performance check had been shown to be adequate for every 6 months for the BTM; however, it seems that the check was done every 3 months (in accordance with DPSOL 105-1885A-PLK [Westinghouse 1989b]) (Merz 1988).

Two more upgraded BTM systems were installed in L-Area (June 1989 and July 1989) and three in KA-Areas (September 1990 and January 1991). The sensitivity quoted for the systems was 2×10^{-8} μCi cm^{-3} in 1-hour measuring time as compared to 1×10^{-6} μCi cm^{-3} for the STM (Ray 1991). In 1989, it was recommended that a third BTM be installed to increase the capabilities of the BTM system by dedicating a monitor to the 148-ft stack. This would allow increased availability for the other existing detector sampling points (Terronez 1989).

By 1991, there were three BTMs in each reactor area monitoring the total stack, at 0-ft elevation for the process and purification areas, and at +40-ft elevation areas. Westinghouse (1991d) provides the instructions for starting up the BTMs. At anytime the process water system contained moderator, at least two tritium monitoring systems should have been operable and monitoring the +148-ft level ventilation exhaust; one of the two monitors should have been a BTM. The third tritium monitoring system should have been operable and monitoring any ventilation exhaust air streams in the 105 building. The three BTMs were connected to six ducts and the atmosphere.

STM and BTM Redundancy

The STMs and BTMs could provide adequate surveillance of the release of process water because the reactor ventilation system was designed so that the process areas were maintained under negative pressure. During reactor operation, the air from the process room, purification, and –40-ft pump room passed through the filter compartments and released to the stack. By monitoring at the exhaust stack, the combined tritium levels from the process areas were measured. By having two tritium monitors at the +148-ft ventilation stream, it was possible to switch one of the detectors to monitor different ventilation streams to determine the source of the releases. It also provided a backup to the +148-ft monitor (Merz 1988).

The STMs were not removed and remained in use because of the broader range over which they could detect tritium in the presence of other radioactive gases. At the low end, this corresponds to very small process leaks throughout the reactor building to small and medium size leaks (the type that normally occur). A very large spill of tritiated water in the reactor building could overwhelm the existing BTM system by exceeding the existing measurement range. The gas flow-through ionization chambers saturate at higher concentrations than the more sensitive proportional chambers of the BTMs. The reported BTM ranged from 2×10^{-8} μCi cm^{-3} air to 2×10^{-3} μCi cm^{-3} air for a 1-hour counting period (Merz 1988).

The results of the STM were compared to the BTM daily (at a minimum of every 48 hours). The STM calibration required that it be within ±10% of the BTM reading. A conservative estimate of the STM accuracy was ±15%, which included ±10% agreement and 5% drift error in the calibration measurement. The STM was to be calibrated every 3 months or whenever the calibration check warranted it (Merz 1988). However, data sheets for the 1990s indicate that in

many cases there were large differences between the STM and BTM results (see Table E-9). This is also true of the dehumidifier results that were reported. More of this duplicate measurement data can be found in Lee (1998).

Table E-9. Data for Tritium Releases from an Unknown Reactor (P, L, or K)[a]

1990 date off 11–11a.m.	Moderator (μCi mL^{-1})	+ 148 Moisture content (g lb^{-1})	+ 148 Tritium content in exhaust air (μCi mL^{-1})	+ 148 Dehumidifier D_2O loss (lb D_2O d^{-1})	+ 148 STM D_2O lost (lb D_2O d^{-1})	+ 148 BTM D_2O lost (lb D_2O d^{-1})	Tritium lost to the environment per day (Ci)
15-Apr	11600	70	0.40	10.0	208.3	21.4	102.5
16-Apr	11600	49	0.42	7.4	210	21.1	101
17-Apr	11600	58	0.26	5.3	198.3	11.7	51.8
18-Apr	11600	32	0.92	10.6	142.9	35.5	170.1
19-Apr	11600	56	0.88	17.9	60.3	37.5	185.9
20-Apr	11600	64	0.53	12.1	40.7	28.9	138.5
21-Apr	11600	10	0.43	1.7	33.3	22.7	108.8
22-Apr	11600	6	0.32	0.5	2.4	22.4	103.76
23-Apr	11600	6	0.36	0.75	9.6	18.4	88.2
24-Apr	11600	6	0.44	1.2	3	18.7	90
25-Apr	11600	6	0.36	0.8	0	17.9	83
26-Apr	11600	6	0.36	0.78	0	17.2	79.7
27-Apr	11600	6	0.40	0.8	25.9	17.5	81.1
28-Apr	11600	12	0.33	1.4	76.8	19.8	94.9
29-Apr	11600	11	0.29	1.1	24.4	17.6	84.3
30-Apr	11600	106	0.26	10.0	23.2	16.9	78.3
1-May	11600	38	0.24	3.4	23.9	18.1	83.9
2-May	11600	110	0.30	10.6	2.9	21	98
3-May	11600	76	0.23	6.4	30	20.5	95
4-May	11600	86	0.26	7.9	46.9	20.1	93.1
5-May	11600	84	NA[b]	NA[b]	184.1	15	71.9

[a] Source: (Westinghouse 1979).
[b] NA = indicated as not available on data sheet.

It is clear from the handwritten data sheets that the BTM data were used to determine moderator and tritium losses. The data sheets from the 1990s (Westinghouse 1990b) used the data from the BTM monitor, multiplied it by the concentration in the moderator, a conversion factor of 0.000413 (for pounds per day × microcuries per day to curies per day), and the air density correction factor (dependent on temperature). The apparent differences in the measurements introduce some doubt as to how well and often the systems were actually calibrated against each other and how the equipment was maintained.

Stack Monitor Integrator

A memo written on 4/2/68 (Du Pont 1958) announced that the prototype SMI was delivered to Building 232-F, and preliminary tests showed it performed in a satisfactory manner. The equipment was designed to eliminate the difficulties encountered while manually integrating the curve drawn by the seven-decade semi-log chart paper recorder set up to record the Kanne chamber measurements. Its features included an instantaneous display (in curies of tritium released since midnight), alarm capabilities with selectable alarm thresholds, and automatic data printouts with time and date. A current-to-frequency conversion technique was used to convert the ionization current from a Kanne chamber into electrical pulses (Figure E-6). Digital techniques were then used to process the signals. The Health Physics group suggested calibration and maintenance procedures to be implemented to test if the integrator could be used to replace manual integration.

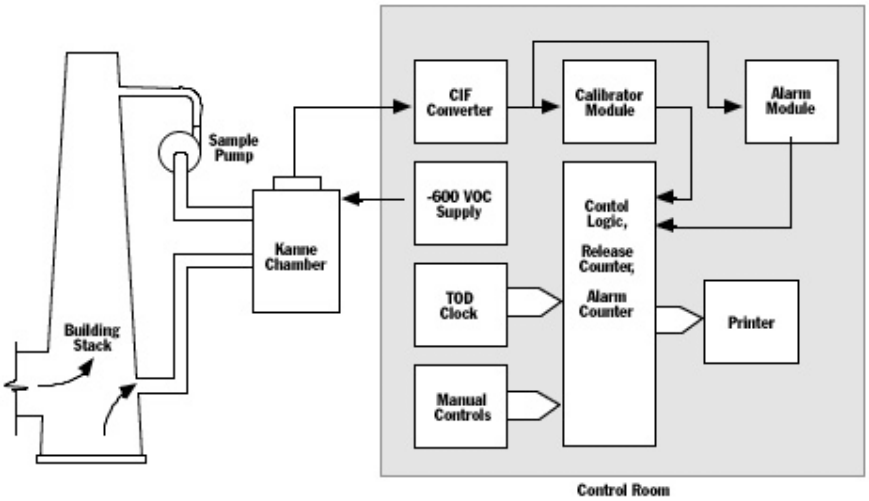

Figure E-6. Schematic of the stack monitor integrator (Reinig 1980).

In 1972, there was a request to get vendor bids to develop three SMI instruments for the current output of ion chambers in the 200-H Area. The instruments were to continuously record the output from the ion chamber and integrate it to be printed every 8 hours and reset after 24 hours. During an alarm, it was to continuously integrate the output and printout at 1-minute intervals. The accumulated reading was not to be reset, and a final printout was to be delivered at the end of the alarm condition (Kilpatrick 1975).

Another memo written 8/6/75 indicates that a prototype SMI was installed in Building 234-H in April 1974 and that field tests showed it accurate and reliable for measuring tritium releases. The long-term accuracy of the integrator electronics that was expected was better than ±5% of reading. However, the overall accuracy of the measured losses was dependent on the calibration of the integrator electronics, the Kanne chamber, and the measurement of the stack airflow. It was recommended that three SMI systems for Buildings 232-H and 234-H be fabricated for the

primary stack monitoring instruments and the existing monitoring systems be retained to keep track of the rate of releases.

A manual for the SMI dated July 2, 1980, (Reinig 1980) indicates that one integrator was installed in Building 234-H in January 1980 and two others were operating in 232-H by late 1980. This was confirmed in a Works Technical Monthly report (Du Pont 1982b). This system was compatible with a proposed FORMS monitoring system that could determine both the amount of HTO and HT released by the stacks. Exhaust airflow up the stack was indicated to be 6.31×10^7 $cm^3\ s^{-1}$. Calibration was required every 6 months.

The calculation to determine the counts per curie released is as follows in Equation E-8 (Gelsie 1981a):

$$R = [x/y] \times [z] \times [1/k] \qquad \text{(E-8)}$$

where

R = counts per curie released

x = Kanne chamber constant = 1.6×10^{-8} A $\mu Ci^{-1}\ cm^{-3}$ (for 18.1 L)

y = stack flow (232-H Line 3 = 131,000 $ft^3\ min^{-1}$ or $6.22 \times 10^6\ cm^3\ s^{-1}$; 232-H Line 1 and 2 = 104,000 $ft^3\ min^{-1}$ or $4.93 \times 10^7\ cm^3\ s^{-1}$; 234-H = 133,000 $ft^3\ min^{-1}$ or $6.31 \times 10^7 cm^3\ s^{-1}$)

z = current-to-frequency transfer function (0.984×10^{11} Hz A^{-1} except for Line 3 in 232-H, which is 0.289×10^{11} Hz A^{-1})

k = normalization constant.

Background must be subtracted from the total tritium released as displayed by the counters. It was necessary to compensate for error caused by the current-to-frequency (C/F) converter module (which linearly converted ionization current from a Kanne chamber into curies of tritium); leakage current; and Kanne chamber background. This was done by running room air through the Kanne chamber and measuring the frequency of the C/F converter. The C/F conversion period was measured automatically to within 100 microseconds by internal times. Background in terms of curies per minute was calculated from this conversion period. The normal background current was about 0.1 Ci min^{-1}. Most of the error was contributed by the C/F converter and was eliminated by subtracting background measured with the integrator (Reinig 1980). The amount of tritium released per minute was calculated per conversion period and the value of K was determined to provide the curies per minute measured by the SMI in C/F conversion. This procedure is described further in a memo dated January 20, 1981 (Gelsie 1981a).

In February 1982, the Works Technical Report (Du Pont 1982b) reported a 100-Ci test release to compare the SMI and charge recorder system in 234-H. A 4.5% difference was seen. Three more controlled release experiments were planned to continue to test the system.

A memo written in April 1981 (Gelsie 1981b) states that the SMI systems had been installed in 232-H and 234-H. A large stack release on March 27, 1981, was recorded by the SMI and provided release data that agreed with the manual integration of recorder traces within 10%. Both rate and quantity during the peak release period were recorded accurately, but there were some problems with the total display counter, which were quickly resolved.

Tritium FORMS Monitor

To monitor the form of the tritium (elemental tritium and tritium oxide form) released from the Tritium Facilities stacks, a monitor was installed in Building 234-H early in 1982. Like the STM, the monitor consisted of two Kanne chambers simultaneously measuring samples of stack effluent. One measured only elemental tritium after the removal of the oxide, and the other measured both forms of tritium (Du Pont 1982b, 1981; Jacobsen 1976).

The monitor used a pressure-swing dryer with self-regulating desiccant beds to remove tritium from the sample. A microcomputer-compatible electrometer measured total and elemental tritium. The difference between the two, as determined by the microcomputer, was available to tritium facility personnel and by telephone link. After a 1-week test in early February, it was determined that total tritium data agreed with Health Protection estimates of the stack release during that time. On February 12, a test release of 100 Ci validated the results with other measurement methods.

The gas stream sample was drawn into the sampler and mixed with tritium-free hydrogen carrier gas. The air moisture (both water and tritiated water vapor) was collected on a molecular sieve (A in Figure E-7). The elemental tritium gas was mixed with the tritium-free hydrogen carrier gas that passed through Column A. A palladium catalyst in Column B converted the hydrogen gas and tritium to oxide form. The water that was formed was collected on the molecular sieve in Column B. The moisture collected from the two columns was kept separate (samples A and B) and analyzed for tritium content. The results for Column A represented the tritium that was in oxide form and the water from Column B was the elemental tritium content in the air. The content of water samples A and B related the tritium concentration in air to the absolute humidity, air sampler flow rate, length of sampling time, and the flow rate of the tritium-free hydrogen gas.

An Air Effluent Monitoring Procedure (Westinghouse 1994, 1991a) instructed facilities personnel that the FORMS measurements should be compared with the manually integrated stack release data from the Kanne chambers and the SMI results for each shift. This comparison was made to verify that the SMI and FORMS data were within 20% of the chart recorder data. If it was not, then the health physics officer responsible for the stack release data was contacted to determine which system was in error. A description of the samplers and flow monitoring devices at SRS (Westinghouse 1991b) indicates that the FORMS and Kanne chambers operated redundantly in the Tritium Facilities stacks. However, there is no indication of which data were used to determine the reported atmospheric tritium releases.

Hold Volume Monitor—Integrator for Tritium Facilities

In June 1982, a 6500-ft^3 hold volume tank was installed in Building 300-H to contain accidental releases of tritium from Building 234-H (Du Pont 1982a). Sensors in the process hoods automatically triggered diversions to the hold volume tank. The monitoring capabilities of tritium diverted to the hold volume before that time were limited to approximately 0.05 g (about 470 Ci). A high-range ionization chamber accurately measures and integrates tritium in the range of 0.1 to 10.0 g. The monitor-integrator was designed to provide sufficient information to allow for decisions on tritium recovery after an accidental release. A C/F converter processed the signal

from the ion chamber to a totaling scale by fiber optic cable. We found only one mention of this tank in the SRS document database and no indication of when or if it was used.

UNCERTAINTY

Uncertainty analysis is an important part of a source term calculation. Because reconstructing the tritium source term at the SRS required using incomplete historical information, we used an uncertainty analysis to account for parameters that were either not completely understood or are defined in the literature with ranges, not specific values.

The uncertainty analysis combines all of the best scientific knowledge available about each parameter in a calculation to provide a distribution of results that is realistic given what we know about each variable. While a distribution is sometimes more difficult to understand, it is certainly a more accurate representation of releases than a single value.

We used the Crystal Ball® uncertainty analysis software package (Decisioneering 1996). The Crystal Ball® package is used within Microsoft Excel and allows the user to define the distribution of possible values for an input parameter about a defined mean. This assumption cell within Excel is used as a parameter in an equation cell. This equation is contained in a forecast cell. The results of the equation are calculated using a Monte Carlo analysis. A Monte Carlo simulation uses randomly generated numbers to measure the effects of the uncertainty expressed in the probability distribution in the assumption cell. A single trial generates a random number based on the probability distribution for each assumption, recalculates the equation within the spreadsheet, and displays the result of the calculation in a forecast chart. This forecast chart represents the results of the many calculations for the series of random numbers as a probability histogram. The more Monte Carlo trials run, the more continuous this histogram appears. This histogram can then be fit to a more conventional distribution, and the statistics describing that distribution are given by the Crystal Ball® software, accounting for the uncertainty in the input variables.

The results of this analysis can be used in the next phase of the dose reconstruction for SRS to estimate the range of possible releases using the source term data presented in this report. We also used the uncertainty analysis to determine whether there was a need to correct the source term data reported by the Site using different types of monitors.

Uncertainty for all monitor types was determined in generic terms. That is, the uncertainty was calculated about a mean of 1 so it could be applied to any value for the tritium released during the time period for which uncertainty was calculated. If the uncertainty for a given calculational technique has two components (each having an uncertainty of ±10%), that uncertainty would be applied to a median of 1, the two factors would be multiplied, and the uncertainty would be calculated using Monte Carlo techniques to produce the total uncertainty for that technique. The final uncertainty can then be applied to all release estimates determined using that measurement technique.

When uncertainty is presented or was discovered in the literature to be represented by plus or minus some percentage value, the distribution was assumed to be normal, with the percentage value representing two standard deviations.

The uncertainty analysis is presented by type of monitor below. Because of the use of similar or identical components in the different equipment, in some cases the discussion of the different monitoring techniques overlap.

Dehumidifiers

In principle, dehumidifier measurements should be a reliable way of collecting tritium water vapor. As long as the coils were kept cold at all times and the sampled air was well mixed, the method should provide reliable and consistent results because it relied on collecting tritium vapor and measuring the amount of tritium in that water. Unfortunately, this method does not collect tritium gas. Therefore, there was no way to estimate total tritium releases to air, only releases that occurred from moderator loss.

The uncertainty inherent in the technique for measuring moderator loss by collecting water vapor with the dehumidifier was subjectively estimated at ±15% (2 standard deviations or 5 and 95% limits on a triangular distribution).

It is nearly impossible to correlate moderator loss to production. In fact, it is possible that moderator loss may have been higher during shutdown or maintenance because the reactor would have been open and moderator would have been more likely to evaporate. Dehumidifier readings were not commonly collected during shutdown because the technique was assumed to be reliable only during routine continuous operations (Caldwell 1958).

Mixing Within the Stack

The aliquot of stack effluent to be passed through the dehumidifier was removed from the exhaust stream. Sufficient mixing should have taken place by the time the exhaust stream reached the level where the sampling takes place because no inflow or outflow points exist in the stack between 0 and 148 ft. This was considered a negligible source of uncertainty.

Calculational Method

The uncertainty related to the tritium release obtained from the dehumidifier measurement was based upon the factors in the following equation used to estimate the tritium release to the atmosphere:

$$H_3 = H \cdot A \cdot (days / month) \cdot K \cdot D \qquad \text{(E-9)}$$

where

H_3 = tritium discharged from stack (Ci)

H = average absolute humidity of stack air (grains lb^{-1})

A = tritium in dehumidifier sample (average daily dehumidifier stack) (μCi HTO l^{-1})

K = conversion factor; obtained from a calibration curve of K factor versus airflow in cfm; converts μCi to Ci, cfm (from the x-axis of the curve) to cfd (unit retained in conversion factor), and grains lb^{-1} to l ft^{-3}

D = air density correction factor.

Absolute Humidity

Absolute humidity was also averaged for use in this equation. Absolute humidity was not usually measured; it was calculated from measurements of temperature and relative humidity. These two quantities were measured three times daily at each change of an 8-hour shift, and averaged before conversion to absolute humidity. As temperature or relative humidity increases, so will absolute humidity. We located humidity data from the SRS corresponding to stack measurements from 1989. These data indicate that humidity in the stack may have been dependent on outside humidity, with higher values for humidity appearing in the summertime. Outdoor temperature may have had a similar impact on stack temperature.

Historical weather data provided by the National Oceanic and Atmospheric Administration (NOAA) from their web site (http://www.noaa.gov) showed that relative humidity during summer months usually reached a maximum of 95%, with evening lows of 55%. During winter months, relative humidity reached an average high of 80% and an average low of 45%. Although these average values changed from day to day, over the course of a month they remain relatively consistent. In fact, NOAA's compilation of weather data from 1961–1990 showed that the average monthly relative humidity at the Augusta, Georgia, airport (the closest NOAA station to the SRS) was consistently around 70%, with the lowest average in April of 64% and the highest average in August of 76%.

Temperature fluctuated by about 20°F throughout the day during all seasons. The variation in temperature in the summer went from a high around 90°F to a low around 70°F, in winter from a high around 56°F to a low around 36°F, and during the spring and fall from a high around 75°F to a low around 55°F. Monthly averages compiled by NOAA from 1895–1996 reflect these same ranges.

Figure E-7 shows the relationships between temperature, relative humidity, and absolute humidity. Absolute humidity varies more at higher temperatures with changes in relative humidity than it does at lower temperatures. During the summer months, with a daily change in relative humidity from 95 to 55% and a temperature change from 90 to 70°F, absolute humidity may have varied from 0.3 to 0.9 g ft^{-3}. For winter months, variation in humidity from 80 to 45% and in temperature from 56–36°F may have varied absolute humidity from 0.05 to 0.2 g ft^{-3}. Using the average value of the extremes may have resulted in uncertainty ranging from 50% to a factor of 2. Because it was not possible to determine at what time of the day measurements of humidity and temperature were taken, the bias in absolute humidity values cannot be determined. The uncertainty distribution was assigned a uniform shape, with minimum and maximum values of 0.05 to 0.2 g ft^{-3}.

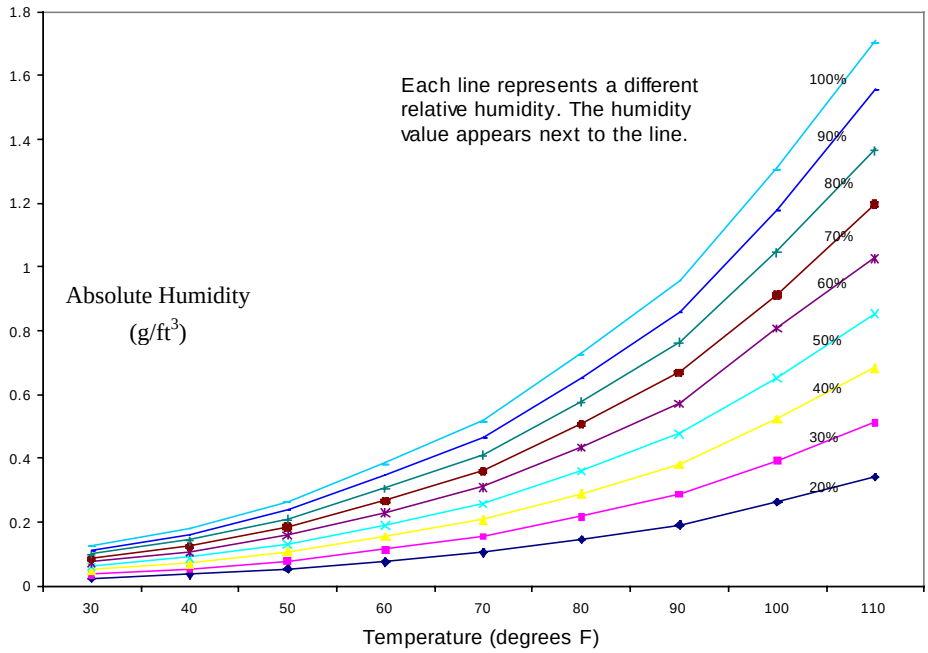

Figure E-7. Temperature, relative humidity, and absolute humidity relationships. Each line on the figure represents a different relative humidity and shows the absolute humidity as it varies with temperature.

Tritium Concentration in Dehumidifier Sample

The average daily dehumidifier stack sample condensate was counted using liquid scintillation counting techniques (μCi HTO L^{-1}). The dehumidifier water was counted in a liquid scintillation counter with a sensitivity of 3×10^{-7} μCi mL^{-1} (±10%) (Longtin et al. 1973; Jacober et al. 1973). Estimates of error from the SRS low-level tritium laboratory predict error in liquid scintillation counting of ±15% (Ostland and Berry 1968). For this uncertainty calculation, liquid scintillation counting uncertainty was estimated at ±20% and assumed to be normal based on indications in the supporting document.

Conversion Factor

The uncertainty in the conversion factor *K* was directly related to the uncertainty in the airflow through the stack, which could have varied up to 20% for any given condition and by over a factor of 3 between shutdown and normal operations. For normal conditions, with airflow variations up to 20%, *K* could have ranged from 7.0×10^{-4} to 1.0×10^{-3}, with a median value of 8.5×10^{-4}. For shutdown conditions, the variation in *K* was from 2×10^{-3} to 2.95×10^{-3}, with a median value of 2.45×10^{-3}. This range in *K* is described graphically in a July 1965 Du Pont operating procedure (Du Pont 1965).

Although each of these uncertainties represents about a ±15% variation from its respective median, it is important to estimate the uncertainty in the total variation in *K*. Monthly tritium release values come from both normal and shutdown conditions, and there is no way of

distinguishing between the conditions. The total uncertainty in *K* is a factor of 4, or approximately a factor of 2 in either direction from the median of the two central values for normal and shutdown conditions. The distribution of *K* was assigned a triangular shape, with minimum, median, and maximum values that represent this factor of 4 of 0.42, 1, and 1.8, respectively. This quantity is unitless and serves only as a calibration factor.

Air Density Correction Factor

The air density correction factor, *D*, is dependent on temperature. For temperatures below 58.5°F, the correction factor is 1.035; for temperatures between 58.5 and 77°F, the correction factor is 1.000; and for temperatures above 77°F, the correction factor is 0.987. To create a change in the air density factor would require a rather large change in temperature. At most, the air density factor resulted in about a 3% change from the median value of 1. We considered this an insignificant source of uncertainty and did not consider it further.

Reporting a Monthly Release Value Based on Selected Daily Measurements

Equation (E-9) calculated monthly tritium release estimates by averaging the dehumidifier measurements taken three times during the week and humidity measurements taken at the same time. Daily measurements would have been available had the dehumidifier been run and collected from each day, but it was difficult to discern whether this was the case. However, there were indications that it was not run during reactor outages. Documents were found that show that the dehumidifier was run only 3 days a week for 24 hours (Caldwell 1958). Data from 1989 were examined and showed that dehumidifier readings did not exist for each day; however, because the dehumidifier was not the preferred measurement technique in 1989, it is difficult to draw any definitive conclusions from this data set. It is clear that these data were probably not used to report releases but merely existed as an emergency measurement backup.

Although Westinghouse (1982) indicates that during later years the dehumidifier was run every day, references from the time period during which dehumidifier measurements were used to estimate releases indicate that once every 3 days was the operating procedure (Caldwell 1958). For this reason, we have chosen to use the conservative assumption that the dehumidifier was only operated once every 3 days and include this source of uncertainty in our calculations.

We examined the 1989 data, however, to explore the effect of averaging a few daily values to obtain monthly totals. If a dehumidifier measurement for each date in a month existed, the monthly average closely reflects a median value between the maximum and minimum value in a month. The average multiplied by the number of days in the month was a good prediction of the sum of the measurements. When 4 or more days of data were blanks, however, the average was generally much smaller than the maximum monthly value. This may indicate that higher dehumidifier readings had a tendency to be damped out when a number of data points during a month were missing. Additionally, nothing was known about the dates for which no measurements are taken. For months during which numerous measurements were missing, little else could have been done other than averaging existing points and multiplying by the number of days in the month.

Using the data from the later years of operations, the month of August 1989 reflected a number of data values that might be expected from the period of time when measurements were taken for 3 days a week. Table E-10 summarizes the data for August 1989.

Table E-10. August 1989 Dehumidifier Data[a]

Number of existing data points	17
Average of existing data points	9.49
Average × number of days in month (31)	294
Sum of existing data points	161
Maximum value	50.3
Minimum value	1.4

[a] Source: (Westinghouse 1979).

For this month, only 17 data points existed. The average of the 17 values clearly reduced the effect of the maximum value during the month, which was 50.3. If 3 of the 14 missing points were in the same range as the maximum, this quantity added to the existing sum of the values would about equal the existing average spread over the month. That still leaves 11 days without data points, indicating that the average may underestimate the sum over all days. Alternately, minimum values could have existed on each day, making the average an overestimate. In the worst case, the uncertainty in the average may have been a factor of 2 in our analysis. This uncertainty was reflected as the maximum and minimum values of a uniform distribution of possible values.

Overall Uncertainty

The total uncertainty in monthly dehumidifier values was calculated using Monte Carlo statistical techniques and 3000 generations of random numbers. The shape of the uncertainty factor results for a monthly source term value was fit to a lognormal distribution with a geometric mean (GM) = 0.99 and a geometric standard deviation (GSD) = 2.3. To determine the range in the monthly source terms that this uncertainty represents, the median source term value should be divided and multiplied by the GSD to obtain one standard deviation on either side of the median value, respectively.

Silica Gel

The operation of the silica gel is quite similar to that of the dehumidifier, and many of the same parameters affect the uncertainty. Moderator loss is calculated from the silica gel using Equation E-10.

$$E = \frac{A \cdot B \cdot D}{100} \qquad \text{(E-10)}$$

where

E = Moderator loss (lb day-1)

A = Absolute humidity (lb water (lb air)-1)
B = Ventilation airflow (lb air day-1)
D = (*C* – 0.0147) 1.11 (weight %)
C = Sample analysis lab results (mol %).

Silica gel samples were taken out of the ventilation airflow of the stack 3 days a week for approximately 3 hours at a time.

Absolute Humidity

The absolute humidity was described in detail in the section above on dehumidifier uncertainty. The major difference between how this parameter affects the moderator loss calculated from the silica gel as opposed to the dehumidifier regards the sample duration. Silica gel samples were only collected for 3 hours on any given day rather than 24 hours at a time.

Data from the NOAA web site were again used to examine the change in temperature and relative humidity over shorter time periods. It appears that it is reasonable to assume that temperature will not vary by more than 20° and humidity by 20% over any 3-hour period. From the NOAA data, this is probably a conservative assumption. Even at high temperatures and relative humidities, the fluctuation in the absolute humidity is no more than about a factor of 2 total. This will be used as the uncertainty estimate for the equation parameter *A*, distributed in a triangular fashion.

Ventilation Airflow

The ventilation airflow has been previously defined to have an uncertainty of ±20% during a given operating condition and by over a factor of 3 between shutdown and normal operations. Because there is no way of knowing for certain during which condition any measurement was made or whether measurements were always taken during a single condition, the uncertainty must be assumed to be a factor of 3 for the parameter *B*, triangularly distributed from the median of the two central values for normal and shutdown conditions.

Sample Analysis Result

The sample analysis result, *C*, is the only parameter with any impact on the uncertainty of parameter *D*. Sample analysis uncertainty is affected by the duration of the sample as it relates to averaging effects and the sampling procedure and analysis uncertainty.

Silica gel moisture collection is still used today as an effective means of collecting water vapor contaminated with tritium. This method is still used because it is accurate when used properly and without exposing the gel to more moisture than it can collect, resulting in missed sample. The sampling procedure and analysis uncertainty was subjectively estimated to be ±20%, normally distributed.

The effect of averaging 3 hours of data to represent a day and 3 of these days a week to represent a month introduces some uncertainty into the calculation. Dehumidifier data were examined to explore the effects of averaging daily values to obtain monthly totals. The uncertainty in this technique was estimated to be a factor of 2. It is more difficult to assess the

impact of averaging 3 hours to represent 24 hours, but we can assume this averaging may have a similar impact to the monthly averaging and assign a factor of 2 uncertainty. This makes total uncertainty in the averaging techniques around a factor of 4. This uncertainty is reflected as the maximum and minimum of a uniform distribution of values.

Total Uncertainty

Total uncertainty in the silica gel values for moderator loss was calculated using Monte Carlo statistical techniques and 3000 generations of random numbers. The shape of the uncertainty factor results for monthly moderator loss values was fitted to a lognormal distribution with a GM =1.15 and a GSD = 2.16.

Kanne Chamber and Stack Monitor Integrator

Kanne Chambers

The underlying principle of a Kanne chamber is good as long as the Kanne chamber's size is sufficiently large to allow an ionization from a low energy beta emission to take place and be absorbed within its volume. The SRS reported the efficiency of this counting system as ±10% for low energy beta emitters such as tritium. This is used as the uncertainty for the basic Kanne chamber measurement technique.

Mixing of Air within the Stack. The aliquot of stack effluent was removed from the exhaust stream at the 148-ft level. Sufficient mixing should have taken place by the time the exhaust stream reaches this level because no inflow or outflow points exist in the stack between 0 and 148 ft. This is a negligible source of uncertainty and is not considered further in the uncertainty estimates.

Calibration. Kanne chambers must be calibrated before use to determine the ionization current produced per unit of radioactivity in the volume of air passing through the chamber. After a calibration was completed, the calibration value should not have changed for the detector. The complicating factor would be if a calibration were performed incorrectly and permanently skewed the results of the chamber.

Hoy (1961) related operational experience with calibrating Kanne chambers at the SRS and identified some possible problems with the calibration and use of Kanne chambers for accurate measurements.

The calibration factor for the Kanne chamber may have been miscalculated because adsorption of tritium onto the walls of the Kanne chamber may not have been accounted for. This had the effect of decreasing the experimental response of the chamber during calibration (less current per unit tritium in air); thus, overpredicting total concentration of tritium in air for the life of the detector. Experimental results from the calibration of Kanne chambers at SRS indicated that wall losses could have been as high as 28% for the smaller chamber or 11.5% for the larger chamber (Hoy 1961). These wall losses would have increased the Kanne chamber measurements by the same amount, requiring a reduction in the calibration factor to achieve the actual result.

Because calibration was done by measuring tritium in the chamber, the general measurement uncertainty of ±10% also applies to the calibration uncertainty. The total uncertainty in calibration was calculated by combining the measurement uncertainty and the wall loss

uncertainty and fit to a normal distribution with a mean of 0.91 and a standard deviation of 0.11. This corresponds to 5 and 95% values -20% and +17% of the mean, distributed relatively normally.

An operational disadvantage of the Kanne chamber was that all molecules creating ionizations in the chamber looked the same because the chamber operated within the ionization region and did not distinguish between energies of the particles deposited. The fraction of the average beta energy deposited in the chamber for different radionuclides and, therefore, the response for each nuclide could be estimated for a Kanne chamber. The fraction of the energy that the ionization chamber captured decreased as the average beta energy increased. The ionization current caused by radioisotopic decay was proportional to the average energy deposited per disintegration. If the relative abundance of each isotope in the gas mixture was known, the amount of ionization because of decay of each isotope could have been estimated. A calibration factor was determined for each beta-emitting gas that was likely to have been present within the Kanne chamber. This method of apportioning the energy deposited was highly dependent on knowing the isotopic ratios of the gases in the chamber, and it involved calculations instead of actual experimental measurements. This source of uncertainty is difficult to quantify.

Some guidance was gained from a 1989 document that revisited the Kanne chamber calibration (Litrell 1989). This work outlined the discovery that the calibration factors established before the use of a Kanne chamber were essentially good values. If the ratio of the gases was known, the impact of noble gases on the measurement for tritium was reasonably well understood. The primary noble gas component in stack air that interfered with tritium was ^{41}Ar. Uncertainty depended on the ability to estimate the ratio of these two gases. This must have been difficult because it was recommended that the Kanne chamber be used only following reactor shutdown or when activity could be attributed to tritium (Caldwell 1958). Supporting documentation seemed to imply that this was a large source of uncertainty and should be avoided if possible. Currently, there is no way of knowing if the Kanne chamber was used under ideal conditions. The uncertainty related to noble gas interference was subjectively estimated as a factor of 2, uniformly distributed about the mean, because no quantitative information was available and some estimate of uncertainty resulting from this factor was necessary.

Temperature and Pressure. Variations in temperature and pressure of the chamber may have had an impact on the consistency of the results. System pressure affected the ionization rate. The calibration memo (Litrell 1989) indicates that this could have easily shifted the Kanne results by ±10%. The variations because of temperature and pressure were quantified as normally distributed about the mean with an uncertainty of 10%.

Relative Humidity. Relative humidity above 95% had the potential to cause condensation within the chamber, but Kanne chambers have been known to operate well even at 100% humidity. Although relative humidity in the SRS area could have been high during the summer, historical records indicated that humidity above 95% was rare, even as a daily high. The condensation factor was assumed to have a negligible effect on the Kanne chambers, and no uncertainty was applied.

Manual Integration. The Kanne chamber measurements were manually integrated to produce tritium stack release results before digital integration techniques were in place. The area under the curve of changes in current with time, as recorded by the chart recorder, was proportional to activity passing through the chamber. The amplifier used on the Kanne system

was logarithmic, as was the paper charting the trace, so Equation (E-11) was used for logarithmic integration of the stack charts:

$$Q = c\left[k_e \sum A_e - A_{bg} T\right] \cdot 1.7 \times 10^6 \qquad \text{(E-11)}$$

where

Q	=	amount of tritium (Ci)
c	=	air exhaust rate during time T (ft^3 min^{-1})
k_e	=	chamber calibration (μCi cm^{-3} A^{-1})
A_e	=	area under curve during time T (A hours)
T	=	release time period (hours)
A_{bg}	=	background air activity (Ci cm^{-3})
1.7×10^6	=	conversion from hours to minutes and from cubic feet to cubic centimeters.

The value of k_e was the chamber calibration for the appropriate Kanne chamber and would have varied with the uncertainty described previously. There should be no additional uncertainty added to the constant than the factor of 2 inherent in the chamber collection of noble gases and the uncertainty related to wall losses.

Air exhaust rate, c, was assumed to have an uncertainty of ±20% at the 95% confidence level as identified in previous sections. For our analysis, a triangular distribution was assumed, with 5 and 95% values set to ±20% of the median value.

The estimate of area under the Kanne chamber current trace had units of ampere hours because the area is calculated by multiplying some vertical distance on the chart (ampere) by the horizontal distance (hours). The uncertainty in area was related not only to the estimate of area using prescribed methods but also to operational uncertainty. This uncertainty was described earlier in this section as ±10% under standard operating conditions and an additional ±10% contributed by variations in pressure within the chamber. The area estimate should have been reasonably good, particularly because the current traces produced by the Kanne chambers were somewhat constant through time. The method used to calculate area under the trace depended on the appearance of the trace. If the trace was quite regular and straight, the straight-line method was used, dividing the trace into a series of rectangles and trapezoids, calculating the area for each section, and adding up the areas of the segments. If the trace was more irregular, the approximate integration method was used. The approximate integration method involved marking off sections of equal length along the x-axis, adding together the heights of the right sides of the segments, and multiplying by the distance (in units of hours) between the reference lines. If the appropriate area method was used with the appropriate graph type, this area estimate should be fairly certain. Because the uncertainty was negligible, we did not make a subjective estimate for this factor.

Stack Monitor Integrator

In later years, the manual integration technique described above was abandoned for a more reliable digital one, referred to as the SMI. The signal from the amplifier was sent directly to a digital processor, and results were available continuously and immediately. The digital signal was converted from current to frequency and normalized to 1 count per curie released so the number

of counts over the integrated period of time was related one-to-one with the curies of tritium released from the stack.

The conversion factor for count per curie released is shown in Equation (E-12). This factor took the chamber calibration, stack flow rate, and the current to frequency transfer function into account.

$$R = \left[\frac{x}{y}\right] \cdot z \cdot \left[\frac{1}{k}\right] \tag{E-12}$$

where

R = counts per curie released
x = Kanne chamber constant (μCi cm^{-3} A^{-1})
y = stack flow (cm^3 s^{-1})
z = current to frequency transfer function (Hz A^{-1})
k = normalization constant.

If R is set to 1, the normalization constant can be solved for. Electronically, normalization was accomplished using a calibration unit attached to the SMI that calibrated the signal and normalized it to 1.

The same uncertainty that applied to the Kanne chamber collection, chamber calibration, and stack flow also applied here. Additionally, electronics used in the digital signal conversion had some associated uncertainty.

The C/F converter linearly converted the ionization current from the Kanne chamber to a user-defined frequency. The input current to the machine charged a capacitor, which produced a voltage ramp. For a given increase in voltage, a trigger was fired that produced a pulse whose width was equal to the user-defined frequency. The trigger fire also discharged the capacitor and restarted the cycle. The uncertainty in this setup was related to the time it took the capacitor to discharge and any current that was lost during that process, as well as the leakage current inherent to the equipment. This source of uncertainty was negligible, and it was not included in the calculation.

In summary, the normalization technique had uncertainty in the airflow and chamber calibration factor, but no additional uncertainty was added for the nominal amount that would have been related to the electronics.

Overall Uncertainty

Monte Carlo techniques were used to combine the different sources of uncertainty in Kanne chamber measurements with 3000 generations of random numbers. Because no uncertainty was applied to the area estimation or electronic integration techniques, the uncertainties for the two different types of Kanne stack chart integration were the same. Data from the uncertainty calculations were widely spread and not well fit by any distribution, but they were reasonably approximated by a lognormal distribution with a GM = 0.9 and a GSD = 1.8. To calculate uncertainty in release estimates, the GM is multiplied by the Kanne chamber estimate to provide a better estimate of a median value. One standard deviation of that median value is calculated by

multiplying and dividing the median by the GSD. Five and ninety-five percentile estimates are obtained by multiplying and dividing by the square of the GSD.

Stack Tritium Monitor

The STM simply adapted the use of the Kanne chamber and the silica gel into a piece of equipment that could more readily distinguish between tritium and noble gases in the reactor areas. Tritium in elemental form was not much of a concern in the reactor areas, and tritium vapor was the release of interest.

Uncertainty in the STM readings inherently had the uncertainty associated with collection of activity using a silica gel column and two Kanne chambers. Although it might appear at first glance that the uncertainties in the three methods might have canceled each other out, they instead became additive and increased the total uncertainty in the final value.

Additionally, the calibration of the STM was more difficult than simple calibration of a Kanne chamber. The increased hardware left extra room for error. The calibration of a Kanne chamber was discussed in a previous section of this appendix. This calibration uncertainty applied to a similar constant used in the STM calculations, called K_1. This constant had units of curies per cubic centimeter per ampere and became a factor in the series of calibration equations described below.

$$A = K_2 \cdot V \tag{E-13}$$

$$K_2 = \frac{F \cdot C}{K_1} \tag{E-14}$$

$$R = \frac{dA}{dt} = K_2 \cdot \frac{dV}{dt} \tag{E-15}$$

where

A	=	total integrated stack activity released (Ci)
V	=	voltage across the capacitor, created by current through capacitor (V)
F	=	flow rate of air out stack ($cm^3\ min^{-1}$)
C	=	capacitance (farads)
		capacitance (in farads) multiplied by voltage (in V) equals current (in A)
K_1	=	chamber calibration ($Ci\ cm^{-3}\ A^{-1}$)
R	=	rate of release of activity ($Ci\ min^{-1}$).

Since K_1 in the above equations depended on the calibration of two Kanne chambers, it is reasonable to assume that uncertainty in K_1 is greater than the uncertainty in the calibration of a single chamber. The measurement uncertainty for a single Kanne chamber, including chamber calibration, is +17% and –20% of the mean (see Kanne chamber section). This uncertainty factor is applied twice in the calculation of uncertainty for the STM by multiplying the total uncertainty by this factor squared.

The other source adding uncertainty to the calibration factor for the rate of release calculation (K_2) is the airflow rate, F. As before, we assigned a flow rate uncertainty of ±20% based on measurement uncertainty (see previous sections).

The silica gel has an uncertainty that is lognormally distributed with a GM = 1.15 and a GSD = 2.16. This uncertainty represents the efficiency with which the silica gel collects total water vapor in the airstream (see silica gel section).

A major source of uncertainty in Kanne chamber measurements is the inability of the chamber to distinguish between noble gas activity and tritium activity. That source of uncertainty is removed by this new experimental setup because subtracting the two Kanne results gives a value for only tritium vapor in the airstream if all sources of uncertainty in collecting activity are accounted for in the calculation.

Pressure fluctuations may still have been an important source of uncertainty because two ionization chambers had to remain under stable pressure throughout measurements. The pressure difference between the two chambers was zeroed before use of the instrument, but that does not imply that pressure remained constant. This source of uncertainty, identified as ±10% earlier, is included twice to account for possible fluctuations in both chambers.

The total uncertainty in STM releases is calculated using Monte Carlo techniques and 3000 trials. The results of the calculation fit a lognormal distribution with a GM = 0.91 and a GSD = 2.19.

A test of the STM system compared to the existing dehumidifier system was performed in 1965 when the STM was first introduced. The test monitored stack releases for 24-hour periods over 1 week. The measured results are shown in Table E-11. The parenthetical values following the measured results are the 5 and 95% uncertainty limits of the values based on the uncertainty factors presented here.

Table E-11. Release Measurements and Uncertainty for a 1965 STM Versus Dehumidifier Test

	Release (Ci d^{-1})	
Day	Stack tritium monitor (5-95% uncertainty limits)	Dehumidifier (5-95% uncertainty limits)
Wednesday	218 (41-950)	336 (64-1800)
Thursday	221 (42-960)	372 (70-2000)
Friday	211 (40-920)	413 (78-2200)
Saturday	162 (31-710)	341 (64-1800)
Sunday	156 (30-680)	346 (65-1800)
Monday	152 (29-660)	321 (61-1700)
Tuesday	240 (46-1000)	390 (74-2100)
Wednesday	240 (46-1000)	320 (60-1700)

For all release days, the uncertainty bounds of the release values overlap, indicating that although the values appear different, they are statistically the same given what was known about the operation of each measurement system.

Berthold Tritium Monitor

The BTM was the most recent evolution of stack monitoring systems at the SRS, and the use of the gas proportional counting principle made it the most robust. In gas proportional counters, like ionization chambers, charged particles were produced by ionization in the gas. Unlike ionization chambers, radiation that interacted in a gas flow proportional counter produced ion pairs in a quantity proportional to the amount of energy it deposited. This allowed the proportional counter to distinguish between tritium oxide and elemental tritium, the forms of tritium released from SRS stacks. The counter not only distinguished between the two, but it also measured the activity of each isotope passing through, something no counter before this one could have done. Two single channel analyzers collected the two different forms of tritium.

General Operation

In the reactor areas, the tritium releases were still estimated by estimating the total amount of moderator lost and then converting back to total curies released. Available documentation referenced two equations that may have been used to calculate total moderator lost from the stack (Westinghouse 1988). However, Westinghouse (1988) identified one equation whose solution was transferred to the data sheet from which tritium loss was calculated (Equation [E-16]).

$$A = C \cdot IB \cdot \frac{F}{T} \tag{E-16}$$

where

A = daily stack loss (lb D_2O day^{-1})
C = conversion factor
IB = daily integrated total counts in channel B, channel which collects tritium oxide ionization (counts)
F = duct flow (cfm)
T = tritium content of moderator from latest lab result (μCi tritium [mL D_2O]$^{-1}$).

The units of the conversion factor were not found in any documentation, but they were determined based on the calculation. The conversion factor presumably converts microcuries of tritium to counts, minutes to day, and milliliters D_2O to pounds D_2O, where 1 mL = 1 g water.

To convert from this value of pounds D_2O per day to curies of tritium per day, the following equation was required:

$$Q = A \cdot T \cdot 0.000413 \cdot D \tag{E-17}$$

where

Q = tritium released to atmosphere (Ci)
A = daily stack loss (lb D_2O d^{-1})
T = tritium content of moderator from latest lab result (μCi tritium [mL D_2O]$^{-1}$)
D = air density correction factor
0.000413 = lb d^{-1} × μCi d^{-1} to Ci d^{-1}.

The air density correction factor, *D*, used in Equation (E-17) is the same factor explained in the dehumidifier section, with uncertainty equal to ±3% of the mean.

Equations (E-16) and (E-17) were combined to remove a source of uncertainty, the tritium content of the moderator. Because this value was only measured once a week, it would have been a source of uncertainty for the stack loss calculation. The same value was used again in the conversion of the moderator loss data back into tritium release, effectively removing this source of uncertainty. Combining the equations produced Equation (E-18).

$$Q = C \cdot IB \cdot F \cdot 0.000413 \cdot D \qquad \text{(E-18)}$$

where all values are as described previously.

The uncertainty in the BTM tritium release values was centered around the stack flow rate; the accuracy of the BTM measurement (resulting in the *IB* value); and the conversion included in the factor *C*, which converted counts from the BTM to microcuries tritium. The uncertainty in *D* is so small as to become negligible in the uncertainty calculation.

Uncertainty in the stack flow rate has been described in all previous sections as ±20% of the mean, with a triangular distribution.

The conversion factor was related to the calibration of the BTM. Because calibration of this instrument was much easier than calibration of the STM or previous stack monitoring equipment, it was probably done more frequently. A small quantity of tritium in nitrogen calibration gas was injected directly into the BTM, and a long check sheet for calibration was established to ensure more accurate stack results. The sensitivity of the BTM was quoted in manufacturers' specifications as much lower than achievable in the field, with a lower limit of detection lower by a factor of 10.

This inconsistency was blamed on the uncertainty in stack flow rate and in moderator tritium content (Merz 1988). Calibration experiments showed that the BTM could measure tritium in the presence of a simulated noble gas to accuracy of less than 5 parts in 1000. Because moderator tritium content had been removed from the tritium release equation and uncertainty in stack flow rate was accounted for, the large inconsistency between predicted sensitivity and achieved sensitivity was ignored. The accuracy of the calibration experiments was noted (0.5%), but we assumed the calibration factor exhibited a small degree of uncertainty, subjectively estimated at ± 10%. During calibration, extreme measures appeared to have been taken to ensure accuracy. Calibration was highly controlled, but not so controlled as to eliminate operator error. A small uncertainty appeared appropriate in this case, however, because the technique was so finely tuned and carefully practiced.

The accuracy of the BTM readings was probably quite good, given operation in ideal conditions, but stack conditions were never ideal and a small degree of operational uncertainty was assumed to account for possible inconsistencies. Radioactive decay is a statistical process, and not all radiation that entered the chamber was likely to have been detected by the equipment. Still, these factors account for a relatively small uncertainty when compared to uncertainties inherent in the other measurement techniques. Operational uncertainty of the BTM was subjectively estimated at ±10%

Total Uncertainty

Total uncertainty in the BTM measurements was calculated using Monte Carlo techniques, with 3000 generations of random numbers. The total uncertainty results were fit to a lognormal distribution with a GM = 0.99 and a GSD = 1.16. To estimate uncertainty in any source term value, the 5 and 95% bounds of the source term estimate can be obtained by multiplying and dividing the median value by the square of 1.16, respectively.

SUMMARY

The accuracy of the atmospheric source term estimates presented in this report is largely dependent on the type of monitors used over time to measure the releases. A detailed description of the equipment and the data used in the uncertainty analysis was presented in this appendix. In reviewing the documentation, it was possible to find dates on equipment operation procedures or reports of plans or prototype tests; however, it was not possible to put together an exact chronological history of monitoring equipment used in each facility. Therefore, there is no clear connection between a reported release value and monitor that was used to measure the release. For example, it has not been possible to determine when the silica gel monitor was replaced by the dehumidifier. Early documentation refers to both monitors.

No clear deficiency in the methodologies used brought us to the conclusion that there was need to apply corrections factors to the data to determine the final source term estimates. To help verify that this was acceptable, we used the results of the uncertainty to evaluate the effectiveness of the monitors. Table E-12 presents a summary of the results of the uncertainty analyses.

Table E-12. Summary of Uncertainty Analysis for Tritium Monitors used at the SRS

Monitor	For results in years	Facilities	Distribution	Range[a]
Silica gel	1954–1958	Reactors	Lognormal	GM = 1.15 GSD = 2.16
Dehumidifier	1954–1970	Reactors	Lognormal	GM = 0.99 GSD = 2.3
Kanne chamber	1954–1974	Tritium facilities	Lognormal	GM = 0.9 GSD = 1.8
Stack Monitor Integrator	1974–present			
Stack tritium monitor	1970–1988	Reactors	Lognormal	GM = 0.91 GSD = 2.19
Berthold tritium monitor	1988–present	Reactors	Lognormal	GM = 0.99 GSD = 1.16

[a] GM = geometic mean, GSD = geometric standard deviation.

Because there were no other technical indications for the need to correct the estimate release data, we think that the results in the column titled “Range” show that the earlier methods, although less accurate, fall within the same range as the later, better methods with a larger uncertainty range. So, *RAC* concludes that the “best estimate” values should be used as the atmospheric tritium source term without correction, and the range of uncertainty should be applied to release data by facility and year as indicated in Table E-12. If more information is found defining the time period of use of the monitors in each facility, these results can be applied more specifically.

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Westinghouse. 1991e. *Savannah River Site Radioactive Air Emission Monitoring.* ESH-EMS-910039, Rev. 0. SRS Phase II Database LEE199407131. March 12.

Westinghouse. 1993. *Operation Of The Tritium Stack Monitor*. Manual 5Q1.5, Procedure 316A, Rev. 3. SRS Phase II Database LEE199407088. August 12.

Westinghouse. 1994. *Determination of Tritium Stack Releases, H-Area.* Air Effluent–Monitoring Procedures. DPSOP-193-3-310. Manual 5Q1.5, Procedure 310, Rev. 5. SRS Phase II Database LEE199407085. February 22.

APPENDIX F

GEOGRAPHIC INFORMATION SYSTEM AND DEMOGRAPHIC DATA COMPILATION

APPENDIX F

GEOGRAPHIC INFORMATION SYSTEM AND DEMOGRAPHIC DATA COMPILATION[a]

INTRODUCTION

This appendix describes the geographic information system (GIS) and demographic data compiled to support the Savannah River Site (SRS) dose reconstruction study. It also briefly describes the management activities for the demographic data at the South Carolina State University (SCSU) GIS Laboratory and the comprehensive database design developed to house data that can support population dose assessments.

GEOGRAPHIC INFORMATION SYSTEM DATA

The GIS component of this project focuses on compiling and formatting spatial data and identifying data sources pertinent to locating contaminant sources (air, water, and land); monitoring locations; contaminant transport pathways (surface water, air, and crops); and population distributions. GIS coverages were also developed to provide a boundary for the study area (Figure F-1) and to support public communication.

One of the objectives for this task was to provide a GIS coverage data directory structure (Figure F-2) and a database that will be compatible with future Centers for Disease Control and Prevention (CDC) GIS technology needs. As GIS technology continues to develop, it will become more important for storage, analysis, and communication of spatial data.

A reduction in the available funding to support GIS data compilation activities limited efforts to compile all relevant spatial data, conduct analyses on imagery, and complete data documentation to meet Federal Geographic Data Committee (FGDC) standards. What was collected, however, provides a solid foundation for future GIS activities and public communication for the project.

Data Acquisition

Data Acquisition Scope

A general study area was defined to focus data acquisition and limit the total amount of data to be obtained. The study area includes the real estate within a 50-mi buffer from the SRS boundary, plus all the counties on either side of the Savannah River downstream from the SRS. Columbia, South Carolina, was also included within the study area (Figure F-1). Development of this boundary is discussed at the end of this section.

The temporal period of interest for data collection was from the early 1950s through the early 1990s. Data for the beginning of each decade were identified if available. Numerous GIS coverages were obtained from various sources; however, documentation did not accompany some of the data and it could not be obtained. GIS data that lacked documentation regarding sources and accuracy are not included.

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[a] Authors of this appendix are Ronald C. Rope of Environmental Perspective, Inc.; Wei Yang of South Carolina State University; and Luke White and Patti McGuire of Teton Geographic Information Systems, Inc.

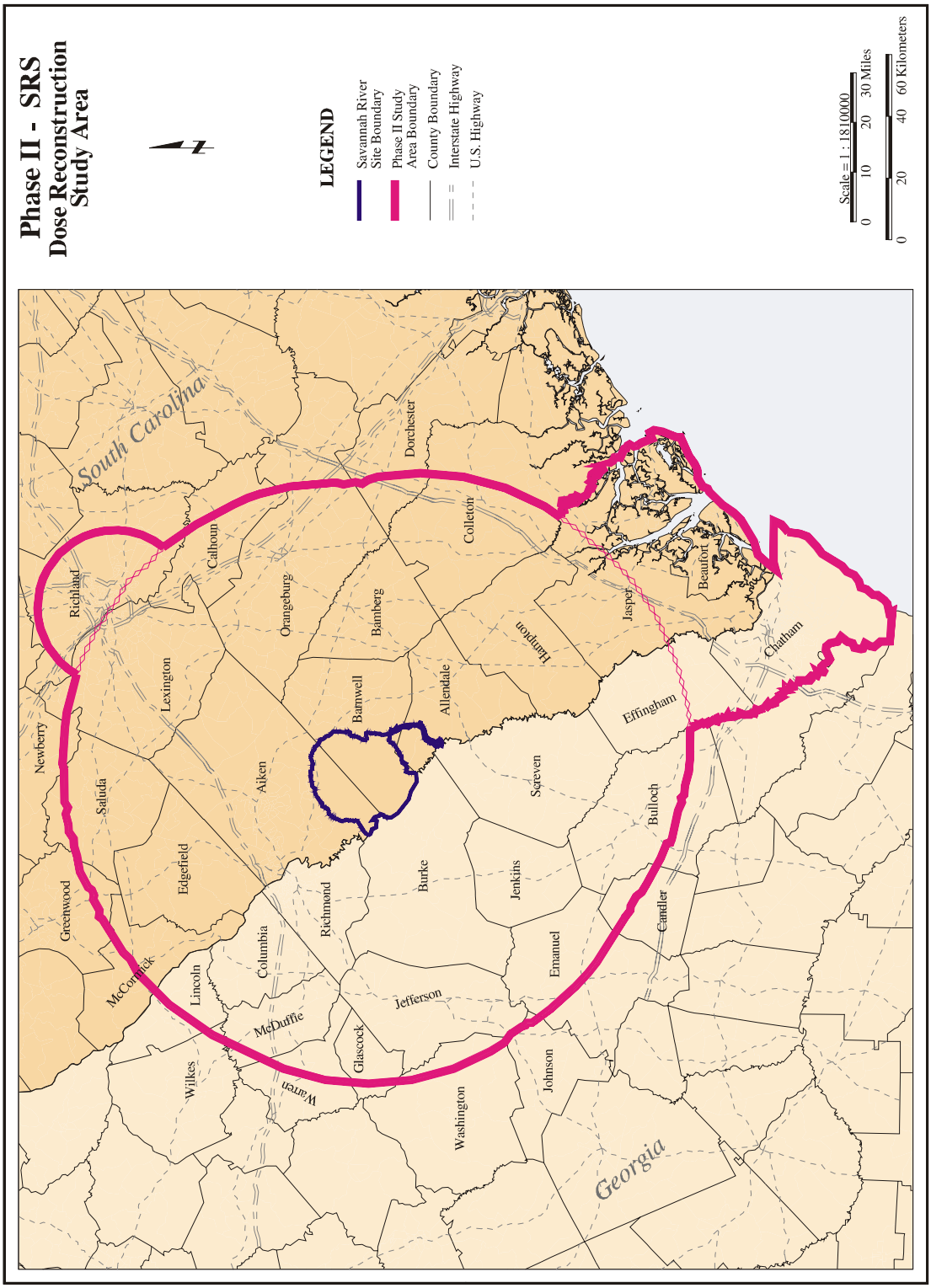


Figure F-1. Map of the project study area.

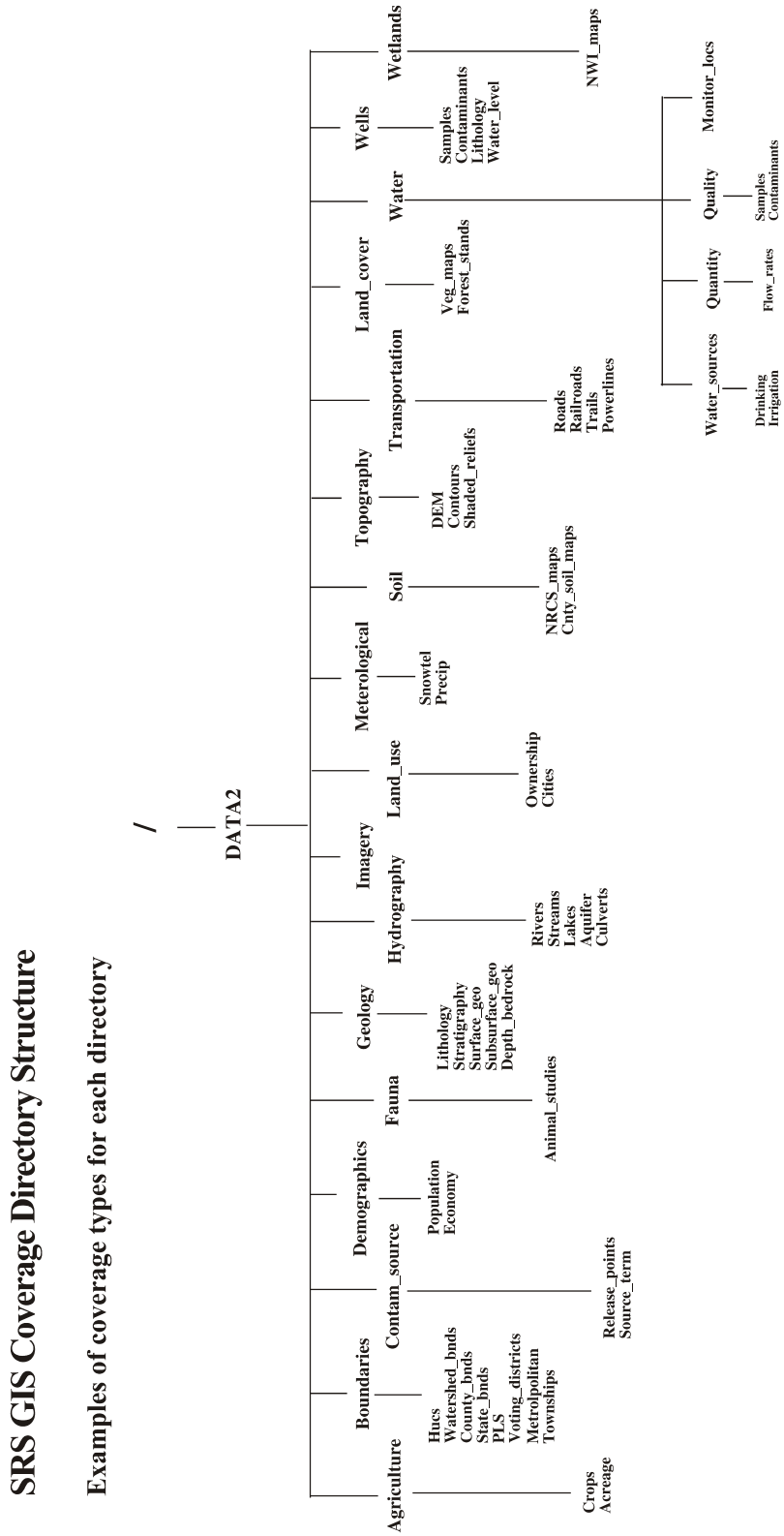


Figure F-2. Proposed GIS data directory structure.

Data Resolution

Generally, 1:100,000 scale data were collected for the area outside the SRS. However, larger scale data for the SRS and some adjacent lands were also obtained. Discussions among project investigators indicated that greater resolutions were not necessary to support analyses.

Data Types

The team attempted to obtain data that would support the following project needs:

1. Locating and mapping locations of chemical and radiological releases (effluents, emissions, spills, and disposal areas)
   - Facility drawings with stacks, drains, effluent pipes
   - Waste disposal areas
   - Storage/loading areas
   - Spill sites
   - Aerial photographs.
2. Identification and analysis of contaminant transport, and exposure pathways (air, surface, ground water, and biota)
   - U.S. Geological Survey (USGS) hydrography
   - SRS drainage patterns
   - United States Fish and Wildlife Service (USFWS) National Wetlands Inventory data
   - Fishing and hunting locations
   - Key habitats for game species
   - Ground cover
   - Digital Elevation Model (DEM) data
   - USGS orthophoto quads
   - Soil data
   - TIGER data or other census data
   - Remotely sensed data (aerial photographs and satellite imagery).
3. Locations where single samples were collected and/or monitoring station locations (historic or current)
   - SRS monitoring sites (current/historic)
   - Other state, federal, and university monitoring sites (EarthInfo data for USGS, NCDC, STORET)
4. General GIS base coverages
   - Transportation (roads, trails, and power lines)
   - Ownership, borders (e.g., counties, cities and towns, forests, recreation areas)
   - Land use.

Sources of GIS Data

The primary sources for GIS data were the SRS Legacy GIS Coverages CD, Version 1 and Version 2, American Digital Cartography Inc. (ADC) Cornerstone Data, U.S. Environmental Protection Agency (EPA) BASINS CD for Region 4, NASA, USGS and EPA North American Landscape Characterization Project.

GIS Coverages Obtained

Table F-1 is a list of the GIS coverages and imagery compiled to date. SRS recently released a new version of the SRS Legacy GIS Coverages that includes both GIS coverages and historic aerial photographs for the SRS. Coverages that were obtained, but do not have appropriate documentation are not included in this table.

Table F-1. GIS Data Compiled

Coverage	Filename	Coverage type	Coverage category	Description	Extent[b]	Source[a]
Bizmuth	Bi91t	Arc	Monitoring	Bismuth present at the SRS, South Carolina in 1991. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Breakwaters	Bkwaters	Arc	Facilities	Twenty-two breakwaters along the Savannah River. Breakwaters are wooden poles placed vertically in a river in a line to divert water flow.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Boundrys	Boundrs	Polygon	Boundaries	Political boundaries	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Buildngs	Buildngs	Polygon	Facilities	Building outlines	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Cemetery	Cemetery	Polygon	Land use	Cemeteries	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Comps	comps	Polygon	Land use	Timber compartments	SRS	SRS Legacy Data, Ver. 1
Contours	contours	Arc	Topography	Contour lines 10-ft interval	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Ecoclass	ecoclass	Polygon	Land_cover	Ecosystem classification	SRS	SRS Legacy Data, Ver. 1
Fences	fences	Arc	Facilities	Fence boundaries	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Gross contours	grosscnt	Arc	Topography	Topographic contours	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1

Table F-1. GIS Data Compiled

Coverage	Filename	Coverage type	Coverage category	Description	Extent[b]	Source[a]
Historic wetlands	Histwet	Polygon	Wetlands	Historical wetlands before 1952 on the SRS, South Carolina	SRS	SRS Legacy Data, Ver. 1
Hollow creek	Hollowcr	Polygon	Geology	Geology map–Hollow Creek 1:24,000 quad	Hollow Creek quad	SRS Legacy Data, Ver. 1
Land cover	Landcov	Polygon	Land_cover	Land cover	SRS	SRS Legacy Data, Ver. 1
Mgmt_ar	mgmt_ar	Polygon	Boundaries	Timber management areas	SRS	SRS Legacy Data, Ver. 1
Npdes96	Npdes96	Point	Contam_source	National Pollutant Discharge Elimination System (NPDES) 1996 permit locations	SRS	SRS Legacy Data, Ver. 1
Openwells	opnwells	Point	Wells	Pre-1951 residential wells	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Pipes	pipes	Arc	Transportation	Pipes and power transmission lines	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Rails	rails	Arc	Transportation	Railroads	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Roads	roads	Arc	Transportation	Roads and trails	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Rsrch_su	rsrch_su	Polygon	Boundaries	Protected forest research	SRS	SRS Legacy Data, Ver. 1
Setaside	Setaside	Polygon	Boundaries	Coverage for Set-Aside boundary and vegetation communities	SRS	SRS Legacy Data, Ver. 1
Site grid	Site_grd	Polygon	Boundaries	Grid over SRS	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Snelling	Snelling	Polygon	Geology	Geology map–Snelling 1:24.000 quad	Snelling Quad	SRS Legacy Data, Ver. 1
Soils	soils	Polygon	Soils	Natural Resource Conservation service (NRCS) soil survey	SRS	SRS Legacy Data, Ver. 1
SRS bays	Srsbays	Polygon	Hydrography	Bay areas on the SRS	SRS	SRS Legacy Data, Ver. 1
SRS boundary	Srsbnd	Polygon	Boundaries	SRS boundary	SRS	SRS Legacy Data, Ver. 1
Stands	stands	Polygon	Land_cover	Timber comps and stand bound	SRS	SRS Legacy Data, Ver. 1

Table F-1. GIS Data Compiled

Coverage	Filename	Coverage type	Coverage category	Description	Extent[b]	Source[a]
Streams	streams	Arc	Hydrography	Single line water courses	SRS and beyond the SRS boundary	SRS Legacy Data, Ver. 1
Trails	trails	Arc	Transportation	Trails maintained by SRFS	SRS	SRS Legacy Data, Ver. 1
Water bodies	waterbds	Polygon	Hydrography	Water bodies and major streams	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Wells95	wells95	Point	Wells	Monitoring wells 2Q 95	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Wells96	wells96	Point	Wells	Monitoring wells 2Q 96	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Willistn	Willistn	Polygon	Geology	Geology map–Williston 1:24,000 quad	Williston quad	SRS Legacy Data, Ver. 1
Wl_comps	wl_comps	Polygon	Land_cover	Wildlife compartments	SRS	SRS Legacy Data, Ver. 1
Wtreg195	wtreg195	Arc	Hydrography	Regional water table 1Q 95	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Wetsoils	Wetsoils	Polygon	Soils	Wetland soils	SRS	SRS Legacy Data, Ver. 1
Cobalt	Co91t	Arc	Monitoring	Cobalt present at the SRS, South Carolina. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Cesium	Cs91t	Arc	Monitoring	Cesium present at the SRS, South Carolina. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Gross counts	Gc91t	Arc	Monitoring	Gross count at the SRS, South Carolina. This coverage contains the gross count at SRS in 1991. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1

Table F-1. GIS Data Compiled

Coverage	Filename	Coverage type	Coverage category	Description	Extent[b]	Source[a]
Potassium	K91t	Arc	Monitoring	Potassium present at the SRS, South Carolina in 1991. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Man made gamma	Mm91t	Arc	Monitoring	Man-made gamma present at the SRS, South Carolina in 1991. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Natural background gamma	Nat91t	Arc	Monitoring	Natural background gamma present at the SRS, South Carolina in 1991. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Protactinium	Pa91t	Arc	Monitoring	Protactinium present at the SRS, South Carolina in 1991. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Thorium	Th91t	Arc	Monitoring	Thorium present at the SRS, South Carolina in 1991. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
Uranium	Ur91t	Arc	Monitoring	Uranium present at the SRS, South Carolina in 1991. An Aerial Radiological Survey of the SRS and Surrounding Area Oct.–Nov. 1991.	SRS and just beyond the SRS boundary	SRS Legacy Data, Ver. 1
1951 aerial photographs	Varies with photo	ERDAS lan files	Imagery	Browse Vertical Aerial Photography 1951	SRS	SRS Legacy Data, Ver. 2
1966 aerial photographs	Varies with photo	ERDAS lan files	Imagery	Browse Vertical Aerial Photography 1966. 301 frames of vertical, false color black and white, aerial photography scanned at browse quality (approximately 70 dots per inch(dpi)) at approximately 10 meter resolution.	SRS	SRS Legacy Data, Ver. 2

Table F-1. GIS Data Compiled

Coverage	Filename	Coverage type	Coverage category	Description	Extent[b]	Source[a]
1974 aerial photographs	Varies with photo	ERDAS lan files	Imagery	Browse Vertical Aerial Photography 1974. 347 frames of vertical, black and white, aerial photography scanned at browse quality (approximately 70 dots per inch(dpi)) at approximately 10 meter resolution	SRS	SRS Legacy Data, Ver. 2
1991 aerial photographs	Varies with photo	ERDAS lan files	Imagery	Browse Vertical Aerial Photography 1991. 25 frames of vertical, normal color, aerial photography scanned at browse quality (approximately 70 dots per inch(dpi)) at approximately 30 meter resolution.	SRS	SRS Legacy Data, Ver. 2
1994 aerial photographs	Varies with photo	ERDAS lan files	Imagery	Browse Vertical Aerial Photography 1994. 55 frames of vertical, false color infrared, NAPP, aerial photography scanned at browse quality (approximately 70 dots per inch(dpi)) at approximately 30 meter resolution.	SRS	SRS Legacy Data, Ver. 2
Gnis	gnis	Point	Annotext	The Geographic Names Information System (GNIS). Features include schools, airports, bridges, canals, churches, dams, hospitals, parks, reservoirs, streams, lakes, etc.	SC and GA[b]	ADC
Hydro	hydro	Polygon, Arc	Hydrology	Includes lakes, reservoirs, streams, swamps, and water (seas). Includes arc and polygon coverages.	SC and GA	ADC
Misc	misc	Arc	Transportation	Airports, power lines, substations, pipelines, etc.	SC and GA	ADC
Quads	quads	Poly	Boundaries	1:100,000 quad boundaries and ID number	SC and GA	ADC
Rail	rail	Arc	Transportation	Railroads and railroad bridges	SC and GA	ADC

Table F-1. GIS Data Compiled

Coverage	Filename	Coverage type	Coverage category	Description	Extent[b]	Source[a]
Roads	roads	Arc, annotext	Transportation	Roads (single line). Features include primary, secondary, under and overpasses, ferry, trails, etc. Includes annotation.	SC and GA	ADC
Roads_d	roads_d	Arc, annotext	Transportation	Roads, both sides of road. Features include primary, secondary, under and overpasses, ferry, trails, etc. Includes annotation.	SC and GA	ADC
Land use and land cover	L_"name"	Polygon	Land_uses	Land use and land cover: Boundaries associated with land use classifications such as residential, deciduous forest land, and forested wetland	SC and GA	EPA - Basins
Urbanized areas	urban	Polygon	Land_use	Urbanized areas: Boundaries of census-defined urbanized areas	SC and GA	EPA - Basins
Populated place locations	urban_nm	Point	Land_use	Populated place locations: locations of populated places as represented on USGS topographic maps	SC, GA	EPA - Basins
Reach File, version 1 (RF1)	rf1	Arc	Hydrology	Reach File, version 1 (RF1): Hydrographic database containing over 68,000 reaches to represent surface waters of the continental U.S.	SC and GA	EPA - Basins
Major roads	roads	Arc	Transportation	Major roads: Interstate and state highway network	SC and GA	EPA - Basins
USGS Hydrologic Unit Boundaries (accounting unit)	acc	Polygon	Boundaries	USGS hydrologic unit boundaries (accounting unit): Nationally consistent delineation of the hydrographic boundaries associated with major U.S. river basins	SC and GA	EPA - Basins
USGS Hydrologic Unit Boundaries (cataloging unit)	cat	Polygon	Boundaries	USGS hydrologic unit boundaries (cataloging unit): Nationally consistent delineation of the hydrographic boundaries associated with major U.S. watersheds	SC and GA	EPA - Basins

Table F-1. GIS Data Compiled

Coverage	Filename	Coverage type	Coverage category	Description	Extent[b]	Source[a]
Drinking water supply (DWS) sites	dws	Point	Wells	Drinking Water Supply (DWS) sites: Location of public water supplies, their intakes, and sources of surface water supply	SC and GA	EPA - Basins
Dam sites	dam	Point	Facilities	Dam sites: Inventory of U.S. dams with associated data such as impoundment volume and maximum depth	SC and GA	EPA - Basins
EPA Region	epa_reg	Polygon	Boundaries	EPA region boundaries: Administrative boundaries	SC and GA	EPA - Basins
State boundaries	st	Polygon	Boundaries	State boundaries: Administrative boundaries	SC and GA	EPA - Basins
County boundaries	cnty	Polygon	Boundaries	County boundaries: Administrative boundaries	SC and GA	EPA - Basins
Water quality monitoring station summaries	wq_stat	Point	Water/quality	Statistical summaries of water quality monitoring for 50 physical and chemical-related parameters. Parameter-specific statistics computed by station for 5-year intervals from 1970 to 1994.	SC and GA	EPA - Basins
Bacteria monitoring station summaries	bac_stat	Point	Water/quality	Statistical summaries of water quality monitoring for 10 bacteria-related parameters. Parameter-specific statistics computed by station for 5-year intervals from 1970 to 1994.	SC and GA	EPA - Basins
National Sediment Inventory (NSI)	nsi	Point	Water/quality	Sediment chemistry, tissue residue, and benthic abundance monitoring data for freshwater and coastal sediments	SC and GA	EPA - Basins
Weather station sites	metpt	Point	Meterology	Location of first-order National Oceanic and Atmospheric Administration (NOAA) weather stations used by the SWRRB model	SC and GA	EPA - Basins
USGS gauging stations	gage	Point	Water/quantity	Inventory of surface water gauging station data including 7Q10 low and monthly mean stream flow	SC and GA	EPA - Basins

Table F-1. GIS Data Compiled

Coverage	Filename	Coverage type	Coverage category	Description	Extent[b]	Source[a]
Permit Compliance System (PCS) sites and computed loadings	pcs	Point	Contam_source	NPDES permit-holding facility information. Contains parameter-specific loadings to surface waters computed using the EPA Effluent Decision Support System (EDSS).	SC and GA	EPA - Basins
Industrial Facilities Discharge (IFD) sites	ifd92	Point	Contam_source	Industrial Facilities Discharge (IFD) sites: Facility information on industrial point source dischargers to surface waters	SC and GA	EPA - Basins
Toxic Release Inventory (TRI) sites, 1992 Release	tri	Point	Contam_source	Facility information from the 1992 TRI public data release. Contains Yes/No flags for each facility indicating media-specific reported releases.	SC and GA	EPA - Basins
Superfund National Priority List Sites	Npl	Point	Contam_source	Superfund National Priority List sites: Location of Superfund National Priority List sites	SC and GA	EPA - Basins
Border Counties	brdrcnty	Arc	Boundaries	SRS P2 study are boundary for the counties downstream of the SRS and adjacent to the Savannah River	Part of the SRS phase 2 study area	RAC
SRS 50 mile buffer	SRS50buf	Arc	Boundaries	Boundary created from 50 mile buffer around the SRS boundary. Also includes Columbia, SC.	Part of the SRS phase 2 study area	RAC
SRS 30 mile circle	SRS30mi	Arc	Boundaries	Approximate 30 mile circle centered on the SRS	Part of the SRS phase 2 study area	RAC
SRS 1 mile buffer	SRS1mi	Arc	Boundaries	Boundary created from 1 mile buffer around the SRS boundary.	Part of the SRS phase 2 study area	RAC
SRS Facilities	SRSfacil	Polygon	Facilities	Polygons for the primary SRS facility areas.	SRS	RAC
USGS GW wells	Usgsgwwq	Point	Wells	USGS ground water monitoring wells within the study area	Entire SRSP2 study area	RAC

Table F-1. GIS Data Compiled

Coverage	Filename	Coverage type	Coverage category	Description	Extent[b]	Source[a]
Counties	Cntys50, cntys50.e00	Polygon	Boundaries	Counties within 50 miles of the center of SRS	Most of the SRSP2 study area	SRS
Polar section grid for SRS area	Srspie, srspie.e00	Polygon	Boundaries	50 mile radius and a 16 section polar grid centered on SRS. See srspiecp.xls for attributes for each section	50 mile from center of SRS	GA Tech. and SRS
Clipped land cover data	Srspieclip, srspieclip.e00	Polygon	Land_use	Clip polygon for crop production within 50 mile buffer of SRS, See srspiecp.xls	50 mi from center of SRS	GA Tech. and SRS
1970 NALC data	102174.lan	Imagery	Imagery	10/21/74 EPA North American Landscape Characterization (NALC) imagery, Landsat MSS data resampled to 60-m resolution	Most of the study area	NASA, USGS, EPA
1980 NALC data	101886.lan	Imagery	Imagery	10/18/86 EPA (NALC) imagery, Landsat MSS data resampled to 60-m resolution	Most of the study area	NASA, USGS, EPA
1990 NALC data	92790.lan	Imagery	Imagery	9/27/90 EPA NALC imagery, Landsat MSS data resampled to 60-m resolution	Most of the study area	NASA, USGS, EPA
NALC DEM	Nalc.dem	DEM	Topography	DEM data for NALC imagery	Most of the study area	NASA, USGS, EPA

[a] SRS Legacy Data = Legacy GIS Coverages, Version 1; Version 2
ADC = GA and SC Cornerstone data obtained from American Digital Cartography, Inc.
EPA Basins = USEPA data from Region 4 BASINS CD, Version 1.
GA Tech. and SRS = Twining 1998
NASA, USGS, EPA = NASA Pathfinder data, North American Landscape Characterization Project
[b] SC = South Carolina, GA = Georgia.

Map Development

A number of maps have been developed to support the project investigators and communication with the public. Table F-2 is a list of these maps.

Table F-2. Maps Produced for the SRS Project

File name	Description of map or figure	Paper Size
census_blk_base_map_a-p.ps	Census blocks: old study area boundary; doesn't include all of Beaufort CO.	a
census_blk_base_map_d-p.ps	Census blocks: old study area boundary; doesn't include all of Beaufort CO.	d
census_blk_base_map_e-p.ps	Census blocks: old study area boundary; doesn't include all of Beaufort CO.	d
feed_past_25mile_a-l.ps	Feedlots/croplands and pasture approximately 25 miles beyond SRS boundary	a-l
med_25mile_gt50k_a-l.ps	SRS with 25 mile buffer; example query of census blocks with median family income greater than $50,000	a-l
pop_25mile_gt3000_a-l.ps	SRS with 25 mile buffer, example query of census blocks with greater than 3,000 people	a-l
sc_road_city-a.ps	SRS and surrounding towns with major roads, 35 mile buffer around SRS	a
sc_road_city-d.ps	SRS and surrounding towns with major roads, 35 mile buffer around SRS	d
Srs_basemap_a-l.ps	B&W basemap of SC and GA, county boundaries, and 50 mile buffer around SRS.	
srs_hydro_wet-a.ps	SRS - hydrography, wetlands, facility areas and major roads	a
srs_hydro_wet-d.ps	SRS - hydrography, wetlands, facility areas and major roads	d
srs_hydro-a.ps	SRS - hydrography, wetlands, facility areas and major roads	a
srs_hydro-d.ps	SRS - hydrography, wetlands, facility areas and major roads	d
srs_landuse_map.ps	Land use for the study area	c
srs_landuse_map_c-l.ps	Land use for the study area	c-l
srs_landuse_map_e-l.ps	Land use for the study area	e-l
srs_road-a.ps	SRS - major roads, facility areas, and hydrography	a
srs_road-d.ps	SRS - major roads, facility areas, and hydrography	d
srs_road2-d.ps	SRS - all roads, buildings, and hydrography	d
srs_samp_grid_map_a-l.ps	SRS map with sample grid	a-l
srs_samp_grid_map_c-l.ps	SRS map with sample grid	c-l
srs_samp_grid_map_e-l.ps	SRS map with sample grid	e-l
srs_shaded_relief_map-24x28.ps	Shaded relief map of SRS	24 × 28 in.

File name	Description of map or figure	Paper Size
srs_shaded_relief_map-e-90.ps	Shaded relief map of SRS	e
srs_study_area_a-p.ps	Entire study area	a
srs_study_area_d-p.ps	Entire study area – old study area map, doesn't include all of Beaufort CO	d
srs_study_area_e-p.ps	Entire study area – old study area map, doesn't include all of Beaufort CO	e

RAC CD-ROM Production

A CD-ROM has been produced that includes data compiled for this study. The list below provides a general description of the GIS and demographic data directories and files housed on the CD. Several GIS data sets compiled by *RAC* are not included on the *RAC* CD because the data are copyrighted; these are the American Digital Cartography Cornerstone Data and the SRS Legacy GIS Coverages. These files were transmitted to the CDC on their original CDs with the Preliminary Draft Report, in September 1998. Other data compiled and reviewed may not be included because there was no documentation for the data. Figure F-3 illustrates the GIS and demographic data directories and files found on the Final SRSDRP2-CD.

Title: "Final SRSDRP2-CD"

GIS and demographic data directories include:

ADCsupport

This folder contains a help file that discusses some of the files obtained from American Digital Cartography. The file provides background information on numerous types of GIS data, only some of which were obtained for this study.

arcexplorer

This folder contains ArcExplorer software developed by Environmental Systems Research Institute, Inc. (ESRI). This freeware executable file will setup an application that allows the user to view the GIS coverages on the *RAC* CD. This software is discussed further in the "readme.doc" file on the CD.

demog

These are the SCSU demographics data collected and entered into Excel tables. The file names used are the same as those in the Demographic Data Table (Table F-3) that documents data types and sources. This directory also includes the food and lifestyle survey data and the oracle database export file.

earthinfo_files

These files are export files obtained from EarthInfo. They are primarily .dbf and .xls files that were extracted to support development of monitoring station GIS coverages. Only the groundwater station location data were developed into a GIS coverage. The remainder of the files can be readily converted to GIS coverages if desired.

map_files

These are map postscript files developed for the project (see Table F-2).

srs_gis_data

This directory has several subdirectories containing GIS data that can be viewed by ArcExplorer software provided on this CD. This entire directory must be copied to the correct drive on your computer for the GIS project .aep files to work correctly. See the "readme.doc" file on the CD for an explanation regarding how to view the GIS data.

aep_files
This folder contains one or more .aep files that will be used to view GIS data.

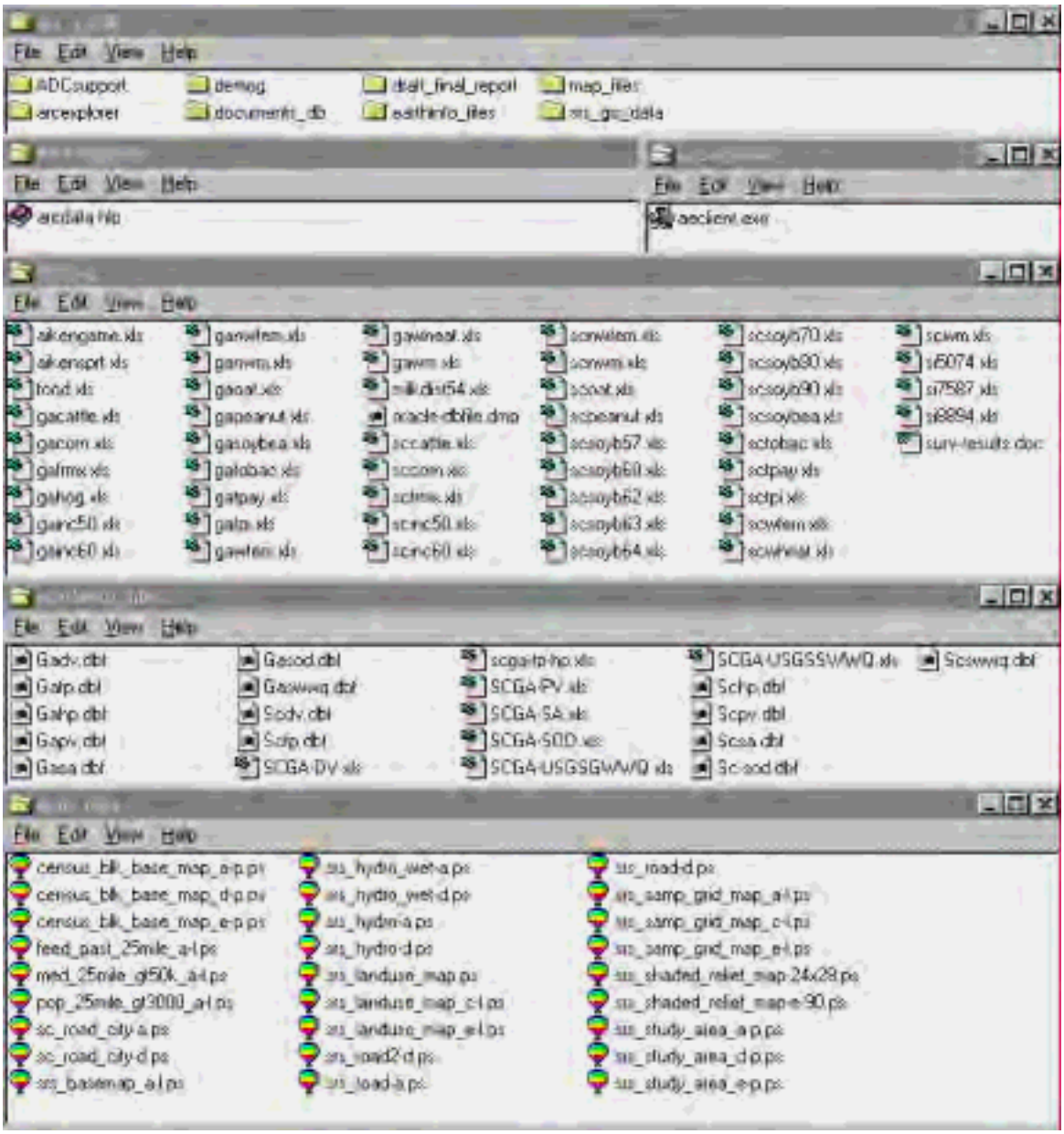
basins-data
These data are from the EPA Region 4, Better Assessment Science Integrating Point and Nonpoint Sources (BASINS)

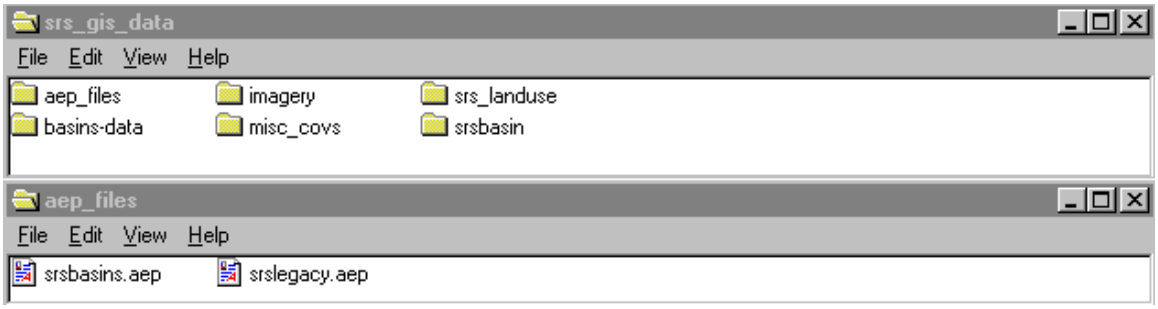
imagery
USEPA-USGS North American Landscape Characterization (NALC) data. This is Landsat MSS data from the early 70's, 80's, and 90's. These data are in ERDAS .lan file format, so they can be viewed using ArcView or ArcExplorer.

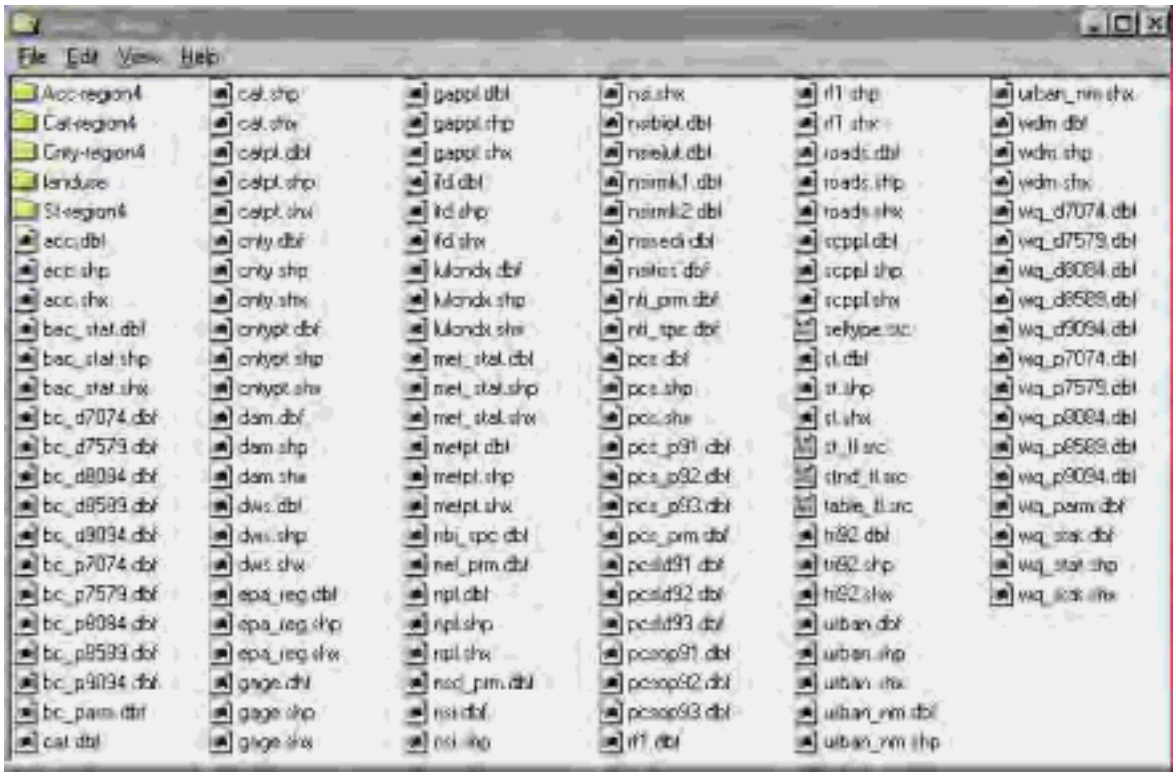
misc_covs
These are several coverages generated by RAC.

srs_landuse
These are files from SRS, originally generated by Georgia Tech to support food production and food chain analysis for the area within fifty miles from the center of SRS.

srsbasin
These are several SRS boundary and other coverages that have been projected to be compatible with the EPA BASINS GIS data.


File Edit View Help
ADCsupport
demog
draft_final_report
map_files
arcexplorer
documents_db
earthinfo_files
srs_gis_data
aeclaim.hlp
aeclient.exe
sikengame.xls
ganwtem.xls
gawnwat.xls
scnwtem.xls
scsoyb70.xls
scwm.xls
sikensport.xls
ganwm.xls
gawm.xls
scnwm.xls
scsoyb80.xls
si6074.xls
food.xls
gaoat.xls
milkdist54.xls
scoat.xls
scsoyb90.xls
si7587.xls
gacattle.xls
gapeanut.xls
oracle-dbfile.dmp
scpeanut.xls
scsoybea.xls
si8894.xls
gacorn.xls
gasoybea.xls
sccattle.xls
scsoyb57.xls
sctobac.xls
surv-results.doc
gafmx.xls
galobac.xls
sccorn.xls
scsoyb58.xls
sctpay.xls
gahog.xls
gatpay.xls
scfmx.xls
scsoyb62.xls
sctpi.xls
ganc50.xls
gatpi.xls
scnc50.xls
scsoyb63.xls
scwtem.xls
ganc60.xls
gawtem.xls
scnc60.xls
scsoyb64.xls
scwnwat.xls
Gadv.dbf
Gasod.dbf
scgatp-hp.xls
SCGA-USGSSWWQ.xls
Scswwq.dbf
Gatp.dbf
Gaswwq.dbf
SCGA-PV.xls
Sctp.dbf
Gahp.dbf
Sodv.dbf
SCGA-SA.xls
Scpv.dbf
Gapv.dbf
Sctp.dbf
SCGA-SOD.xls
Scsa.dbf
Gasa.dbf
SCGA-DV.xls
SCGA-USGSGWWQ.xls
Sc-sod.dbf
census_blk_base_map_a-p.ps
srs_hydro_wet-a.ps
srs_road-d.ps
census_blk_base_map_d-p.ps
srs_hydro_wet-d.ps
srs_samp_grid_map_a-l.ps
census_blk_base_map_e-p.ps
srs_hydro-a.ps
srs_samp_grid_map_c-l.ps
feed_past_25mile_a-l.ps
srs_hydro-d.ps
srs_samp_grid_map_e-l.ps
med_25mile_gt50k_a-l.ps
srs_landuse_map.ps
srs_shaded_relief_map-24x28.ps
pop_25mile_gt3000_a-l.ps
srs_landuse_map_c-l.ps
srs_shaded_relief_map-e-90.ps
sc_road_city-a.ps
srs_landuse_map_e-l.ps
srs_study_area_a-p.ps
sc_road_city-d.ps
srs_road2-d.ps
srs_study_area_d-p.ps
srs_basemap_a-l.ps
srs_road-a.ps
srs_study_area_e-p.ps


srs_gis_data
File Edit View Help
aep_files
imagery
srs_landuse
basins-data
misc_covs
srsbasin
aep_files
File Edit View Help
srsbasins.aep
srslegacy.aep


File Edit View Help
Acc-region4
Cat-region4
Cnty-region4
landuse
St-region4
acc.dbf
acc.shp
acc.shx
bac_stat.dbf
bac_stat.shp
bac_stat.shx
bc_d7074.dbf
bc_d7579.dbf
bc_d8084.dbf
bc_d8589.dbf
bc_d9094.dbf
bc_p7074.dbf
bc_p7579.dbf
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bc_p9094.dbf
bc_parm.dbf
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cat.shx
catpt.dbf
catpt.shp
catpt.shx
cnty.dbf
cnty.shp
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epa_reg.shp
epa_reg.shx
gage.dbf
gage.shp
gage.shx
gappl.dbf
gappl.shp
gappl.shx
ifd.dbf
ifd.shp
ifd.shx
kilcndx.dbf
kilcndx.shp
kilcndx.shx
met_stat.dbf
met_stat.shp
met_stat.shx
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metpt.shp
metpt.shx
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nel_prm.dbf
npl.dbf
npl.shp
npl.shx
nsd_prm.dbf
nsi.dbf
nsi.shp
nsi.shx
nsibipt.dbf
nsieiut.dbf
nsimk1.dbf
nsimk2.dbf
nsiedi.dbf
nsiles.dbf
nti_prm.dbf
nti_spc.dbf
pcs.dbf
pcs.shp
pcs.shx
pcs_p91.dbf
pcs_p92.dbf
pcs_p93.dbf
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pcsld91.dbf
pcsld92.dbf
pcsld93.dbf
pcsop91.dbf
pcsop92.dbf
pcsop93.dbf
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roads.shp
roads.shx
scppl.dbf
scppl.shp
scppl.shx
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st.shp
st.shx
st_ll.inc
stnd_ll.inc
table_ll.inc
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tri92.shp
tri92.shx
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urban.shp
urban.shx
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urban_nm.shp
urban_nm.shx
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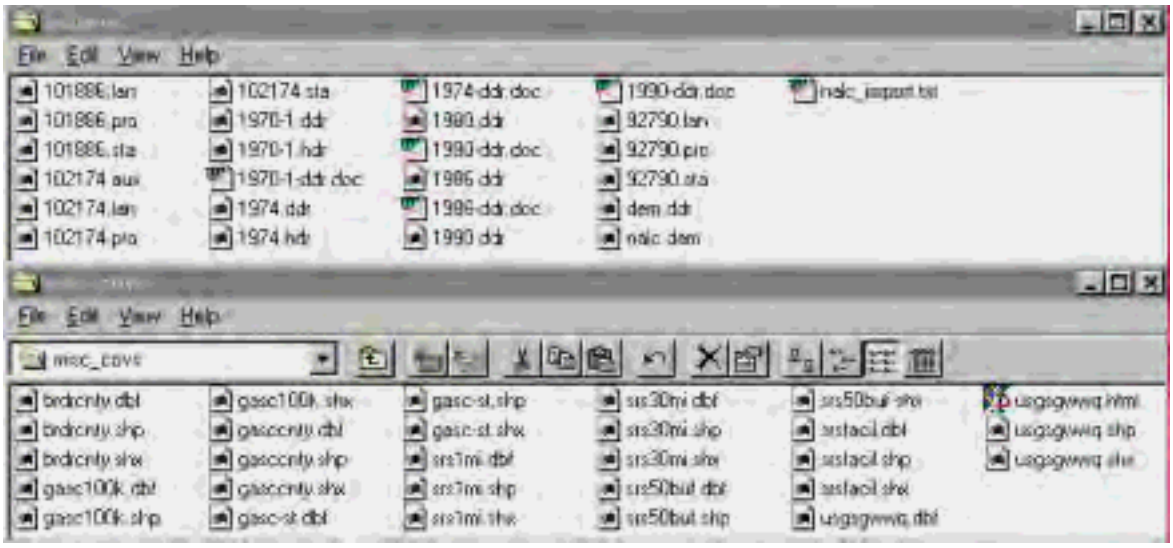
File Edit View Help
101886.lan
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1970-1.ddr
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1974.ddr
1974.hdr
1974-ddr.doc
1980.ddr
1980-ddr.doc
1986.ddr
1986-ddr.doc
1990.ddr
1990-ddr.doc
92790.lan
92790.pro
92790.sta
dem.ddr
nsic.dem
nsic_import.txt
misc_covs
brdrcnty.dbf
brdrcnty.shp
brdrcnty.shx
gasc100k.dbf
gasc100k.shp
gasc100k.shx
gasccnty.dbf
gasccnty.shp
gasccnty.shx
gasc-st.dbf
gasc-st.shp
gasc-st.shx
srs1mi.dbf
srs1mi.shp
srs1mi.shx
srs30mi.dbf
srs30mi.shp
srs30mi.shx
srs50bui.dbf
srs50bui.shp
srs50bui.shx
srstacil.dbf
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srstacil.shx
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usgsgwwq.html
usgsgwwq.shp
usgsgwwq.shx

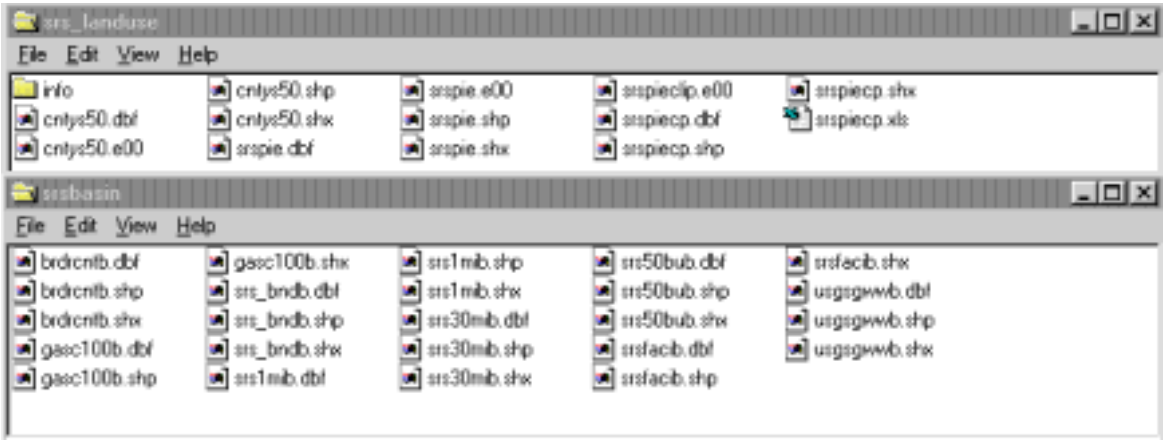


Figure F-3. Directories and files found on the Final SRSDRP2-CD.

Selecting the Study Area Extent

Phase II of the SRS dose reconstruction study required reviewing the thousands of documents collected during the study's first phase and collecting the information to be used to estimate offsite historical radiation and chemical doses and risks. Much of the information collected was geographically based (map-based). We needed to find information showing where groups of people lived while SRS releases were occurring and what food crops and animals were being grown in the region. We also needed to know what water supplies were being used and in which directions the winds were blowing during the years of the releases. Much of this information can be recorded as numbers on a map (for example, the number of people living in a certain town or county during 1960). *RAC* has collected such information in great detail, and we decided, with the help of the SRS Citizens' Health Effects Subcommittee, how large the area being studied should be. *RAC* recommended to the CDC and to the HES that the study area for such data collection be limited to a 50-mi distance from the center of the SRS. While it would be reasonable to select either larger or smaller study areas, our experience performing similar studies in Ohio, Colorado, Utah, and Washington State lead us to recommend a limited radius for several reasons.

One of these reasons has to do with the ways in which past radiation and chemical exposures of people who lived near the Site will be determined. There are several methods used to make such calculations, and some are much more accurate than others. The better methods require more detailed information than is usually available, however.

Radiation or chemical doses are best reconstructed from a series of measurements of the concentrations of specific radionuclides or chemicals in the bodies of those people exposed. If the quantity of a radioactive material in a person's body over time is known, a reasonably accurate calculation of the radiation dose can be made and the risk that material would produce over time. While such measurements are occasionally available (perhaps for some time after an accidental release occurred), such information is not routinely collected when exposures are likely to be low.

In the absence of such data, scientists responsible for estimating historical doses use other, less accurate approaches. For example, there may be measurements from air samplers running at various offsite locations that recorded the concentrations of airborne radionuclides or chemical to which people near the samplers were exposed. There may also be similar information from the

analysis of water, soil, vegetation, crops, animals, and animal products. Such information may be used to estimate the human uptake (from breathing, eating, or drinking) of the measured radionuclides. Once estimates of uptake are made, we can calculate how much of the radionuclide or chemical accumulated within people living in the area and then estimate dose and risk. Estimates of accumulation in the body are, in general, less accurate using this approach than if direct, in-the-body measurements were available.

When measurements of radionuclides or chemicals in air, water, and foods are not available, it is usually necessary to use estimates of the facility's releases, both routine and accidental, to calculate offsite human exposure. These release estimates (called the source term) can be used by computer models of the movement of materials in the environment to estimate offsite air, water, and food concentrations of released materials. Calculations of exposure, dose, and risk can then be made; however, they are less accurate than would be the case if actual, offsite measurements had been taken. Most radiation and chemical dose assessments rely heavily on this last approach because better information is rarely available over an extended period of time.

While dose calculations could be made for people living at any distance from a site, the estimates are usually limited to a specific area. This is because as released chemicals or radionuclides releases drift away from a site, they disperse, deposit, and become less concentrated in the air. Smaller doses are, thus, calculated to occur at increasing distances. The decision is usually made to limit a study's area to avoid spending time calculating very small doses at very large distances. For specific cases, doses might be calculated for greater distances, such as when an accident results in the release of larger than usual quantities of radionuclides for a short period.

This general decrease in dose at increasing distance has been studied carefully and is well understood. For example, researchers have performed experiments to examine the accuracy of computer models used to predict doses at increasing distances from a release site (Fields, et al. 1984). One conclusion of these studies is that doses are relatively small and increasingly inaccurate at great distances from the site of release. It is reasonable to exclude from the study area those regions in which very small dose and risk are likely.

The large set of detailed, geographically based data and satellite imagery necessary to support this study requires a significant amount of time to acquire, format, and document properly. Developing information sets for regions beyond a 50-mi radius from the SRS would add little value to the overall dose assessment and would use resources (staff, time, and money) better spent focusing closer to the Site.

For these reasons, *RAC* recommended limiting the assessment area for the SRS dose reconstruction research to a 50-mi buffer from the Site's boundary. Communities located outside this radius but potentially using drinking water from the Savannah River are also included in the study area. Figure F-1 shows this recommended study area.

DEMOGRAPHIC DATA COMPILATION

Introduction

The ultimate goal of a dose reconstruction project is an estimation of expected numbers of health effects that would be associated with exposure of members of a particular population to contaminants from the facility or process under study. The purpose of establishing a demographic database is to compile information for those populations that may have been affected by releases of chemicals and/or radionuclides from the SRS. The task for the GIS Laboratory at SCSU is to design and implement a relational database showing where to find and how to retrieve information about the population dynamics, agricultural practices, and human lifestyles for all counties in the Savannah River region.

The data collection process is relatively straightforward for the years since 1980 because the data are available and accessible in a digital format. However, before 1965 demographic and agricultural data are incomplete, documented on paper and, therefore, more difficult to retrieve. Some statistical methods have been used to generate the estimates for data before 1965.

It has been difficult identifying lifestyle data consisting of eating habits, time spent outdoors, and time spent indoors for the population living in the Savannah River region from the 1950s to the present. To obtain the most accurate account of such lifestyle data, a survey was conducted to determine the most appropriate and valid estimates for this area. All the data are for the targeted counties within South Carolina and Georgia within the study area.

To document data sources and provide easy access to the metadata, a data set catalogue containing the names and phone numbers of each individual that aided in collecting the data sets was developed. This catalogue is maintained and updated upon the arrival and retrieval of new data. Each data set includes a data dictionary that allows easy access to the tables and their field names.

Demographic Database

The data sets in the database include agriculture, economic, lifestyle, and demographic data for counties within the study area. The information has been acquired from various sources but primarily from government agencies that are believed to be reliable.

The data collection and database development followed a systematic and functional approach. Generally, the following components were considered during the database development:

- The project tasks
- Logical translation into a GIS database
- GIS system standards (hardware and software)
- GIS database design and standards.

The following components and factors were considered during the database design process:

- File system organization
- Naming conventions
- Spatial data automation standards
- Coordinate system and scale

- Data description
- Access and security
- Maintenance and updates
- Integration with GIS.

System Standards

Software

ESRI's Arc/INFO, GRID, TIN, NETWORK, ARCEDIT, ARCPLOT, TABLES, INFO, ARC Macro language (AML), DATABASE INTEGRATOR, IMAGE INTEGRATOR, ArcStorm, ArcView, ArcDoc, ArcPress, ArcScan, and MapObjects are standard vector based GIS software products for the database.

ERDAS IMAGINE is raster based, standard GIS and image processing software for the database.

Oracle, dBASE, FoxPro are standard database management systems (RDBMS) used to support the database and application development.

Microsoft NT, Window95, and Solaris 2.5 are standard operation systems.

Naming Conventions

1. The demographic database for the project in the GIS Laboratory at SCSU is designed to ensure compatibility with DOS and CD-ROM drives. It is defined as up to eight characters, followed by a period and up to three characters as a file extension.
2. The database has been designed by assigning an abbreviation code for each data set. It is used in associated names. Normally, the code is a combination of the state name and the first character of each word of the data set's names; they are designed to be as meaningful as possible. The abbreviation code may vary from two to seven characters depending on practical situations. A typical example is GAFPT, a code for a data set called "Georgia Full-Part Time Employment."
3. A two-digit number is used to describe the temporal data sets. For example, "1980 Population Dynamics for South Carolina" has an abbreviation of "scpop80" and "1950 Population Dynamics for Georgia" is abbreviated "gapop50."
4. Standard file extensions are used. This includes Oracle file extensions, dBASE file extensions, Microsoft Excel extension, Arc/INFO related file extension, ArcView file extensions, ERDAS IMAGINE extensions, and ordinary ASCII text file extensions.

Data Automation Standards

The SCSU GIS Laboratory automates data with three approaches: data collection, data conversion, and data entry. The data automation plan usually follows this order for a new data set generation:

- Identify automation need based on the tasks
- Determine automation methods (conversion, collection, or entry)
- Implement data quality control procedure.

Data Collection, Conversion, and Entry

The GIS Laboratory is responsible for identifying automation and update needs for new and existing data and their contents. This effort implements standards for quality, content, and transferability. The GIS Laboratory is also responsible for determining data automation methods. For example, data conversion from existing ASCII text, data collection using government files, field survey or other statistical and mathematical methods, and data entry by scan or manual. The Laboratory is responsible for implementing the automation plan and performing the data quality control procedures.

Data Quality Control Procedures

The GIS Laboratory implements a combined quality assurance (QA) procedure for data QA and is responsible for performing and overseeing all data –quality control procedures and activities for the SRS dose reconstruction project at SCSU.

The GIS Laboratory uses the following quality control (QC) procedures to verify accuracy and completeness of the data collected:

- Assess the quality of data if it is in hard copy format
- Check for omissions, incomplete numbers, and copying visibility
- Scan the hard copy data
- Review the scanned data for accuracy in the scan process
- Arrange data in the appropriate format to be entered
- Double check data for errors and accuracy against the original hard copy
- Construct the Database structure
- Enter the completed data into the database table
- Document the source from which the data was obtained
- Keep tracking and reporting progresses of data automation process, make necessary data quality upgrades, enforce consistently the data quality control procedures.

Data and File System Backup

The GIS Laboratory is responsible for the custodianship, retention, protection and backup of all the collected data and file systems currently. To ensure the protection of all data, the Laboratory follows the GRANDFATHER, FATHER, SON scheme for the backup of both file system and data files.

Each Monday through Wednesday and Friday, all the data that have been created or modified will be backed up using the appropriate tape backup unit (TBU). This backup tape is referred to as the SON. Each SON tape will be kept for a minimum of 1 month. Each Thursday a complete backup of all system and data files is completed using the appropriate TBU. This backup tape is referred to as the FATHER. Each FATHER tape will be kept for a minimum of 1 month. On the last Thursday of the month, a complete backup of all system and data files is to be completed using the appropriate TBU. This backup tape is referred to as the GRANDFATHER. Each GRANDFATHER tape will be kept for a minimum of 2 years. All GRANDFATHER tape(s) are stored at an approved offsite location.

Data Sets in the Database

In the dose reconstruction database, data sets are grouped into four primary groups: agricultural data, economic data, demographic data, and lifestyle data. Summary information for each data set is provided in Table F-3, which includes a description of items listed in the data table and source contacts. These files are found in the "demog" directory on the Final SRSDRP2-CD.

Table F-3. Description of Demographic Data Files

File Name (xls)	Data Description	Data Source	Analyses Performed on Data
Aikengame	Hunting Practices in Crackerneck Wildlife Management Area (Aiken County)	Mike Caudell SC Department of Natural Resources (803) 734-3886	
Aikensprt	Sportsman Type in Crackerneck Wildlife Management Area (Aiken County)	Mike Caudell SC Department of Natural Resources (803) 734-3886	
Food	Results of Food Surveys Taken in 10 Targeted Counties for SC & GA	Wei Yang/Leonard Gore 1890 Research & Extension SC State GIS Lab (803) 536-8461	Statistical (frequency distributions)
Gacattle	Cattle Inventory for Georgia	Al Smith, Univ. of GA (Athens), College of Agriculture and Environmental Sciences (706) 542-0900 Jim Brueggen, National Agriculture Statistics Service (202) 720-488	
Gacorn	Corn Grain Harvested And Yield	Al Smith, Univ. of GA (Athens), College of Agriculture and Environmental Sciences (706) 542-0900 Jim Brueggen, National Agriculture Statistics Service (202) 720-488	
GAFRMX	GA Econ Data (Farm Expenditures)	N/A U.S. Bureau of the Census- Bureau of Economic Analysis (202) 606-5360	

File Name (xls)	Data Description	Data Source	Analyses Performed on Data
Gahog	Georgia Hog Inventory	Al Smith, Univ. of GA (Athens), College of Agriculture and Environmental Sciences (706) 542-0900 Jim Brueggen, National Agriculture Statistics Service (202) 720-488	
Gainc50	1950 Econ and Estimates for GA	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	Yes - Estimates are interpolated. The estimates were derived from economic formulation recommended by SC State Univ. School of Business.
Gainc60	1960 Econ Data and Estimates for GA	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	Yes - Estimates are interpolated. The estimates were derived from economic formulation recommended by SC State Univ. School of Business.
Ganwfem	Population Dynamics for Georgia non-white Female by age	Marty Sik Georgia Office of Planning and Budget (Office of Research and Statistical Services) (404) 656-0911 Wei Yang 1890 Research GIS Lab (803) 536-8921	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10. The number of years for current estimate is then added to prior decennial census year
Ganwm	Population Dynamics for Georgia non-white Male by age	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10. The number of years for current estimate is then added to prior decennial census year
Gaoat	Georgia Oats Harvested and Yield	Al Smith, Univ. of GA (Athens), College of Agriculture and Environmental Sciences (706) 542-0900 Jim Brueggen, National Agriculture Statistics Service (202) 720-488	

File Name (xls)	Data Description	Data Source	Analyses Performed on Data
Gapeanut	Peanut Harvested And Yield	Al Smith, Univ. of GA (Athens), College of Agriculture and Environmental Sciences (706) 542-0900 Jim Brueggen, National Agriculture Statistics Service (202) 720-488	
Gasoybea	Georgia Soybeans Harvested and Yield	Al Smith, Univ. of GA (Athens), College of Agriculture and Environmental Sciences (706) 542-0900 Jim Brueggen, National Agriculture Statistics Service (202) 720-488	
Gatobac	Georgia Tobacco Harvested and Yield	Al Smith, Univ. of GA (Athens), College of Agriculture and Environmental Sciences (706) 542-0900 Jim Brueggen, National Agriculture Statistics Service (202) 720-488	
GATPAY	GA Econ Data (Transfer of Payments)	N/A U.S. Bureau of the Census- Bureau of Economic Analysis (202) 606-5360	
GATPI	GA Econ Data (Total Personal Income)	N/A U.S. Bureau of the Census- Bureau of Economic Analysis (202) 606-5360	
Gawfem	Population Dynamics for Georgia white female by age	Marty Sik Georgia Office of Planning and Budget (Office of Research and Statistical Services) (404) 656-0911	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10. The number of years for current estimate is then added to prior decennial census year

File Name (xls)	Data Description	Data Source	Analyses Performed on Data
Gawheat	Georgia Wheat Harvested and Yield	Al Smith, Univ. of GA (Athens), College of Agriculture and Environmental Sciences (706) 542-0900 Jim Brueggen, National Agriculture Statistics Service (202) 720-488	
Gawm	Population Dynamics for Georgia white Male by age	Marty Sik Georgia Office of Planning and Budget (Office of Research and Statistical Services) (404) 656-0911 Wei Yang 1890 Research GIS Lab (803) 536-8921	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10. The number of years for current estimate is then added to prior decennial census year.
Milkdist	Milk Distribution in 1950's for Georgia and South Carolina	Andrew Bouville National Cancer Institute (301) 496-9326	
Sccattle	Cattle Inventory for South Carolina	Steve Pavlesek S.C Agricultural Statistics Service (803) 765-5333 Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
Sccorn	South Carolina Corn Harvested and Yield	Steve Pavlesek S.C Agricultural Statistics Service (803) 765-5333 Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
SCFRMX	SC Econ Data (Farm Expenditures)	N/A U.S. Bureau of the Census- Bureau of Economic Analysis (202) 606-5360	
Scinc50	1950 Econ and Estimates for SC	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10

File Name (xls)	Data Description	Data Source	Analyses Performed on Data
		(803) 536-8461	years and divides them by 10. The number of years for current estimate is then added to prior decennial census year.
Scinc60	1960 Econ and Estimates for SC	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10. The number of years for current estimate is then added to prior decennial census year.
Scnwfem	Population Dynamics for South Carolina non-white female by age	Diane Tester South Carolina Budget and Control Board (Census Data Center) (803) 734-3782 Wei Yang 1890 Research GIS Lab (803) 536-8921	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10. The number of years for current estimate is then added to prior decennial census year.
Scnwm	Population Dynamics for South Carolina non-white by age	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10. The number of years for current estimate is then added to prior decennial census year.
Scoat	South Carolina Oat Harvested and Yield	Steve Pavlesek S.C. Agricultural Statistics Service (803) 765-5333 Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
Scpeanut	South Carolina Peanut Harvested and Produced	Steve Pavlesek S.C Agricultural Statistics Service (803) 765-5333 Jim Brueggen National Agriculture Statistics Service (202) 720-4889	

File Name (xls)	Data Description	Data Source	Analyses Performed on Data
Scsoyb57	South Carolina Soybeans Harvested and Yield in 1957	Steve Pavlesek S.C Agricultural Statistics Service (803) 765-5333 Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
Scsoyb60	South Carolina Soybeans Harvested And Yield in 1960	Steve Pavlesek S.C Agricultural Statistics Service (803) 765-5333 Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
Scsoyb62	South Carolina Soybeans Harvested And Yield in 1962	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	
Scsoyb63	South Carolina Soybeans Harvested and Yield in 1963	Marty Sik Georgia Office of Planning and Budget (Office of Research and Statistical Services) (404) 656-0911 Wei Yang 1890 Research GIS Lab (803) 536-8921	
Scsoyb64	South Carolina Soybeans Harvested and Yield in 1964	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	
Scsoyb70	South Carolina Soybeans Harvested and Yield in 1970	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	

File Name (xls)	Data Description	Data Source	Analyses Performed on Data
Scsoyb80	South Carolina Soybeans Harvested and Yield in 1980	Diane Tester South Carolina Budget and Control Board (Census Data Center) (803) 734-3782 Wei Yang 1890 Research GIS Lab (803) 536-8921	
Scsoyb90	South Carolina Soybeans Harvested and Yield in 1990	Wei Yang 1890 Research & Extension SC State GIS Lab (803) 536-8461	
Scsoybea	South Carolina soybean data, all years	Steve Pavlesek S.C Agricultural Statistics Service (803) 765-5333 Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
Sctobac	South Carolina Tobacco Harvested and Yield	Steve Pavlesek S.C Agricultural Statistics Service (803) 765-5333 Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
SCTPAY	SC Econ Data (Transfer of Payment)	N/A U.S. Bureau of the Census- Bureau of Economic Analysis (202) 606-5360	
SCTPI	SC Econ Data (Total Personal Income)	N/A U.S. Bureau of the Census- Bureau of Economic Analysis (202) 606-5360	
Scwfem	Population Dynamics For South Carolina White female by age	Diane Tester South Carolina Budget and Control Board (Census Data Center) (803) 734-3782 Wei Yang 1890 Research GIS Lab (803) 536-8921	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10. The number of years for current estimate is then added

File Name (xls)	Data Description	Data Source	Analyses Performed on Data
		(803) 536-8921	to prior decennial census year.
Scwheat	South Carolina Wheat Harvested and Yield	Steve Pavlesek S.C Agricultural Statistics Service (803) 765-5333 Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
Scwm	Population Dynamics for South Carolina white male by age	Diane Tester South Carolina Budget and Control Board (Census Data Center) (803) 734-3782	Yes - Estimates are interpolated. The formula for deriving estimates subtracts the two decennial census years and divides them by 10. The number of years for current estimate is then added to prior decennial census year.
Si5074	Corn Grain Silage Yield and Production (1950-1974)	Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
Si7587	Corn Grain Silage Yield and Production (1975-1987)	Jim Brueggen National Agriculture Statistics Service (202) 720-4889	
Si8894	Corn Grain Silage Yield and Production (1988-1994)	Jim Brueggen National Agriculture Statistics Service (202) 720-4889	

Database Security and Access

The dose reconstruction project database security involves protecting the whole database file system. Generally, there is no direct public access to the project database file system. The access to the data, as stored in the project file system, is restricted using operating system security tools.

The file system access rights are controlled by the host computer (UNIX, NT) administrator. Usually, there is no limitation for any internal GIS staff who have access to the UNIX and NT environment and want to view the file system. However, access (write and execute) rights are

restricted to the limited staff working on the dose reconstruction project. The access rights to the user account area is defined based on the operating system security by the system administrators.

There are four levels of access rights to the *RAC* project database file system:

- *Administrator* (GIS manager and host computer administrator): Can create, destroy, or modify the file system; define database structure and tile system; begin and end transactions; and set user access rights.
- *Manager* (GIS manager): Has read and write access to the database; controls transactions; creates and updates data; and can reassign transactions owned by an operator.
- *Operator* (GIS developers and analysts): Has read and limited write access to the database.
- *None* (all other internal and external users): No access to the database and file system.

Food and Lifestyle Survey

A food and lifestyle survey was conducted to learn about the dietary habits of people in the Georgia and South Carolina study area. The survey was designed to collect information on lifestyle activities (food intake, leisure time and work obligations). It is important to survey dietary habits so that estimates of human doses or exposure from ingestion can be based on where food was grown and how much was consumed. For example, for a particular population group (i.e., black males) within the study area, surveys can provide information on their eating habits and about the specific foods they may have consumed. This information is important because eating habits of certain population groups may yield a greater chance of exposure to potentially contaminated foods. The specific results of the survey are in surv-results.doc.

Approach and Methods

Numerous surveys have been conducted to study food habits and consumption of particular food groups (Rupp 1980; Pao 1982; Pennington 1983; EPA 1984a, 1984b). Some studies have involved assessing radionuclide intake. These studies have included distributions by race, sex, age, and occasionally, income levels. The USDA (USDA 1987, 1993) and USDA and U.S. Department of Health, Education, and Welfare (HEW) conducted these surveys (USDHEW 1963). Literature reviews were conducted to determine appropriate survey methods for the target population. Food and nutrition scientists were contacted from South Carolina State University Family Consumer Sciences Department, U.S. Department of Agriculture (USDA) Food & Nutrition Information Center, and USDA Agriculture Research Service to gather information to help design the consumption survey. After reviewing literature and obtaining information from these agencies, the lifestyle and food survey questionnaire for local populations was produced. The following steps were used to estimate food consumption:

1. Determine the most appropriate previously produced estimates
2. Use sub-population distribution from data obtained to weight for the local population
3. Adjust estimates with supplementary local data
4. Create an annual index of change based on USDA-ERS per capita food consumption data
5. Use the series to create annual estimates from 1950 onward.

Previous study estimates that were reviewed contained detailed information on bioclinical and clinical data. They focused more on actual chemical nutrients consumed than eating habits broken down by age, race, and gender. In addition, many of these estimates were regional or statewide. Based on the available information, obtaining local data through surveys was determined to be the most direct and practicable approach and the surveys would satisfy steps 1 through 3 above.

In producing the surveys, the distribution of specific ages (people 18-years old and older), genders, and races in the sample for the 50-county area were determined. Public locations, such as grocery stores, were chosen as good areas to conduct the surveys because the locations could be categorized (e.g., county/town, urban/rural, and affluent/not so affluent) and provide a good survey distribution. After the surveys were produced, student workers went in pairs to each of the targeted counties. Surveys were conducted in Georgia and South Carolina counties. Five counties for each state served as sample areas. The counties for South Carolina were Aiken, Allendale, Barnwell, Edgefield, and Hampton. Georgia counties included Burke, Columbia, Effingham, Richmond, and Screven. A sample size of 500 (250 for South Carolina and 250 for Georgia) was used to represent the population of all targeted counties. These counties were chosen because of their proximity to the SRS, and their location adjacent to the Savannah River. It was felt that people living in these counties would be most likely to have the greatest chance of exposure in the study area. Using the survey forms, the surveyors collected the name of the main contact, name of person conducting interview, date of interview, time of interview, and county of residence for each respondent. The questionnaire contained two main sections: one section focused on food intake and the other focused on demographic information. Each respondent was informed that their personal answers would be kept confidential and used in combination with other survey data only.

Results

A large majority (72.5%) of the respondents used a community water supply, while only (19%) used private wells. The main source of drinking water came from the community water supply. A very small amount (4.4%) of respondents drank bottled water. Over 75% of the respondents ate green vegetables such as broccoli, okra, and spinach. Only 30% of the individuals ate asparagus. Over 70% of the respondents ate yams and turnips, while 47% ate winter squash. Of the melons consumed, 70% of the respondents ate cantaloupes and watermelons. The vast majority of respondents ate pears and plums. There was a big disparity in the type of meat eaten. Individuals chose to consume beef, veal, or pork (77%) and chicken liver (64%) over lamb (31%). As for fish, 76% of the respondents ate fish and 24% did not. Of those eating fish, 37% stated that the fish they ate, were caught by someone they knew, and therefore, they probably came from a local source. Twenty-four percent ate fish from an unknown source, and 38% of the respondents did not answer this question.

Of the demographic responses obtained, approximately 32.5% were white, 57.2% were black, 2.6% were American Indian, 1.8% were Asian or Pacific Islander, and 3.0% were other. Approximately 51% of the respondents were in the 20 through 35 years category and 30% were 36-years old or older. A majority of the respondents worked full-time. Thirty-three percent had a

high school degree and 26% had college degrees. Respondents spent most of their time outdoors in their yards. A very small number spent time hunting and camping.

Discussion

The data that were gathered in the food and lifestyle surveys appear to be a good source of information to understand food distribution patterns assuming local demand is satisfied with local production. However, the data are only useful for the year 1997. In addition, statistical breakdowns will be needed to estimate percentages for each individual county surveyed. Estimates for prior years will have to be derived by using advised sources of prior surveys. Diet variation through time is a complicated problem because the two regional surveys that have been conducted do not cover the time period of interest for this study. Most of the data available from these surveys are from 1980 onward. USDA-ERS data are available for food consumption and food spending. The years for these data are only 1977–1978 and 1987–1988. Sub-populations from past estimates need to be identified and used with local data (including the data from this survey). An annual index of change needs to be developed and used to create annual local estimates from 1950 onward. Additional expertise in the area of food consumption will be sought for this effort. Other food groups including milk products and leafy vegetables need to be considered further along with more information on home-grown food. After estimates of food consumption are derived for the years of interest, they will serve as a primary source to identify populations that may have ingested radionuclides or chemicals released from the Site.

Survey Form

LIFESTYLE SURVEY FOR DOSE RECONSTRUCTION
AT SAVANNAH RIVER SITE

Main Contact: GIS Lab
South Carolina State University
1890 Research and Extension
300 College Street
Orangeburg, SC 29115
536-8921 or 7175

Name of Person(s) Conducting Interview: _____

Date of Interview: _____ Time of Interview: _____ am _____ pm

County of Residence for respondent (please circle one):
1) South Carolina - Aiken, Allendale, Barnwell, Edgefield, Hampton
2) Georgia - Burke, Columbia, Effingham, Richmond, Screven

Purpose: This survey is designed to collect information on lifestyle activities (food intake, leisure time, work obligations). The information will help us identify pathways for radioactive elements from the Savannah River Site nuclear facility. Your responses to this survey are important for establishing which exposure pathways might have affected air, water, and land area. In order to estimate the radioactive exposure to human populations, data for food consumption, outdoor activities and work obligations are necessary. This survey will be used to collect information from surrounding counties of the Savannah River Site.

Your cooperation and participation in this survey are very important. The information that you provide in this survey will be kept confidential and will be used in combination with other survey data only. Should you have concerns or questions, please feel free to contact the SCSU GIS Lab.

FOOD INTAKE

1) What is the main source of the water used for cooking in your home? Is it:

the community water supply, …… 01
your own well or rain cistern , …….. 02
your own spring or a public spring, ….. 03
bottled water your purchase, or …… 04
something else? (SPECIFY) …… 05
_____

2) What is the main source of the water used in your home for preparing beverages such as coffee, tea, juices, and baby formula? Is it:

the community water supply, …….. 01
your own well or rain cistern,……..02
your own spring or a public spring, … 03
bottled water you purchase, or …… 04
something else? (SPECIFY)
_____

3) What is the main source of plain drinking water in your home? Is it:

the community water supply, …… 01
your own well or rain cistern, …… 02
your own spring or a public spring, …… 03
bottled water you purchase, or
something else? (SPECIFY)…… 04
_____

SUGGESTED COMMENT: [You may paraphrase this] Now let's go to yesterday and talk about where you, or other people who live here, obtained the food you ate and where you ate it. (Ask following questions)

4) Where did you obtain most of the ingredients for this food? (please circle all that apply)

01 Store, such as
Supermarket, Grocery Store, or Warehouse,
Convenience Store, Drug Store, or Gas Station
Specialty Store such as:
Bakery, Deli, Seafood, Ethnic Food, Health Food
Commissary
Produce Stand or Farmer's Market
02 Restaurant with waiter/waitress
Service
03 Fast Food Place, Pizza Place
04 Bar Tavern
05 School Cafeteria
06 Other Cafeteria
07 Vending Machine
08 Child Care Center, Family Day
Care Home, Adult Day Care
09 Soup Kitchen Shelter, Food Pantry
10 Meals on Wheels
11 Other Community Food Program
12 Grown or *Caught by you or someone you know

* IF FISH OR SEAFOOD, ASK: Did it come from a:
41 Freshwater lake, pond, or river
42 The ocean, or
43 A bay, sound, or estuary?
44 Don't know type of water

13 Someone else/gift

Some Other Place (please circle)
14 Mail Order Purchase
15 Common Coffee Pot Or Snack Tray
16 Residential Dining Facility
17 Other (Specify)
98 Don't know

5) Did you eat/drink this food/beverage at your home?

Yes …….. 01
No …… 02

6) Before you ate/drank this particular food/beverage, was it ever at your home?

Yes …….. 01
No ……02

7) Was the amount of food that you ate yesterday about usual, less than usual, or more than usual?

Usual ……. 01
Less Than Usual …….. 02
More Than Usual …… 03

8) What is the main reason the amount you ate yesterday was less than usual? (please circle all that apply)

Sickness …… 01
Short Of Money ……. 02
Traveling …… 03
At a Social Occasion Or
On a Special Day …… 04
On Vacation …… 05
Too Busy …… 06
Not Hungry ……. 07
Dieting …… 08
Fasting …… 09
Bored Or Stressed …… 10
Some Other Reason (Specify)
_____

9) What is the main reason the amount you ate yesterday was more than usual? (please circle all that apply)

Traveling …….. 01
At a Social Occasion Or
On a Special Day …… 02
On Vacation …….. 03
Very Hungry …… 04
Bored Or Stressed…… 05
Some Other Reason

(SPECIFY) _____

SUGGESTED COMMENT: Now I'd (or we) would like you to think about all the plain drinking water that you had yesterday, regardless of where you drank it. By plain drinking water, I mean tap water or any bottled water that is not carbonated, with nothing added to it, not even lemon.

10) How many glasses of plain drinking water did you drink yesterday?

# of glasses _____
None …… 000

11) How much of this plain drinking water came from your home? Would you say all, most, some, or none?

All …… 01
Most …… 02
Some …… 03
None ……. 04

12) What was the main source of plain drinking water that did not come from your home?
Was it tap water, water from a drinking fountain, bottled water, or something else?

Tap Water And/Or Drinking 01
Bottled Water ……..……. 02
Other Source……..…… 03
(SPECIFY) _____
Don't Know…..……. 04

13) During the past 12 months, that is, since last June, have you eaten any of these foods listed below? Please circle all that apply.

	YES	NO
Artichokes …… ……	1	2
Asparagus …….	1	2
Broccoli ……	1	2
Brussels sprouts ……	1	2
Cauliflower ……	1	2
Eggplant …….	1	2
Kale ……	1	2
Swiss chard ……	1	2
Okra ……	1	2

Spinach ……	1	2
Summer squash (thin skin) …….	1	2
Winter squash …….	1	2
Sweet potato or yams …….	1	2
Turnips, other than greens …….	1	2
Avocado or guacamole ……..	1	2
Grapefruit, other than juice ……	1	2
Cantaloupe …….	1	2
Honeydew melon ……	1	2
Watermelon ……	1	2
Nectarines ……..	1	2
Pears ……..	1	2
Plums …….	1	2
Rhubarb …….	1	2
Chicken liver …….	1	2
Beef, veal or pork liver …….	1	2
Lamb …….	1	2
Shellfish ……	1	2
Fish, other than shellfish or canned fish ……	1	2
IF YES: Was any of the fish you ate caught by you or someone you know? …….	1	2

DEMOGRAPHIC INFORMATION

1) Circle gender of individual.

Male…….. 01
Female…….. 02

2) What race do you identify yourself as? (Circle one)

White…… 01
Black…… 02
American Indian, Eskimo, or Aleutian.. 03
Asian or Pacific Islander.. 04
Other…… 05

3) What is your age range? (Circle one)
0-5 yrs 6-10 yrs 11-15 yrs 16-20 yrs 21-25 yrs 26-30 yrs 31-35 yrs 36-40 yrs
41-45 yrs 46-50 yrs 51-55 yrs 56-60 yrs 61-65 yrs 66-70 yrs 71-75 yrs 76+

4) What is the highest level of education that you obtained? (Circle response)

Never Attended School Or Kindergarten : 00

Elementary:
01 02 03 04 05 06 07 08

High School:
09 10 11 12 or GED

College:
01 02 03 04+

5) Are you presently: (Skip questions 5-9, if response is given for this question)

Looking For Work ....…… 01
Going To School ...…… 02
Keeping House ....…….. 03
Retired …...……. 04
Unable To Work….……. 05
Other (SPECIFY) …… 06
_____

6) Last week, did you work at all at a paid job or in your own business or farm?

Yes…… 1
No…….. 2

7) Do you have a paid job from which you were temporarily absent?

Yes…… 1
No…….. 2

8) How many hours did you work at all jobs in the last week? Include all overtime hours that you worked and hours on any part-time jobs as well as your principal job.

Of Hours _____

9) How many hours a week do you usually work?

OF Hours _____

10) In your best estimation, how many hours did you spend outdoors? Include outside related work and leisure.

OF Hours _____

11) Of your time spent outdoors, what is it mostly in?

Working (job occupation) …….. 01
Yard activities (gardening, mowing, cleaning, etc…) …… 02
Athletic events or games ……. 03
Hunting/Fishing …… 04
Camping/Hiking …… 05

12) How active do you consider yourself?
Extremely active …… 01
Somewhat active …… 02
Not active …… .. 03

13) In regard to this household, is the property owned or being bought by:
someone living in this household …….. 01
rented with payment required, or …….. 02
occupied without payment of rent
required …….. 03

SUGGESTED ENDING COMMENT: Thank you kindly for allowing me (if applicable, us) this time to interview you. The information that you have provided is of extreme importance to our project. We need this information, along with others, in order to do our calculations. No person will be identified. Again, thank you, and have a nice (morning/afternoon/evening).

Structures for Demographic Database Tables

Structures for all the demographic database tables are provided below.

Structure for database: **food**
Description: Results of Food Survey taken in 10 Targeted Counties for SC & GA (5 in GA / 5 in SC)

Item_Name	Width	Output	N.Dec	Type	Description
STATE	2	2		Numeric	State
COUNTY	2	2		Numeric	County
SEX	4	4	2	Numeric	Sex
AGE	4	4	2	Numeric	Age
RACE	4	4	2	Numeric	Race
EDUC	4	4	2	Numeric	Education
NFRUITS	1	1		Numeric	Number of People Who Eat Fruits
NMEATS	1	1		Numeric	Number of People Who Eat Meats
NVEGS	1	1		Numeric	Number of People Who Eat Vegetables
NFISH	1	1		Numeric	Number of People Who Eat Fish
NCOMM1		1		Numeric	"Community Water, Main Source"
NWELL	1			Numeric	"Well Water, Main Source"
NSPRING	1	1		Numeric	"Spring Water, Main Source"
NBOTTLE	1	1		Numeric	"Bottle Water, Main Source"
NSELSE	1	1		Numeric	"Something Else, Main Source"

NOTES:

THE CODES FOR THE DEMOGRAPHIC VARIABLES ARE AS FOLLOWS:

STATE	13 ='GEORGIA' 45 ='SOUTH CAROLINA'
COUNTY	3 ='AIKEN' 5='ALLENDALE' 11='BARNWELL' 33='BURKE' 37='EDGEFIELD' 49='HAMPTON' 73='COLUMBIA' 103='EFFINGHAM' 245='RICHMOND' 251='SCREVEN'
SEX	1='MALE' 2='FEMALE'
AGE	1='0-5 YEARS' 2='6-20 YEARS' 3='21-35 YEARS' 4='36-65 YEARS' 5='OVER 65'
RACE	1='WHITE' 2='BLACK' 3='AMERICAN INDIAN' 4='ASIAN OR PACIFIC ISLANDER' 5='OTHER'
EDUC	1='ELEMENTARY' 2='HIGH SCHOOL' 3='COLLEGE' 0 - 8 YEARS 9 - 12 YEARS 13 YEARS & OVER

Source of data is GIS Laboratory (South Carolina State University, 1890 Research & Extension)

Structure for Database: **Gacattle**
Description: Cattle Inventory for Georgia

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year

FIPS	5	5		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
CTLCVIN	6	6		Numeric	Cattle-calf inventory
BFCOWIN	6	6		Numeric	Beef-cow inventory
MKCOWIN	5	5		Numeric	Milk-cow inventory
OTCTIN	6	6		Numeric	Other-cattle excluding calves

NOTES:
- The source for data is Georgia Agricultural Statistics Service County Estimates
- Years included are 1962, 1963, 1964, 1970, 1980, and 1990

Structure for Database: **Gacorn**
Description: Corn Grain Harvested and Yield

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
FIPS	5	5		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
CNGRHAR	6	6		Numeric	Corn Grain Harvested
CNGRYIEL	6	6		Numeric	Corn Grain Yield
CNGRPROD	8	8		Numeric	Corn Grain Produced

NOTES:
- The sources for data are Georgia Agricultural Statistics Service County Estimates and NASS
- Years included are 1950, 1957, 1960, 1962, 1963, 1964, 1970, 1980, 1990

Structure for Database: **Gahog**
Description: Georgia Hog Inventory

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
FIPS	5	5		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
HGPGINV	5	5		Numeric	ALL Hog and Pig Inventory

NOTES:
- The source for data is Georgia Agricultural Statistics Service County Estimates
- Years included are 1960, 1962, 1963, 1964, 1970, 1980, and 1990

Structure for Database: **Gaoat**
Description: Georgia Oats Harvested and Yield

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
FIPS	5	5		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
PLPRAC	5	5		String	Acres Planted for All Purposes
GRHRAC	5	5		Numeric	All Grain Harvested Acres
GRYLAC	5	5	2	Numeric	All Grain Yield/Harvested Acre (Bushel)
GRPRAC	7	7		Numeric	All Grain Production Bushels

NOTES:
- The source for data is Georgia Agricultural Statistics Service County Estimates and NASS
- Years included are 1980 and 1990

Structure for Database: **Gapeanut**
Description: Peanut Harvest and Yield

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
FIPS	5	5		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
ALLHVAC	7	7		Numeric	All Harvested Acres
ALPHVAC	7	7		Numeric	All Per Harvested Acres
GRPRDAC	10	10		Numeric	All Per Harvested Acre (lbs)

NOTES:
- The sources for data are Georgia Agricultural Statistics Service County Estimates and NASS
- Years included are 1950, 1957, 1960, 1962, 1963, 1964, 1970, 1980, 1990

Structure for Database: **Gasoybea**
Description: Georgia Soybeans Harvested and Yield

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
FIPS	7	7		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
BEHRVAC	6	6		Numeric	All Beans Harvested Acres

BEYLAC	5	5	2	Numeric	All Beans Yield/Harvested Acres
BEPRDAC	6	6		Numeric	All Beans Production Bushels

NOTES:
- The source for data is Georgia Agricultural Statistics Service County Estimates and NASS
- Years included are 1950, 1957, 1960, 1962, 1963, 1964, 1972, 1980, 1990

Structure for Database: **Gatobac**
Description: Georgia Tobacco Harvested and Yield

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
FIPS	5	5		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
ALHVAC	4	4		Numeric	All Type-14 Harvested Acres
ALHVACP	4	4		Numeric	All Type-14 Per Harvested Acres (lb)
ALPRD	7	7		Numeric	All Type-14 Production Pounds

NOTES:
- The sources for data are Georgia Agricultural Statistics Service County Estimates and NASS
- Years included are 1950, 57, 60, 62, 63, 64, 70, 80, and 90

Structure for Database: **Gawheat**
Description: Georgia Wheat Harvested and Yield

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
FIPS	5	5		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
GRHAAC	5	5		Numeric	All Grain Harvested Acres
GRYIEL	5	5	2	Numeric	All Grain Yield/Harvested Acres (bushels)
GRPROD	7	7		Numeric	All Grain Produced (bushels)

NOTES:
- The sources for data are Georgia Agricultural Statistics Service County Estimates and NASS
- Years included are 1950, 1957, 1960, 1962, 1963, 1964, 1970, 1980,and 1990

Structure for database: **milkdist54**
Description: Milk Distribution in 1950s for Georgia and South Carolina

Item_Name	Width	Output	N.Dec	Type	Description
SURST	2	2		String	Surplus State
SURNUM	2	2		Numeric	Surplus Region Number
DEFST	2	2		String	Deficit State
DEFNUM	2	2		Numeric	Deficit Region Number
ETR	5	5		Numeric	Estimated Transfer Rate (kL y-1)
SOI	27	27		String	Source of Information
DR	1	1		Numeric	Degree of Reliability

NOTES:

- Milk Region # - South Carolina Counties

76 Anderson Co., Cherokee Co., Greenville Co., Laurens Co., Oconee Co., Pickens Co., Spartanburg Co., Union Co.
77 Chester Co., Fairfield Co., Kershaw Co., Lancaster Co., York Co.
78 Chesterfield Co., Darlington Co., Dillon Co., Florence Co., Georgetown Co., Horry Co., Marion Co., Marlboro Co., Williamsburg Co.,
79 Calhoun Co., Clarendon Co., Lee Co., Lexington Co., Orangeburg Co., Sumter Co., Richland Co.,
80 Abbeville Co., Aiken Co., Edgefield Co., Greenwood Co., Laurens Co., McCormick Co., Saluda Co.
81 Allendale Co., Bamberg Co., Barnwell Co., Beaufort Co., Berkeley Co., Charleston Co., Colleton Co., Dorchester Co., Hampton Co., Jasper Co.,

- Milk Region # - Georgia Counties

82 Banks Co., Elbert Co., Franklin Co., Habersham Co., Hart Co., Lincoln Co., Madison Co., Oglethorpe Co., Rabun Co., Stephens Co., Wilkes Co.,
83 Barrow Co., Cherokee Co., Clarke Co., Cobb Co., Dawson Co., Dekalb Co., Fannin Co., Forsyth Co., Gilmer Co., Gwinnett Co., Hall Co., Jackson Co., Lumpkin Co., Oconee Co., Pickens Co.,Towns Co., Union Co., Walton Co.
84 Burtow Co., Catoosa Co., Chattooga Co., Dade Co., Floyd Co., Gordon Co., Murray Co., Paulding Co., Polk Co., Walker Co., Whitfield Co.
85 Fulton Co.
86 Appling Co., Brantley Co., Bryan Co., Bulloch Co., Burke Co., Camden Co., Candler Co., Charlton Co., Chatham Co., Columbia Co., Effingham Co., Emanuel Co., Evans Co., Glascock Co., Glynn Co., Jefferson Co., Jenkins Co., Liberty Co., Long Co., McDuffie Co., McIntosh Co., Pierce Co., Richland Co., Screven Co., Tatnall Co., Toombs Co., Ware Co., Warren Co.
87 Baldwin Co., Bibb Co., Bleckney Co., Butts Co., Crawford Co., Dodge Co., Greene Co., Hancock Co., Houston Co., Jasper Co., Johnson Co., Jones Co., Laurens Co., Monroe Co., Montgomery Co., Morgan Co., Newton Co., Peach Co., Pulaski Co., Putnam Co., Rockdale Co., Taliaferro Co., Truetlen Co., Twiggs Co., Washington Co., Wheeler Co., Wilkinson Co.
88 Carroll Co., Chattahoochee Co., Clayton Co., Coweta Co., Douglass Co., Fayette Co., Haralson Co., Harrison Co., Heard Co., Henry Co., Lamar Co., Macon Co., Marion Co., Meriwether Co., Muscogee Co., Pike Co., Schley Co., Spaulding Co., Talbot Co.,

Taylor Co., Troup Co., Upson Co.
89 Atkinson Co., Ben Hill Co., Berrien Co., Brooks Co., Clinch Co., Coffee Co., Colquitt Co., Cook Co., Crisp Co., Dooly Co., Echois Co., Irwin Co., Jeff Davis., Lanier Co., Lowndes Co., Telfair Co., Tift Co., Turner Co., Wilcox Co., Worth Co.
90 Baker Co., Calhoun Co., Clay Co., Decatur Co., Dougherty Co., Early Co., Grady Co., Lee Co., Miller Co., Mitchell Co., Randolph Co., Seminole Co., Stewert Co., Sumter Co., Terrell Co., Thomas Co., Webster Co., Quitman Co.

Milk marketing orders (mmo's) constituted main source of information.
Georgia MMO: Atlanta, Columbus, and Savannah
South Carolina MMO: Anderson, Columbia, Florence, Greenville, and Spartanburg

Structure for table: **Sccattle**

Description: Cattle Inventory for South Carolina

Item_Name	Width	Output	N.Dec	Type	Description
YEAR	5	5		Numeric	Year
FIPS	5	5		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
CTLCVIN	6	6		Numeric	Cattle-calf inventory

NOTES:

The source for data is SC Agricultural Statistics Service County Estimates

Structure for Database: **Sccorn**
Description: South Carolina Corn Harvested and Yield

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
FIPS	5	5		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
CNGRHAR	6	6		Numeric	Corn Grain Harvested
CNGRYIEL	4	4		Numeric	Corn Grain Yield
CNGRPROD	7	7		Numeric	Corn Grain Produced

NOTES:
- The sources for data are S.C. Agricultural Statistics Service, Clemson Dept. of Agricultural Economics, and NASS
- Years included are 1950, 1957, 1960, 1962, 1963, 1964, 1970, 1980, and 1990

Structure for Database: **aikengame**
Description: Hunting Practices in Crackerneck Wildlife Management Area (Aiken County)

Item_Name	Width	Output	N.Dec	Type	Description
YEAR	5	5		String	YEAR
DEER	3	3		Numeric	DEER
HOGS	2	2		Numeric	HOGS
GSQUIR	3	3		Numeric	Gray_Squirrels
FSQUIR3	3			Numeric	Fox_Squirrels
BOBWHITE	3	3		Numeric	Bobwhite
WOODCOCK3	3			Numeric	Woodcock
DUCKS	3	3		Numeric	Ducks
RABBIT	3	3		Numeric	Rabbit
RACCOON	3	3		Numeric	Raccoon
TURKEY	3	3		Numeric	Turkey
DOVE	3	3		Numeric	Dove
COYOTE	3	3		Numeric	Coyote
FISH	3	3		Numeric	Fish

NOTES:
- The source for data is South Carolina Dept. of Natural Resources
- Division of Wildlife and Freshwater Fisheries

Structure for Database: **Scoat**
Description: South Carolina Oats Harvested and Yield

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	4	4		Numeric	Year
ACRHRV	5	5		Numeric	Acres harvested
YLDACRBU	5	5	2	Numeric	Yield per acre bushels
PROD	8	8		Numeric	Production

Structure for Database: **Scpeanut**
Description: South Carolina Peanuts Harvested and Produced

Item_Name	Width	Output	N.Dec	Type	Description

YR	5	5	Numeric	Year
FIPS	5	5	Numeric	Federal Information Processing Standard
COUNTY	15	15	String	County
ALLHVAC	4	4	Numeric	Peanuts Harvested
ALLPHVAC	5	5	Numeric	Peanuts harvested in Pounds
GRPRDAC	7	7	Numeric	Peanuts Produced

NOTES:

- The sources for data is S.C. Agricultural Statistics Service, Clemson Univ. Dept. of Agricultural Economics and NASS
- Years included are 1950, 1957, 1960, 1962, 1963, 1964, 1970, 1980, 1990

Structure for table: **Scsoyb57**
Description: South Carolina Soybeans Harvested and Yield in 1957

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
ACRES57	8	8		Numeric	All Planted All Purpose Acres
PERYIELD57	7	7		Numeric	Yield Per Acres
PROD57	9	9		Numeric	Total Production

Notes:

"The source for data is S.C. Agricultural Statistics Service, Clemson Univ. Dept. of Agricultural Economics and NASS"

Structure for table: **Scsoyb60**
Description: South Carolina Soybeans Harvested and Yield in 1960

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
ACRES60	8	8		Numeric	All Planted All Purpose Acres
PERYIELD60	7	7	2	Numeric	Yield Per Acres
PROD60	9	9		Numeric	Total Production

NOTES:

"The source for data is S.C. Agricultural Statistics Service, Clemson Univ. Dept. of Agricultural Economics and NASS

Structure for table: **Scsoyb62**
Description: South Carolina Soybeans Harvested and Yield in 1962

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
ACRES62	8	8		Numeric	All Planted All Purpose Acres
PERYIELD60	7	7	2	Numeric	Yield Per Acres
PROD60	9	9		Numeric	Total Production

NOTES:

"The source for data is S.C. Agricultural Statistics Service, Clemson Univ. Dept. of Agricultural Economics and NASS"

Structure for table: **Scsoyb63**
Description: South Carolina Soybeans Harvested and Yield in 1963

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
ACRES63	8	8		Numeric	All Planted All Purpose Acres
PERYIELD63	7	7		Numeric	Yield Per Acres
PROD63	9	9		Numeric	Total Production

NOTES:

"The source for data is S.C. Agricultural Statistics Service, Clemson Univ. Dept. of Agricultural Economics and NASS"

Structure for table: **Scsoyb64**
Description: South Carolina Soybeans Harvested and Yield in 1964

Item_Name	Width	Output	N.Dec	Type	Description

FIPS	5	5		Numeric	Federal Information Processing Standards
ACRES64	8	8		Numeric	All Planted All Purpose Acres
PERYIELD64	7	7	2	Numeric	Yield Per Acres
PROD64	9	9		Numeric	Total Production

NOTES:

"The source for data is S.C. Agricultural Statistics Service, Clemson Univ. Dept. of Agricultural Economics and NASS"

Structure for table: **Scsoyb70**
Description: South Carolina Soybeans harvested and Yield in 1970

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standards
ACRES70	8	8		Numeric	All Planted All Purpose Acres
PERYIELD70	7	7	2	Numeric	Yield Per Acre
PROD70	9	9		Numeric	Total Production

NOTES:

"The source for the data is S.C. Agricultural Statistics Service, Clemson Univ. Dept. of Agricultural Economics and NASS"

Structure for table: **Scsoyb80**
Description: South Carolina Soybeans Harvested and Yield in 1980

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
ACRES80	8	8		Numeric	All Planted All Purpose Acres
PERYIELD80	7	7	2	Numeric	Yield Per Acres
PROD80	9	9		Numeric	Total Production

NOTES: The source for data is S.C. Agricultural Statistics Service, Clemson Univ. Dept. of Agricultural Economics and NASS"

Structure for table: **Scsoyb90**
Description: South Carolina Soybeans Harvested and Yield in 1990

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
ACRES90	8	8		Numeric	All Planted All Purpose Acres
PERYIELD90	7	7	2	Numeric	Yield Per Acres
PROD90	9	9		Numeric	Total Production

Structure for Database: **Aikensptmn**
Description: Sportsman Type in Crackerneck Wildlife Management Area (Aiken County)

Item_Name	Width	Output	N.Dec	Type	Description
Year	5	5		Numeric	Year
HUNTERS	4	4		Numeric	Number of Hunters
FISHERMEN	3	3		Numeric	Number of Fishermen

NOTES:

The source for data is South Carolina Dept. of Natural Resources
Division of Wildlife and Freshwater Fisheries

Structure for Database: **Scwheat**
Description: South Carolina Wheat Harvested and Yield

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
FIPS	7	7		Numeric	Federal Information Processing Standard
COUNTY	15	15		String	County
GRHAAC	6	6		Numeric	All Grain Harvested Acres
GRYIEL	5	5	2	Numeric	All Grain Yield/Harvested Acres (bushels)
GRPROD	7	7		Numeric	All Grain Produced (bushels)

NOTES:
- The sources for data is S.C. Agricultural Statistics Service, Clemson Dept. of Agricultural Economics, and NASS
- Years included are 1950, 1957, 1960, 1962, 1963, 1964, 1970, 1980, and 1990

Structure for Database: **si5074**
Description: Corn Grain Silage Yield and Production (1950–1974)

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
STATE	2	2		String	Federal Information Processing Standard
PLPRAC	5	5		Numeric	Planted for All Purposes Acres
GRHRAC	7	7		Numeric	Harvested Grain Acres
YLDPAC	5	5	1	Numeric	Yield Per Acre (bushels)
PROD	9	9		Numeric	Production Bushels
HRSIAC	5	5		Numeric	Harvested Silage Acres
YLDPACT	5	5	1	Numeric	Yield Per Acre Tons
PRODT	5	5		Numeric	Production Tons

NOTES:
- The source for data is the National Agriculture Statistics Service
- Years included are 1950 through 1974

Structure for Database: **si7587**
Description: Corn Grain Silage Yield and Production (1975–1987)

Item_Name	Width	Output	N.Dec	Type	Description
YR	5	5		Numeric	Year
STATE	2	2		String	Federal Information Processing Standard
PLPRAC	5	5		Numeric	Planted for All Purposes Acres
GRHRAC	7	7		Numeric	Harvested Grain Acres
YLDPAC	5	5	1	Numeric	Yield Per Acre (bushels)
PROD	9	9		Numeric	Production Bushels
HRSIAC	4	4		Numeric	Harvested Silage Acres
YLDPACT	5	5	1	Numeric	Yield Per Acre Tons
PRODT	5	5		Numeric	Production Tons

NOTES:
- The source for data is the National Agriculture Statistics Service
- Years included 1975 through 1980

Structure for Database: **si8894**
Description: Corn Grain Silage Yield and Production (1988–1994)

Item_Name	Width	Output	N.Dec	Type	Description

YR	5	5		Numeric	Year
STATE	4	4		String	Federal Information Processing Standard
PLPRAC	5	5		Numeric	Planted for All Purposes Acres
GRHRAC	7	7		Numeric	Harvested Grain Acres
YLDPAC	5	5	2	Numeric	Yield Per Acre (bushels)
PROD	9	9		Numeric	Production Bushels
HRSIAC	4	4		Numeric	Harvested Silage Acres
YLDPACT	5	5	2	Numeric	Yield Per Acre Tons
PRODT	5	5		Numeric	Production Tons

NOTES:
- The source for data is the National Agriculture Statistics Service
- Years included are 1988 through 1994

Structure for Database: **GAFRMX**
Description: Farm expenses for Georgia

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
ITEM	4	4		String	Item
CRFM	5	5		Numeric	Cash receipts from marketings
TLP	5	5		Numeric	Total livestock and products
TC	5	5		Numeric	Total crops
OI	5	5		Numeric	Other income
GP	5	5		Numeric	Government payments
IIRR	5	5		Numeric	Imputed income and rent received
PE	5	5		Numeric	Production expenses
FP	5	5		Numeric	Feed purchased
LP	5	5		Numeric	Livestock purchased
SP	5	5		Numeric	Seed purchased
FL	5	5		Numeric	Fertilizer and lime (incl. chem.)
PPRR	5	5		Numeric	Petroleum products purchased
HFLE	5	5		Numeric	Hired farm labor expenses
AOPE	5	5		Numeric	All other production expenses
VIC	5	5		Numeric	Value of inventory change
LV	5	5		Numeric	Livestock
CR	5	5		Numeric	Crops
TCR	5	5		Numeric	Total cash receipts
LTPE	5	5		Numeric	Less: total prod. expense
RNI	5	5		Numeric	Realized net income
PVIC	5	5		Numeric	Plus: value of inventory change

TNIC	5	5		Numeric	Total net income incl. corporate
LCF	5	5		Numeric	Less: corporate farms
PSA	5	5		Numeric	Plus: farm wages and prerequisite
TNFP	5	5		Numeric	Total Net Farm proprietors
PFWP	5	5		Numeric	Plus: Farm wages and prerequisite
PFOL	5	5		Numeric	Plus: farm other labor income
TFLP	5	5		Numeric	Total farm labor and proprietors

Structure for Database **GATPAY**
Description: Farm Expenses for Georgia

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
TTP	6	6		Numeric	Total transfer payments
GPI	6	6		Numeric	Government payments to individuals
RDIP	6	6		Numeric	Retired & disabled insurance benefits payments
ODIP	6	6		Numeric	"Old-age, surveillance & disability insurance payment"
RRDP	5	5		Numeric	Railroad retirement & disability payments
FCED	5	5		Numeric	Federal civil employee retirement payment
MRP	5	5		Numeric	Military retirement payments
SLED	5	5		Numeric	State & local govt. employee retirement payments
WCP	5	5		Numeric	Workers comp. payments (federal & state)
OGIP	5	5		Numeric	Other govt. disability insurance & retirement payments
MP	6	6		Numeric	Medical payments
IMBP	5	5		Numeric	Income maintenance benefit payments
SSIP	5	5		Numeric	Suppl. security income (SSI) payments
AFDC	5	5		Numeric	Aide to families with dependent children (AFDC)
FS	5	5		Numeric	Food stamps
OIM	5	5		Numeric	Other income maintenance
UIBP	5	5		Numeric	Unemployment insurance benefit payments
SUIC	5	5		Numeric	State unemployment insurance compensation

UCFE	5	5		Numeric	Unemp. comp for fed civilian employees (UCFE)
UCRE	5	5		Numeric	Unemp. comp for railroad employees
UCX	5	5		Numeric	Unemp. comp for veterans (UCX)
VBP	5	5		Numeric	Veteran benefit payments
VPCP	5	5		Numeric	Veteran pensions & compensation payments
EVDS	5	5		Numeric	"Education. asst to vets, dependant, and survivors"
VLBP	5	5		Numeric	Veterans life insurance benefit payments
OAV	5	5		Numeric	Other assistance to veterans
FETP	5	5		Numeric	Fed. educ. & trng. asst. pay (excl. vets)
OPL	5	5		Numeric	Other payments to individuals
PNI	5	5		Numeric	Payment to nonprofit institutions
FGP	5	5		Numeric	Federal govt. payments
SGP	5	5		Numeric	State and local govt. payments
BP	5	5		Numeric	Business payments
BPI	5	5		Numeric	Business payments to individuals

Structure for Database **GATPl**
Description: Total Personal Income for Georgia

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
TPI	8	8		Numeric	Total personal income
NPI	7	7		Numeric	Nonfarm personal income
FI	6	6		Numeric	Farm income
POP	6	6		Numeric	Population
PCPI	6	6		Numeric	Per capita personal income
EPW	8	8		Numeric	Earnings by place of work
LPSI	6	6		Numeric	Less: Personal cont. for social security insurance
PAR	7	7		Numeric	Plus: adjustment for residence
ENPR	8	8		Numeric	Equals: Net earn. by place of residence
PDIR	8	8		Numeric	Plus: Dividends, interest and rent
PTP	6	6		Numeric	Plus: Transfer payments
WS	8	8		Numeric	Wages and salaries
OLI	6	6		Numeric	Other labor income
PI	6	6		Numeric	Proprietors income
FARM	6	6		Numeric	Farm

NF	6	6		Numeric	Nonfarm
FXX	6	6		Numeric	Farm
NFXX	8	8		Numeric	Nonfarm
PRIVATE	8	8		Numeric	Private
ASFO	6	6		Numeric	Ag., Serv., fish and other
MIN	6	6		Numeric	Mining
CNST	6	6		Numeric	Construction
MANU	6	6		Numeric	Manufacturing
NDG	6	6		Numeric	Nondurable goods
DG	6	6		Numeric	Durable goods
TPU	6	6		Numeric	Transportation and public utilities
WT	6	6		Numeric	Wholesale trade
RT	6	6		Numeric	Retail trade
FRE	6	6		Numeric	Finance., insurance and real estate
SERV	6	6		Numeric	Services
GGE	6	6		Numeric	Government and government enterprises
FC	6	6		Numeric	Federal, Civilian
MIL	6	6		Numeric	Military
SLC	6	6		Numeric	State and local

Structure for Database **SCFRMX**
Description: Farm Expenses for South Carolina

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
CRFM	5	5		Numeric	Cash receipts from marketings
TLP	5	5		Numeric	Total livestock and products
TC	5	5		Numeric	Total crops
OI	5	5		Numeric	Other income
GP	5	5		Numeric	Government payments
IIRR	5	5		Numeric	Imputed income and rent received
PE	5	5		Numeric	Production expenses
FP	5	5		Numeric	Feed purchased
LP	5	5		Numeric	Livestock purchased
SP	5	5		Numeric	Seed purchased
FL	5	5		Numeric	Fertilizer and lime (incl. chem)
PPRR	5	5		Numeric	Petroleum products purchased
HFLE	5	5		Numeric	Hired farm labor expenses
AOPE	5	5		Numeric	All other production expenses
VIC	5	5		Numeric	Value of inventory change
LV	5	5		Numeric	Livestock
CR	5	5		Numeric	Crops

TCR	5	5		Numeric	Total cash receipts
LTPE	5	5		Numeric	Less: total prod. expense
RNI	5	5		Numeric	Realized net income
PVIC	5	5		Numeric	Plus: value of inventory change
TNIC	5	5		Numeric	Total net income include corporate
LCF	5	5		Numeric	Less: corporate farms
PSA	5	5		Numeric	Plus: farm wages and prerequisite
TNFP	5	5		Numeric	Total Net Farm proprietors
PFWP	5	5		Numeric	Plus: Farm wages and prerequisite
PFOL	5	5		Numeric	Plus: farm other labor income
TFLP	5	5		Numeric	Total farm labor and proprietors

Structure for Database **SCTPAY**
Description: Farm Expenses for South Carolina

Item_Name	Width	Output	N.Dec	Type	Description
YEAR	5	5		Numeric	Year
TTP	8	8		Numeric	Total transfer payments
GPI	8	8		Numeric	Government payments to individuals
RDIP	8	8		Numeric	Retired & disabled insurance benefits payments
ODIP	8	8		Numeric	Old-age, surveillance & disability insurance payment
RRDP	8	8		Numeric	Railroad retirement & disability payments
FCED	8	8		Numeric	Federal civil employee retirement payment
MRP	8	8		Numeric	Military retirement payments
SLED	8	8		Numeric	State & local govt. employee retirement payments
WCP	8	8		Numeric	Workers comp. payments (federal & state)
OGIP	8	8		Numeric	Other govt. disability insurance & retirement payments
MP	8	8		Numeric	Medical payments
IMBP	8	8		Numeric	Income maintenance benefit payments
SSIP	8	8		Numeric	Suppl. security income (SSI) payments
AFDC	8	8		Numeric	Aide to families with dependent children (AFDC)
FS	8	8		Numeric	Food stamps
OIM	8	8		Numeric	Other income maintenance
UIBP	8	8		Numeric	Unemployment insurance benefit payments
SUIC	8	8		Numeric	State unemployment insurance compensation
UCFE	8	8		Numeric	Unemp. comp for fed civilian employees (UCFE)

UCRE	8	8		Numeric	Unemp. comp for railroad employees
UCX	8	8		Numeric	Unemp. comp for veterans (UCX)
VBP	8	8		Numeric	Veteran benefit payments
VPCP	8	8		Numeric	Veteran pensions & compensation payments
EVDS	8	8		Numeric	"Education. asst. to vets, dependant, and survivors"
VLBP	8	8		Numeric	Veterans life insurance benefit payments
OAV	8	8		Numeric	Other assistance to veterans
FETP	8	8		Numeric	Fed. educ. & trng. asst. pay (excl. vets)
OPL	8	8		Numeric	Other payments to individuals
PNI	8	8		Numeric	Payment to nonprofit institutions
FGP	8	8		Numeric	Federal govt. payments
SGP	8	8		Numeric	State and local govt. payments
BP	8	8		Numeric	Business payments
BPI	8	8		Numeric	Business payments to individuals

Structure for Database: **SCTPl**
Description: Total Personal Income for South Carolina

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
IPR	5	5		Numeric	Income by place of residence
TPI	7	7		Numeric	Total personal income
NPI	7	7		Numeric	Nonfarm personal income
FI	7	7		Numeric	Farm income
POP	8	8		Numeric	Population
PCPI	7	7		Numeric	Per capita personal income
EPW	7	7		Numeric	Earnings by place of work
LPSI	7	7		Numeric	Less: Personal contribution for social insurance
PAR	9	9		Numeric	Plus: adjustment for residence
ENPR	7	7		Numeric	Equals: Net earnings by place of residence
PDIR	7	7		Numeric	Plus: Dividends, interest, and rent
PTP	7	7		Numeric	Plus: Transfer payments
WS	7	7		Numeric	Wages and salaries
OLI	7	7		Numeric	Other labor income
PI	7	7		Numeric	Proprietors income
FARM	7	7		Numeric	Farm
NF	7	7		Numeric	Nonfarm

FXX	7	7		Numeric	Farm
NFXX	7	7		Numeric	Nonfarm
PRIVATE	7	7		Numeric	Private
ASFO	7	7		Numeric	Ag., Serv., fish and other
MIN	7	7		Numeric	Mining
CNST	7	7		Numeric	Construction
MANU	7	7		Numeric	Manufacturing
NDG	7	7		Numeric	Nondurable goods
DG	7	7		Numeric	Durable goods
TPU	7	7		Numeric	Transportation and public utilities
WT	8	8		Numeric	Wholesale trade
RT	7	7		Numeric	Retail trade
FRE	7	7		Numeric	Finance, insurance and real estate
SERV	7	7		Numeric	Services
GGE	7	7		Numeric	Government and government enterprises
FC	7	7		Numeric	Federal, Civilian
MIL	7	7		Numeric	Military
SLC	7	7		Numeric	State and local

Structure for database: **Gawmal**
Description: Population Dynamics for Georgia white male by age

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
ALLAGE	6	6		Numeric	Number of male for all ages
GUN5	5	5		Numeric	Number of male under 5
G5T9	5	5		Numeric	Number of male age 5-9
G10T14	4	4		Numeric	Number of male age 10-14
G15T19	4	4		Numeric	Number of male age 15-19
G20T24	5	5		Numeric	Number of male age 20-24
G25T29	4	4		Numeric	Number of male age 25-29
G30T34	4	4		Numeric	Number of male age 30-34
G35T39	4	4		Numeric	Number of male age 35-39
G40T44	4	4		Numeric	Number of male age 40-44
G45T49	4	4		Numeric	Number of male age 45-49
G50T54	4	4		Numeric	Number of male age 50-54
G55T59	4	4		Numeric	Number of male age 55-59
G60T64	4	4		Numeric	Number of male age 60-69
G65T69	4	4		Numeric	Number of male age 65-69
G70T74	4	4		Numeric	Number of male age 70-74
G75T84	4	4		Numeric	Number of male age 75-84

G85OV	4	4		Numeric	Number of male age over85

Structure for database: **Gawfmal**
Description: Population Dynamics for Georgia white female by age

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
ALLAGE	6	6		Numeric	Number of female for all ages
GUN5	4	4		Numeric	Number of female under 5
G5T9	4	4		Numeric	Number of female age 5-9
G10T14	4	4		Numeric	Number of female age 10-14
G15T19	4	4		Numeric	Number of female age 15-19
G20T24	4	4		Numeric	Number of female age 20-24
G25T29	4	4		Numeric	Number of female age 25-29
G30T34	4	4		Numeric	Number of female age 30-34
G35T39	4	4		Numeric	Number of female age 35-39
G40T44	4	4		Numeric	Number of female age 40-44
G45T49	4	4		Numeric	Number of female age 45-49
G50T54	4	4		Numeric	Number of female age 50-54
G55T59	4	4		Numeric	Number of female age 55-59
G60T64	4	4		Numeric	Number of female age 60-64
G65T69	4	4		Numeric	Number of female age 65-69
G70T74	4	4		Numeric	Number of female age 70-74
G75T84	4	4		Numeric	Number of female age 75-84
G85OV	4	4		Numeric	Number of female age over85

Structure for database: **Ganwmal**
Description: Population Dynamics for Georgia non-white male by age

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
ALLAGE	6	6		Numeric	Number of male for all ages
GUN5	5	5		Numeric	Number of male under 5
G5T9	5	5		Numeric	Number of male age 5-9
G10T14	4	4		Numeric	Number of male age 10-14
G15T19	4	4		Numeric	Number of male age 15-19
G20T24	5	5		Numeric	Number of male age 20-24
G25T29	4	4		Numeric	Number of male age 25-29
G30T34	4	4		Numeric	Number of male age 30-34

G35T39	4	4		Numeric	Number of male age 35-39
G40T44	4	4		Numeric	Number of male age 40-44
G45T49	4	4		Numeric	Number of male age 45-49
G50T54	4	4		Numeric	Number of male age 50-54
G55T59	4	4		Numeric	Number of male age 55-59
G60T64	4	4		Numeric	Number of male age 60-64
G65T69	4	4		Numeric	Number of male age 65-69
G70T74	4	4		Numeric	Number of male age 70-74
G75T84	4	4		Numeric	Number of male age 75-84
G85OV	4	4		Numeric	Number of male age over85

Structure for database: **Ganwfmal**
Description: Population Dynamics for Georgia non-white female by age

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	11	11		Numeric	Year
ALLAGE	5	5		Numeric	Number of male for all ages
GUN5	5	5		Numeric	Number of male under 5
G5T9	5	5		Numeric	Number of male age 5-9
G10T14	4	4		Numeric	Number of male age 10-14
G15T19	4	4		Numeric	Number of male age 15-19
G20T24	5	5		Numeric	Number of male age 20-24
G25T29	4	4		Numeric	Number of male age 25-29
G30T34	4	4		Numeric	Number of female age 30-34
G35T39	4	4		Numeric	Number of female age 35-39
G40T44	4	4		Numeric	Number of female age 40-44
G45T49	4	4		Numeric	Number of female age 45-49
G50T54	4	4		Numeric	Number of female age 50-54
G55T59	4	4		Numeric	Number of female age 55-59
G60T64	4	4		Numeric	Number of female age 60-64
G65T69	4	4		Numeric	Number of female age 65-69
G70T74	4	4		Numeric	Number of female age 70-74
G75T84	4	4		Numeric	Number of female age 75-84
G85OV	4	4		Numeric	Number of female age over85

Structure for database: **Scwmal**
Description: Population Dynamics for South Carolina white male by age

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing

					Standard
YEAR	5	5		Numeric	Year
ALLAGE	6	6		Numeric	Number of male for all ages
SCUN5	4	4		Numeric	Number of male under 5
SC5T9	4	4		Numeric	Number of male age 5-9
SC10T14	4	4		Numeric	Number of male age 10-14
SC15T19	6	6		Numeric	Number of male age 15-19
SC20T24	6	6		Numeric	Number of male age 20-24
SC25T29	4	4		Numeric	Number of male age 25-29
SC30T34	4	4		Numeric	Number of male age 30-34
SC35T39	4	4		Numeric	Number of male age 35-39
SC40T44	4	4		Numeric	Number of male age 40-44
SC45T49	4	4		Numeric	Number of male age 45-49
SC50T54	4	4		Numeric	Number of male age 50-54
SC55T59	4	4		Numeric	Number of male age 55-59
SC60T64	4	4		Numeric	Number of male age 60-64
SC65T69	4	4		Numeric	Number of male age 65-69
SC70T74	4	4		Numeric	Number of male age 70-74
SC75T84	4	4		Numeric	Number of male age 75-84
SC85OV	4	4		Numeric	Number of male age over85

Structure for database: **Scwfmal**
Description: Population Dynamics for South Carolina white female by age

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
ALLAGE	6	6		Numeric	Number of female for all ages
SCUN5	4	4		Numeric	Number of female under 5
SC5T9	4	4		Numeric	Number of female age 5-9
SC10T14	4	4		Numeric	Number of female age 10-14
SC15T19	4	4		Numeric	Number of female age 15-19
SC20T24	5	5		Numeric	Number of female age 20-24
SC25T29	4	4		Numeric	Number of female age 25-29
SC30T34	4	4		Numeric	Number of female age 30-34
SC35T39	4	4		Numeric	Number of female age 35-39
SC40T44	4	4		Numeric	Number of female age 40-44
SC45T49	4	4		Numeric	Number of female age 45-49
SC50T54	4	4		Numeric	Number of female age 50-54
SC55T59	4	4		Numeric	Number of female age 55-59
SC60T64	4	4		Numeric	Number of female age 60-64
SC65T69	4	4		Numeric	Number of female age 65-69
SC70T74	4	4		Numeric	Number of female age 70-74

SC75T84	4	4		Numeric	Number of female age 75-84
SC85OV4	4	4		Numeric	Number of female age over85

Structure for database: **Scnwmal**
Description: Population Dynamics for South Carolina non-white male by age

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
ALLAGE	6	6		Numeric	Number of male for all ages
SCUN5	4	4		Numeric	Number of male under 5
SC5T9	4	4		Numeric	Number of male age 5-9
SC10T14	4	4		Numeric	Number of male age 10-14
SC15T19	4	4		Numeric	Number of male age 15-19
SC20T24	4	4		Numeric	Number of male age 20-24
SC25T29	4	4		Numeric	Number of male age 25-29
SC30T34	4	4		Numeric	Number of male age 30-34
SC35T39	4	4		Numeric	Number of male age 35-39
SC40T44	4	4		Numeric	Number of male age 40-44
SC45T49	4	4		Numeric	Number of male age 45-49
SC50T54	4	4		Numeric	Number of male age 50-54
SC55T59	4	4		Numeric	Number of male age 55-59
SC60T64	4	4		Numeric	Number of male age 60-64
SC65T69	4	4		Numeric	Number of male age 65-69
SC70T74	4	4		Numeric	Number of male age 70-74
SC75T84	4	4		Numeric	Number of male age 75-84
SC85OV4	4	4		Numeric	Number of male age over85

Structure for database: **Scnwfmal**
Description: Population Dynamics for South Carolina non-white female by age

Item_Name	Width	Output	N.Dec	Type	Description
FIPS	5	5		Numeric	Federal Information Processing Standard
YEAR	5	5		Numeric	Year
ALLAGE	6	6		Numeric	Number of female for all ages
SCUN5	4	4		Numeric	Number of female under 5
SC5T9	4	4		Numeric	Number of female age 5-9
SC10T14	4	4		Numeric	Number of non-white female age 10-14
SC15T19	4	4		Numeric	Number of non-white female age 15-19
SC20T24	4	4		Numeric	Number of non-white female age 20-24

SC25T29	4	4	Numeric	Number of non-white female age 25-29
SC30T34	4	4	Numeric	Number of non-white female age 30-34
SC35T39	4	4	Numeric	Number of non-white female age 35-39
SC40T44	4	4	Numeric	Number of non-white female age 40-44
SC45T49	4	4	Numeric	Number of non-white female age 45-49
SC50T54	4	4	Numeric	Number of non-white female age 50-54
SC55T59	4	4	Numeric	Number of non-white female age 55-59
SC60T64	4	4	Numeric	Number of non-white female age 60-64
SC65T69	4	4	Numeric	Number of non-white female age 65-69
SC70T74	4	4	Numeric	Number of non-white female age 70-74
SC75T84	4	4	Numeric	Number of non-white female age 75-84
SC85OV	4	4	Numeric	Number of non-white female age over85

ENTITY RELATIONSHIPS DIAGRAM, PHYSICAL DATA DIAGRAM, AND DATA DICTIONARY

Introduction

This section describes a general data model for building a GIS-based data repository for use in computing dose-to-man amounts. The model is based on the simplest form of dose-to-man: source term is transported via pathways to the target receptor, man. The logic is graphically represented in level 0.0 of the included entity-relationship diagrams. The first level below this (0.1, 0.2 etc.) reference a rough physical model for each major component.

This data model is high level, only. Additional work needs to be done to refine the data elements within each entity to assure that temporal and geographic scale descriptors are adequate for the scale of the project. For instance, the entities currently contain a reference to area or acreage location. This may become a county reference, if that is appropriate to the modeling scale and the available data. It may become more highly focused if a census-tract identifier is deemed usable for the particular data element. Note that the groundwater pathway is not modeled in this design.

Implicit Relationships

The next section describes, generically, the data layers that are contained in the GIS repository. The collected entity-relationship (ER) diagrams and data dictionary (Table F-4) represent the underlying data layers that are associated with but not part of the GIS data collection. The underlying data layers can be collected and used without involving the GIS model. As well, any GIS data layers may be used in spatial analysis without invoking other database engines.

The term “clipping,” used in describing the data layers below, refers to the GIS process of excluding all data not within a specified study area. A base data layer determining this study area is implied in the other data set descriptions.

Part of the power of the GIS approach, however, is that the data can be selected from the database using implicit relationships: primarily geographic coordinate and secondarily time. Most of the tables modeled in the collected ER diagrams have both spatial and temporal components; each has been keyed to the GIS data layers where appropriate.

Base Geographic Data Layers

Census Tract Data

This data layer consists of a corrected Topologically Integrated Geographic Encoding and Referencing (TIGER) line file clipped to the extent of the study area, as provided by the U.S. Census Bureau. TIGER line files are known to have internal consistency errors, and the layer must be, at minimum, built as a polygon layer with no label errors. Corrected TIGER line files may be available commercially, and this alternative should be considered before attempting to manually correct the errors.

With the TIGER line files built consistently, the TIGER database is accessible for the area of interest. The census database has many useful statistics about the demographics of an area. These statistics are represented by the entity labeled receptor in the receptor ER diagram, 0.4. Additional information will be keyed to this file.

Boundary

This data layer consists of, at minimum, the USGS 1:24000 scale boundary information from the Digital Line Graph (DLG) database clipped to the area of interest. Boundaries have a strong temporal component, and each useful time frame should be represented in a separate data layer. Modelers will find hunting unit boundaries and forest service boundaries to be useful, and these may need to be acquired and added to this data layer in addition to the USGS data.

Migratory Ranges

As available, migratory ranges of large game animals will be captured to be compatible with 1:24000 scale base layers. These ranges will be used primarily as a reference to determine the likelihood of wild game uptake by local population.

Land Cover Data Layer

This file consists of a polygonal representation of the all types of land cover in the study area. This data layer can be extracted from image classification if the data set has not been developed. The strength of the temporal component of this data set is unknown. A brief overview of agricultural practices and development in the study area should indicate the importance of the temporal component. Gradients of urbanization and pasture and grazing lands should be identified on this data layer. The scale of the data available for modeling is not known, and the key value between the GIS data layer and, thus, the tabular data is not clearly defined.

Surface Hydrology

These files consist of, at minimum, the hydrology DLG data from the USGS 1:24000 scale maps, clipped (as appropriate) to the study area; any additional standing water body information as polygons; and the National Wetlands Inventory information represented at the same scale. Stream segments and associated information (flow direction and stream reach magnitude) are also useful for hydrologic modeling, if available. Flow direction can be corrected using utilities provided with ArcINFO. These data are keyed to information in the ER model through the entity RIVERS or the entity LAKES, using the arc and polygon identifiers for the keys to the underlying data.

Gauging Station

This information is often available from the USGS as a point coverage that can be subsequently clipped to the study area. This is really the accurate representation of the water balance numbers provided by the USGS. These data are keyed to the tabular data in the ER diagram through the GAUGING STATION entity.

Meteorology Stations

A data set similar to the gauging stations data set can be acquired through the National Oceanic and Atmospheric Administration (NOAA), indicating where the meteorology stations are located and keying their station identifiers. These data are keyed to the database through the entity MET STATION.

Transportation Data Layer

This data layer consists of the best representation of the public roadways available in the study area. These data may be extracted from the TIGER line files, but much additional information needs to be appended to the data layer to make this a useful approach. Other sources may provide a more useful data layer. These data are useful to define old political boundaries and historical development. The data are represented in the ER model as the ROUTES entity.

Releases Data Layer

This layer will consist of point and polygon releases as they are identified by the research team. These data are related to the ER model through the SOURCE TERM tables.

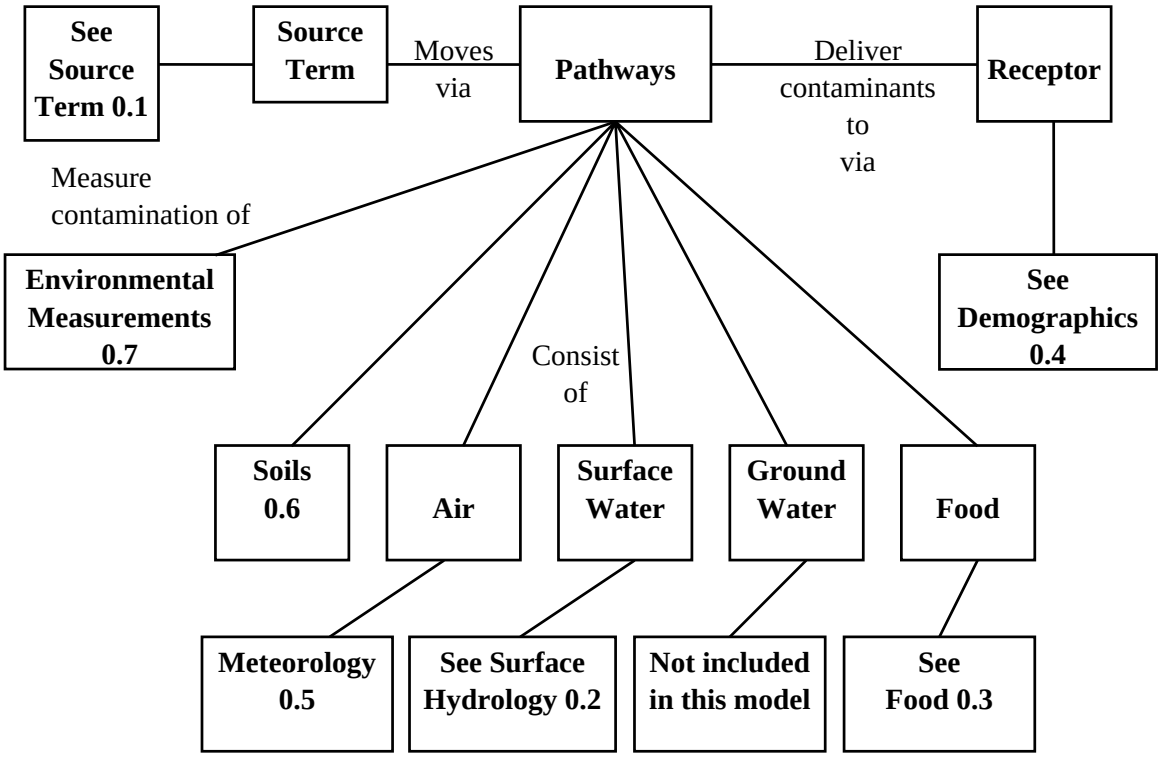
Available Photographs

Available photographs, such as soil conservation service aerials, will be digitized and stored online if deemed necessary for ongoing research. There will be no attempt to georeference this data until the photos are actually used.

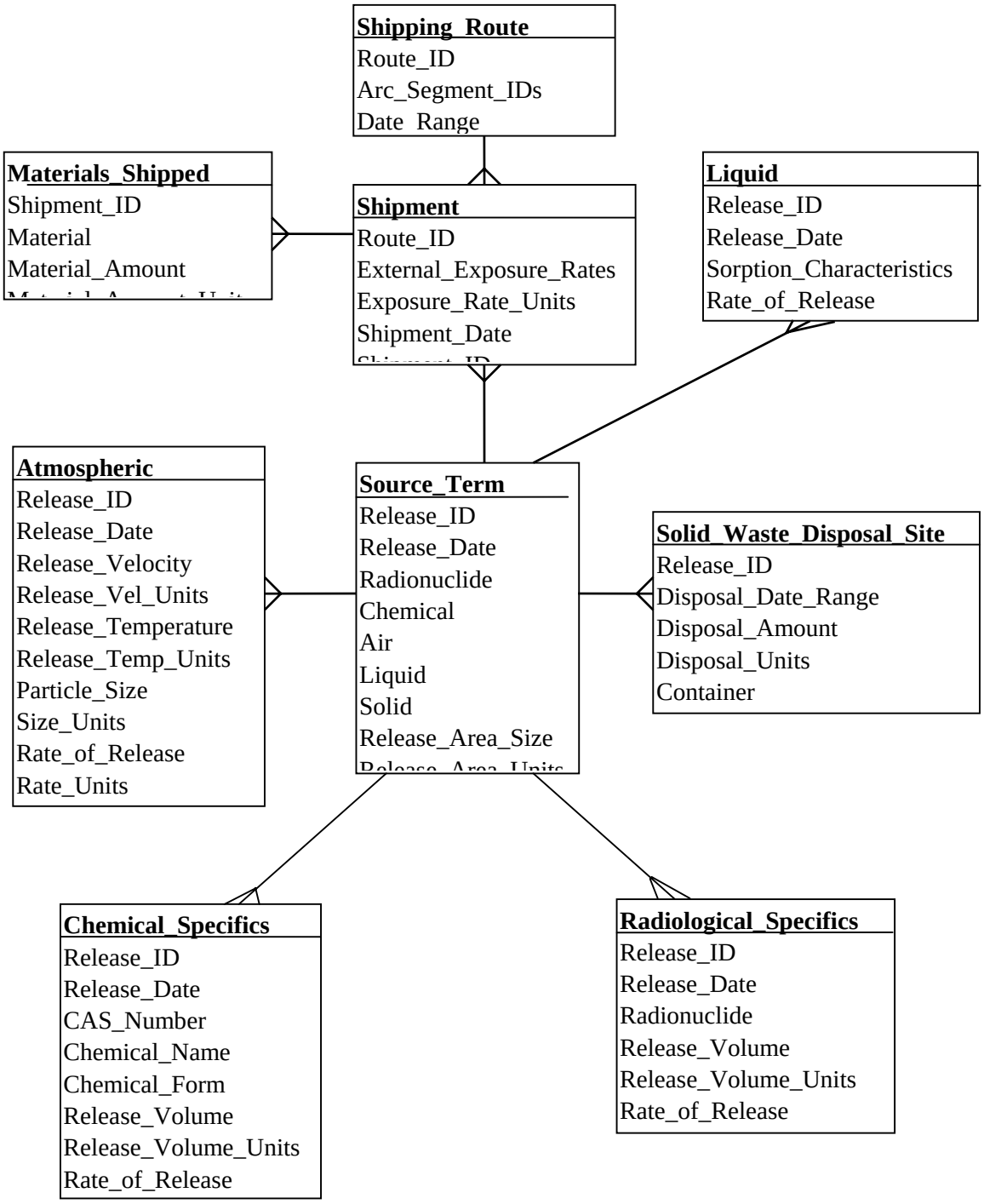
Entity Relationship Diagrams

The following section details the ER diagrams, beginning with level 0.0 and extending down vertically one level. The physical implementation of this model would require two or more additional vertical levels to account for cross-reference tables and foreign key relationships.

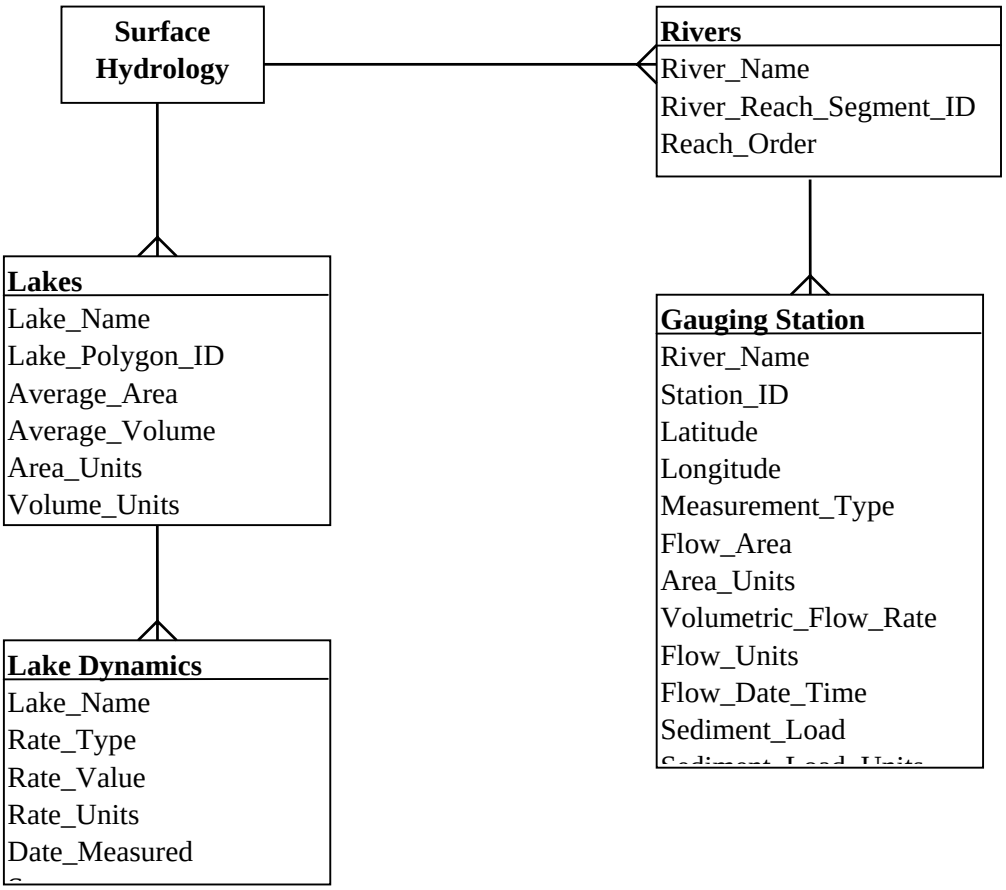
Dose to Man Data Model 0.0



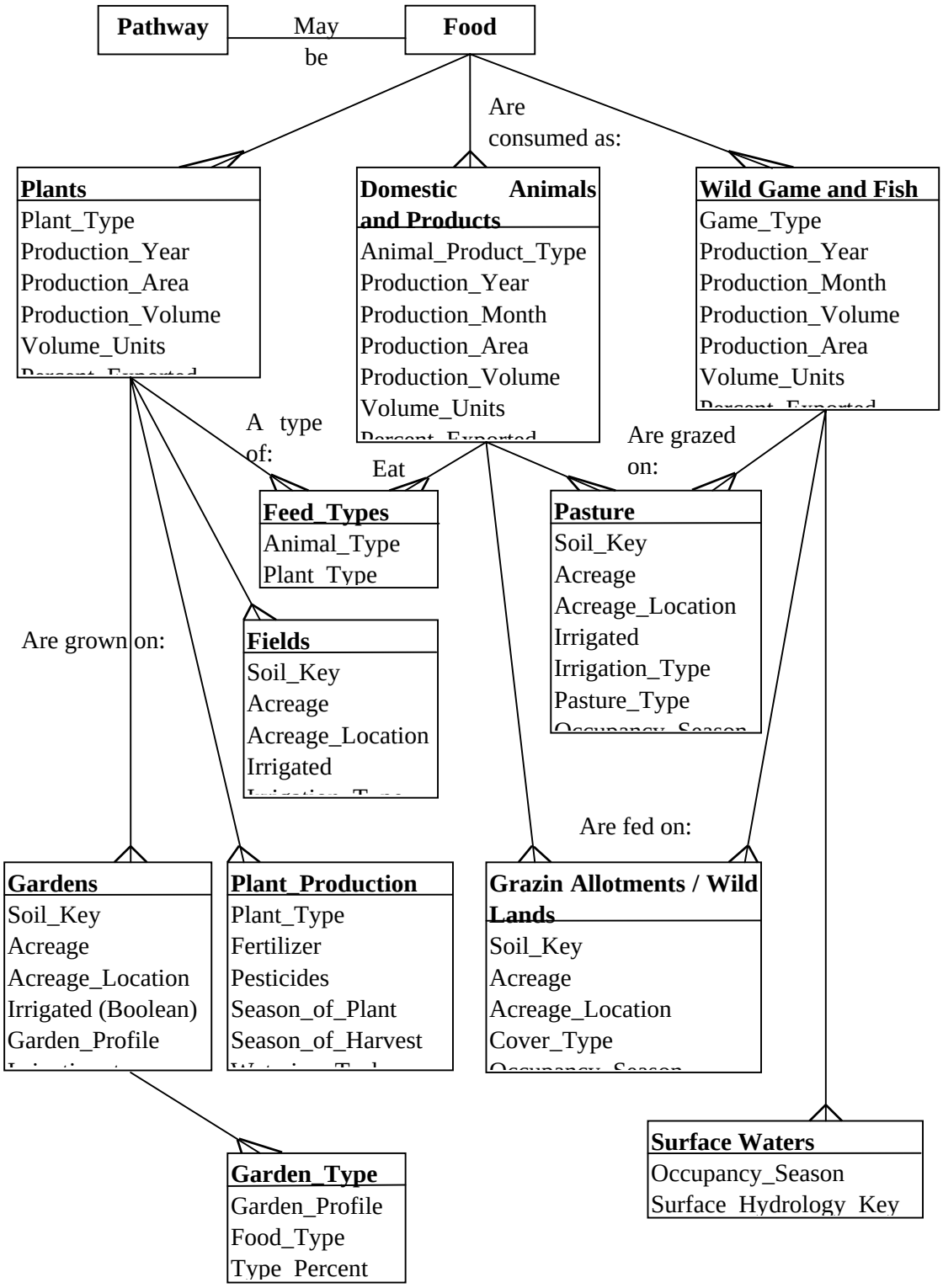
Dose to Man - Source Term - 0.1



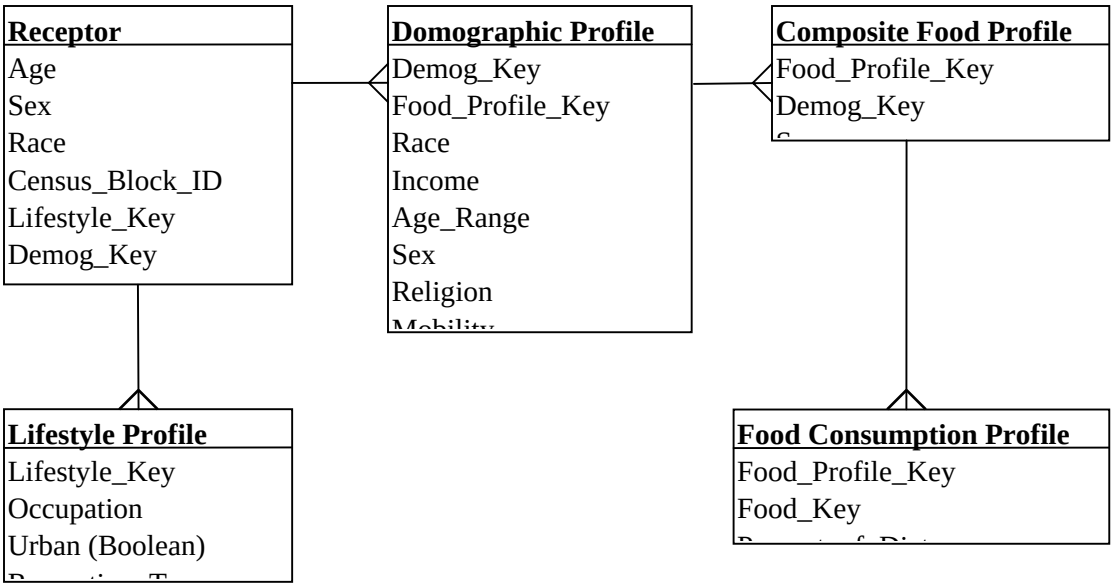
Surface Hydrology - 0.2



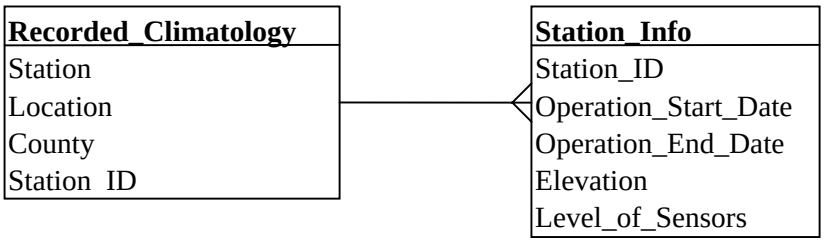
Dose to Man - Food - 0.3



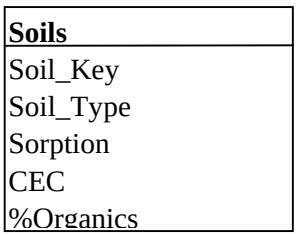
Dose to Man - Receptor - 0.4



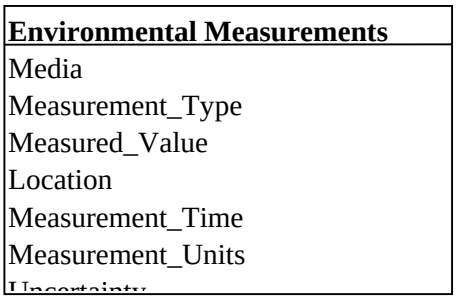
Dose to Man - Meteorology - 0.5



Soils - 0.6



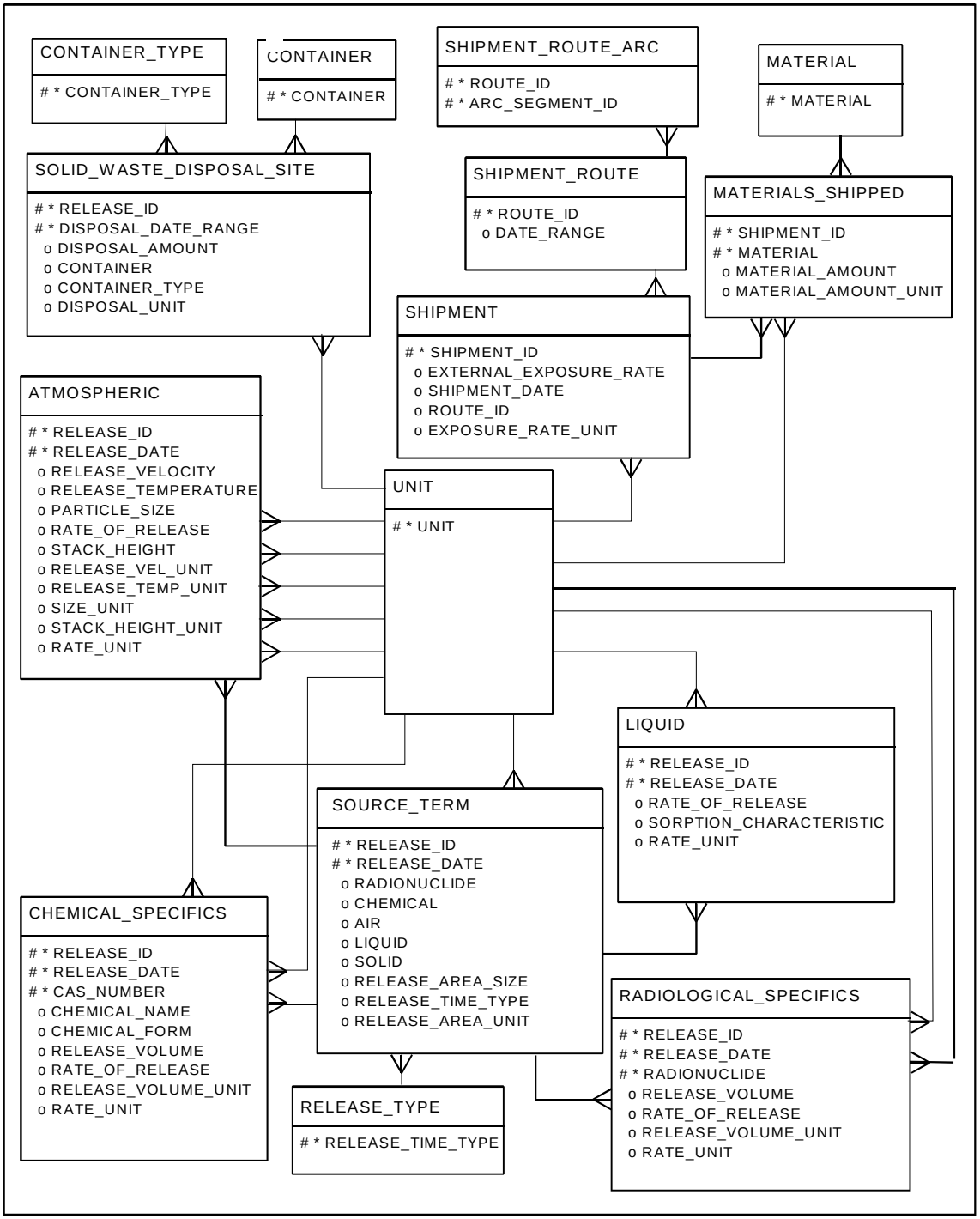
Environmental Measurements - 0.7



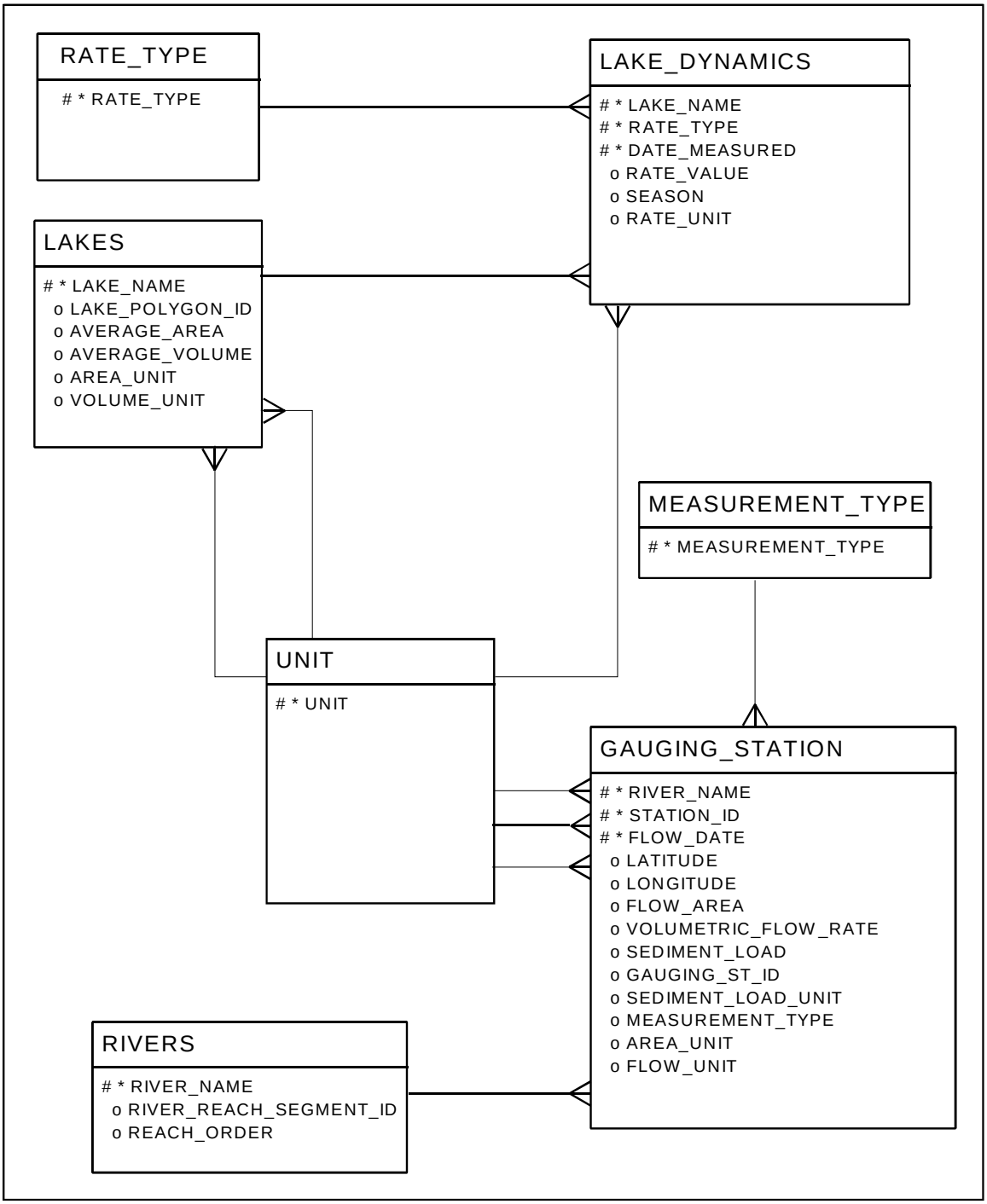
Physical Data Diagrams

This section contains the physical data for the ER diagram presented above.

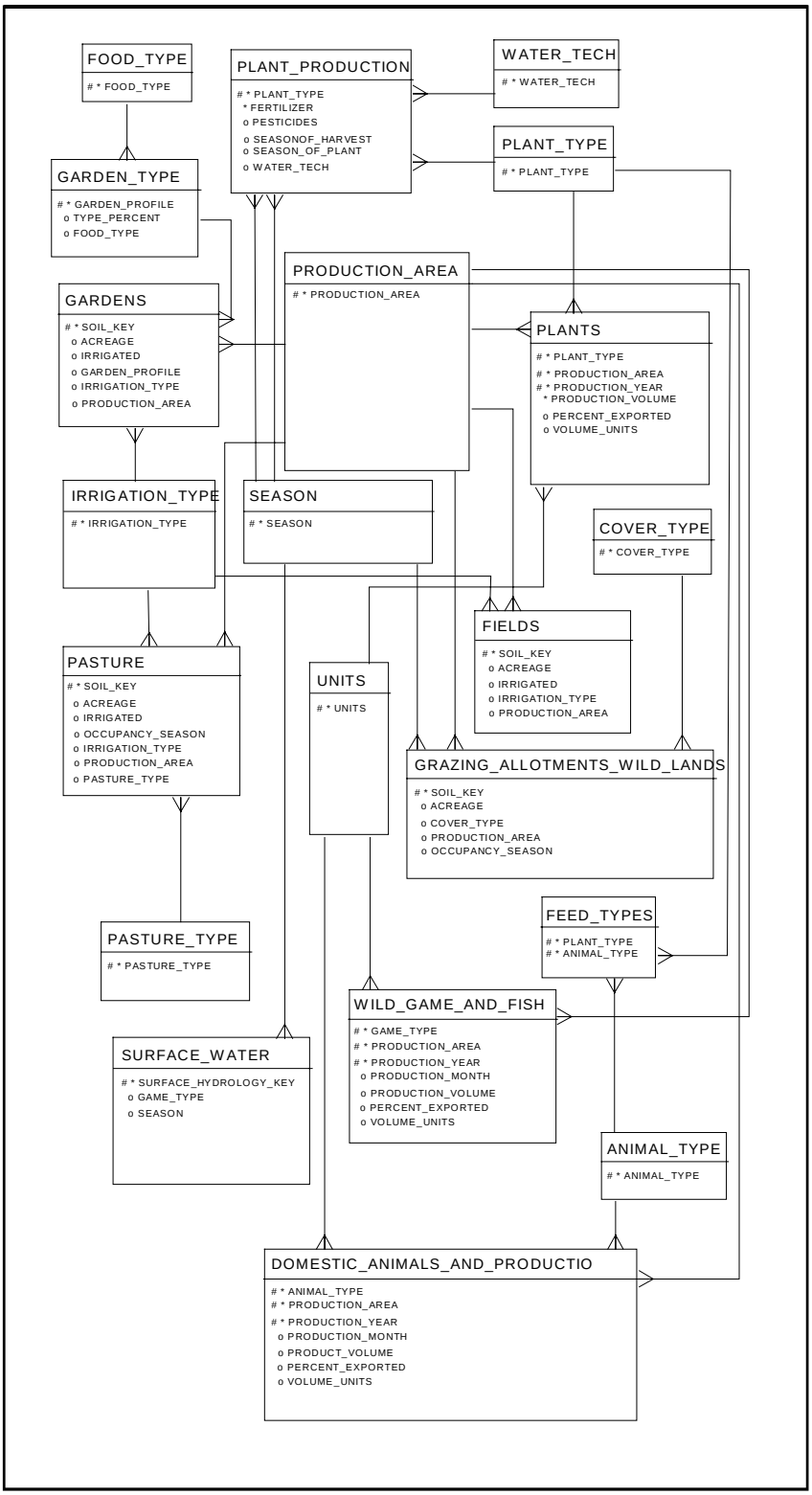
Dose to Man - Source4 Term 0.1



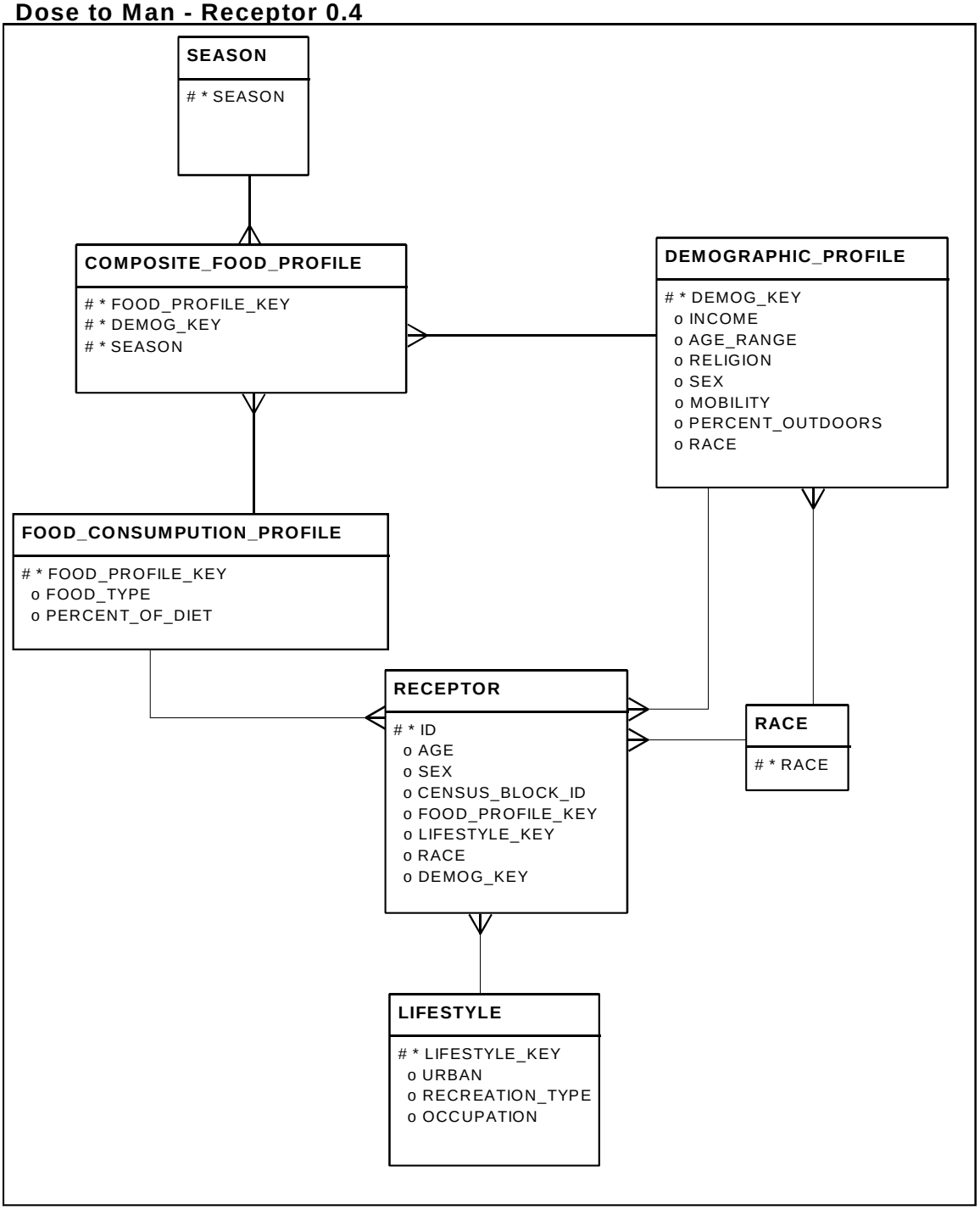
Surface Hydrology - 0.2



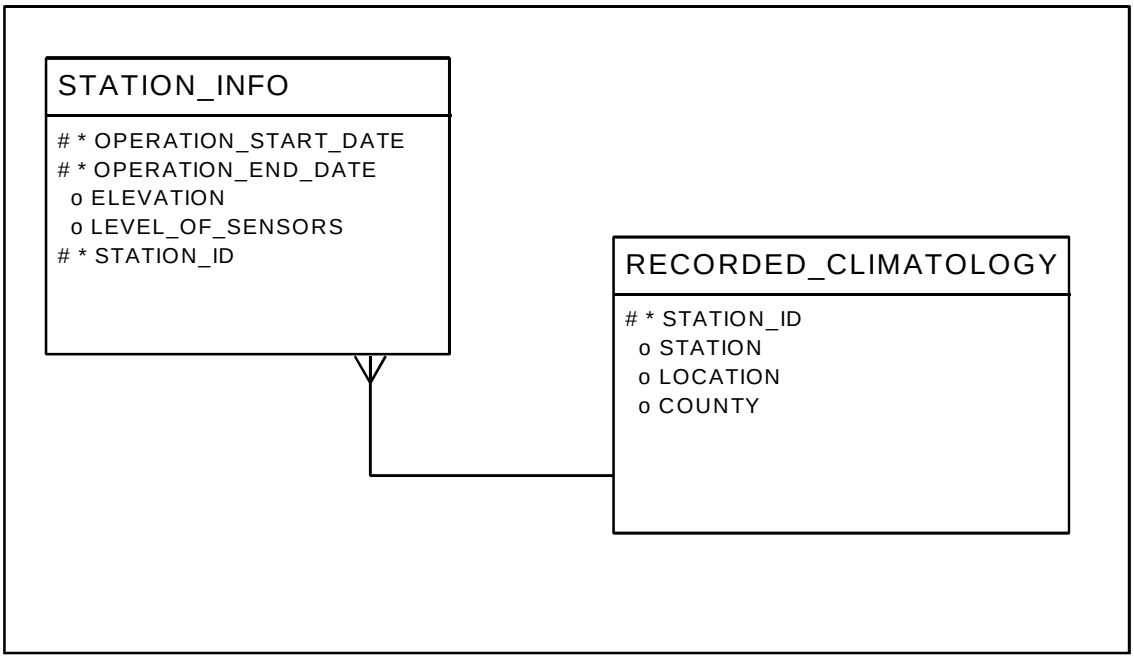
Dose to Man - Food - 0.3



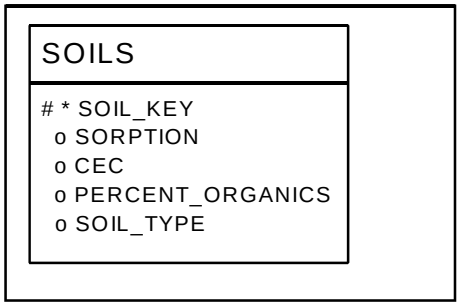
Dose to Man - Receptor 0.4



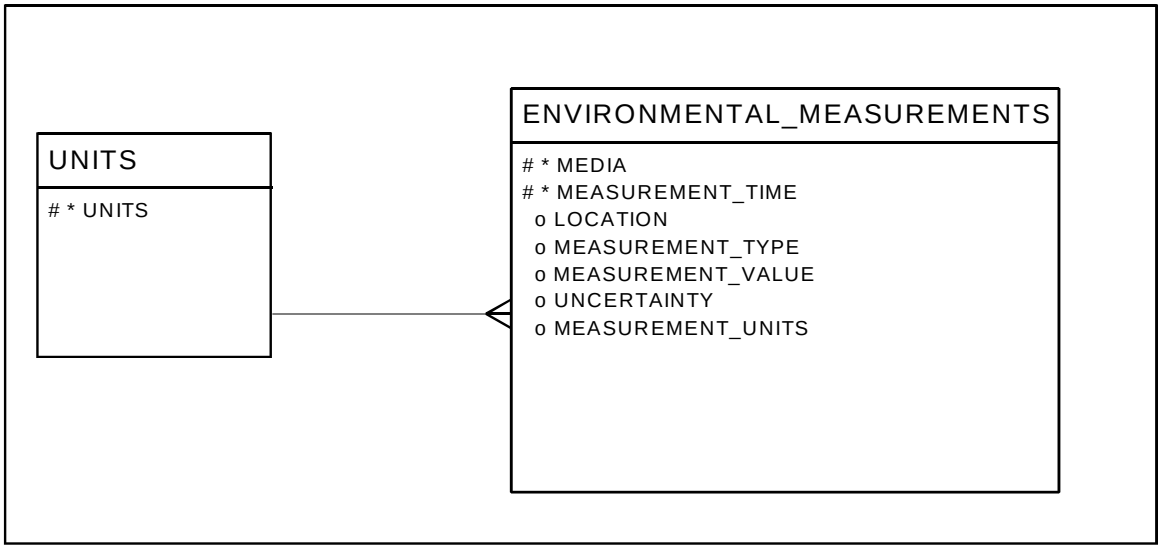
Does to Man Meteorology - 0.5



Soils - 0.6



Environmental Mesurements - 0.7



Data Dictionary

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
LAKES TBL			
LAKE_NAME	C	40	Official USGS lake name, if available. This is the generic key value for all still bodies of water (exclusive of smaller wetlands).
LAKE_POLYGON_ID	I	10	User-assigned feature number in the Arc/INFO coverage used to relate the table with the coverage. Must be defined the same in the table and in the coverage.
AVERAGE_AREA	N		Arc/INFO computed area of the polygon defined as the lake, based on the average area determined from the USGS topography
AVERAGE_VOLUME	N		Arc/INFO computed volume of the defined lake if bottom profiles are available
AREA_UNITS	C	10	Unit of measure used to determine area (meters, feet, etc.)
VOLUME_UNITS	C	10	Unit of measure used to determine volume (cubic meters, acre feet, etc.)
RIVERS TBL			

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
RIVER_NAME	C	40	Official USGS river name, as available. This is the generic key value for an identified river, stream, or branch.
RIVER_REACH_SEGMENT_ID	I	10	User-assigned feature number in the Arc/INFO coverage used to relate the table with the coverage. Must be defined the same in the table and in the coverage. A river consists of many riv_seg_ids.
REACH_ORDER	N		River segment reach (USGS-assigned reach number)
LAKE_DYNAMICS TBL			
LAKE_NAME	C	40	Official USGS lake name. Key value, relates to Lakes tbl above.
RATE_TYPE	C	50	Rate category, such as sedimentation, evaporation, or infiltration rates, used to define water balance and sedimentation dynamics (in- and out-flow rates)
RATE_VALUE	N		The rate of elevation change in the lake
RATE_UNITS	C	10	Unit of measure associated with rate measurements
DATE_MEASURED	D	8	Date the rate was calculated or measured
SEASON	C	12	Time of year (fall, spring, etc.) the data were gathered
GAUGING_STATION TBL			
RIVER_NAME	C	40	Official USGS river name. Key value.
STATION_ID	C	40	A unique string identifying the gauging station (name of the gauging station)
LATITUDE	N		Geographic coordinate of the gauging station on the surface of the earth
LONGITUDE	N		Geographic coordinate of the gauging station on the surface of the earth
MEASUREMENT_TYPE	C	40	Text string describing measurement categories, for example, flow meter, measured cross-section and strip chart, etc.
FLOW_AREA	N		Area of the measurement cross-section as determined by the measuring entity
AREA_UNITS	C	10	Unit of measure used to determine area
VOLUMETRIC_FLOW_RATE	N		Flow volume as measured for this gauging stream and this record

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
FLOW_UNITS	C	10	Unit of measure associated with volumetric flow rate
FLOW_DATE	D	8	Date the data were gathered from the gauging station. Date format MM/DD/YY and time.
SEDIMENT_LOAD	N	10	The sediment load measured for this reach
SEDIMENT_LOAD_UNITS	C	10	Unit of measure used to determine sediment load
GAUGING_ST_ID	I	10	User-assigned feature number in the Arc/INFO coverage used to relate the table and the coverage. The ID must be defined the same in the table and in coverage.
PLANTS TBL			
PLANT_TYPE	C	60	Text string describing crop type for this record
PRODUCTION_YEAR	I	4	Year of crop harvest for this record
PRODUCTION_AREA	C	50	Text string describing crop location place (farm, county, etc.)
PRODUCTION_VOLUME	N		Amount of crop harvested in this location and during this harvest year
VOLUME_UNITS	C	10	Production unit of measure
PERCENT_EXPORTED	N		Percentage of crop which left the county or other specified production area
FEED_TYPES			
PLANT_TYPE	C	60	Text string describing crop type for this record
ANIMAL_TYPE	C	60	The type of animal
PLANT_PRODUCTION TBL			
PLANT_TYPE	C	60	Text string describing crop type for this record
FERTILIZER	C	40	Fertilizers used on the plants
PESTICIDE	C	4	Pesticides used on the plants
SEASON_OF_PLANT	C	10	Season the crop was planted
SEASON_OF_HARVEST	C	10	Season the plant was harvested
WATERING_TECH	C	40	Watering technique used on the crop
GARDEN_TYPE TBL			

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
GARDEN_PROFILE	I	5	Key value identifying a particular garden profile (many records compose a garden profile). The sum of the type percent should equal 100 for each garden type.
FOOD_TYPE	C	40	Plant type grown (carrot, peas, etc.)
TYPE_PERCENT	N		Percentage of each type of plant grown
FIELDS TBL			
SOIL_KEY	I	5	The key to the soil table
ACREAGE	N		Number of acres farmed with consumable products.
PRODUCTION_AREA	C	50	May contain city, county, or state
IRRIGATED	B	1	Contains Y for irrigated, N for not irrigated
IRRIGATION_TYPE	C	40	The type of irrigation used on the land
DOMESTIC_ANIMAL_AND_PRODUCTION TBL			
ANIMAL_PRODUCT_TYPE	C	50	Type of animal product (cow, eggs, chicken, etc.)
PRODUCTION_YEAR	I	4	Year the product was processed for food consumption
PRODUCTION_MONTH	I	2	Month the product was processed for food consumption
PRODUCTION_AREA	C	50	Identifies place of product origin (farm, county, etc.)
PRODUCTION_VOLUME	N		Volume associated with product for this record (may be head of cattle, gallons of milk, dozens of eggs, etc.)
VOLUME_UNITS	C	10	Animal type
PERCENT_EXPORTED	N		Percentage of this animal product which left the production area
WILD_GAME_AND_FISH TBL			
GAME_TYPE	C	50	Text string containing type of wild game, fowl, or fish (deer, bear, elk, etc.). Key value.
PRODUCTION_YEAR	I	4	Year the animal was processed for food consumption
PRODUCTION_MONTH	I	2	Month the animal was processed for food consumption
PRODUCTION_AREA	C	50	Describes the place of harvest (state, forest, county, etc.)

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
PRODUCTION_VOLUME	N		Number of animals referenced by this record at this specified time they were processed for food consumption
VOLUME_UNITS	C	10	Units of measure for harvest type
PERCENT_EXPORTED	N		Percentage of the animal type that was exported from the harvest area
PASTURE TBL			
SOIL_KEY	I	5	The key to the soil table
ACREAGE	N		Number of acres used as pasture
PRODUCTION_AREA	C	50	Contains location information: city, county, or state
IRRIGATED	B	1	Contains Y for irrigated, N for not irrigated
IRRIGATION_TYPE	C	40	The type of irrigation used on the land
PASTURE_TYPE	C	40	Type of feed on the pasture: grass, hay, etc.
OCCUPANCY_SEASON	C	30	Time of year the pasture is used by animals
GRAZING_ALLOTMENTS_WILD_LANDS TBL			
SOIL_KEY	I	5	The key to the soil table
ACREAGE	N		Number of acres used as grazing or wild lands for this record (and this location)
PRODUCTION_AREA	C	50	Contains a description of the location of the acreage for this record. May be city, county, forest, or state, etc.
COVER_TYPE	C	40	Type of vegetation on the acreage
OCCUPANCY_SEASON	C	30	Time of year the land is used by grazing animals
SURFACE_WATER TBL			
GAME_TYPE	C	50	Species using this water body
SEASON	C	10	Period of time that the waters are used by this species
SURFACE_HYDROLOGY_KEY	N		Polygon or arc linking this record to the information contained in the appropriate Arc/INFO data layer
RECORDED_CLIMATOLOGY TBL			
STATION	C	40	Meteorology station name as assigned by NOAA or other owning agency. This field must be unique.

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
LOCATION	C	40	Description of the physical location of the station
COUNTY	C	40	County name in which the station is located
STATION_ID	C	10	Point identifier from the Arc/INFO coverage. This both assures uniqueness and allows access to the information contained in the INFO database (location, topology, etc.).
STATION_INFO TBL			
OPERATION_START_DATE	D		Start of operation
OPERATION_END_DATE	D		End of operation
LEVEL_OF_SENSORS	C	50	The level of the sensors
ELEVATION	N		Elevation of station
STATION_ID	C	10	Point identifier from the Arc/INFO coverage. This both assures uniqueness and allows access to the information contained in the INFO database (location, topology, etc.).
SOURCE_TERM TBL			
RELEASE__ID	C	20	The release identifier. This is a user-generated, unique key that will be used to relate all of the tables in the source term relation. This field must be unique for each record. Release ID is the key into the Arc/INFO coverage that has coordinates for the release.
RELEASE_DATE	D		The date and time stamp that specifies when the release information for this record was collected.
RADIONUCLIDE	C	1	A Boolean indicating whether there was one or more radionuclide components for this release identifier
CHEMICAL	C	1	A Boolean indicating whether there was one or more chemical components for this release identifier
AIR	C	1	A Boolean indicating whether this release has an atmospheric component
LIQUID	C	1	A Boolean indicating whether this release has a liquid component
SOLID	C	1	A Boolean indicating whether this release has a solid component

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
RELEASE_AREA_SIZE	N		For nonpoint source releases, a measurement of the release area is referenced by this field
RELEASE_AREA_UNITS	C	10	The associated units for the release area size field
RELEASE_TIME_TYPE	C	20	This field describes the type of release with respect to time: periodic, episodic, single event, etc.
MATERIALS_SHIPPED TBL			
SHIPMENT_ID	C	20	The shipment identifier. This is a user-generated key that will be used to relate all of the shipment-oriented tables in the source term relation.
MATERIAL	C	40	For each shipment_id there may be several materials, one for each record
MATERIAL_AMOUNT_UNITS	C	20	The units associated with the amount value for this record
MATERIAL_AMOUNT	N		The quantity of material associated with this record and this shipment
SHIPMENT_ROUTE TBL			
ROUTE_ID	C	20	A user-assigned identifier that references a collection of roadways that is repeatedly used for shipping waste products to waste repositories or temporary storage sites
DATE_RANGE	C	30	Range of dates that this route was used for shipping waste
SHIPMENT_ROUTE_ARC			
ARC_SEGMENT_IDS	N		Each route_id is composed of several arc_segment identifiers from the Arc/INFO data layer
ROUTE_ID	C	20	The shipment route arc
SHIPMENT TBL			
ROUTE_ID	C	20	A user-assigned identifier that references a collection of roadways that is repeatedly used for shipping waste products to waste repositories or temporary storage sites
EXTERNAL_EXPOSURE_RATE	N		Measured exposure rate for the route identified by this record

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
EXPOSURE_RATE_UNITS	C	20	Units associated with the exposure rate units measured for this record
SHIPMENT_DATE	D		The date and time stamp that specifies when this shipment was moved from start
SHIPMENT_ID	C	20	Shipment identifier as defined in the MATERIALS_SHIPPED.TBL relation
LIQUID TBL			
RELEASE_ID	C	20	The release identifier. This is a user-generated, unique key that will be used to relate all of the tables in the source term relation.
RELEASE_DATE	D		The date and time stamp that specifies when this release occurred
SORPTION_CHARACTERISTIC	C	40	Sorption characteristics associated with the material identified in this record
RATE_OF_RELEASE	N		Measured rate of release for the release identified in this record
RATE_UNITS	C	20	Units associated with the rate of release measurement
ATMOSPHERIC TBL			
RELEASE_ID	C	20	The release identifier. This is a user-generated, unique key that will be used to relate all of the tables in the source term relation.
RELEASE_DATE	D		The date and time stamp that specifies when this release occurred
RELEASE_VELOCITY	N		Measured stack release velocity
RELEASE_VEL_UNITS	C	20	Units associated with the release velocity from the stack
RELEASE_TEMPERATURE	N		Temperature of the exit gas
RELEASE_TEMP_UNITS	C	10	Units of measure for the release temperature for this record
PARTICLE_SIZE	N		Associated particulate size, if this record documents a particulate release
SIZE_UNITS	C	10	The units of measure associated with the particulate size measurement for this record
STACK_HEIGHT	N		The height of the stack from the ground
STACK_HEIGHT_UNITS	C	10	The units of measure associated with the stack height
RATE_OF_RELEASE	N		Release volumetric rate measurement

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
RATE_UNITS	C	20	Units associated with the volumetric release measurement
SOLID_WASTE_DISPOSAL_SITE TBL			
RELEASE_ID	C	20	The release identifier. This is a user-generated, unique key that will be used to relate all of the tables in the source term relation. Several records may be associated with this field.
DISPOSAL_DATE_RANGE	C	30	The range of dates that this site was in operation. mm/dd/yy-mm/dd/yy
DISPOSAL_AMOUNT	N		Amount of the material that is disposed at this repository.
DISPOSAL_UNITS	C	20	Units associated with this volume or amount measurement
CONTAINER	C	20	Type of container for this site
CONTAINMENT_TYPE	C	20	Type of containment (barrier, clay liner, etc.) in place for this disposal site
CHEMICAL_SPECIFICS TBL			
RELEASE_ID	C	20	The release identifier. This is a user-generated, unique key that will be used to relate all of the tables in the source term relation. Several fields may be associated with this field.
RELEASE_DATE	D		The release date for this chemical type if different than the parent source term information
CAS_NUMBER	C	15	The chemical abstract system number associated with this material, as released
CHEMICAL_NAME	C	80	The chemical name, as identified by the researchers, for the chemical material released at this release point, for this record
CHEMICAL_FORM	C	20	Form of the chemical material released
RELEASE_VOLUME	N		Volume of the released material corresponding to this record
RELEASE_VOLUME_UNITS	C	20	Units associated with the release volume for this record
RATE_OF_RELEASE	N		Release volumetric rate measurement for this material for this release episode
RATE_UNITS	C	20	Units associated with the rate of release measurement for this record

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
RADIOLOGICAL_SPECIFICS TBL			
RELEASE_ID	C	20	The release identifier. This is a user-generated, unique key that will be used to relate all of the tables in the source term relation. Several fields may be associated with this field.
RELEASE_DATE	D		The release date for this chemical type if different than the parent source term information
RADIONUCLIDE	C	20	Radionuclide name for the material described in this record
RELEASE_VOLUME	N		The volume of material released in this event
RELEASE_VOLUME_UNITS	C	20	The units associated with the release volume
RATE_OF_RELEASE	N		The volumetric release rate for the event referenced by this record
RATE_UNITS	C	20	The volumetric release rate units
RECEPTOR TBL			
ID	N	10	Unique id
AGE	N		Age of receptor
SEX	C	1	Sex of receptor
RACE	C	15	Race of receptor
CENSUS_BLOCK_ID	C	20	Census block identifier associated with this record. This value is carried as an attribute in the Arc/INFO data layer and allows the tabular information in these relations to be spatially related.
LIFESTYLE_KEY	C	15	This is the key value associated with the lifestyle relation
DEMOG_KEY	C	15	This is the key value associated with the demographic profile relation
FOOD_KEY	C	15	This is the key value associated with the food profile relation
DEMOGRAPHIC_PROFILE TBL			
RACE	C	15	Race associated primarily with this demographic profile
INCOME	C	15	Income range associated with this demographic profile

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
AGE_RANGE	C	8	Range of ages associated with this demographic profile
SEX	C	1	The primary sex of this demographic profile, if applicable
RELIGION	C	20	The primary religion of this demographic profile
MOBILITY	N		Mobility of the typical constituent of this demographic profile, as measured in years residence
PERCENT_OUTDOORS	N		Percent of time spent outdoors, by the typical constituent of this profile
COMPOSITE_FOOD_PROFILE TBL			
FOOD_PROFILE_KEY	C	15	This is the key value that relates this entity with the food consumption profile entity
DEMOG_KEY	C	15	The key value that associates this record with the demographics profile
SEASON	C	10	The keys in this table relate the food profile to the demographic group depending on the season indicated in this record
FOOD_CONSUMPTION_ PROFILE TBL			
FOOD_PROFILE_KEY	C	15	This is the food_profile key, a user-defined unique value that is used to indicate the composite food group profile
FOOD_TYPE	C	30	This is the food type (milk, cheese, wheat products, etc.)
PERCENT_OF_DIET	N		This is the percent of the food profile that consists of this food_type
LIFESTYLE TBL			
LIFESTYLE_KEY	C	15	The user-defined, unique key value that defines this lifestyle profile record
OCCUPATION	C	80	The predominant occupation, or class of occupation, that is found in this lifestyle profile
URBAN	C	1	A Boolean indicating urban (= T) or rural (= F)

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
RECREATION_TYPE	C	80	Categories of recreation activity associated with this lifestyle profile
SOILS TBL			
SOIL_KEY	I	5	The key to the soil table
SOIL_TYPE	C	40	The type of soil
SORPTION	N		
CEC	N		
PERCENT_ORGANICS	N		
ENVIRONMENTAL_ MEASUREMENTS TBL			
MEDIA	C	40	The media of the measurement
MEASUREMENT_TYPE	C	40	
MEASUREMENT_VALUE	N		
MEASUREMENT_UNITS	C	10	The units associated with the measurement value
LOCATION	C	40	Description of the physical location
MEASUREMENT_TIME	D		The time of the measurement
UNCERTAINTY	N		The uncertainty of the measurement value
LOOKUP TABLES			
ANIMAL_TYPE TBL			
ANIMAL_TYPE	C	50	The list of animal types
CONTAINER TBL			
CONTAINER	C	20	The list of containers
CONTAINER_TYPE TBL			
CONTAINER_TYPE	C	20	The types of containers
COVER_TYPE TBL			
COVER_TYPE	C	40	The types of covers
FOOD_TYPE TBL			
FOOD_TYPE	C	40	Types of foods
GAME_TYPE TBL			
GAME_TYPE	C	50	Types of game
GARDENS TBL			
SOIL_KEY	I		
ACREAGE	N		
IRRIGATED	C	1	
GARDEN_PROFILE	I		
IRRGATION_TYPE	C	40	
PRODUCTION_AREA	C	50	
IRRIGATION_TYPE TBL			
IRRIGATION_TYPE	C	40	Irrigation methods

Table F-4. Data Dictionary

Table or Field name	Type	Length	Description
MEASUREMENT_TYPE TBL			
MEASUREMENT_TYPE	C	40	Types of measurements
MATERIAL TBL			
MATERIAL	C	40	Materials that can be shipped
PASTURE_TYPE TBL			
PASTURE_TYPE	C	40	Types of pastures
PLANT_TYPE TBL			
PLANT_TYPE	C	60	Plants
PRODUCTION_AREA TBL			
PRODUCTION_AREA	C	50	Production areas
RACE TBL			
RACE	C	15	Race
RATE_TYPE TBL			
RATE_TYPE	C	50	Rate types
RELEASE_TYPE TBL			
RELEASE_TIME_TYPE	C	20	Release time types
SEASON TBL			
SEASON`	C	10	Seasons
UNITS TBL			
UNITS	C	10	Units of measure
WATER_TECH TBL			
WATER_TECH	C	40	Watering techniques

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APPENDIX G

PHASE II RECORDS REVIEW

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PHASE II RECORDS REVIEW

INTRODUCTION

This appendix is a report on the methods and results of review of records routinely received by Savannah River Site Document Control during Phase II of the SRS dose reconstruction study. It covers efforts extended from the end of Phase I to the end of April, 1997, when the backlog of boxes had been evaluated.

A large number of records boxes of unclassified material had accumulated at the SRS Document Control vaults since the completion of the Phase I records review in June, 1995. Site visits in the months following Phase I completion indicated that few of the records accumulated since that time were likely to be valuable to the dose reconstruction project, and that the cost and time needed for total box review would be extremely high.

Radiological Assessments Corporation (RAC), with the assistance of the CDC and the Citizens' Health Effects Subcommittee, developed a methodology to quickly and efficiently review these records. This process involved the use of detailed printouts describing records box contents, along with a program of onsite random checks of the accuracy of the printouts. The methodology and its application are described below. Also during the Phase II work, RAC personnel examined accumulated boxes of classified records in their entirety.

INITIAL EFFORTS AT BOX REVIEW USING ACCEPTED PROTOCOL

Radiological Assessments Corporation received from SRS staff extensive computer printouts of 8,698 unclassified boxes (some were microfilmed, these were also examined) held under the authority of Document Control at SRS. On the basis of box content descriptions, RAC initially assigned category values to the contents; annotations were made on the printouts as to category assigned. In July, 1996 a RAC team member traveled to SRS with the main purpose to begin the work to clarify the contents of 363 boxes which RAC could not categorize based on the printouts. This effort was valuable in that, of the 363 boxes, all were given *RAC* classifications. Only three were deemed to be category 2 records of high potential value to the Project, the rest were category 0 or 1.

During that trip, we also began looking at samples of boxes that had been initially labeled as category 0 or 1, for the purpose of checking our ability to judge box content value via printout examination. This latter work gave us confidence that box content descriptions were generally adequate.

During November, 1996 a second trip to SRS allowed RAC to complete work with some 8,698 backlogged records boxes. As the result of the two visits, 93 of the 133 boxes that had been labeled via the printouts as potentially category 2 were downgraded upon physical examination of box contents. All of the initial group of 18 category 3 boxes were downgraded to 1 or 0. No boxes were upgraded.

RESULTS OF CONSIDERATION OF THE NEXT GROUP OF BOXES

After finishing inspection of boxes from the printout of 8,698 backlogged records boxes, RAC was provided with a computerized listing of about 2,000 additional boxes that had been received by SRS records staff more recently. Prescreening of the computer printout was done before travel to SRS in February and March, 1997. A random number table was employed for selection of boxes to be physically inspected. The results of examination indicated that no boxes fell into the category of 2 or higher. Thirty boxes were identified as category 1, and the remaining as category 0. Also, during this trip, examination of classified materials began, as described below.

APRIL 1997 BOX EVALUATION AT SRS

Three analysts from RAC made a box evaluation trip to SRS in April 1997. This work also allowed RAC to bring up to date the review of unclassified SRS records boxes. No computer printout was provided prior to the Site visit, but one was made available upon arrival. This listing of boxes of paper or microfilm indicated 3,325 additional boxes in storage. Again, a random number table was used to select over 220 boxes of microfilm for examination. Results revealed no material of high potential value to the study. Thirty-four items were considered to be of category 1 status, the rest were category 0.

CLASSIFIED MATERIALS

During February and April 1997, it was also necessary to examine the contents of 458 boxes of classified materials stored in the 773A vault. These efforts required that all boxes be opened, and all contents scanned. These were largely recent materials, and of little value to Dose Reconstruction. Of this group, 55 were labeled as category 1 and the rest were category 0. At that time, RAC had completed review of all classified materials in the 773A vault. This work concluded the process of additional SRS Phase II document review at the site.

APPENDIX H

THE PHASE II DOCUMENT DATABASE

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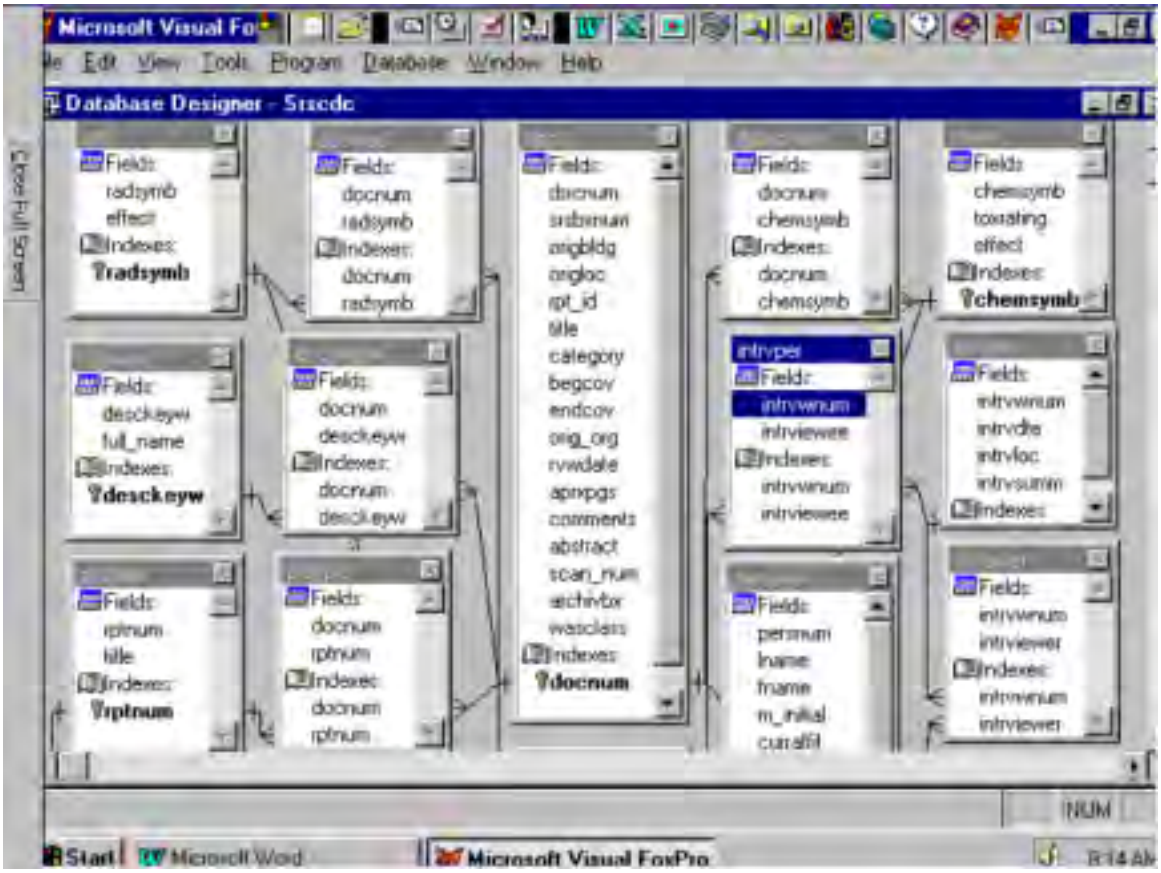
THE PHASE II DOCUMENT DATABASE

Purpose – The purpose of this application is to provide a user friendly method for entering, viewing, and printing data associated with the CDC SRS Dose Reconstruction Project for Phase II. The application's main focus stems from the docs table. Parent to child relationships among the associated tables have been established. Referential integrity rules between parent and child tables have been established to maintain validity of the defined relationships. Validation rules and triggers have been set at the record and field levels to ensure data integrity and consistency. These rules will be applied for update, change, delete, or insert values in parent or child tables. This Application runs in Visual FoxPro Version 3 only.

Entity Relationship Diagram



Data Dictionary/Data Structure

Table Name: DOCS.DBF

Table Description: Stores the core information for records reviewed for Dose Reconstruction

Field	Field Name	Type	Description	Width
1	docnum	Character	Unique number automatically assigned	16
2	srsbxnum	Character	SRS Box where document was found	20
3	origbldg	Character	Building where document was found	15
4	origloc	Character	Location in building where document was found	20
5	rpt_id	Character	Memo #, report #, publication #, Library of Concress ID etc.	20
6	title	Character	Title of the document	254
7	category	Numeric	0=No Value (used in Phase 1 review) 1=Potential value 2=Significant value/Document copied 3=Highly significant value/Document copied and abstracted.	1
8	begcov	Date	Document's coverage beginning date as identified during document inspection	8
9	endcov	Date	Document's coverage ending date as identified during document inspection	8
10	orig_org	Character	Organization that produced the document	25
11	rvwdate	Date	Date reviewer first reviewed the document and initiated entry into database	8
12	aprxpgs	Numeric	Approximate page count for document – Large documents assigned 1000 to indicate significant copy effort	5
13	comments	Memo	Comments that do not logically fit elsewhere but may useful information	4
14	abstract	Memo	If this was a category 3 document, an abstract was written for the document	4
15	scan_num	Character	If the document was scanned and saved in a word processing document, the wp document name is identified here.	12
16	archivbx	Character	Indicates the specific offsite box/microfilm that contains copies of category 2 & 3 documents	15
17	wasclass	Logical	Indicate whether the document was originally a classified document	1
18	initials	Character	Reviewer's initials	10

Table Name: KEYWORD.DBF
Table Description: Stores keyword abbreviations and full descriptions. Keywords are used for search criteria.

Field	Field Name	Type	Description	Width
1	desckeyw	Character	Short name for keyword	20
2	full_name	Character	Full name and description of keyword	50

Table Name: DOCSKEYW.DBF
Table Description: Linking table. Connects a document with its keyword(s).

Field	Field Name	Type	Description	Width
1	docnum	Character	Record's document number	16
2	desckeyw	Character	Keyword associated with record	20

Table Name: CHEM.DBF
Table Description: Stores chemical abbreviations and full descriptions. Chemicals are used for search criteria.

Field	Field Name	Type	Description	Width
1	chemsymb	Character	Short name for chemical	30
2	toxrating	Character	Toxin rating	50
3	effect	Memo	Effects of toxin	4

Table Name: DOCSCHEM.DBF
Table Description: Linking table. Connects a document with its chemical(s).

Field	Field Name	Type	Description	Width
1	docnum	Character	Record's document number	16
2	chemsymb	Character	Chemical associated with record	30

Table Name: RAD.DBF
Table Description: Stores radionuclide description and it's effect. Radionuclides are used for search criteria.

Field	Field Name	Type	Description	Width
1	radsymb	Character	Name/symbol for radionuclide	30
2	effect	Memo	Effects of radionuclide	4

Table Name: DOCSRAD.DBF
Table Description: Linking table. Connects a document with its radionuclide(s).

Field	Field Name	Type	Description	Width
1	docnum	Character	Record's document number	16
2	radsymb	Character	Radionuclide associated with record	30

Table Name: PH2TITLE.DBF
Table Description: Stores names of chapters and appendices created for the Phase 2 Dose Reconstruction.

Field	Field Name	Type	Description	Width
1	rptnum	Character	Number assigned to a chapter or appendix when it is created. Linked to projrpts.dbf.	20
2	title	Character	Title of chapter or appendix	254

Table Name: PROJRPTS.DBF
Table Description: Linking table. Connects referenced document(s) to the chapters and appendices of the Phase 2 Dose Reconstruction report

Field	Field Name	Type	Description	Width
1	docnum	Character	Document # for document used as reference. Linked to docs.dbf.	16
2	rptnum	Character	Chapter or appendix number for the Phase 2 report	20

Table Name: PERSONS.DBF
Table Description: Table that contains the core information for personnel such as authors and reviewers.

Field	Field Name	Type	Description	Width
1	persnum	Character	Automatically assigned unique identification number	10
2	lname	Character	Last name	15
3	fname	Character	First name	15
4	m_initial	Character	Middle initial	2
5	curraffil	Character	Currant affiliation (such as place of employment)	30
6	street	Character	Street address	30
7	city	Character	City	15
8	county	Character	County	15
9	state	Character	State	2
10	zip	Character	Zip code	10
11	country	Character	Country	15
12	phone	Character	Phone number	12
13	initials	Character	Initials	3

Table Name: PREFAFIL.DBF
Table Description: Table that contains information about the previous affiliation(s) of the persons listed in the persons.dbf table.

Field	Field Name	Type	Description	Width
1	personum	Character	Person ID number linked to persons.dbf	10
2	prevafil	Character	Previous affiliation of person	30
3	begdate	Date	Date affiliation such as employment began	8
4	enddate	Date	Date affiliation such as employment ended	8

Table Name: AUTHORS.DBF
Table Description: Linking table that connects author(s) with identified SRS documents

Field	Field Name	Type	Description	Width
1	docnum	Character	Document # link to docs.dbf	16
2	persnum	Character	Number assigned to person entered in person.dbf. Link to person.dbf.	10

Table Name: RPTPERS.DBF
Table Description: Table that identifies authors of Phase 2 reports.

Field	Field Name	Type	Description	Width
1	rptnum	Character	Phase 2 report number. Link to ph2title.dbf	20
2	persnum	Character	Number assigned to personnel entered in person.dbf. Link to person.dbf.	10

Table Name: LOCATION.DBF
Table Description: GIS Location table

Field	Field Name	Type	Description	Width
1	idgisloc	Character	GIS Location identification number	30
2	locdesc	Character	Description of GIS location.	45

Table Name: LOCREL.DBF
Table Description: Linking table that connects document(s) with its chemical(s) and/or radionuclide(s) and the toxin's GIS location

Field	Field Name	Type	Description	Width
1	idgisloc	Character	GIS identification number	30
2	docnum	Character	Document number.	16
2	toxsymb	Character	Radionuclide or chemical symbol or short name	30

Table Name: INTRVIEW.DBF
Table Description: Table contains information about interviews conducted.

Field	Field Name	Type	Description	Width
1	intrvwnum	Character	Automatically assigned unique interview number	10
2	intrvdte	Date	Date interview was conducted	8
3	intrvloc	Character	Location where interview was conducted	30
4	intrvsumm	Memo	Summary of the interview	4

Table Name: INTRVPER.DBF
Table Description: Linking table that links person(s) interviewed with the interview.

Field	Field Name	Type	Description	Width
1	intrvwnum	Character	Interview number that links to table intrview.dbf	10
2	intrviewee	Character	Person interviewed. Link to persnum in table persons.dbf	10

Table Name: INVIEWER.DBF
Table Description: Linking table that links person(s) conducting interview with the interview.

Field	Field Name	Type	Description	Width
1	intrvwnum	Character	Interview number that links to table intrview.dbf	10
2	intrviewer	Character	Person who conducted interview. Link to persnum in table persons.dbf	10

Table Name: INITIALS.DBF
Table Description: Holds initials and names for persons who have names loaded into the welcome screen of the application.

Field	Field Name	Type	Description	Width
1	initials	Character	Person's initials. Linked to initials in docs.dbf.	3
2	name	Character	Full name of person	25

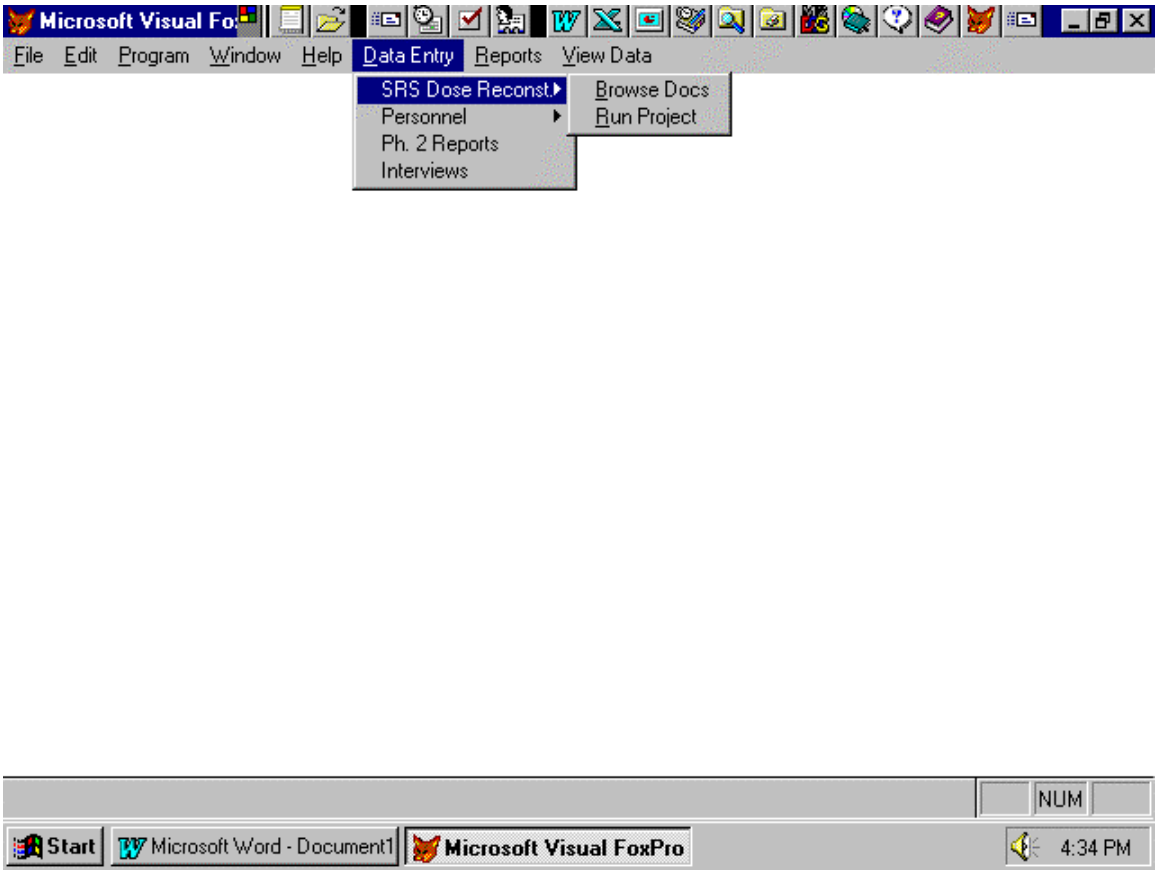
Loading Software

1) Create a subdirectory called C:\vfp\srsph2\.
2) Copy all files into this subdirectory.
3) If you do not have a copy of Visual FoxPro 3.0, move the VFP300.ESL file from the C:\vfp\srsph2\ folder to the C:\vfp\ folder.

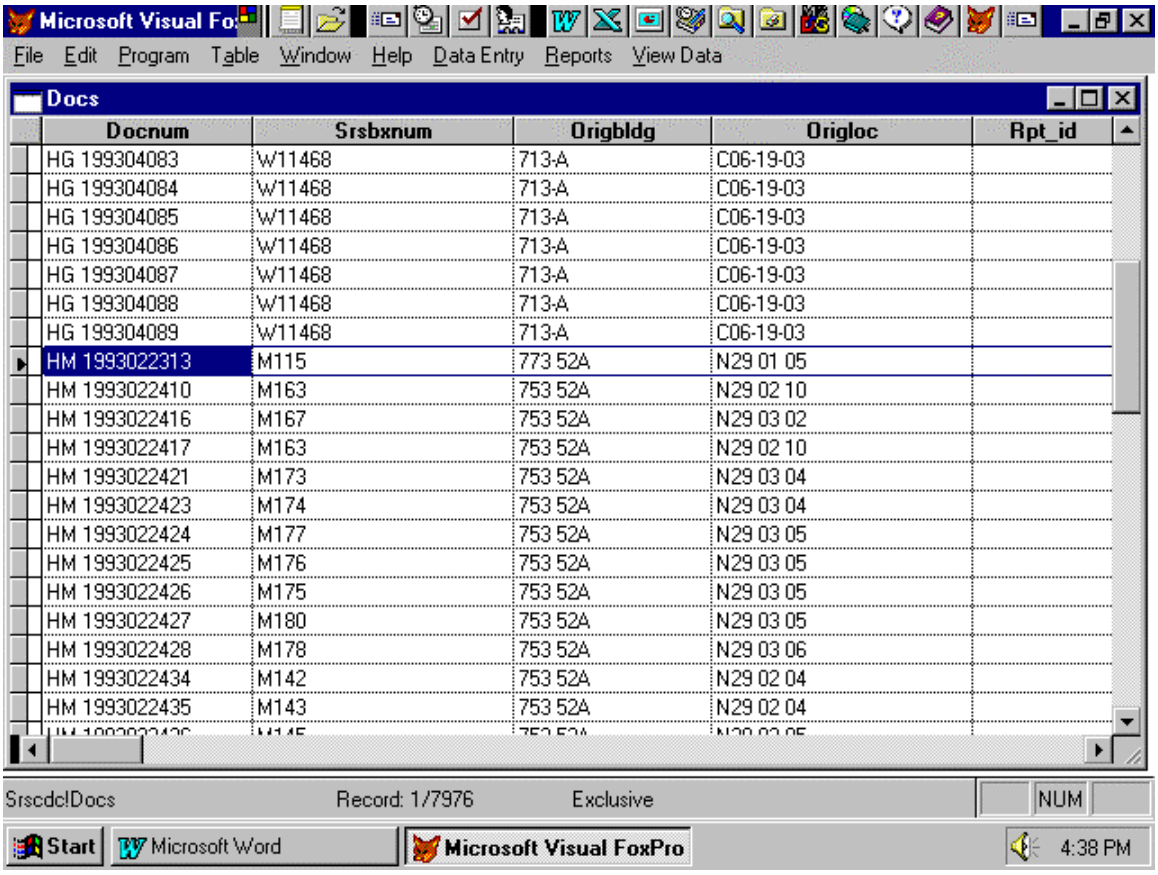
Running application:

This application was created using Visual FoxPro 3.0.
1) If you have a copy of Visual FoxPro Version 3.0 (the Application requires Version 3.0):
From within FoxPro choose Do then choose SRSDOSE.APP or from the Windows environment choose Run and then C:\vfp\srsph2\srsdose.app
2) If you do not have a copy of Visual FoxPro Version 3.0:
From the Windows environment choose Run and then type C:\vfp\srsph2\srsdose2.exe
(you must have a copy of file VFP300.ESL in the VFP subdirectory to run the .exe file). All of the software must be installed on your C: drive, for the simplest use of this application.
*Remember that you may have been given the application for the purpose of viewing the existing documents. Changes that you make will not be reflected in the Master document. All needed changes to existing documents must be reported as described in the Backup section of this document. If you have been given the application for the purpose of making additions, any additions you make must be sent to the Master document holder as described in the Backup section of this document.

Main Menu - The Main Menu is the Entry into the Application.

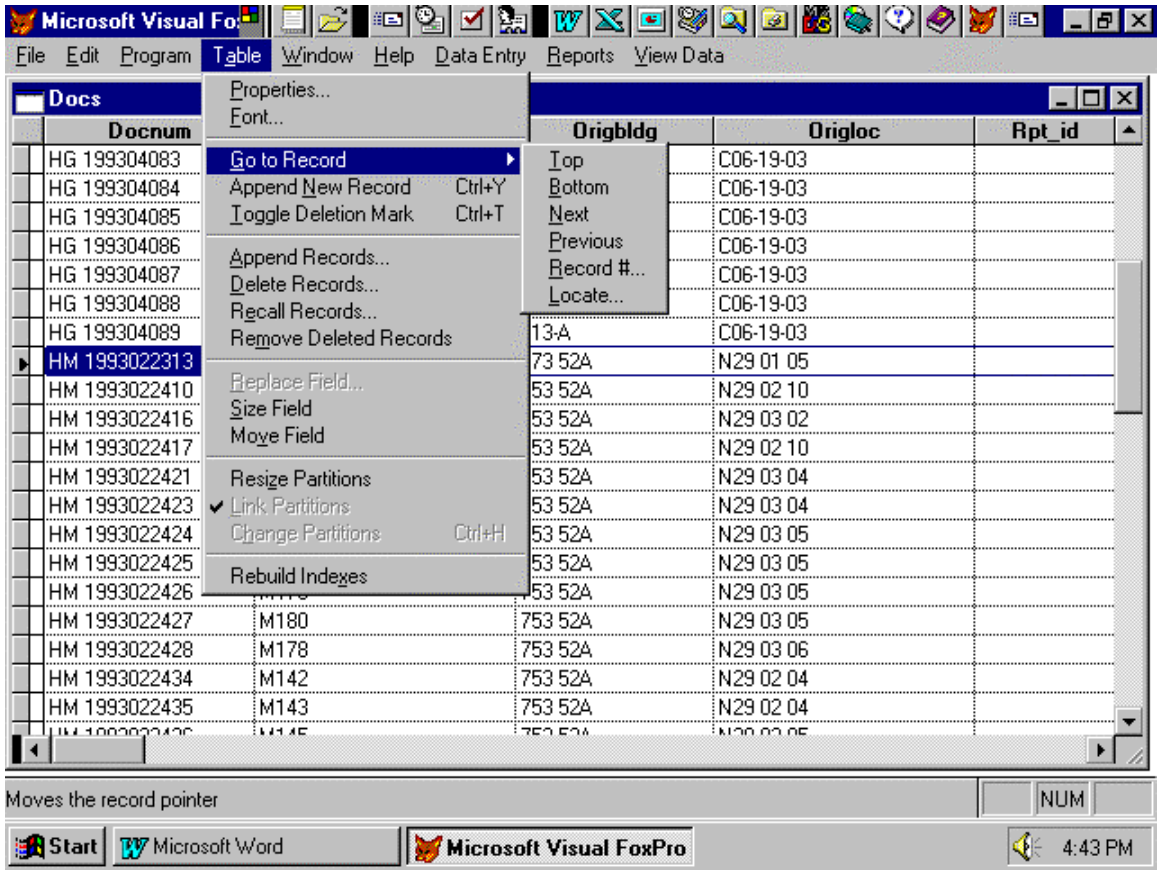


Browse - Choosing Browse brings up the SRS documents table in a spreadsheet format.

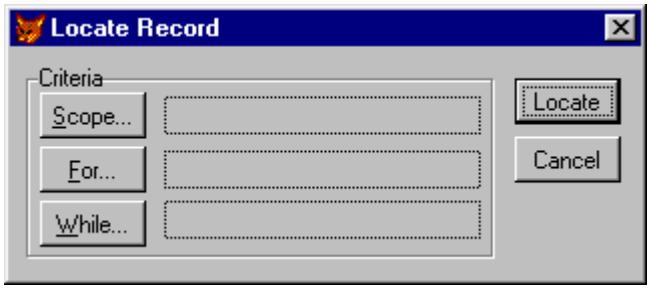


Docnum	Srsbxnum	Origbldg	Origloc	Rpt_id
HG 199304083	W11468	713-A	C06-19-03	
HG 199304084	W11468	713-A	C06-19-03	
HG 199304085	W11468	713-A	C06-19-03	
HG 199304086	W11468	713-A	C06-19-03	
HG 199304087	W11468	713-A	C06-19-03	
HG 199304088	W11468	713-A	C06-19-03	
HG 199304089	W11468	713-A	C06-19-03	
HM 1993022313	M115	773 52A	N29 01 05	
HM 1993022410	M163	753 52A	N29 02 10	
HM 1993022416	M167	753 52A	N29 03 02	
HM 1993022417	M163	753 52A	N29 02 10	
HM 1993022421	M173	753 52A	N29 03 04	
HM 1993022423	M174	753 52A	N29 03 04	
HM 1993022424	M177	753 52A	N29 03 05	
HM 1993022425	M176	753 52A	N29 03 05	
HM 1993022426	M175	753 52A	N29 03 05	
HM 1993022427	M180	753 52A	N29 03 05	
HM 1993022428	M178	753 52A	N29 03 06	
HM 1993022434	M142	753 52A	N29 02 04	
HM 1993022435	M143	753 52A	N29 02 04	

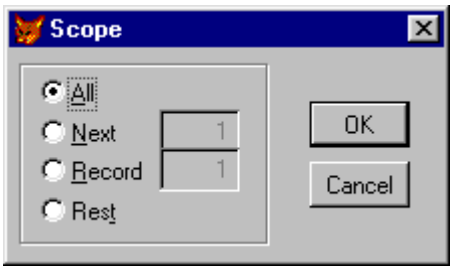
Pulldown menus - allow user to do such things as asking the system to Locate a set of records meeting defined criteria.



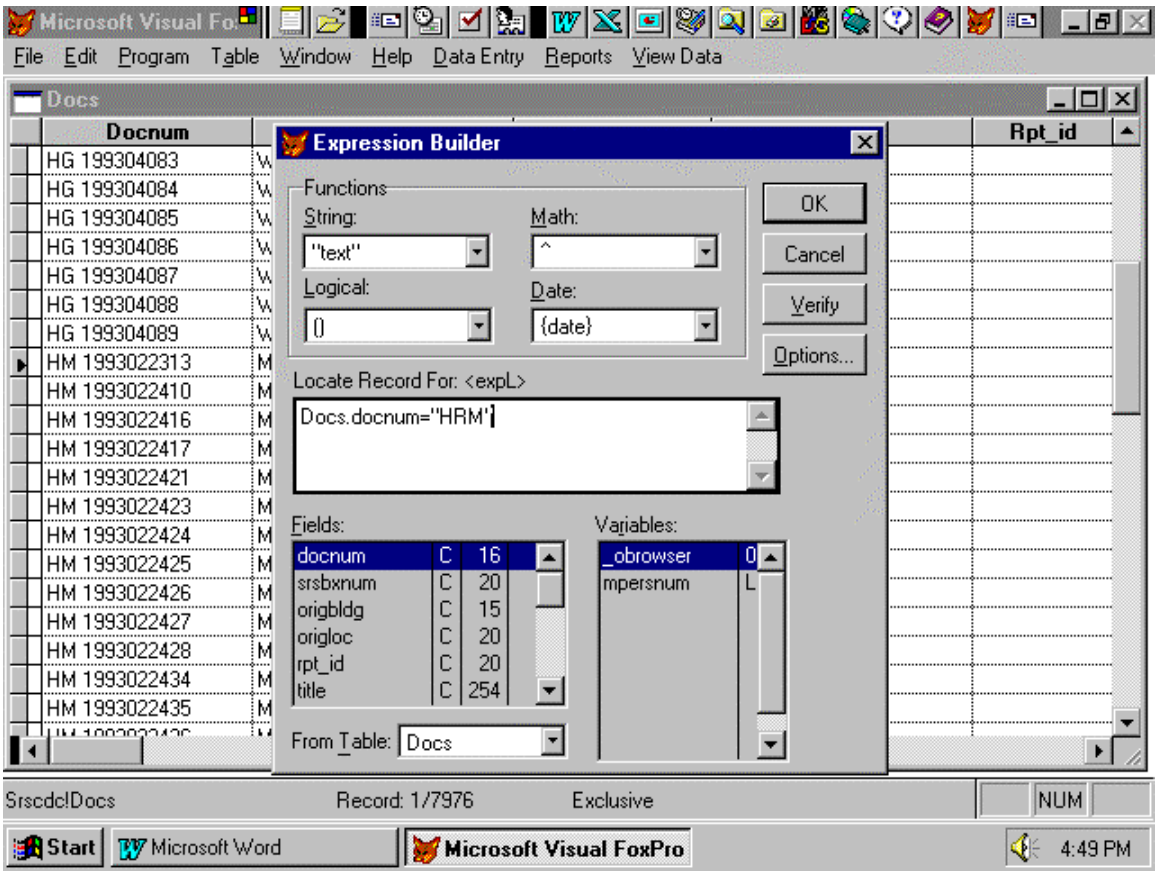
Locate – To locate records choose locate from the pull-down menus as shown above. You will get the following locate record box. From this box you can enter criteria for your search.



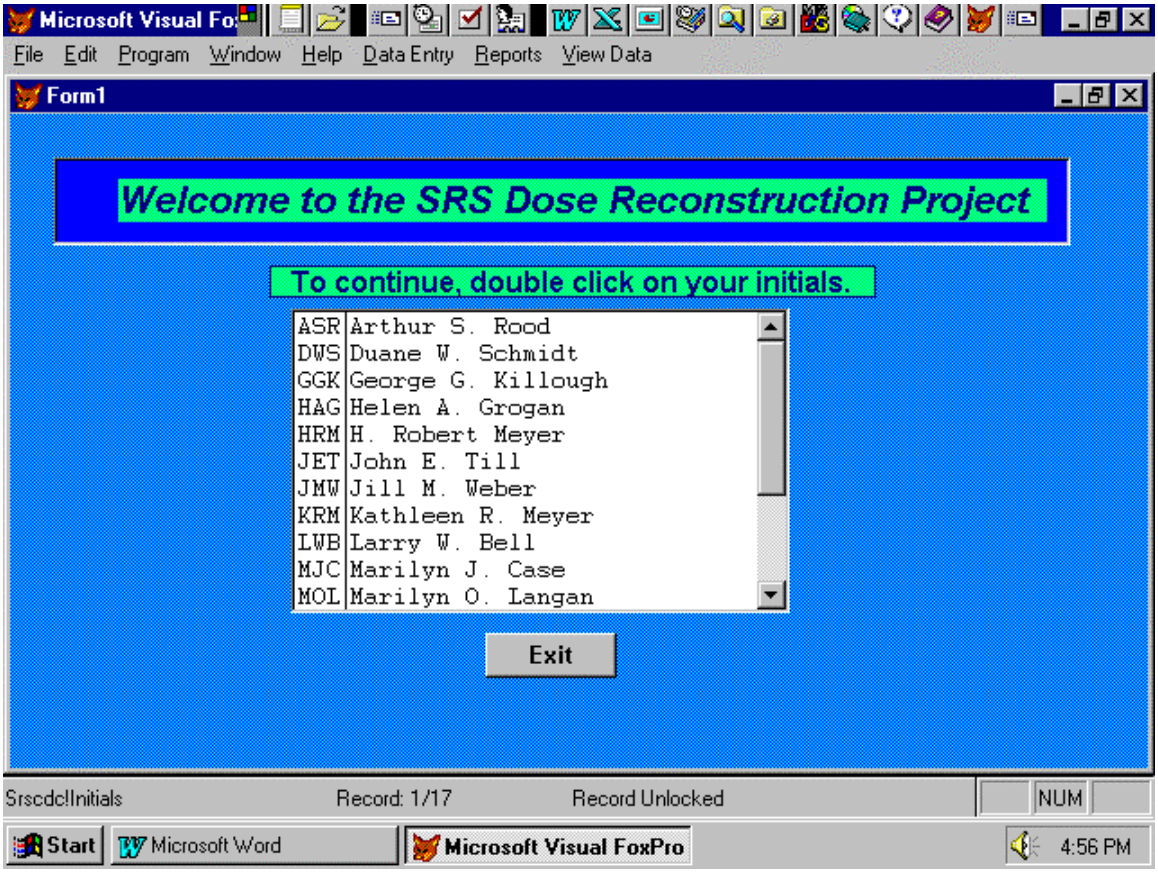
Scope – Pressing the scope button will bring up a box that allows you to specify what records you wish to be searched.



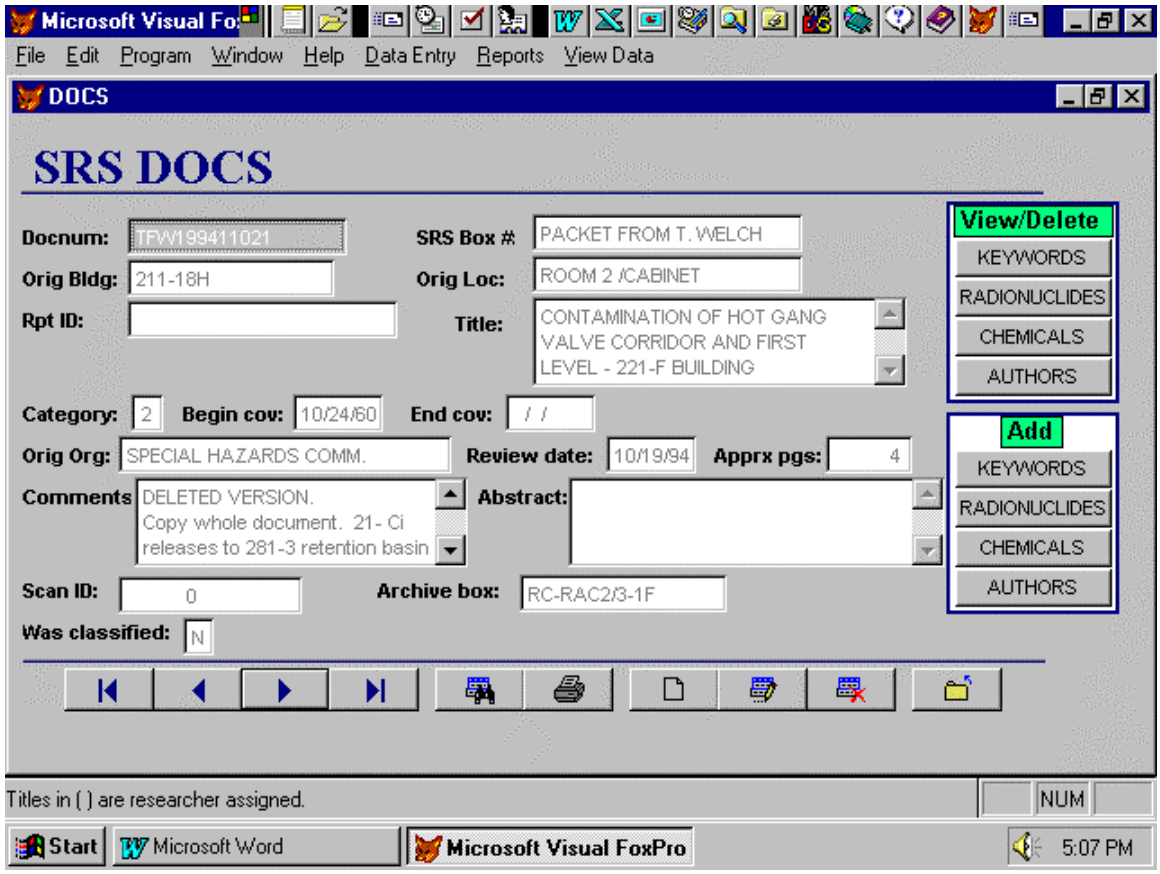
Expression Builder/For or While – Pressing the for button or the while button will bring up the Expression Builder used to define criteria when requesting system to pull particular information. The sample below will pull all records whose document number begins with the initials HRM.



Welcome – Choosing Run Report from the Data Entry/SRS Dose Reconst menu (See Main Menu) pick will bring up the Welcome Screen for the main branch of the application, the SRS Dose Reconstruction Project entry/modification screens.

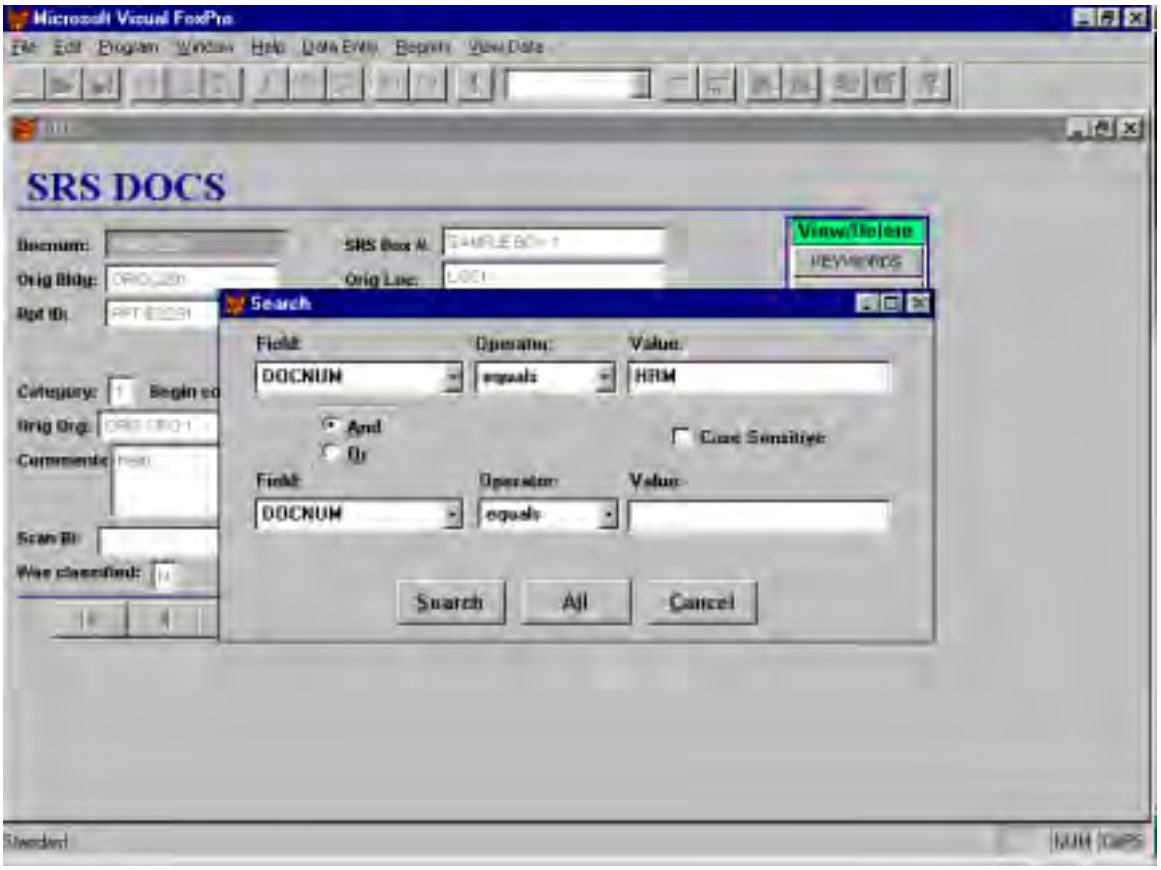


Document Screen - This is the main screen for viewing records and updating the data.

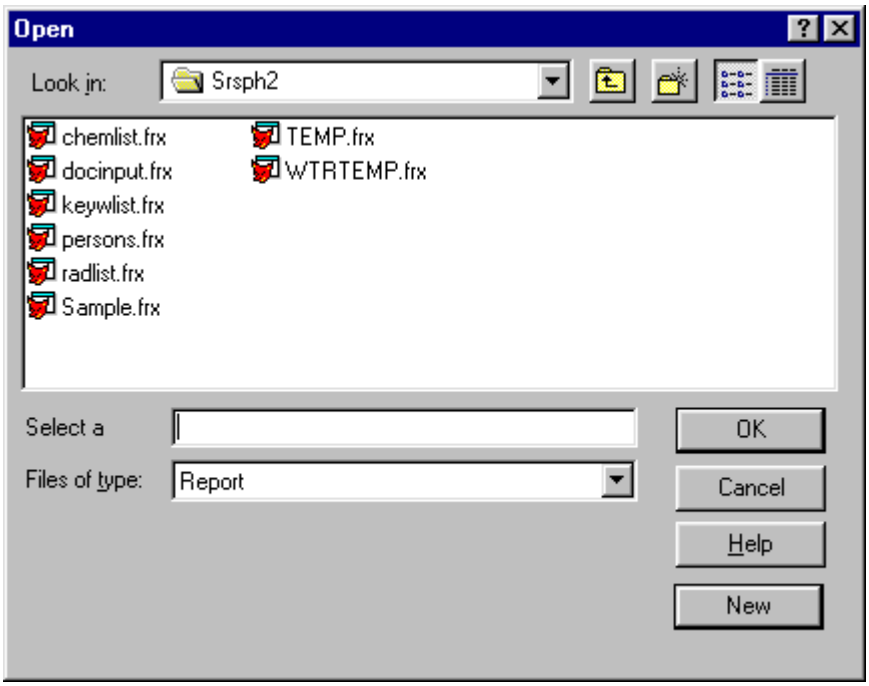


Picture Buttons – The picture buttons at the bottom of the document screen are fairly self-explanatory. Tips will appear if you move your cursor over a button. The first set of 4 buttons are used for moving the record pointer from record to record. They are First Record, Previous Record, Next Record, Last Record. The second set of 2 buttons include a button for searching and one for printing. The third set of 3 buttons include Add/save new record, Edit/Revert Record, Delete Record. The last set of one button is the button to exit this screen and return to the Main Menu.

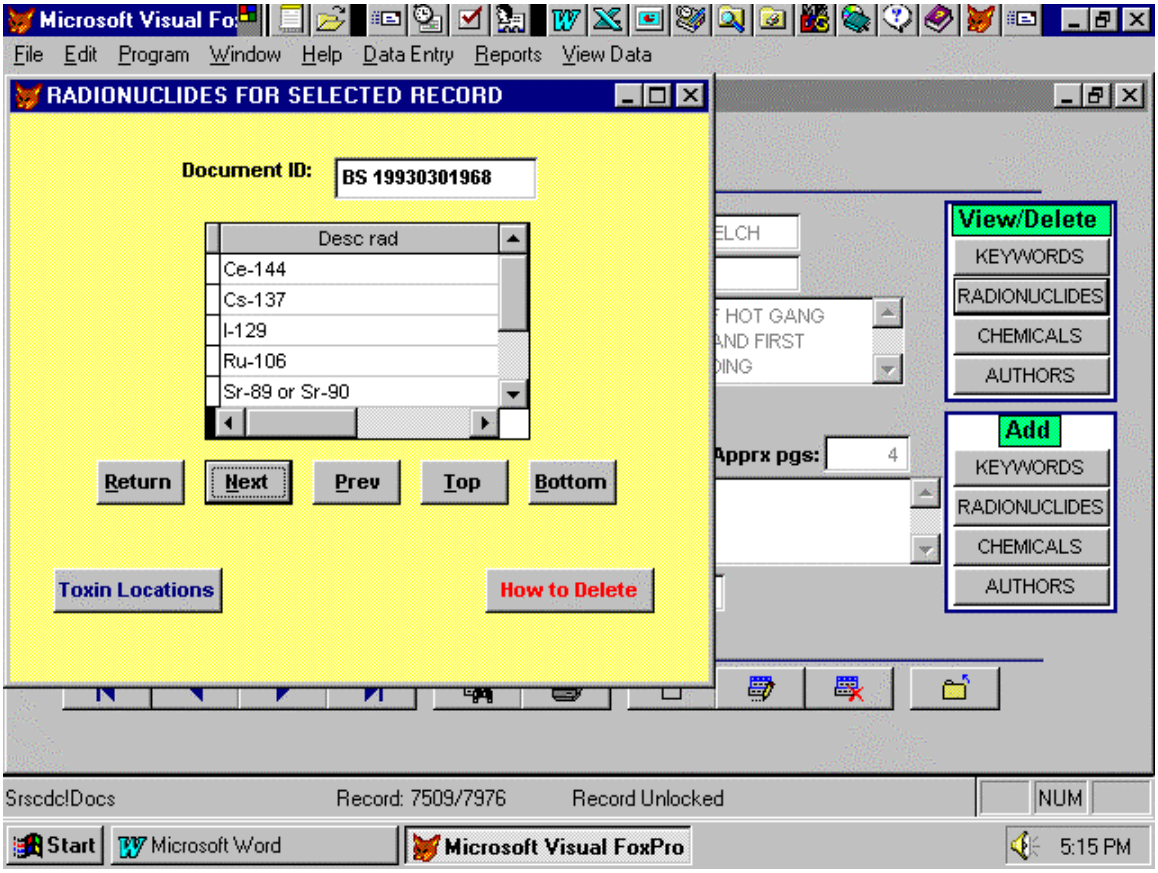
Searching – If you choose the search button you can specify criteria for viewing specific records. In the example below we are searching for documents beginning with the letters HRM.



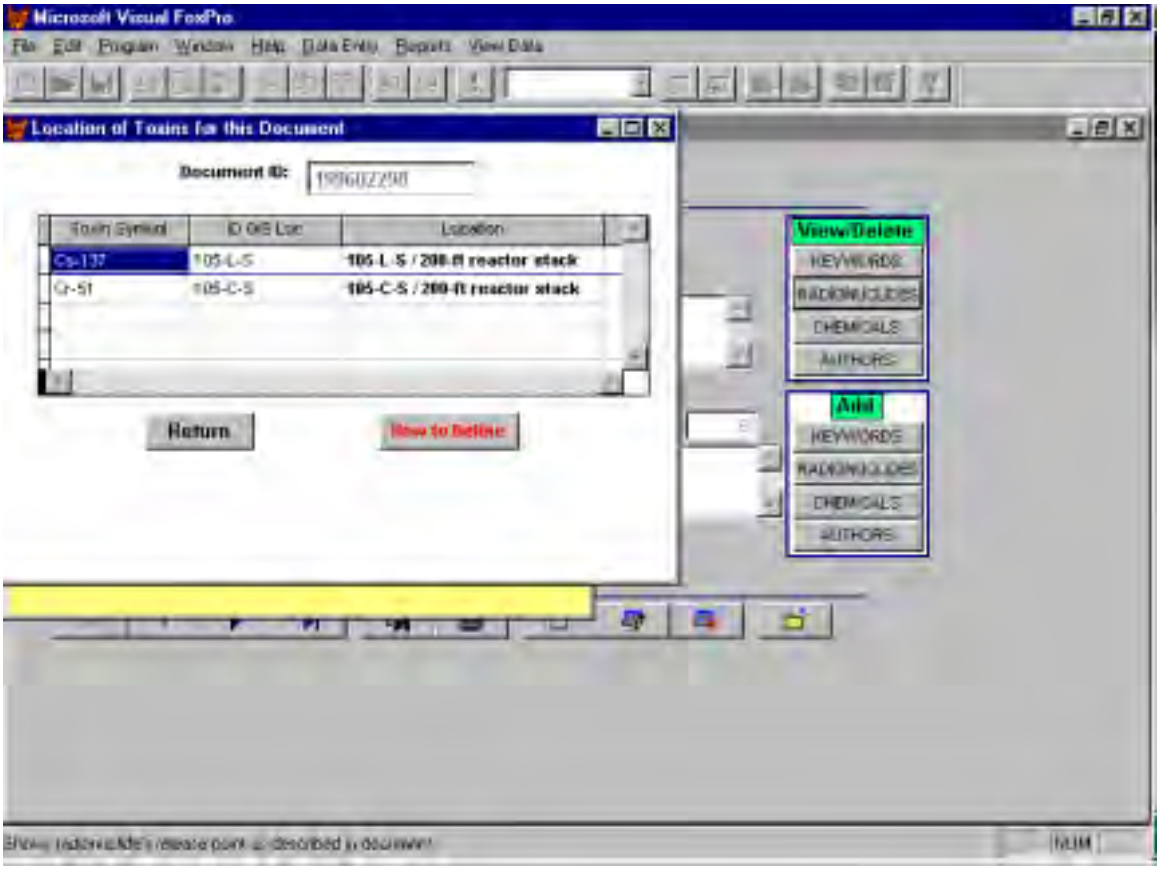
Print Report – Choosing the print report button will give you a list of existing reports that you can use to print. Existing reports can be modified to suit your application. You can also create new reports for printing.



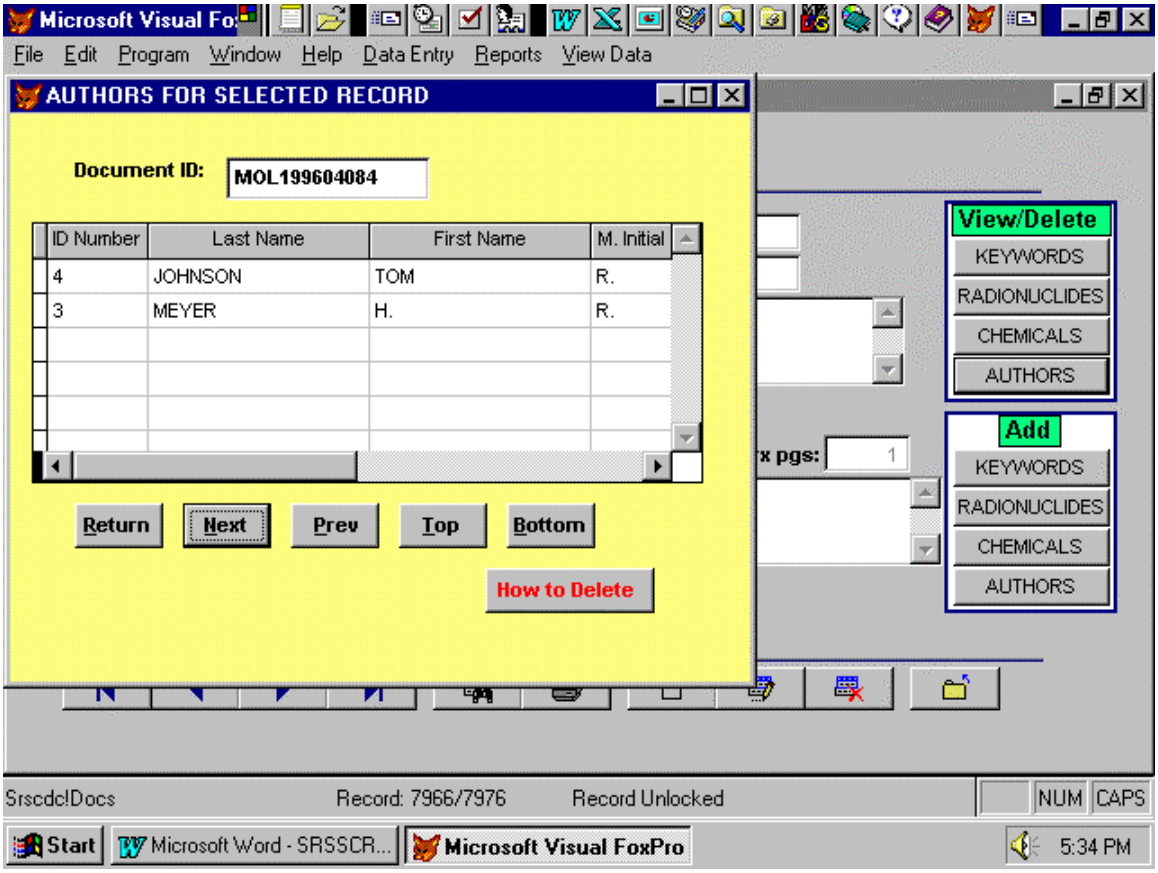
Viewing Keywords, Radionuclides, Chemicals, and Authors - Pressing the buttons in the View/Delete box (above) will bring up a list of the Keywords, Radionuclides, Chemicals, or Authors associated with the selected document.



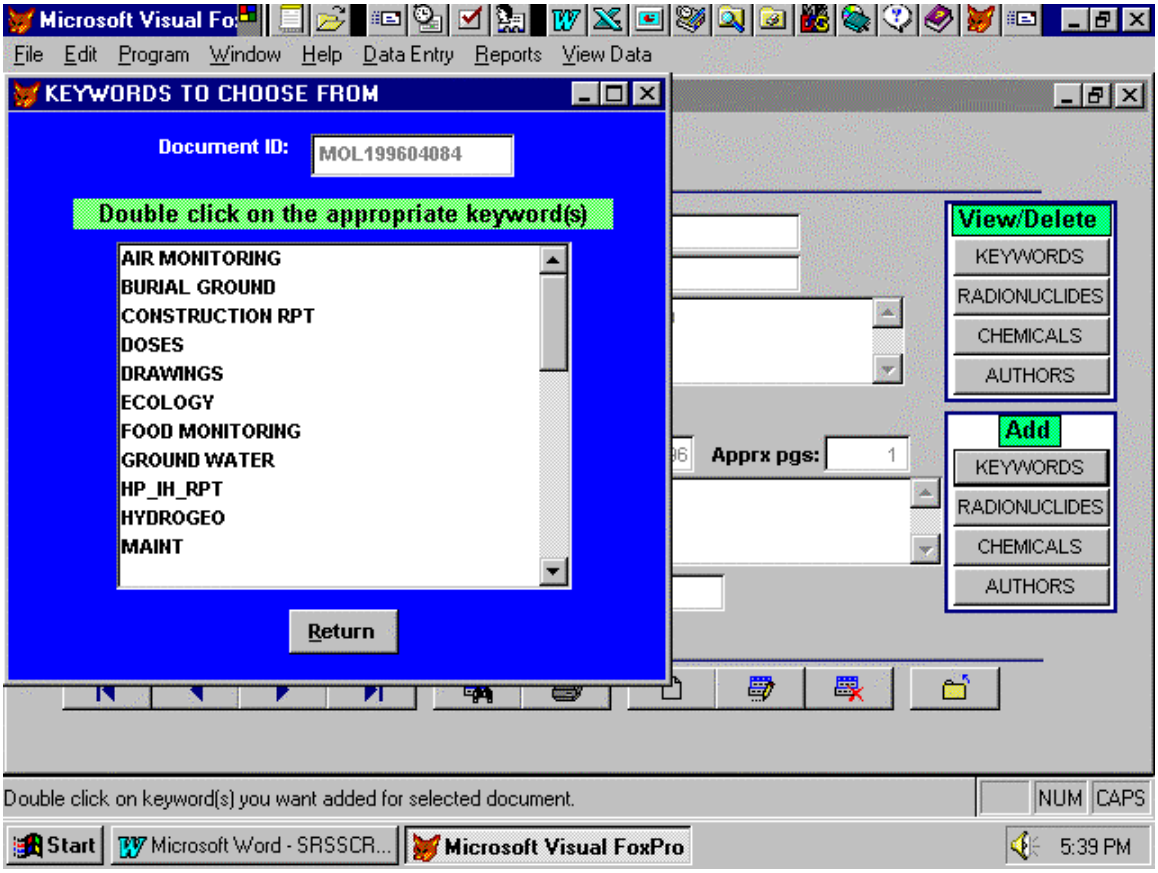
Toxin Locations - While Viewing the Radionuclides or Chemicals you can press the Toxin Locations button to bring up a screen that shows the GIS ID Location as well as the SRS location where the toxin was found. This information is not available for all documents.



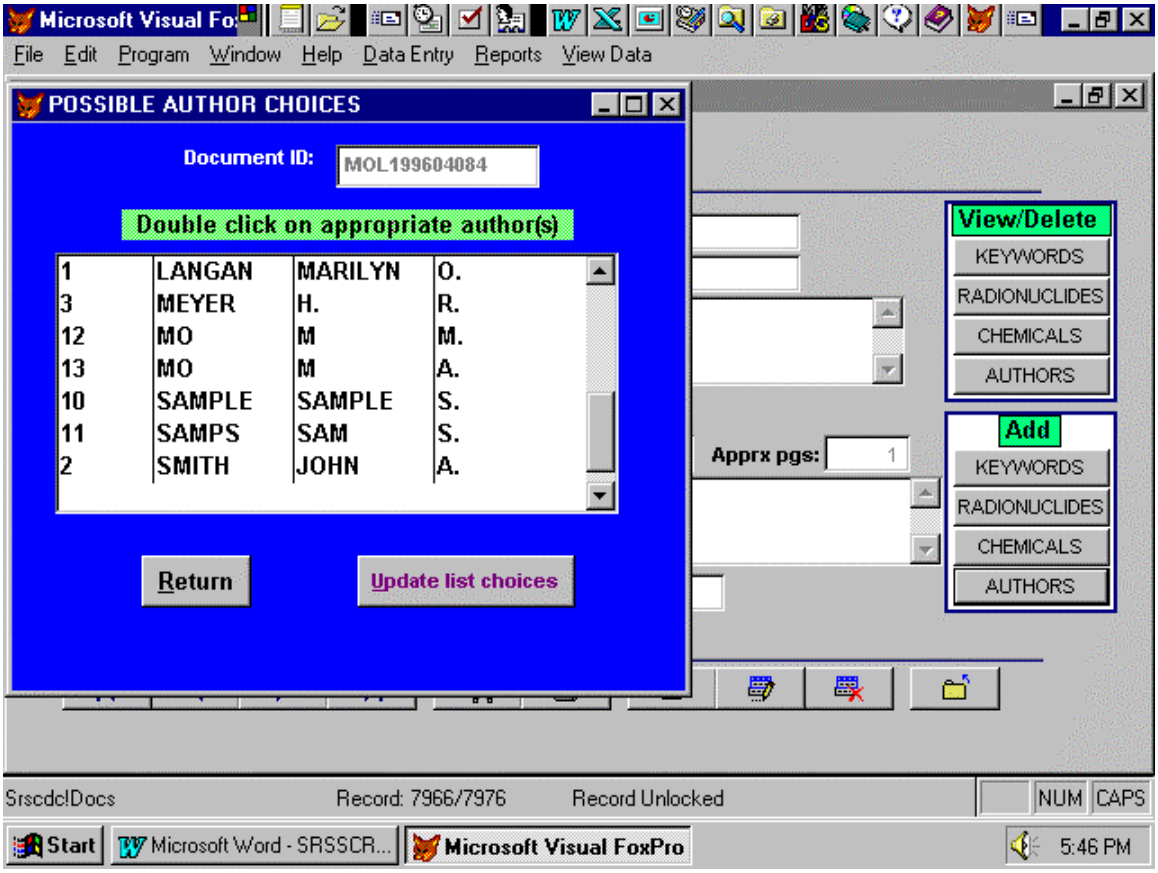
Authors - This screen comes up when the Authors button is pressed in the View/Delete Box. This shows the author(s) for the selected document. (The example is a sample document.) Authors for Phase 1 documents can be entered at a later date.



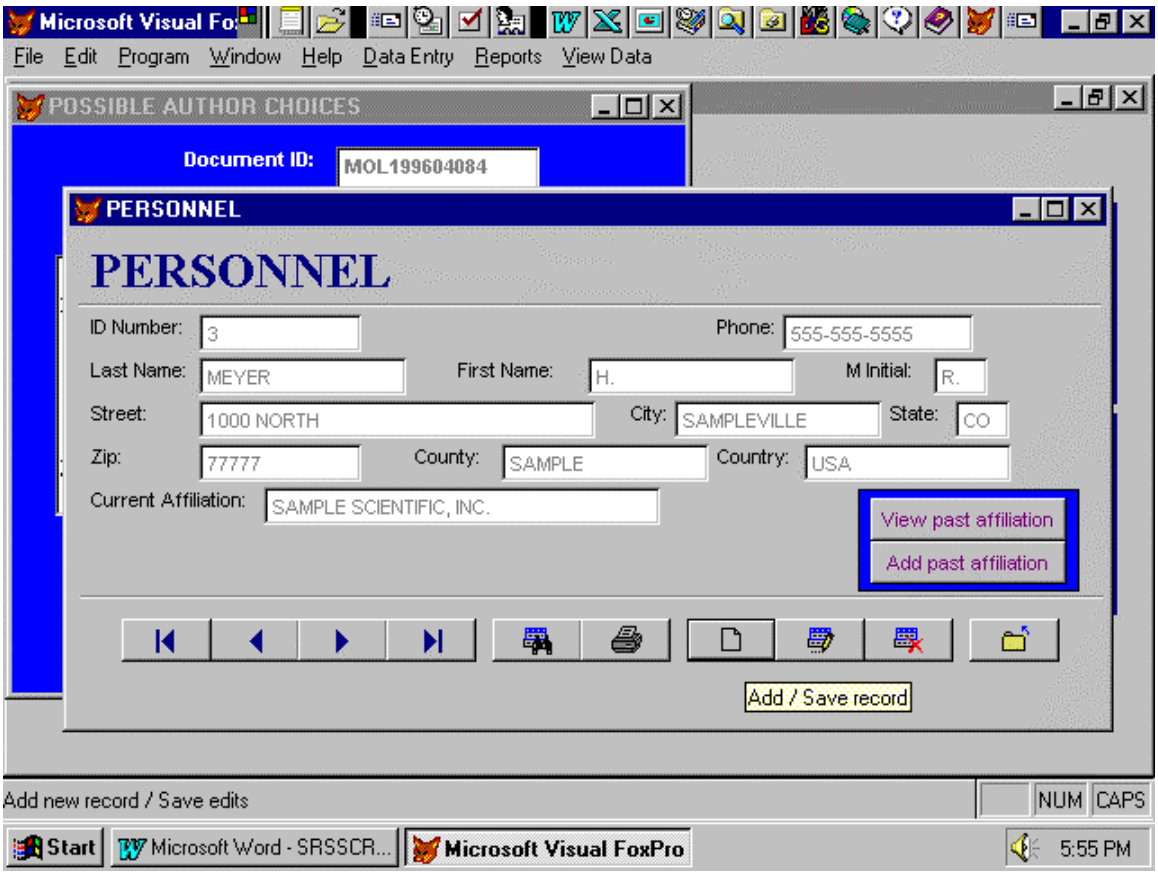
Adding Keywords, Radionuclides, Chemicals, & Authors - When a document needs to be updated to add Keywords, Radionuclides, Chemicals, or Authors the user chooses the appropriate button in the Add box. A screen similar to the one below will appear. Keywords, Radionuclides, and Chemicals show preset choices for the user to choose from. Simply **double click** on the item to be added. A message will appear in the upper right corner of your screen indicating that your request has been added.



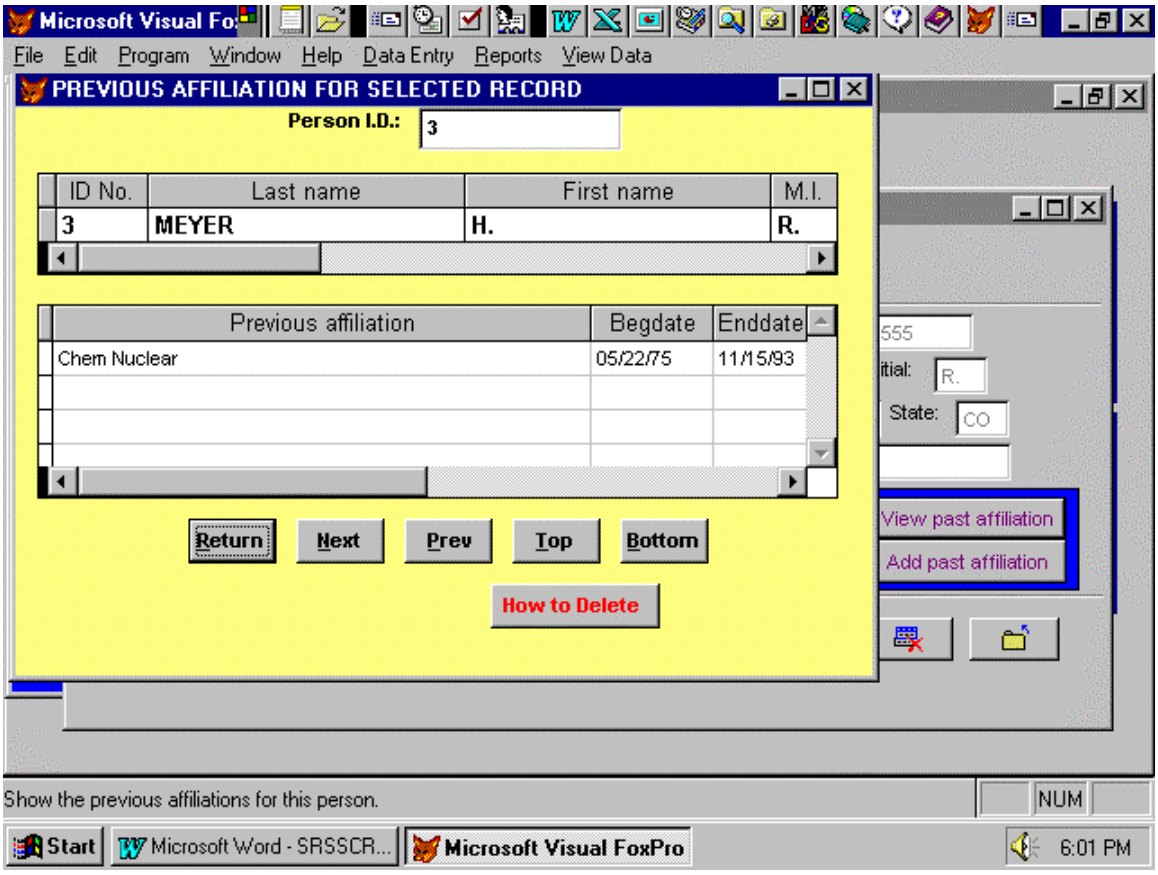
Personnel List Update - Due to the dynamic nature of the Authors list a button has been included to allow the addition of Authors not already on the existing list. If you need to add an author not already on the list, press the Update list choices button shown below.



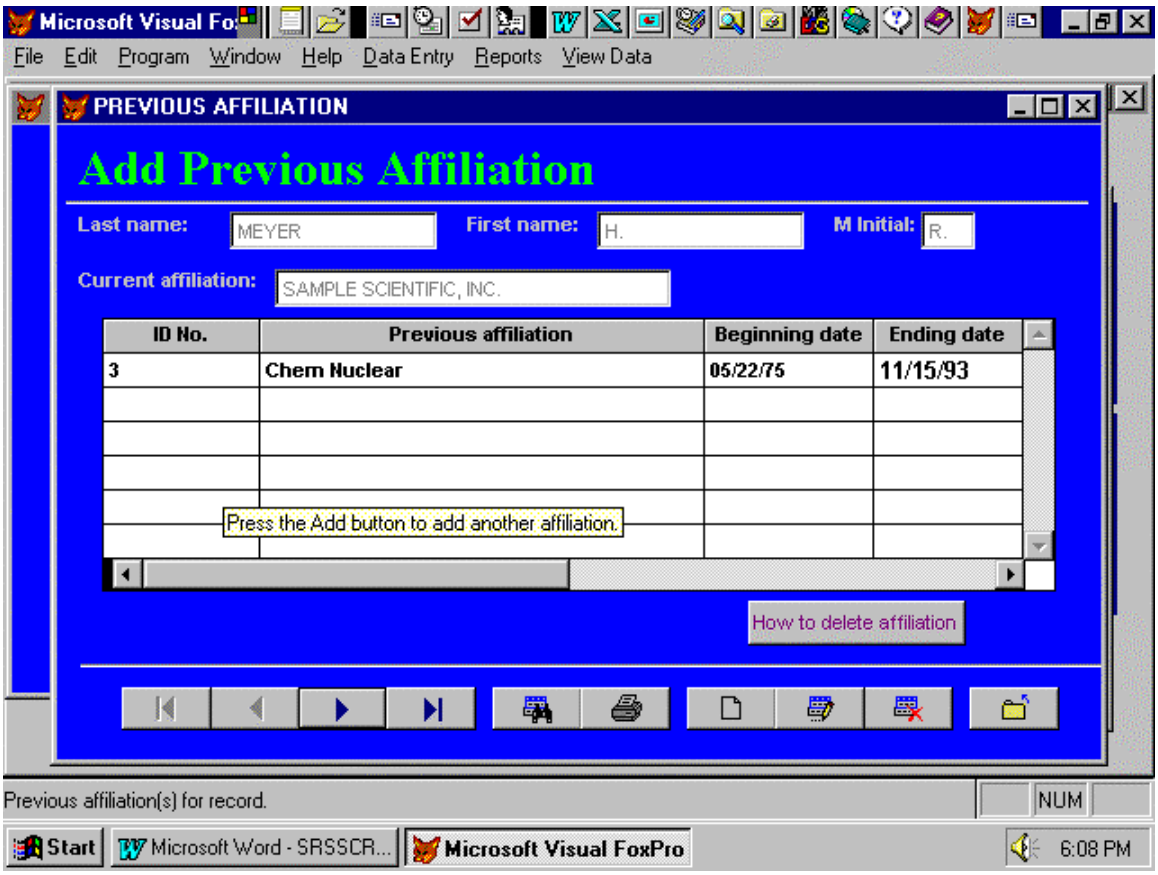
Personnel Screen - This is the Personnel input screen. It appears when the Update List Choices button is pressed as shown in the figure above. It can also be accessed from the Main Menu under Data Entry/Personnel.



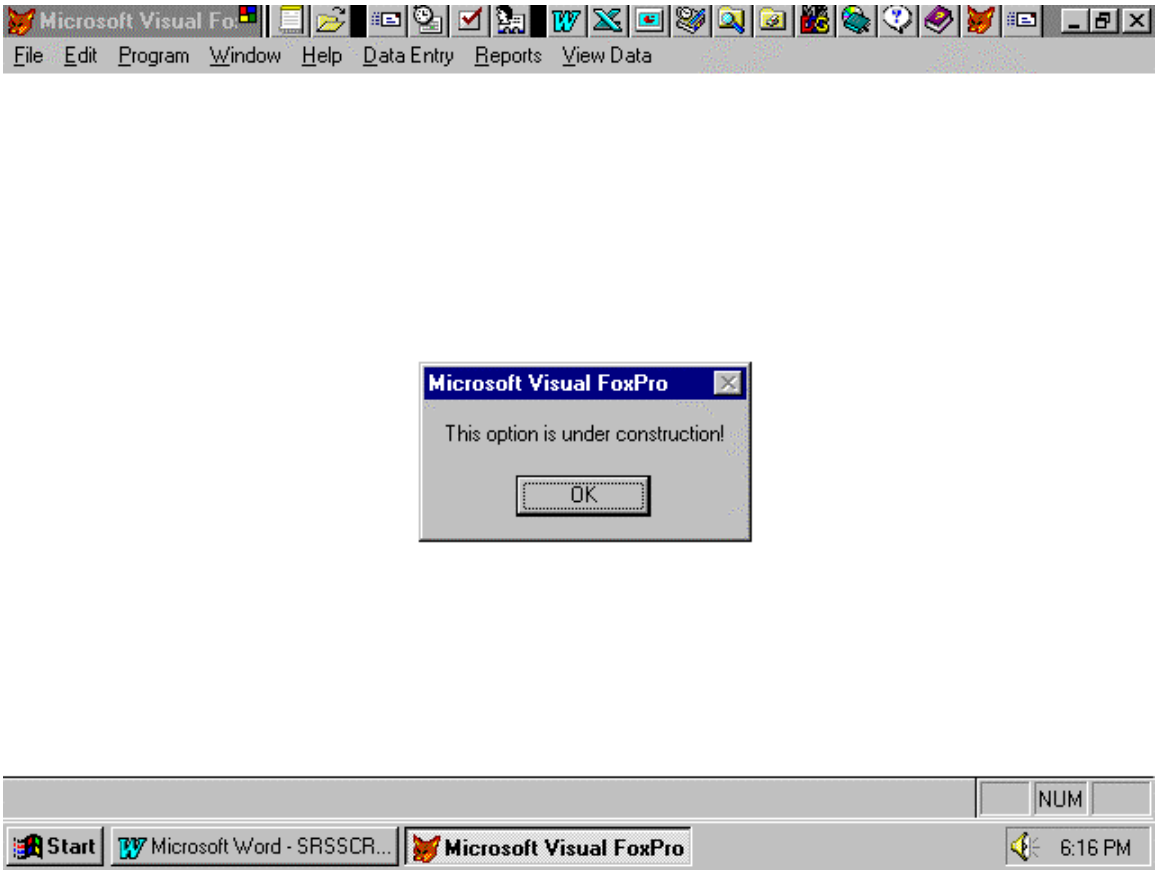
Affiliation Screen - The person's current affiliation is shown on the personnel screen. To see the previous affiliation for the selected document, press the View past affiliation button as shown in the figure above and the following screen will appear showing the organizations that the person has been associated. (The example is a sample only.)



Add Previous Affiliation Screen - When an affiliation for a person needs to be added to the list, the Add past affiliation button is pressed. The following screen appears to accommodate the addition.



Under Construction - A message is displayed for options on the Main Menu that are in developmental stages or are for future updates.

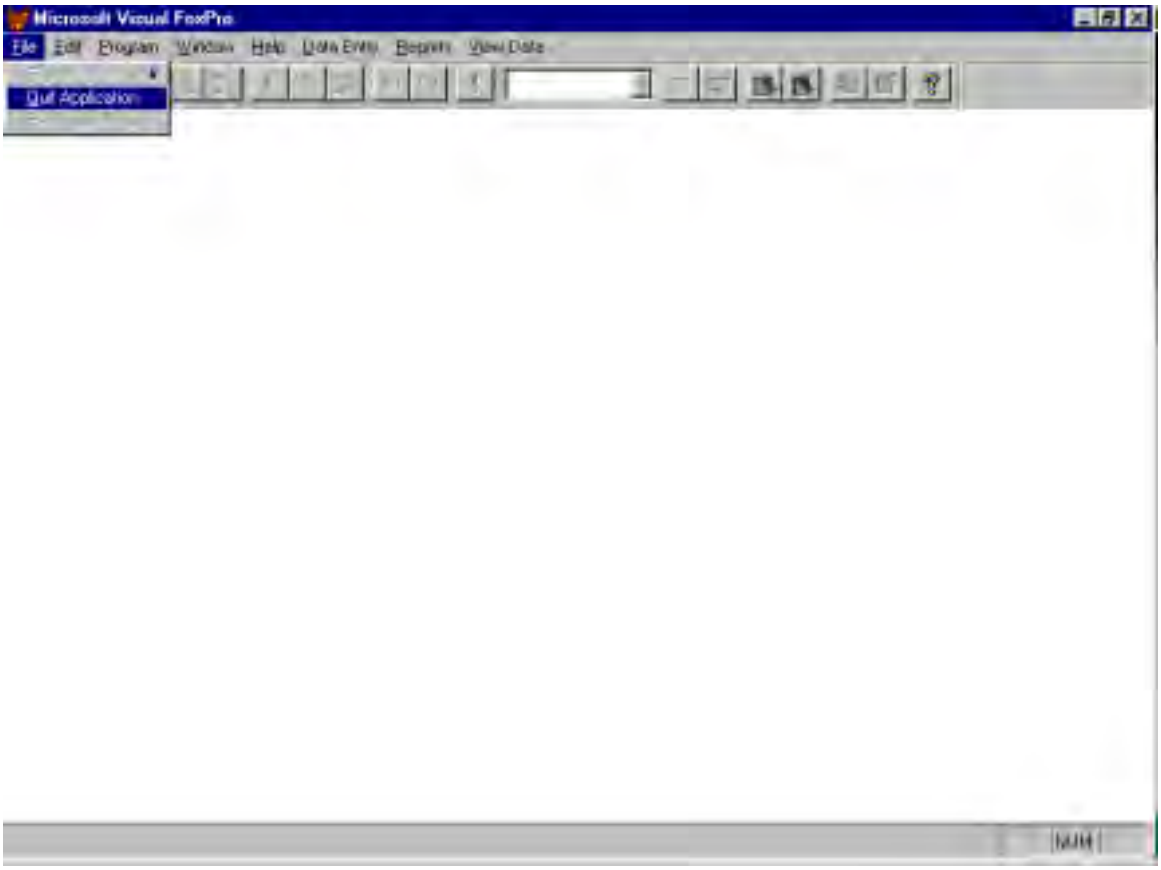


Backup

All files ending in the extensions .dbf, .fpt, and .cdx need to be backed up after making **additions**. Periodically these files need to be added to the master data file. Any changes to existing records need to be reported to the master file coordinator. These will **NOT** be made to master unless they are reported in writing. Remember, while this is not on a network you must be careful about the way changes are made.

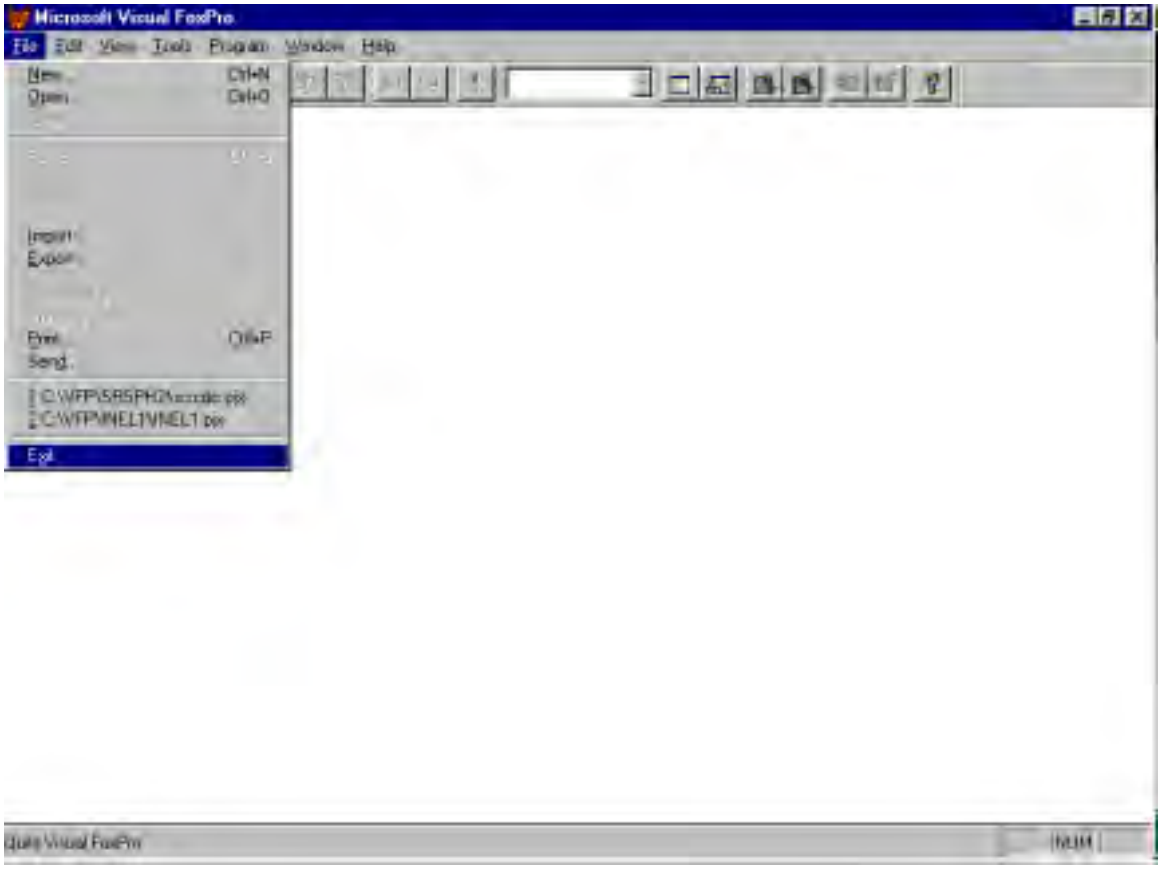
Exit Application

Exit each screen by clicking on the Return or Exit button. Once you have pressed the Exit button to exit from the Welcome screen you will return to the application's Main Menu. From the Main Menu choose File. Then choose Quit Application.



+See note on next page

+If you entered the application from within Foxpro you will now need to choose File and then choose Exit to actually exit the Foxpro program.



APPENDIX I-1

SOFTWARE REQUIREMENTS AND DESIGN SPECIFICATIONS FOR THE DOCUMENT DATABASE

APPENDIX I-1

SOFTWARE REQUIREMENTS AND DESIGN SPECIFICATIONS FOR THE DOCUMENT DATABASE

PURPOSE

This document provides the software requirements and design specifications for the Document Database as described in Task 3 of the *Radiological Assessments Corporation* Technical Proposal for Source Term Calculation and Ingestion Pathway Data Retrieval for the Savannah River Site (SRS) Environmental Dose Reconstruction Project, May 11, 1995 (RAC 1995). This document is intended for review by personnel at the Centers for Disease Control and Prevention (CDC) and software developers under subcontract with the *Radiological Assessment Corporation* (*RAC*).

GENERAL REQUIREMENTS

The Document Database was developed for the SRS Environmental Dose Reconstruction Project to track documents, which are defined as any material that provides information or data used in the project. The database tracks

- Published internal documents and reports,
- Journal articles and books,
- Unpublished reports, memos, and letters,
- Field or office notebooks and log entries,
- Laboratory analysis reports and associated quality assurance documentation,
- Databases, electronic or otherwise, containing information used in the study (e.g. monitoring data),
- Documented information and data obtained from interviews or phone conversations, and
- other documented information.

QUALITY ASSURANCE PROGRAM REQUIREMENTS

Software design, development, testing, and implementation is subject to the quality assurance (QA) requirements stated in Task 6 of the RAC Technical Proposal. These include the following:

1. A software requirements specification document,
2. A software design document,
3. A software test plan (verification and validation test plan),
4. Verification and validation test results and,
5. Software user's manual.

This document provides the software requirements and design specifications. All database requirements as specified in the contract will be me via a program of review and comparison carried out by at least two *RAC* reviewers. In addition, at least two *RAC* reviewers, and the CDC and external reviewers, will examine the database to ensure that: 1) test data, audio files and other nonessential elements are removed from the database before delivery to CDC; 2) all citations in

RAC reports are correctly listed in the database; 3) cross references between *RAC* reports and source documents are correct; and 4) source document authors are listed correctly.

HOST SYSTEM REQUIREMENTS

The document database software will use the Foxpro© relational database language and will require a microcomputer operating under MS DOS©, Version 6.0 or higher, Windows© Version 3.1 or higher, and at least 8MB of extended random access memory, although 16MB is preferable. About 200MB of free hard disk space will be required to load and operate the software.

GENERAL USER INTERFACE REQUIREMENTS

The software will provide a user interface to allow a trained nondeveloper to add, subtract, edit, or query fields in the database. The user interface will be written in the Visual Foxpro programming language. The user will be presented with a typical Windows based interface menu. Under each main menu item, the user will be able to select options such as data entry and report generation. The current menu options are shown in Figure I1-1.

Typically, opening, closing, saving, and exiting a Windows application is accomplished through the file menu option. However, this software does not include open, close, and save menu options because the database files will automatically be opened or saved when opening or exiting the application. The *Edit* menu contains the typical Windows options such as *Cut*, *Copy*, *Paste*, and *Select*.

The core of the application is the *Data Entry* menu (Figure I1-1). The user may search for records and view their contents, add records, delete records, and print selected records. Records can be retrieved, added, deleted, edited, or printed. There are four areas of data entry:

1. Those that refer to SRS Environmental Dose Reconstruction discovery documents (*Documents*),
2. Those that contain information of persons involved in the dose reconstruction (*Personnel*),
3. Those that describe documents created for Phase II of the SRS Environmental Dose Reconstruction Project (*Project Reports*), and
4. Those that describe interviews that have been conducted (*Interviews*).

Printed reports can be obtained from the *Reports* menu. Reports may be printed for all the categories listed under the *Data Entry* menu. The *Utilities* menu offers options to export data to other databases via the DBS format, or export the data for use in a Geographical Information System (GIS). The *Utilities* menu also allows access to tables so the lookup table can be updated throughout the life of the project.

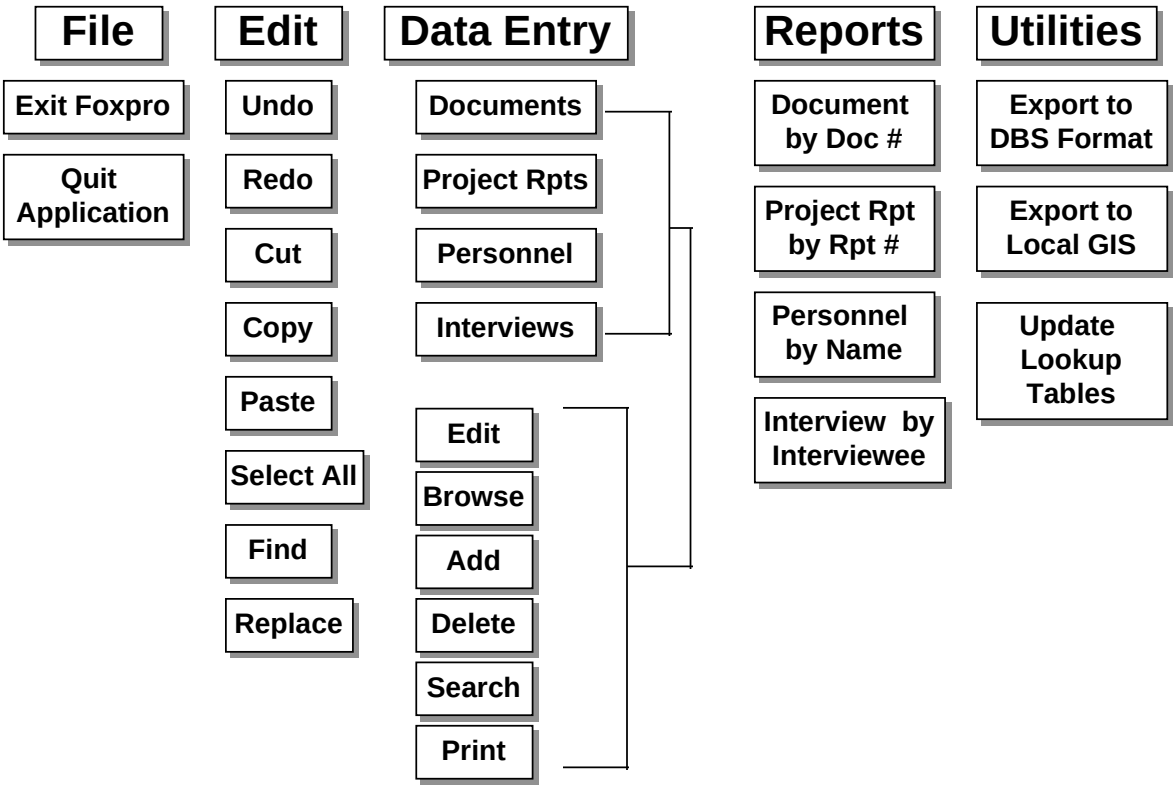


Figure I1-1. User interface menus for the Document Database. The *Documents* submenu under the *Data Entry* menu refers to documents found during document searches. *Project Reports* refers to reports generated during Phase II of the SRS Dose Reconstruction Project. The GIS designation under the *Utilities* menu refers to a Geographical Information System.

DESIGN SPECIFICATIONS

The software will use the Visual Foxpro relational database software and programming language. Foxpro databases (designated with the .DBC extension) are composed of tables that are designated with a .DBF file extension. Tables will be created for each *data entity*. Each entity may contain numerous data *fields*. A field contains the information that is either entered or retrieved by the user, or defined based on a logical expression. A field is designated as a character field, numeric field, date field, logical field, or memo field. Character, date, memo, and numeric fields have lengths defined by the developer. That is, the field has a limited number of characters that may be entered in the field by the end-user. Logical fields have a predefined length of 1. Tables are usually linked to other tables by fields of the same name and data type and length. In this way, the information presented to the end-user may originate from several tables that are related by fields of the same type and length.

The user interface will access the tables to enter data, edit existing data, query existing data, and generate reports. The program will be compiled into an executable file for distribution to end-users.

Entity Relationship Specifications

Data entities defined in the Data Dictionary (see Table 1 at the end of this document) are related to one another by the Foxpro generated data entity relationship diagram as illustrated in Figure I1-2 and I1-3.

Referential Integrity

The referential integrity of the database defines how changes made to one table are carried through to all other related tables. Tables are usually defined as parent or child. The parent table is usually where records are added, deleted, or edited. Changes made to the parent table are passed to the child table according to the rules defined in the Foxpro referential integrity builder (Figures I1-3, I1-4, and I1-5). The columns labeled Parent Tag and Child Tag fields on the Foxpro referential integrity builder are the parent and child tables that are related. Note that in most cases, the parent and child tag have the same name. The parent and child tags do not have to share the same name but the tags must be of the same data type and length. The Update, Delete, and Insert columns refer to the actions a user may take when editing a record. Within these columns, the integrity rules, *Cascade*, *Ignore*, and *Restrict* define the action taken when changes made to the parent table are passed to the child table. If *Cascade* is selected, then all changes to the parent table are passed to the child table. If *Ignore* is chosen, then changes made to the parent table are ignored in the child table. If *Restrict* is chosen, then changes to the parent table are restricted if there are related records in the child table.

Data Dictionary

The Data Dictionary (Table 1) describes each data entity and the fields that make up that entity. Each field is described by its type (e.g. character, or numeric), number of characters in the field (field width or length), format of data, and description of information the field is to contain.

Interface with Previous Database Specifications

The original database developed in Phase I of the SRS Environmental Dose Reconstruction Project will be incorporated into the new database.

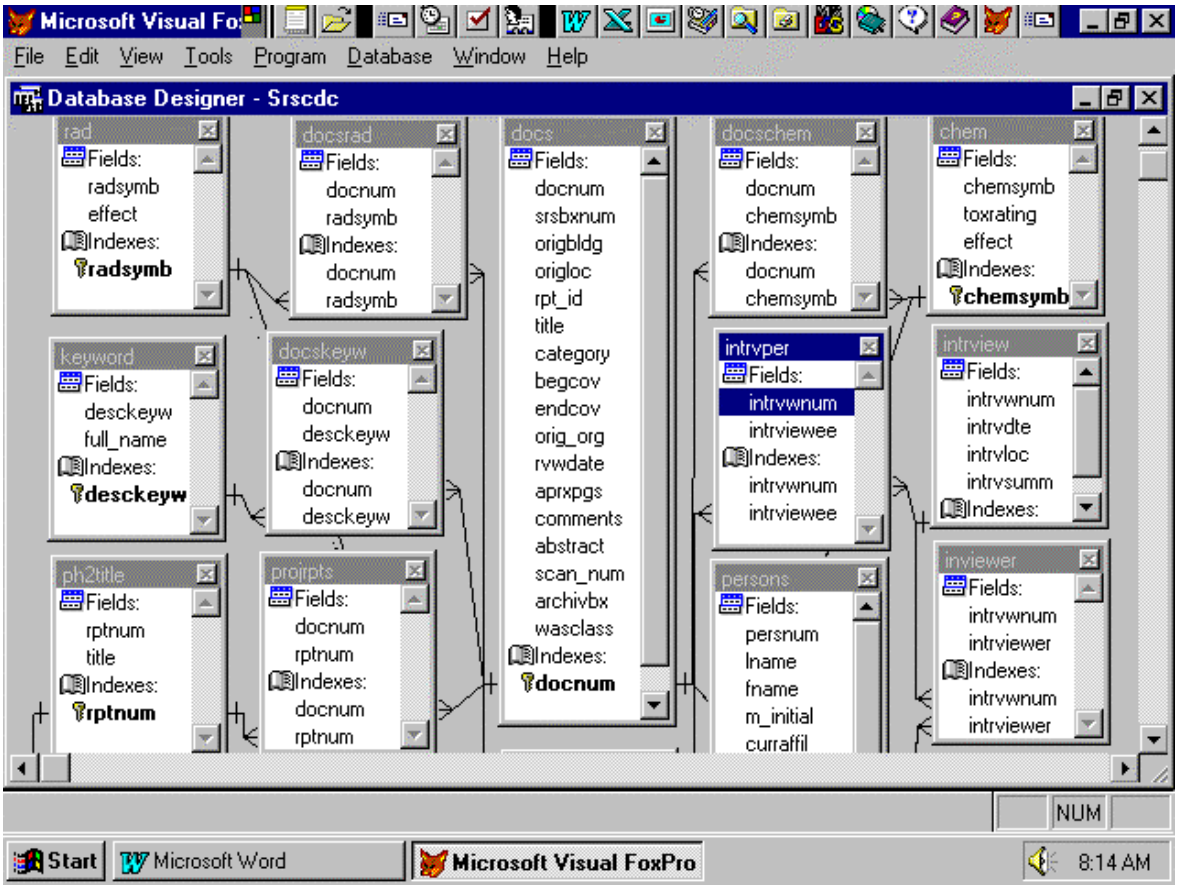


Figure I1-2. Data entity relationship diagram generated by Foxpro. The shaded area represents individual data tables. Within each table are the fields that comprise the table, and indexes used to sort and retrieve field entries.

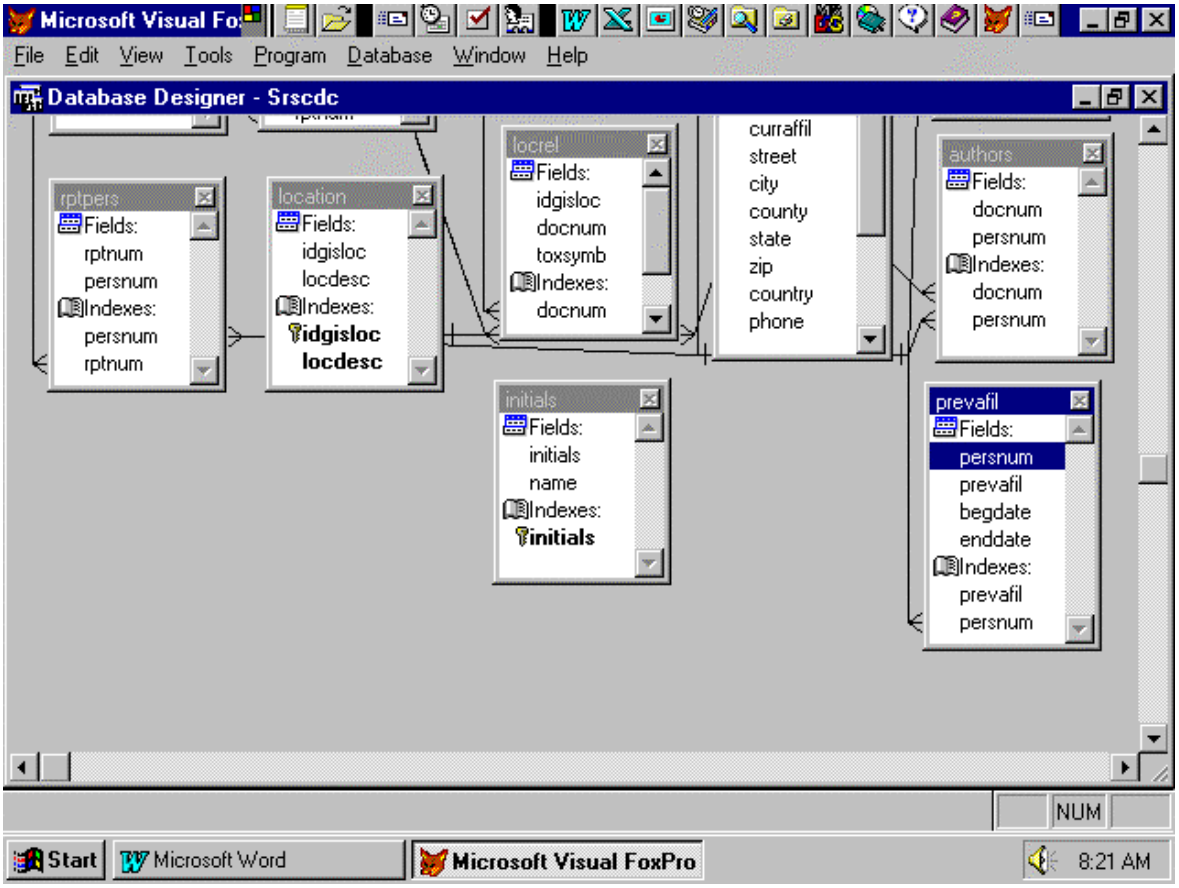


Figure I1-3. Data entity relationship diagram generated by Foxpro. The shaded area represents individual data tables. Within each table are the fields that comprise the table, and indexes used to sort and retrieve field entries.

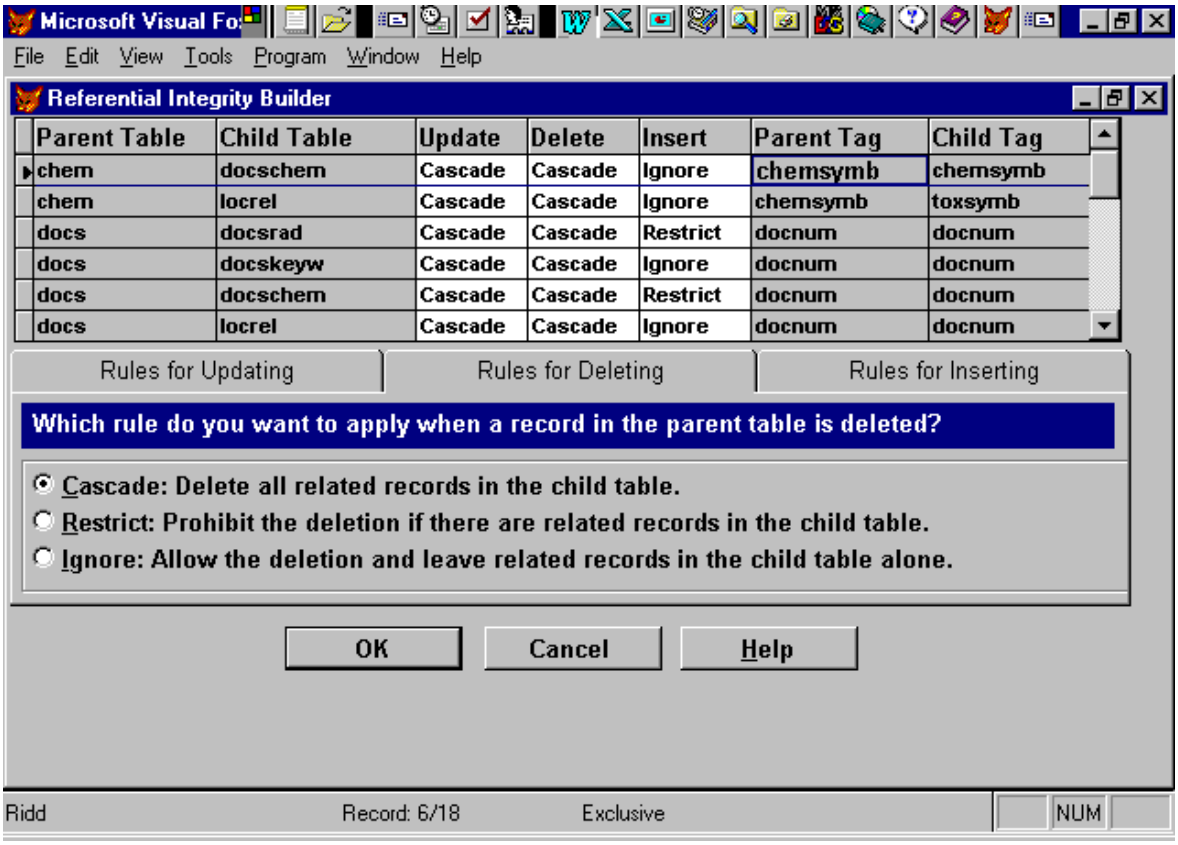


Figure I1-4. Referential integrity for the *chem* and *docs* tables.

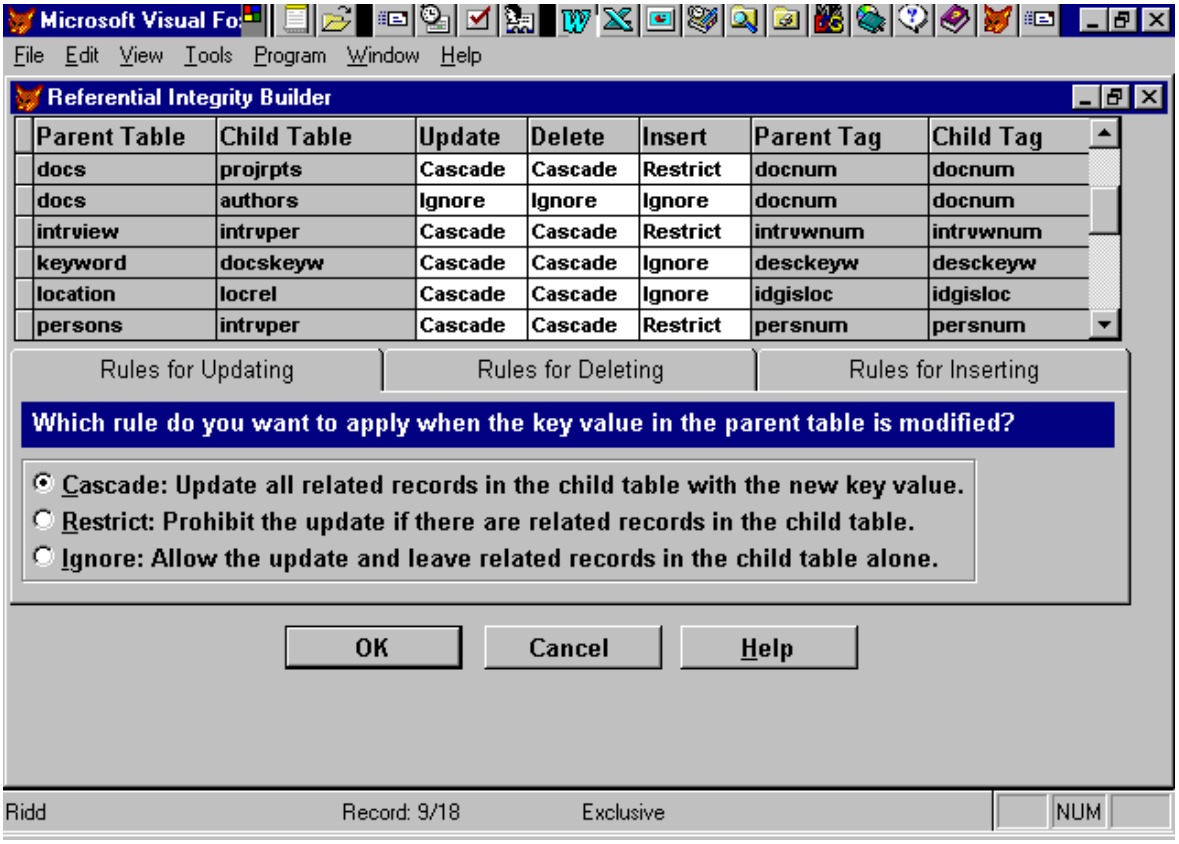


Figure I1-5. Referential integrity for the *docs*, *intrview*, *keyword*, *location*, and *persons* tables.

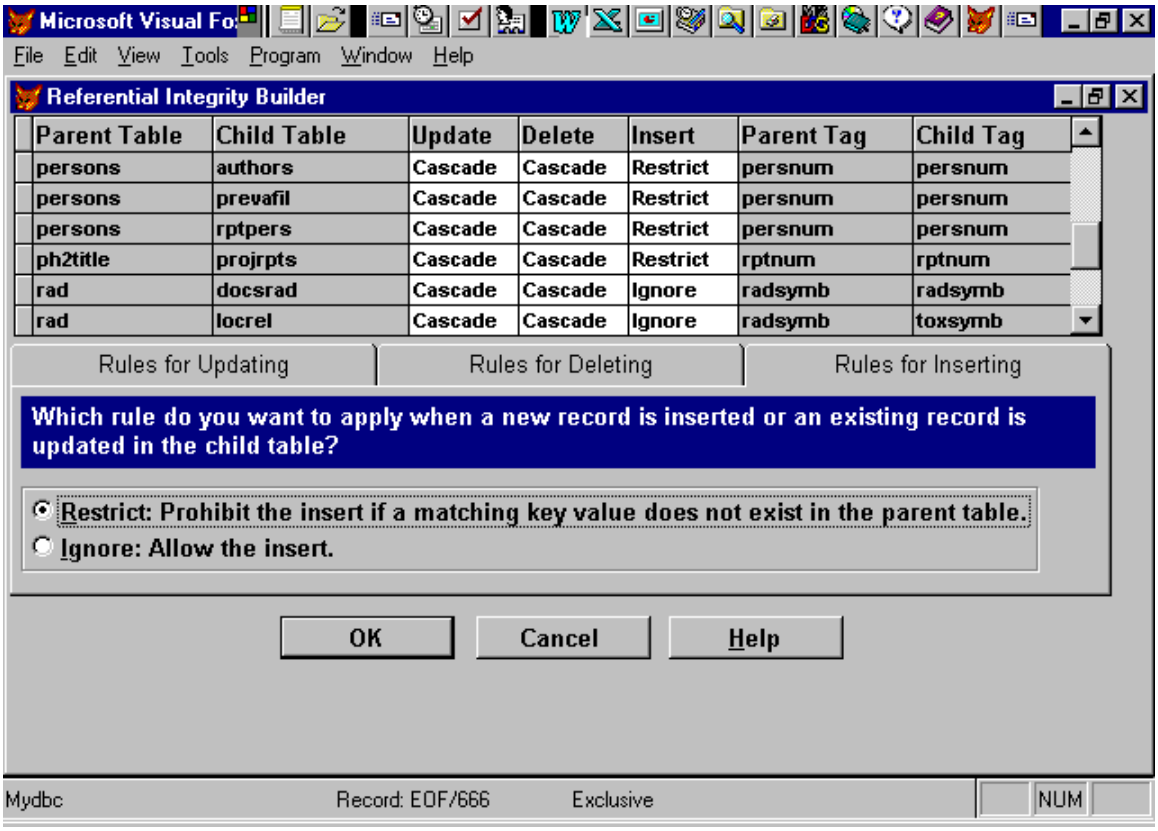


Figure I1-6. Referential integrity for the *persons*, *ph2title*, and *rad* tables.

Table I1-1. SRS Phase II Document Database Data Dictionary

Table or Field Name	Field Name[a]	Field Width	Field Description
DOCS.DBF			
DOCNUM	C	16	A unique string automatically assigned by the database to each document entered. The number is automatically created from: the reviewer's initials; the PC's current date value; and an auto-incremented serial number for each reviewer, reset to zero each day. The format of DOCNUM is: Reviewer initials, year, month, day, and the serial number, e.g. HRM 1993 7 25 1; HRM 1993 7 25 2, etc.
SRSBXNUM	C	20	Boxes stored in the repositories at SRS have been assigned box numbers by SRS records personnel, recorded in this field. Not all records in the database will have a box number, because some were obtained from places other than the official SRS repositories, including file cabinets in staff offices. SRSBXNUM is not useful as a separate table, since SRSBXNUMs are not unique identifiers. Linking documents to non-unique box numbers via a table might create confusion during attempted retrieval of original documents. Also includes identifying characters assigned to records discovered in boxes, file cabinets and other locations offsite.
ORIGLOC	C	20	Original location of SRS document box. The location of an SRS box is subject to repeated change over time. SRS maintains a computerized tracking system capable of locating its records boxes upon request. In the event that Category 0 or 1 records must be relocated during Phase II and later work, the SRS tracking system will be utilized. Category 2 or 3 documents are copied and retained external to the SRS system, in *RAC* archive boxes.
ORIGBLDG	C	15	SRS building in which records box was originally stored.
RPT_ID	C	20	May include document's memo number, report number, publication (journal), Library of Congress ID, or other identification, if applicable.
TITLE	C	254	The official document title. If untitled, researcher creates a title based on document content. A created title is placed in parentheses.
CATEGORY	N	1	Category **2** and **3** documents are likely to be of significant value to the study. Both Category 2 and 3 documents are copied to CDC's contractor and the Aiken reading room. Category **3** documents have been determined by the discovering researcher to be of sufficient potential value to the dose reconstruction that they should be abstracted into the database. Category **1** records are of potential value, but are likely to be available in more original form elsewhere. Category **0** includes documents of no value to dose reconstruction.

Table or Field Name	Field Name[a]	Field Width	Field Description
			Reference documents used during the Phase II research may be copyright-protected and not copied to the Project's archive boxes. They are defined in this database as Category 1 records. CATEGORY is a required entry field.
BEGCOV	D	8	The document's coverage beginning date, identified during inspection of the document.
ENDCOV	D	8	The document's coverage ending date, identified during inspection of the document.
ORIG_ORG	C	15	Organization that produced the document
RVWDATE	D	8	The date that the Dose Reconstruction Project researcher first encountered the document and initiated its entry into the database. Not related to the original date of the document creation, or to the date abstracted by an assistant, or to later reviews by other researchers.
APRX_PGS	N	5	Approximate page count of the document. The default value for this field is 1. Large documents were often assigned the value 1000, to indicate significant copying effort, during Phase I research.
COMMENTS	M	10	Contains previous contents of Phase I SRSDOSE Comments memo fields; contains comments from Phase II research which don't fit logically elsewhere in the database.
ABSTRACT	M	10	Contains abstract if document is Category 3
SCAN_NUM	N	12	If some documents are eventually scanned and made available in electronic format, this will serve as a link to the scanned data.
ARCHIVBX	C	15	All copied documents are in Project possession offsite. ARCHIVBX indicates the specific *RAC* box in which the copied Category 2 or 3 document is stored. This ID also points to the specific microfilm cassette containing the document.
WASCLASS	L	1	Indicates whether the document was originally classified (Y) or not (N). All documents used during dose assessment will have been declassified. A document's original classification level (CONFIDENTIAL, SECRET, or TOP SECRET), or special caveats (NOFORN, RESTRICTED DATA, FORMERLY RESTRICTED DATA, NAVAL NUCLEAR PROPULSION INFORMATION, etc.) is not generally noted in the database. Default is N.
INITIALS	C	3	Initials of document reviewer

PROJRPTS.DBF			(SRS Dose Reconstruction Project Reports)
DOCNUM	C	14	(See DOCS.DBF)

Table or Field Name	Field Name[a]	Field Width	Field Description
RPTNUM	C	15	Number assigned to chapter or appendix
PH2TITLE.DBF			
RPTNUM	C	15	(See PROJRPTS.DBF)
TITLE	C	254	Title of chapter or appendix
RPTPERS.DBF			
RPTNUM	C	15	(See PROJRPTS.DBF)
PERSNUM	C	10	(See PERSONS.DBF) As used in PROJRPTS.DBF, these are the authors of Phase II project reports, only.
INTRVPER.DBF			(Persons interviewed or doing interview)
PERSNUM	C	10	(See PERSONS.DBF)
INTRVWNUM	C	10	(See INTRVW.DBF
INTRVIEWEE	L	1	Yes or no (is the person an interviewee?)
INTRVIEWER	L	1	No or yes (is the person an interviewer?)
INTRVW.DBF			
INTRVWNUM	C	10	Unique ID number
INTRVDTE	D	8	Date interview held.
INTRVLOC	C	30	Location of interview
INTRVSUMM	M	10	Summary of interview.
DOCSKEYW.DBF			
DOCNUM	C	16	(See DOCS.DBF)
DESCKEYW	C	20	(See KEYWORD.DBF)
KEYWORD.DBF			
DESCKEYW	C	20	Word or phrase to quickly characterize document content
FULL_NAME	C	40	Definition of descriptive keyword
DOCSCHEM.DBF			
DOCNUM	C	16	(See DOCS.DBF)
CHEMSYMB	C	30	Unique symbol identifying toxic agent
CHEM.DBF			
CHEMSYMB	C	30	Unique chemical ID (e.g. CCl4)
TOXRATING	C	50	Toxin's hazard rating
EFFECT	M	10	Summary of potential effects of exposure to toxin
AUTHORS.DBF			
DOCNUM	C	16	(See DOCS.DBF)

Table or Field Name	Field Name[a]	Field Width	Field Description
PERSNUM	C	10	(See PERSONS.DBF)
PERSONS.DBF			
PERSNUM	C	10	Unique ID
LNAME	C	15	Last name
FNAME	C	15	First name
M_INITIAL	C	2	Middle initial
CURRAFFIL	C	30	Current affiliation
STREET	C	30	Mailing address
CITY	C	15	
COUNTY	C	15	
STATE	C	2	
ZIP	C	10	
COUNTRY	C	15	Defaults to "U.S.A."
PHONE	C	12	
PREVAFIL.DBF			
PERSNUM	C	10	Unique ID
PREVAFIL	C	30	Previous affiliation of individual
BEGDATE	D	8	Date previous affiliation began
ENDDATE	D	8	Date previous affiliation ended
DOCSRAD.DBF			
DOCNUM	C	16	(See DOCS.DBF)
RADSYMB	C	30	(See RAD.DBF)
RAD.DBF			
RADSYMB	C	30	Unique ID character set for radionuclide (e.g., Pu-238)
EFFECT	M	10	Discussion of radionuclide's principal characteristics
LOCREL.DBF			(Main locations for toxic releases: buildings., facilities, etc.)[b]
LOCDESC	C	30	Unique reference number
DOCNUM	C	16	(See DOCS.DBF)
TOXSYMB	C	30	Radioactive material or toxic chemical symbol
LOCATION.DBF			
LOCDESC	C	30	Unique reference number
LOCDESCR	C	30	Brief descriptions of possible release locations. Used as a lookup table.

Table or Field Name	Field Name[a]	Field Width	Field Description

[a] Field Type: C = character, N = numeric, D = date, M = memo, L = logical. Mandatory data entry field if the character in this column is boldfaced.

[b] Detailed location data (e.g., sample and, sampler latitude/longitude) are maintained in the Phase II Geographic Information System database.

REFERENCE

Radiological Assessments Corporation (*RAC*), 1995. *Technical Proposal: Source Term Calculations and Ingestion Pathway Data Retrieval for the Savannah River Site (SRS) Environmental Dose Reconstruction Project*., May 11, 1995.

APPENDIX I-2

SOFTWARE TEST PLAN FOR THE DOCUMENT DATABASE

APPENDIX I-2

SOFTWARE TEST PLAN FOR THE DOCUMENT DATABASE

PURPOSE AND BACKGROUND

This document provides the testing plan to verify and validate the Document Database software as described in Task 3 of the *Radiological Assessments Corporation* Technical Proposal for Source Term Calculation and Ingestion Pathway Data Retrieval for the Savannah River Site (SRS) Environmental Dose Reconstruction Project, May 11, 1995 (RAC 1995). Verification is defined as the process of determining whether or not the products of a given phase of the software development cycle fulfill the requirements established during the previous phase. Validation is defined as the process of evaluating the software at the end of the software development process to ensure compliance with the software requirements.In this project, individual phases for developing of the Document Database were not clearly delineated. Therefore, this test plan covers verifying of each of the code modules and validating that the code as a whole fulfills the requirements stated in the software requirements and design specifications document. This document is intended for review by personnel at the Centers for Disease Control and Prevention (CDC) and software developers under subcontract with the *Radiological Assessment Corporation*.

The Document Database was developed for the SRS Environmental Dose Reconstruction Project to track documents, which are defined as any material that provides information or data used in the project. The database tracks

- Published internal documents and reports,
- Journal articles and books,
- Unpublished reports, memos, and letters,
- Field or office notebooks and log entries,
- Laboratory analysis reports and quality assurance documentation,
- Databases, electronic or otherwise, containing information used in the study (e.g. monitoring data,
- Documented information and data obtained from interviews or phone conversations, and
- other documented information.

The software will use the Foxpro© relational database software and programming language. Foxpro databases, or tables (designated with a .DBF file extension), will be created for each *data entity*. Each entity may contain numerous data *fields*. A field contains the information that is either entered or retrieved by the user. A field is designated as a character, numeric, date, logical, or memo field. Character, date, memo, and numeric fields have lengths defined by the developer. That is, the field has a limited number of characters that may be entered in the field by the end user. Logical fields have a predefined width of 1. Tables are usually linked to other tables by fields of the same name and data type. In this way, the information presented to the end user may originate from several tables that are related by fields of the same name and type.

The Foxpro programming language will be used to create a user interface to enter or query the database. The program will be compiled into an executable file for distribution to end users.

OBJECTIVES

This test plan for the Document Database will accomplish the following objectives:

- Detail the activities required to prepare for and conduct the software test,
- Communicate to all responsible parties the tasks that they are to perform and the schedule to be followed when performing the tasks,
- Define the sources of information used to prepare the plan, and
- Define the operating environment needed to conduct the test.

SCOPE

This test plan covers the testing of the Document Database software by independent reviewers. The developers may also use the procedures defined in this document for internal testing before release. The testing will cover referential integrity of the tables and the application software designed to manage and control input, output, and editing of the tables.

TEST ITEMS

The testing of a software product should include four components. These are

1. Functional coverage,
2. Topological coverage,
3. Component interface,
4. Performance measures.

Functional coverage refers to how many of the code processes and data (if applicable) are exercised during the testing. An example of a process is the editing and retrieval features built into the Document Database. Data refers to the pieces of information stored, retrieved, and manipulated by the software.

Topological coverage refers to the logic path that the code takes to determines if a particular function, task, or feature is executed. For example, the logic path the code may take to allow record retrieval may involve first checking the user identification and then checking if the record exists.

Component interface refers to how well each of the code components or modules work together as a whole. Each module or component may work fine independently. Testing the component interface verifies that the code logic linking each component operates correctly.

Performance measures refer to measures that quantitatively describe how well the code performed during testing.

In this test plan, we have attempted to address all four of these components by designating a test database that will include entries in all data entities and data fields, and testing all code features that have been written and implemented. Neither functional or topological coverage is 100% assured, but through the methods outlined in this test plan, we can be reasonably assured that most situations a user may encounter during normal operation of the code are tested.

The software tested will be designated the Beta Test Version until successful completion of the testing procedures.

FEATURES TO BE TESTED

User interface application modules are written under the Visual Foxpro object-oriented programming language. The user is presented a series of menu items that perform specific tasks. These items have methods and properties attached to them and it is these features that will be tested, along with the relational integrity of the data tables that are linked to their operation. A main menu is presented first and includes the items typically found on most Windows -based applications. Under each main menu item is a series of submenus that exercises different options, depending on the needs of the user. The main menu items shown in Figure I2-1 will be tested. The referential integrity of each table will also be tested as stated in the software requirements and design specifications document (Rood et al. 1996).

APPROACH

A test database will be used that will include entries for all data tables and lookup tables. The test database will be relatively small; therefore, output from the tables can be easily checked against the data in the test database. The test database will be developed before beginning any testing. The hardcopy printout of the test database will be used to measure software performance.

A person designated as the test technician will perform the actual software testing. The test technician will perform tasks such as data entry, retrieval, and report generation. Results from each test will be provided to the test analyst who will determine if the test was performed correctly and if the results are satisfactory. The test analyst will provide written documentation that the test was completed satisfactorily. This documentation may take the form of a letter report. The test technician and test analyst will be independent of the development team. We suggest the CDC staff perform these duties. The developer may use the testing procedure outlined in this document to test the software before release, but is not required to provide these results to the CDC. Referential integrity diagrams, data dictionary, and database design diagrams will be provided to the test analyst before testing begins. These items are described in Rood et al. (1996). The database design diagram consists of a listing of each of the tables contained within the database application. Under each table, the fields associated with that table and indexes are listed. The referential integrity diagrams define how changes made to one table are passed to other tables.

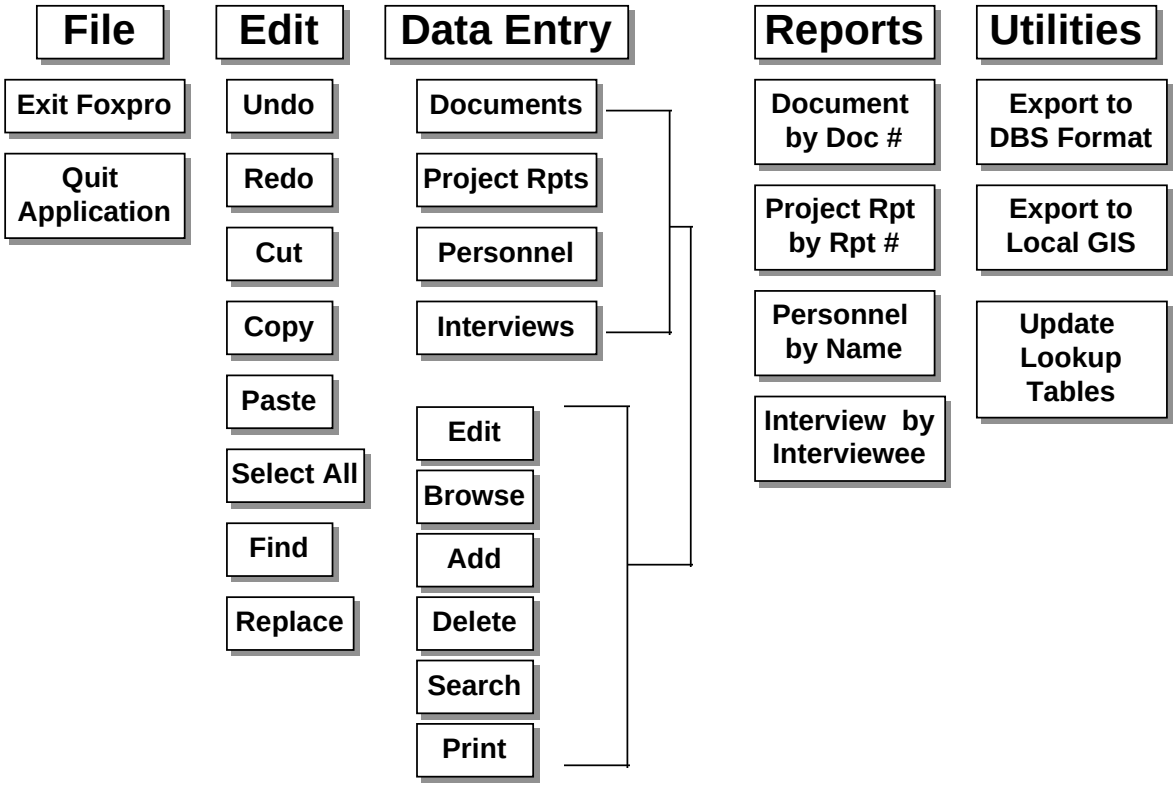


Figure I2-1. User interface menus for the Document Database. The GIS designation under the utilities menu refers to a Geographical Information System.

Testing will be performed on the Document Database software identified as the Beta Test Version 1.0. The testing procedure will be composed of three phases. In the first phase, data entry will be tested, in the second phase, document query, additions, deletions and editing will be tested, in the third phase, report generation, utilities, lookup tables, and housekeeping options will be tested. Each phase is discussed below.

Phase 1 Testing: Data Entry

Phase 1 testing will exercise the applications written to enter data from the keyboard into the various data tables. Menu items exercised in this phase will include the Add sub-menu under Data Entry in the main menu. It will also test the entity relationships between data tables. The test database will be entered manually and reviewed for accuracy by the test technician. A hardcopy printout of the database will be provided to the test analyst who will determine if the data present on the hardcopy printout is indeed in the database and that it was placed in the proper table. A query of each individual table performed in the interactive mode of Foxpro and outside the query applications shall be performed in order to verify the information presented on the hardcopy is present in the table or tables designated to store that information.

Phase 2 Testing: Document Query and Editing

Phase 2 testing will exercise the Documents, Project Reports, Personnel, and Interviews sub-menu options under Data Entry in the main menu. In each of the sub-menus, the search, delete, edit, and print options will be exercised. A subset of the test database will be selected for searches, employing the user interface to perform the searches. The subset of the test database will be selected by the test analyst. The subset will include entries from each of the data categories (Documents, Project Reports, Personnel, and Interviews). The test technician will query the database for each record in the subset and print the results. The hardcopy printout will be provided to the test analyst to verify each record was retrieved and printed properly. The subset of the entries will be selected for deletion by the test technician. The test analyst will then query the database for each record in the subset to verify that the records were deleted. These records will then be added back by the test technician.

Each record in the subset of the test database will be selected for editing. Each record will be retrieved and edited using the user interface by the test technician. The test technician will provide the test analyst a record of all editing changes made to the database. The records will then be retrieved by the test analyst and verified to make sure the changes were saved and that referential integrity of the table is intact. That is, the updates, deletions, and inserts are passed from the parent table to the child table correctly.

Phase 3 Testing: Report Generation, Utilities, Lookup Tables, and Housekeeping

Phase 3 testing will exercise the report generation options of the user interface. The test technician will generate reports for the entire test database by document number, project report number, personnel, and interviewee. The reports will be reviewed by the test analyst and visually compared with the hardcopy printout of the database to assure all records that should have been printed were printed.

The test technician will then export the entire test database to both DBS format and local Geographic Information System (GIS) format. The DBS-formatted export file will then be imported into another database application by the test technician and provided to the test analyst. The test analyst will then check each record in the other database application to assure records were exported from the Document Database correctly. The GIS-formatted export file will then be imported into a local GIS application by the test technician and provided to the test analyst. The test analyst will then check the GIS database to verify each record was exported correctly from the document database.

PERFORMANCE MEASURES

Performance measures used during testing will be pass, fail, and conditional pass. Each item described in the three test phases previously described will be assigned one of these measures. A pass will be assigned to a test if items selected for editing, retrieval, printing, or export match *exactly* what is present in the test database hardcopy printout. A fail will be assigned to a test if either the software fails to operate or the software generates results that are inconsistent with the hardcopy printout of the test database. A conditional pass may be assigned to a test if the software functioned properly during the test, but the style and format of the results are undesirable.

REPORTING

The test analyst should document the test results and provide any pertinant information (such as deficiencies in performance), either verbally or in writting, to the software developer. Documentation should include all test results and any abnormalities noted by the test analyst. This documentation should be saved by the test analyst. If during the testing procedure a portion of the code does not operate correctly and makes continued testing impossible, then the test analyst will notify the software developer of the problem immediately. The software developer will make the necessary corrections and re-issue a new version of the software. The new software will be designated Beta Test Version 1.1 for the first occurrence and 1.2, 1.3 etc. for any future occurrences. All testing done prior to the occurrence will be redone using the new the software to assure corrections did not impact other subroutines in the software.

The test report should not include recommendations for improvements to the code. However, these may be requested separately. The report should include results from each of the tests identified in this document. All tests should be performed using the same Beta Test Version number.

After the software developer receives verbal test results, appropriate corrections to the software will be made, and the software will then be released to the CDC, identifying it as Version 1.0.

OPERATING ENVIRONMENT FOR TESTING

The software will use the Foxpro relational database language and will require a microcomputer operating under MS DOS©, Version 6.0 or higher, Windows Version 3.1 or higher, and at least 8MB of extended random access memory although 16MB is preferable. About 200MB of free hard disk space will be required to load and operate the software. A copy of Visual Foxpro should be loaded on the computer performing the testing.

REFERENCES

Radiological Assessments Corporation (RAC), 1995. *Technical Proposal: Source term Calculations and Ingestion Pathway Data Retrieval for the Savannah River Site (SRS) Environmental Dose Reconstruction Project*. Neeses, South Carolina, May 11, 1995

Rood, A. S., M. O. Langan, H. R. Meyer, 1996. *Software Requirements and Design Specifications for the Document Data Base. Radiological Assessments Corporation* DRAFT Technical Memorandum, June19-CDH-SRS-1996.

APPENDIX J

GROUNDWATER CONTAMINATION AND THE POTENTIAL FOR OFFSITE MIGRATION

APPENDIX J

GROUNDWATER CONTAMINATION AND THE POTENTIAL FOR OFFSITE MIGRATION

ABSTRACT

This appendix describes the various geological formations and associated aquifers that direct the flow of groundwater in the environs of the Savannah River Site (SRS). It also discusses the potential for offsite migration of groundwater and associated contaminant transport based on water flow in the aquifers underlying the SRS. Although groundwater contamination from SRS releases may be a potential exposure pathway for the future, the evidence suggests that it did not impact offsite residents before 1992.

INTRODUCTION

Operations at the SRS have resulted in groundwater contamination at several locations around the Site, including areas below seepage and retention basins. However, the accumulated groundwater monitoring data suggest that the potential for contamination of offsite groundwater is limited. Most of the impacted areas are near the interior of the Site, and the underlying groundwater outcrops at seep lines along the various Site streams eventually traveling offsite to the Savannah River. Surface water release estimates made for the Site streams (discussed in Chapter 5) are based on water concentrations measured at the location where the streams cross Road A. This sampling location is downstream from most of the impacted groundwater below seepage and retention basins at separations and reactor areas. The measurements result in surface water estimates that include potential contributions from the groundwater pathway that may be related to Site activities.

Groundwater below the M-Area, A-Area, D-Area, and TNX is closer to the Site boundary. Measurements made in 1993 indicate the plume of contamination in groundwater below the M-Area and A-Area to be near the Site boundary (Arnett et al. 1994). While this plume may eventually contaminate offsite groundwater, groundwater does not appear to be an exposure pathway to people living offsite before 1992. If this is the case, groundwater contamination is not an important factor in estimating historical dose from past SRS releases, but groundwater data may require further evaluation to estimate potential current or future offsite exposures. Figure J-1 shows the primary operational onsite areas and major Site streams and roads.

SAVANNAH RIVER SITE GEOLOGY

The geologic and hydrogeologic systems that control groundwater flow in the vicinity of the SRS are complex. Siple (1967) established three roughly separable systems consisting of (1) crystalline basement rocks composed of metamorphic and intrusive igneous rocks, (2) hardened Triassic-aged sediments, and (3) overlapping, weakly consolidated Cretaceous-aged and more recently deposited coastal plain sediments.

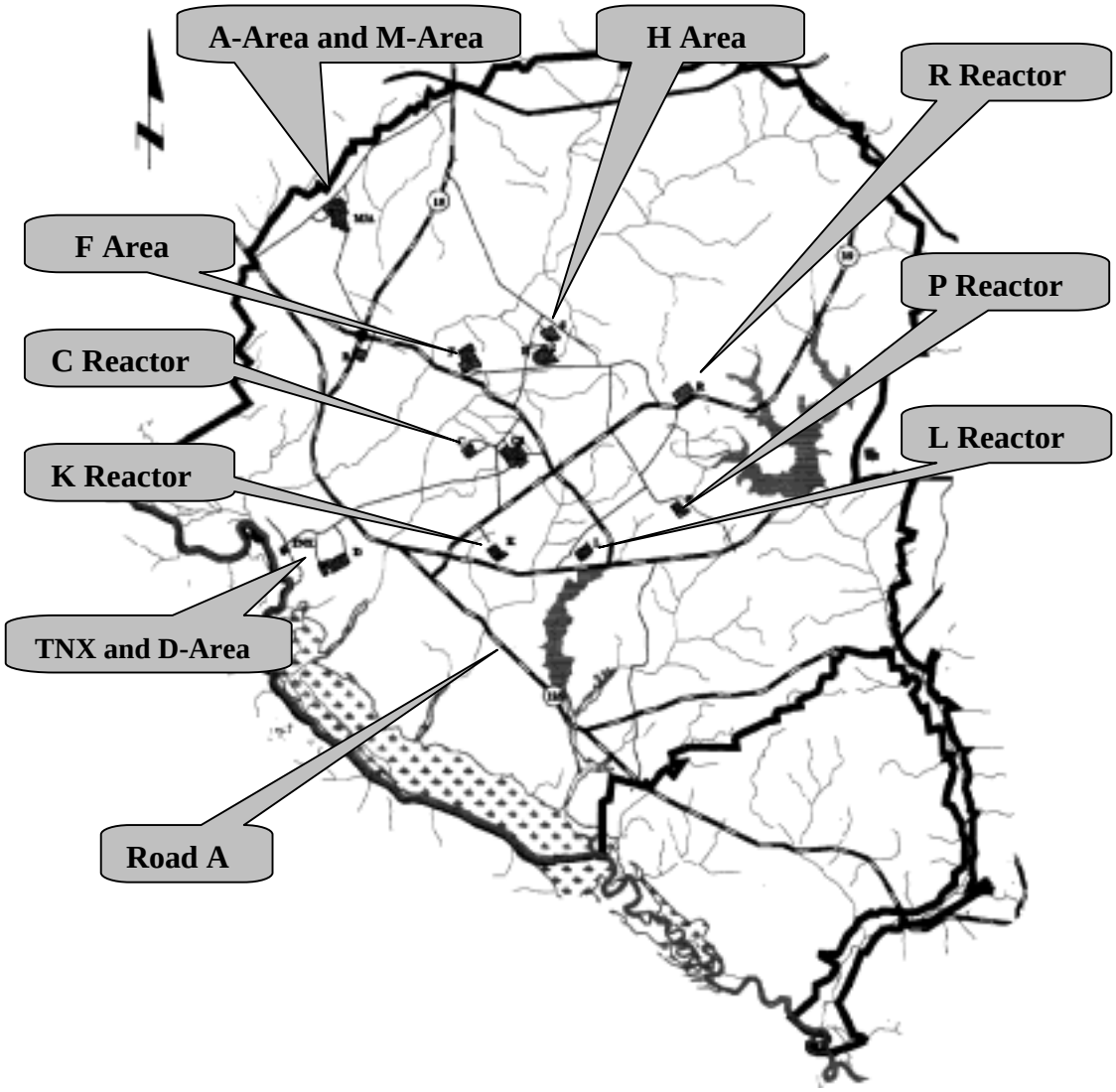


Figure J-1. The SRS showing the reactor (C-Area, K-Area, L-Area, P-Area, and R-Area) and processing (F-Area and H-Area) areas, TNX, D-Area, M-Area, and A-Area. The Site occupies approximately 300 mi^2 at the boundary of Georgia and South Carolina near Augusta, Georgia, and Aiken, South Carolina.

The permeability of the basement and Triassic-aged rocks is likely low, and test wells have shown that the water they do contain in joints and fractures is geopressured with a high hydraulic head. Marine (1974) attributed this overpressuring to osmotic pressure across the overlying impermeable aquitards, but its origin is uncertain. Overlying the basement and Triassic-aged rocks is a blanket of hardened, poorly sorted clayey sediments that hydraulically separates and isolates the younger, overlying sedimentary materials from the Triassic-aged and basement rocks (Carlton et al. 1993).

The nomenclature for the various aquifers, aquitards, and confining systems has changed significantly within the past 30 years. The layer that separates the older basement and Triassic-aged rocks from the more recently deposited sediments is referred to by Aadland et al. (1992) as the Appleton Confining System, but it has also been called the Cape Fear Formation. The

Cretaceous-aged formations are referred to by Aadland et al. (1992) as the Dublin-Midville Aquifer system. This system includes the deepest aquifers and is the source of most of the pumped groundwater at the SRS. The system includes aquifers in the Peedee, BlackCreek, and Middendorf Formations, which have also been referred to as the Upper and Lower Tuscaloosa Aquifers. Aadland et al. (1992) refers to the upper Tertiary-aged formations as the Floridian Aquifer system, which is separated from the lower system by confining beds of the Black Mingo Group, particularly the New Ellenton Formation. Also included in the upper system are the Tobacco Road, Dry Branch, McBean, Congaree, and Williamsburg Formations. The upper aquifers have historically been referred to as the Hawthorn, Barnwell, and Congaree Aquifers. Figure J-2 shows a geologic cross section of the SRS.

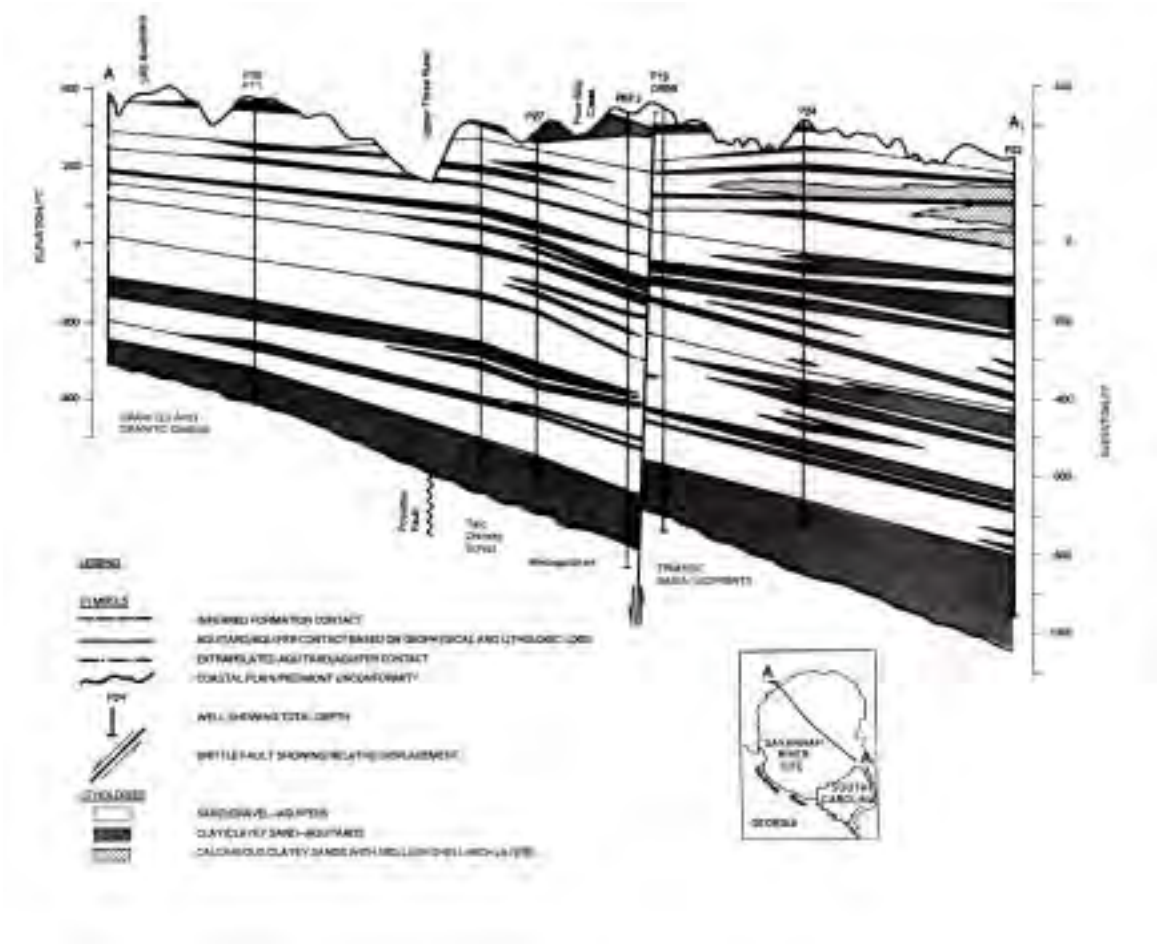


Figure J-2. Geologic cross section of the Savannah River Site. Sedimentary deposits of the Upper Coastal Plain near SRS consist primarily of alternating clay- and sand-rich layers with local carbonate-rich horizons. From Cummins et al. (1990).

Groundwater is used as a domestic, municipal, and industrial water supply throughout the Upper Coastal Plain. In Aiken County, municipal and industrial water supplies are primarily developed from the Cretaceous (lower) zone, and most domestic water supplies are developed from the Congaree-Fourmile and upper saturated zones in the Tertiary-aged sediments (Arnett et al. 1994). Potential contamination of surface water drinking supplies is discussed in Chapter 13

and Chapter 18 for radionuclides and chemicals, respectively. Figure J-3 shows the different stratigraphic units at the SRS. The Upper Cretaceous formations include what are know as the Upper and Lower Tuscaloosa Aquifers.

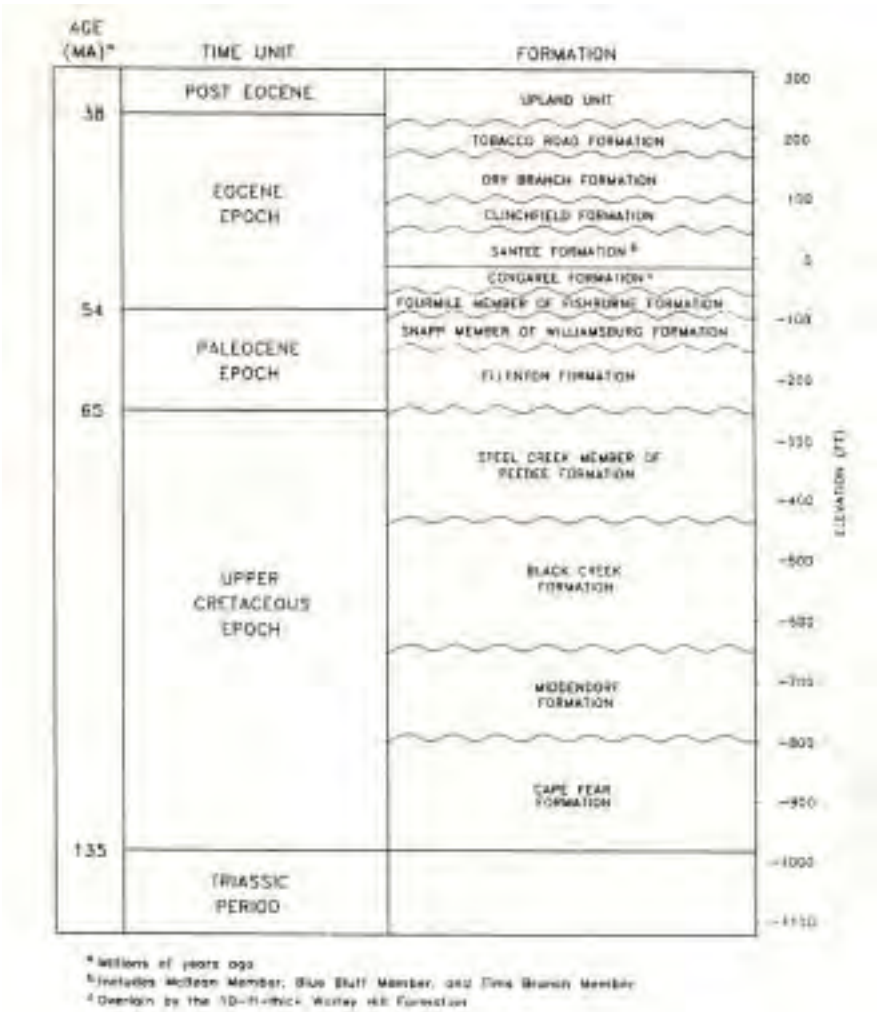


Figure J-3. Stratigraphic units at the SRS. Two regionally important aquifers, including the Upper and Lower Tuscaloosa Aquifers, occur in the Upper Cretaceous-age sandy sediments of the Middendorf, Black Creek, and Peedee formations. Aquifers in Eocene-age sediments are locally important but yield lower amounts of water. From Cummins et al. (1990).

The aquifers in the Tertiary-aged sediments receive local recharge, and flow at the water table is toward minor tributaries. Deeper aquifers generally flow toward the major Site streams. The deepest of the aquifers, those in the Cretaceous-aged sediments, including the Upper and Lower Tuscaloosa Aquifers, receive recharge at outcrop areas to the north of the Site, and groundwater flow is generally toward the Savannah River (Carlton et al. 1993).

The direction of vertical groundwater flow, and therefore contaminant transport, at any locality may change or even reverse in successively deeper aquifers. Beneath much of the SRS, hydraulic head decreases with depth, and the vertical flow of water is downward. This is the case

in the A-Area and M-Area, where discontinuous aquitards and downward-decreasing hydraulic head combine to allow water movement from the water table to deeper zones. R, P, and L reactor areas are also located in downward gradient areas, or recharge zones, for the lower aquifers. An upward gradient dominates at other areas, inhibiting downward groundwater flow to the deeper aquifers and directing flow to the upper aquifers in the Tertiary zone. This is the case in the F-Area and H-Area (separations areas), C and K reactor areas, D-Area, and TNX.

A-Area and M-Area, located approximately 0.5 mi from the nearest SRS boundary, are on a watertable mound. Horizontal groundwater flow is east toward Tim's Branch, southwest toward the Savannah River, and north and west toward drainage into lower topographic zones. The D-Area and the TNX are located approximately 0.75 and 0.25 mi from the nearest SRS boundary, respectively, and horizontal groundwater flow in these areas is toward the Savannah River and the nearby swamp. Horizontal groundwater flow at other SRS areas, including F-Area, H-Area, and the five reactor areas, is toward tributary streams draining into the major Site streams and eventually to the Savannah River.

GROUNDWATER MONITORING

The SRS has been concerned with the potential for radionuclide contamination of groundwater since the early-1950s and began monitoring a number of onsite wells shortly after operations began. During the late 1960s, groundwater monitoring in the vicinity of the F-Area and H-Area seepage basins intensified to determine the impact of the seepage basins on radionuclides, nitrates, and pH in groundwater and in Four Mile Creek, which receives input from groundwater in this area (Fenimore and Horton 1973). The monitoring network has been expanded since then to include about 1200 monitored wells. Monitoring has been most extensive at onsite locations, and groundwater monitoring by the SRS has been limited at offsite locations. Figure J-4 shows the locations of groundwater monitoring wells in the vicinity of the SRS during 1995. Monitoring wells are densely populated around the separations, fabrication, and reactor areas onsite, but offsite wells are significantly less numerous.

The groundwater monitoring program for nonradioactive materials or chemicals (discussed in Chapter 19) was established in 1982 and was rather limited until the mid-1980s when extensive groundwater sampling and analysis began. Much of the groundwater data has been collected to support waste characterization and cleanup activities. Improvements were made to groundwater sampling and sampling preservation techniques in 1983, including better flushing of wells before sampling and sample filtration for metal analysis (Zeigler et al. 1985).

The Environmental Monitoring Section (EMS) of the Environmental Protection Department maintains the monitoring program, which includes wells at various onsite locations, particularly around waste disposal areas and seepage and retention basins. The EMS currently maintains both a radioactive and nonradioactive monitoring program. Two additional SRS organizations also monitor groundwater: the Raw Materials Engineering and Technology Department monitors for volatile organics in A-Area and M-Area, and the Interim Waste Technology Division of the Savannah River Laboratory monitors selected burial ground wells (Murphy et al. 1991).

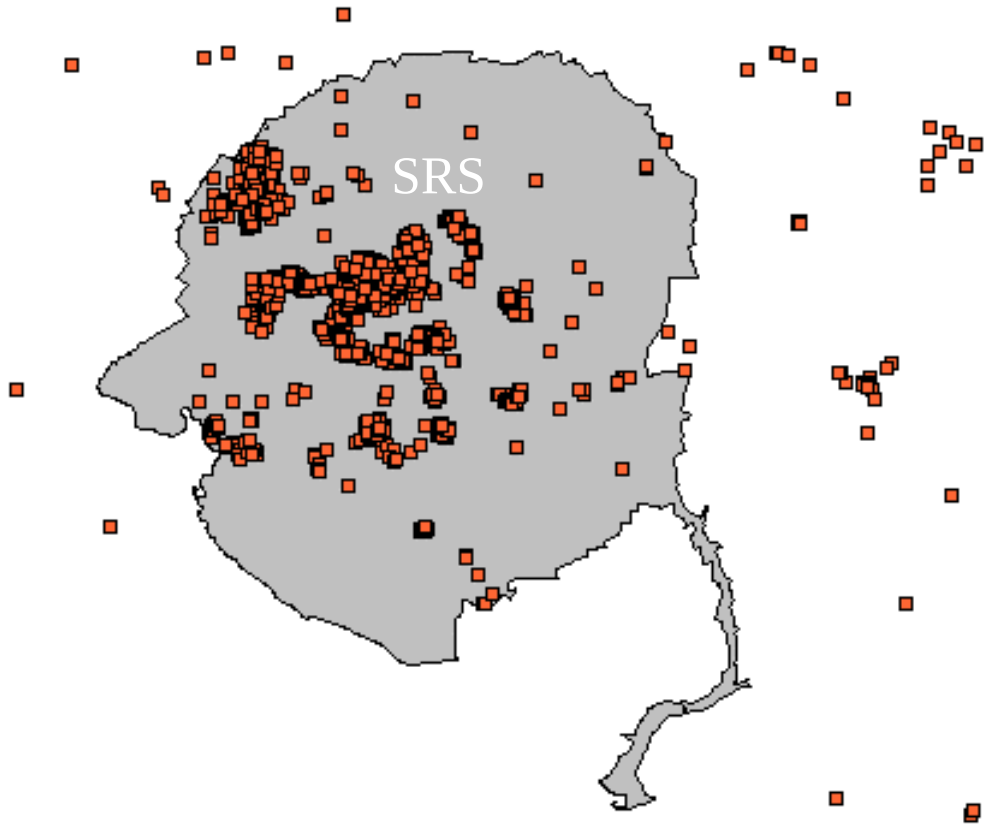


Figure J-4. Locations of SRS-maintained groundwater monitoring wells during 1995. These data are part of the Geographic Information System (GIS) coverages that are described in detail in Appendix F.

The EMS samples groundwater as part of the ongoing routine groundwater monitoring program or in response to specific requests from SRS personnel. The routine program schedules wells to be sampled semiannually, annually, or biannually, but requests by personnel outside the EMS result in monthly or quarterly sampling for many wells. New wells added to the program are initially sampled for four consecutive quarters for a comprehensive list of constituents. Sampling for this comprehensive list of constituents is also carried out for all active wells biannually (except for several older wells not properly constructed for such extensive sampling). Those wells with measured constituent concentrations exceeding a certain level are routinely sampled either annually or semiannually depending on the measured concentrations. All active wells are monitored quarterly for pH, temperature, specific conductance, alkalinity, and water level (Cummins et al. 1991).

PLUME DEFINITION WELLS

In addition to the network of routine groundwater monitoring wells, the SRS has established numerous plume definition wells to track the movement of contaminant plumes in the A-Area and M-Area, the F-Area and H-Area, and the TNX. Contaminant plumes in these areas are discussed in the following three sections.

M-Area and A-Area

The plume definition wells at A-Area and M-Area were installed after volatile organic contamination of the underlying groundwater was discovered in June 1981. The principal sources of contamination appear to be the solvent storage tank area, the M-Area settling basin, and the A-14 sewer outfall (Marine and Bledsoe 1984). The contamination plume, defined as water with contaminant concentrations above the primary drinking water standard (which consists primarily of trichloroethylene and, to a lesser extent, tetrachloroethylene and other chlorinated solvents), extended to wells within approximately 2000 ft of the nearest SRS boundary in 1993 (Arnett et al. 1994). Based on data provided by Marine and Bledsoe (1984), concentrations of trichloroethylene and tetrachloroethylene above the drinking water standard extended to wells within approximately 5000 ft from the plant boundary in 1984. This suggests that the contaminant plume moved toward the Site boundary at a rate of more than 250 ft y^{-1} between 1984 and 1993. It should be noted that reported distances from the leading edge of the contaminant plume to the nearest plant boundary appear to be general approximations, and this report does not make conclusions about their accuracy. Marine and Bledsoe (1984) reports a horizontal flow velocity of 20 to 25 ft y^{-1}, which is more than an order of magnitude lower based on flow velocity, water table gradient, effective porosity, and hydraulic conductivity.

The offsite wells (Figure J-4) are positioned too far from the SRS boundary to determine conditions near the boundary. However, recently installed wells close to the Site boundary near Green Pond Road are located between the leading edge of the contaminant plume and the Site boundary and do not show contamination at the Site boundary (Heffner 1998). Additionally, the residences nearest the Site boundary were connected to the New Ellenton municipal water system some time ago. Therefore, they do not rely on water supplied from the underlying aquifers, which may be impacted by the contaminant plume (Heffner 1998).

Changes in analyte concentration over time are also difficult to interpret because of groundwater remedial activities, which have significantly impacted groundwater flow in this area (Arnett et al. 1994). A groundwater remediation program has been in place at A-Area and M-Area since April 1985, and approximately 300,000 lb of solvents had been removed from nearly 1.75 billion gallons of groundwater as of September 1993 (Arnett et al. 1994). Groundwater contamination by chlorinated solvents resulting from M-Area operations and subsequent remedial air stripping are also addressed in Chapter 17.

Groundwater contamination extended vertically downward to the aquitard separating the upper aquifer system (Tertiary-aged formations) from the lower aquifer system (Cretaceous-aged formations) in 1984 based on M-Area well monitoring (Marine and Bledsoe 1984). Contamination extending into the lower system (frequently referred to as the Tuscaloosa Aquifer) was not evident, but the potential for downward migration exists because of the downward hydraulic head gradient in this area. Arnett et al. (1994) reported that trichloroethylene was detected above the drinking water standard (0.005 mg L^{-1}) in two wells in the Black Creek Unit, which is part of the Cretaceous-aged formations. Therefore, there is some indication that contamination is migrating downward into the lower aquifer system (Tuscaloosa Aquifer). The aquitards separating the lower aquifers of the upper aquifer system do, however, retard downward migration into the upper aquifers of the lower aquifer system. Additionally, the remedial efforts at M-Area and A-Area in the upper aquifer system have likely reduced the hydraulic drive toward

the deeper formations, as well as helped direct the lateral flow away from the Site boundary (Marine and Bledsoe 1984).

Although some contamination of the lower aquifer system does apparently exist, the leading edge of the contaminant plume, as discussed previously, was positioned approximately 2000 ft from the Site boundary in 1993. Furthermore, recently installed wells close to the Site boundary do not indicate local groundwater contamination (Heffner 1998). Therefore, it does not appear to represent an exposure pathway to members of the public before 1992.

Separations Areas

A number of plume definition wells were installed in the F-Area and H-Area (separations areas) in 1951 and 1952. These wells historically were used to monitor for radioactive constituents, and they were monitored for chemical constituents for the first time in 1993 (Arnett et al. 1994). Groundwater in the southern portion of these areas discharges to Four Mile Creek and its tributaries; in the northern portion of these areas, groundwater discharges to Upper Three Runs Creek and its tributaries (Cummins et al. 1991).

Vertical flow of groundwater contaminants is restricted by an aquitard, commonly referred to as the "green clay," and is generally confined to the shallower aquifers in the upper aquifer system. Additionally, the upward hydraulic head gradient in this area inhibits contaminant migration to the lower aquifer system. Both sets of seepage basins are on the slopes of the water table, and underlying groundwater is directed by the horizontal flow pattern toward Four Mile Creek, so the potential for offsite groundwater contamination is low.

TNX

TNX groundwater has been designated as a Resource Conservation and Recovery Act/Comprehensive Environmental Response, Compensation and Liability unit because of a contaminant plume consisting primarily of volatile organics that extends toward the Savannah River. Based on data from plume definition wells, the trichloroethylene plume in the Savannah River swamp extended to within approximately 500 ft of the Savannah River in 1990 (Cummins et al. 1991). Groundwater in this area discharges to the Savannah River and the nearby swamp.

Only formations in the upper aquifer system between elevations (above sea level) of about 150 and 100 ft have the potential for contamination, which is confined to the immediate vicinity of the basins. The upward hydraulic head gradient in this area prohibits downward migration of groundwater, and the potential for contamination of the deeper sediments is extremely low (Marine and Bledsoe 1984). The nearest Site boundary, the Savannah River, is approximately 1000 ft to the west of TNX, and contaminant migration through the intervening Savannah River swamp appears to be slow.

OTHER POTENTIAL SOURCES OF CONTAMINATION NEAR THE SRS BOUNDARY

The Savannah River Laboratory (SRL) seepage basins are located east of the A-Area, approximately 4000 ft southeast of the nearest Site boundary. Groundwater sampling indicates low-level contamination in the upper aquifer system, and, as in A-Area, the potential for

downward contaminant migration exists because of the downward hydraulic head gradient in this area. Based on water table elevations, horizontal movement appears to be toward the Site boundary. The relatively flat water table in this area results in a small horizontal gradient, however, and horizontal movement is likely slow (Marine and Bledsoe 1984).

The Silverton Road waste site is located about 1.5 mi west of M-Area, approximately 3200 feet from the nearest Site boundary (on U.S. Forest Service land). However, the nearest boundary down the groundwater gradient is approximately 10,500 ft to the southwest. Monitoring indicates volatile organic contamination in the upper aquifer system, and, as in M-Area, the potential for downward migration exists. The horizontal groundwater gradient at this location is toward the Site boundary, so there is potential for eventual offsite migration. However, the rate of migration is likely slow because of the relatively flat water table (Marine and Bledsoe 1984).

AVAILABLE DATA

A number of groundwater-related datasets are available and are being provided to the Centers for Disease Control and Prevention with this report, including several geographic information system coverages that are described in detail in Appendix F. The SRS groundwater monitoring locations shown in Figure J-4 are examples of the types of data that are available.

Additionally, coverages are being provided for U.S. Geological Survey groundwater quality data and water table elevation contours. Figure J-5 shows U.S. Geological Survey groundwater monitoring well locations in the vicinity of the SRS. The wells are clustered most heavily along the eastern edge of the Site, and there are relatively few wells in the vicinity of TNX, D-Area, M-Area, and A-Area.

CONCLUSIONS

Although groundwater contamination from SRS releases may be a potential exposure pathway for the future, the evidence suggests that it did not impact offsite residents before 1992. Most of the groundwater flow in contaminated areas at the SRS is toward Site streams and eventually the Savannah River. It does not appear that the groundwater monitoring data could be used to increase the accuracy of our surface water release estimates. These estimates, which are based on water concentrations measured in Site streams at Road A, include possible contributions from contaminated groundwater in the separations and reactor areas.

A potential exists for future contamination of offsite groundwater resulting from activities in A-Area and M-Area. As of 1993, the contamination plume beneath these areas, characterized by chlorinated solvent concentrations above the primary drinking water standard, extended to wells within about 2000 ft of the nearest Site boundary. However, remedial programs have significantly decreased the amounts of contamination, and the plume does not appear to have reached the Site boundary. However, evidence suggests that the contaminant plume is migrating downward into the lower aquifer system. There is also the potential for the contaminant plume in groundwater below the TNX to eventually extend and discharge to the Savannah River, and groundwater below the SRL seepage basins and the Silverton Road waste site appears to be migrating toward the Site boundary.

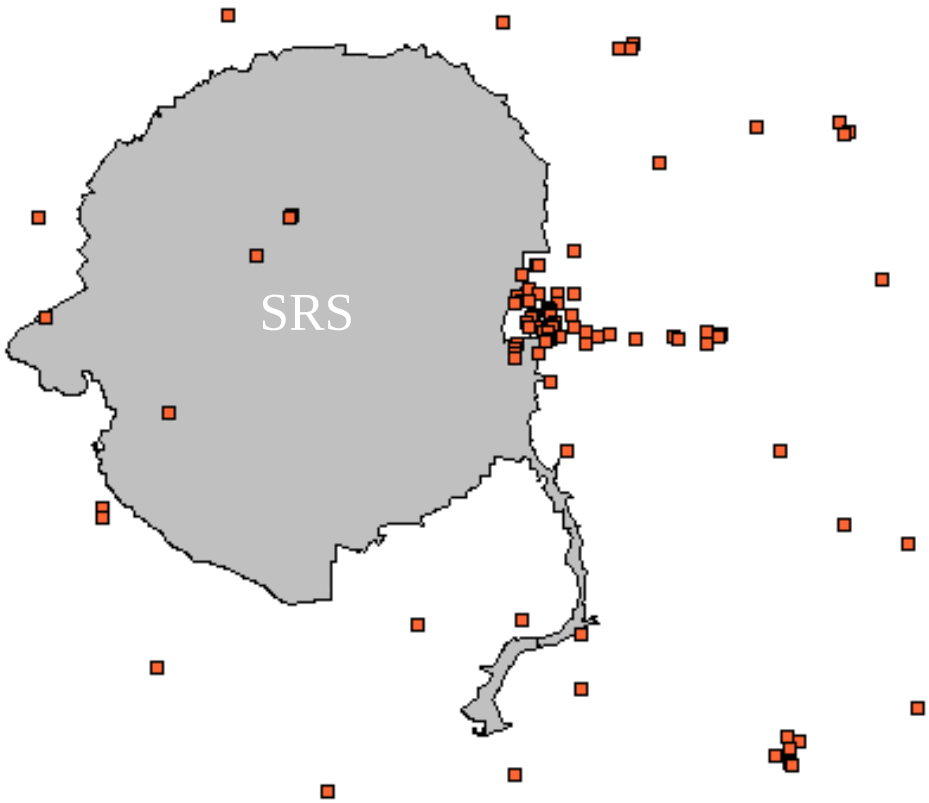


Figure J-5. Locations of groundwater monitoring wells maintained by the U.S. Geological Survey.

Additional groundwater data may be necessary to thoroughly examine the potential for current or future public exposure. However, it does not appear that these data are necessary for establishing historical offsite exposure through contaminated groundwater, which does not appear to have been a complete pathway during the 1953–1992 period covered by this historical dose reconstruction study.

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APPENDIX K

POTENTIAL USES FOR ENVIRONMENTAL MONITORING DATA

APPENDIX K

POTENTIAL USES FOR ENVIRONMENTAL MONITORING DATA

INTRODUCTION

The environmental monitoring data we have compiled have a number of potential uses during subsequent phases of the Savannah River Site (SRS) dose reconstruction project. These uses include source term verification, model validation and parameter development, and direct exposure assessment. Additionally, comparing monitoring data for different media can assist with establishing data quality and verifying reported values. This appendix discusses and illustrates some of the potential uses for the environmental monitoring data compiled, but it is not intended to include all potential uses. Other potential uses for specific media data are discussed in Chapters 8, 9, 10, 11, 12.1, 12.2, 13, and 14.

Varying temporal and spatial resolution for different media and for release estimates complicates comparisons and frequently limits the usefulness of the data. Another factor that complicates these types of comparisons is that the SRS is not the only source of environmental contamination. It is often difficult to conclusively identify the origin of environmental contamination, and background concentrations can be difficult to establish. In the context of historical dose reconstruction for the SRS, background refers to contaminants present in the environment that did not originate as a result of SRS activities. Other factors that may preclude or complicate data analyses for specific media are discussed in detail in Chapters 8, 9, 10, 11, 12.1, 12.2, 13, and 14.

ESTABLISHING THE SOURCE OF CONTAMINATION

Atmospheric weapons testing resulted in widespread deposition of radionuclides, particularly during the early 1960s, and it is often difficult to distinguish between contamination resulting from weapons testing and contamination resulting from SRS operations. Evaluating concentrations measured at different distances from the Site and comparing concentrations with deposition trends can help establish the source of contamination.

Figure K-1 shows ^{90}Sr deposition from atmospheric weapons testing fallout (discussed in Chapter 6) measured in Columbia, South Carolina, and ^{137}Cs concentrations measured in vegetation (Chapter 9) collected at the SRS plant perimeter and 25-mi radius locations. Fallout deposition was generally greatest between 1962 and 1964, and ^{137}Cs concentrations measured in vegetation follow the same general trend seen for ^{90}Sr deposition in Columbia, South Carolina, which would not have been impacted by Site releases. Vegetation concentrations are similar at the plant perimeter and 25-mi radius locations, which also suggests weapons testing as the primary source of the cesium.

Figure K-2 compares monthly average ^{131}I concentrations measured in air (Chapter 8) at the plant perimeter and 25-mi radius locations from 1959 through 1964. Concentrations at both locations appear generally well correlated throughout much of this time period. Concentrations at the plant perimeter are elevated compared to 25-mi radius locations during July 1959 and June 1961, suggesting impact from radioiodine releases from the Site extending to at least the plant perimeter during these time periods. Similar concentrations at plant perimeter and 25-mi radius

locations during certain months of 1961 and 1962 suggest that fallout from global nuclear weapons testing is the primary source of contamination.

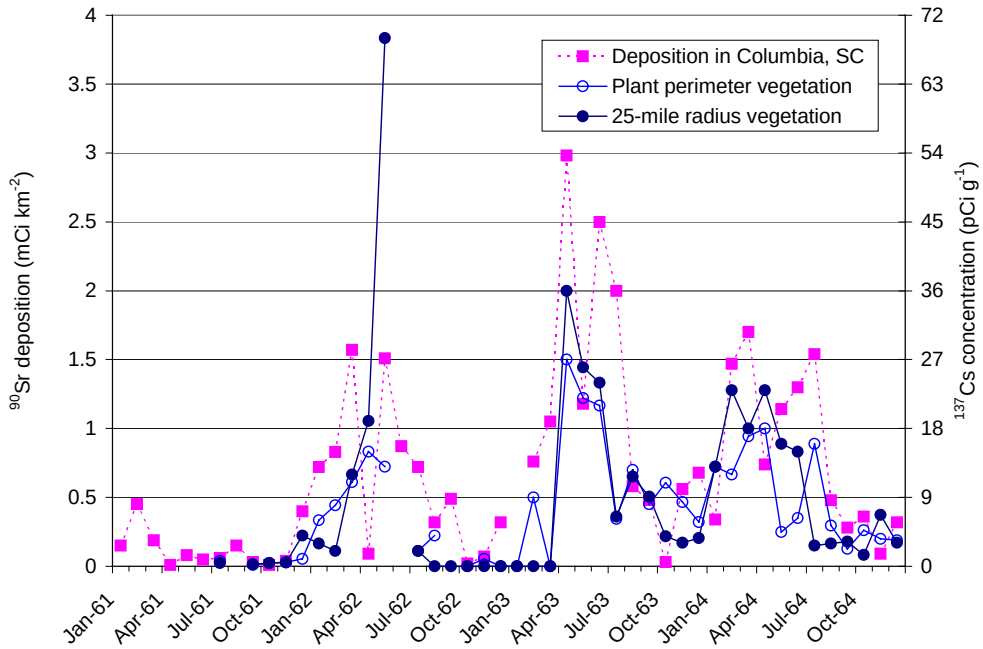

Figure K-1. Monthly average ^{90}Sr deposition in Columbia, South Carolina, and monthly average ^{137}Cs concentrations measured in vegetation at the plant perimeter and 25-mi radius locations from 1961 through 1964. Link to tabulated data.

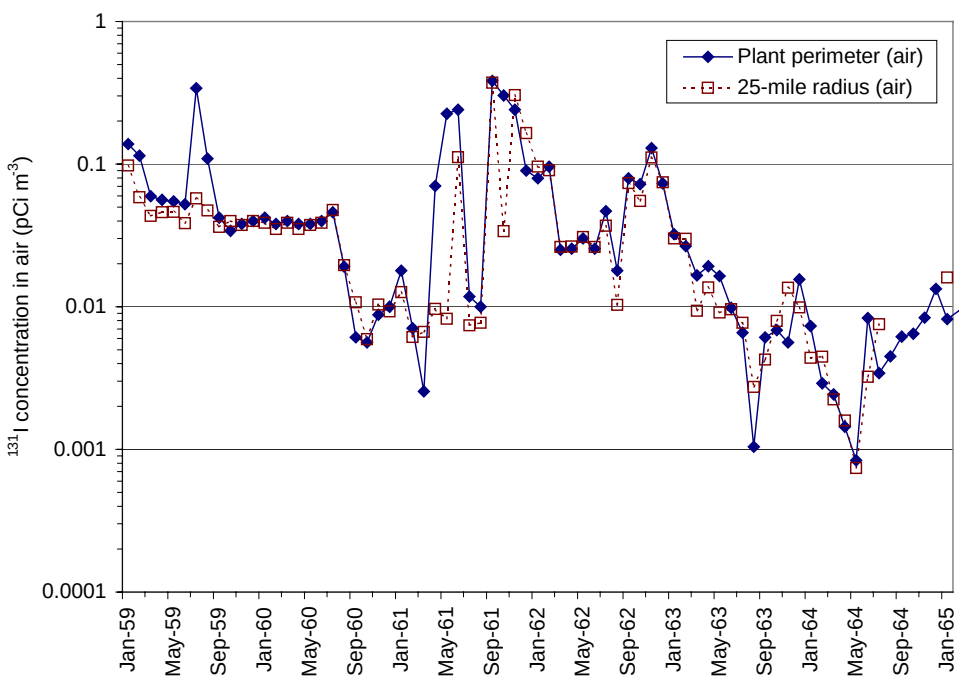

Figure K-2. Monthly average ^{131}I concentrations measured in air collected at plant perimeter and 25-mi radius locations from 1959 through 1964. Link to tabulated data.

Figure K-3 shows monthly average ^{131}I concentrations measured in vegetation at the plant perimeter and 25-mi radius locations from 1966 through 1973 (Chapter 9). Concentrations are similar at both locations and are near the detection limit since 1969, suggesting little Site impact beyond the plant perimeter during this time period. Concentrations near the detection limit are also seen during this time period for other media, such as air and milk (see Chapters 8 and 10).

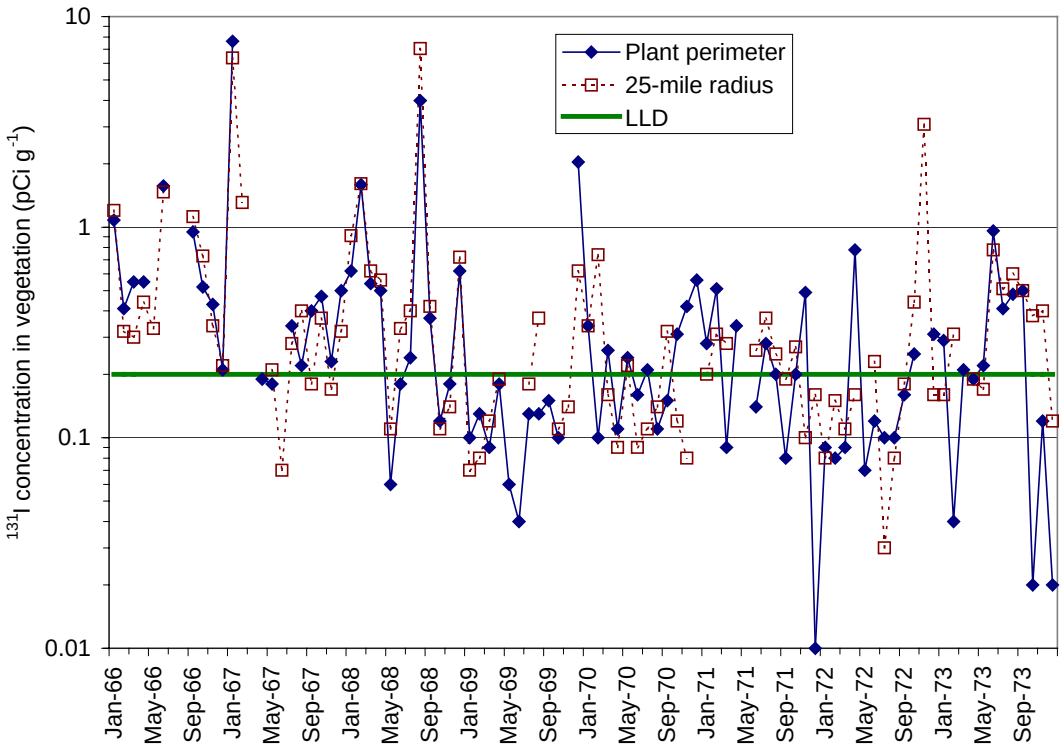

Figure K-3. Iodine-131 concentrations measured in vegetation collected from plant perimeter and 25-mi radius locations from 1966 through 1973 shown with the lower limit of detection (LLD). Link to tabulated data.

Figure K-3 shows the annual average tritium concentrations for the years 1974 through 1991. It appears clear that Site tritium releases have contributed to elevated concentrations measured in vegetation samples at offsite locations extending at least to the 25-mi radius locations. Concentrations measured at the F-Area and H-Area are clearly greater than those measured at the plant perimeter, 25-mi, and 100-mi locations. The plant perimeter concentrations are consistently higher than 25-mi radius concentrations, which are slightly higher than 100-mi radius concentrations. Concentrations for all locations decreased significantly from 1974 through 1977 and have fluctuated somewhat since that time.

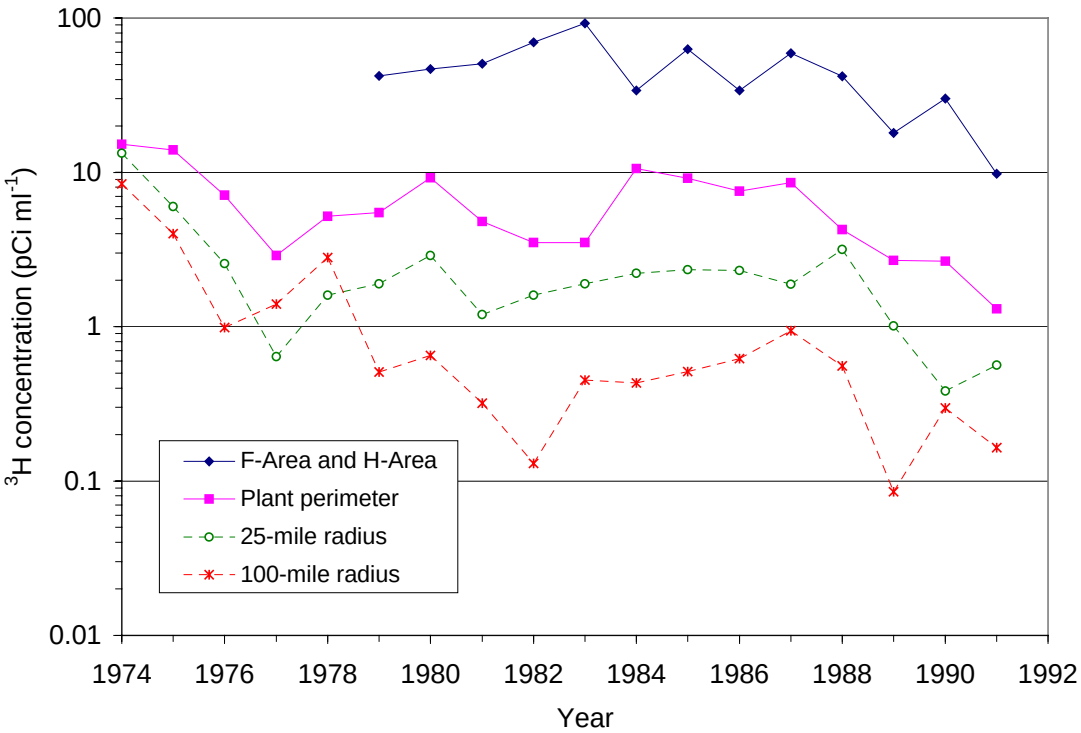

Figure K-3. Tritium concentrations measured in water extracted from vegetation at the F-Area and H-Area, plant perimeter, 25-mi radius, and 100-mi radius locations. Link to tabulated data.

In general, offsite (i.e., extending to at least the plant perimeter) impacts are consistently evident for atmospheric tritium releases. Potential offsite impacts resulting from radioiodine atmospheric releases are evident for several time periods before 1962. Fallout from global nuclear detonations appears to be the primary source for all other atmospherically deposited fission products beyond the plant perimeter. See Chapters 8, 9, 10, 11, 12.1, 12.2, 13, and 14 for additional details regarding concentrations measured in various environmental media.

VERIFYING SOURCE TERM ESTIMATES

Environmental data are useful for comparison to source term estimates and can assist with verifying the estimated release amounts. Figure K-5 compares atmospheric elemental ^{131}I release estimates (Chapter 4.2) with ^{131}I concentrations measure in air (Chapter 8) at onsite (F-Area and H-Area) and plant perimeter locations. Onsite air concentrations are clearly impacted during periods of increased releases, particularly during January, February, and July 1959; January 1960; June 1961; and May, June, and July 1962. Plant perimeter concentrations also appear to be impacted during July 1959 and June 1961, suggesting impact beyond the plant perimeter during these periods of elevated releases. Elevated concentrations are also evident during the latter part of 1961, but similar increases at both onsite and plant perimeter locations and lower release amounts during this time suggest increased weapons testing as the probable source of these elevated concentrations. Although monthly monitoring data are not available for air before 1959,

higher relative release amounts, particularly during 1956, suggest that offsite impact was likely. Some summary data are available (for example, see Chapter 8, Figure 8-5 and Table 8-6) that confirm the highest releases occurred before 1960, particularly during 1956.

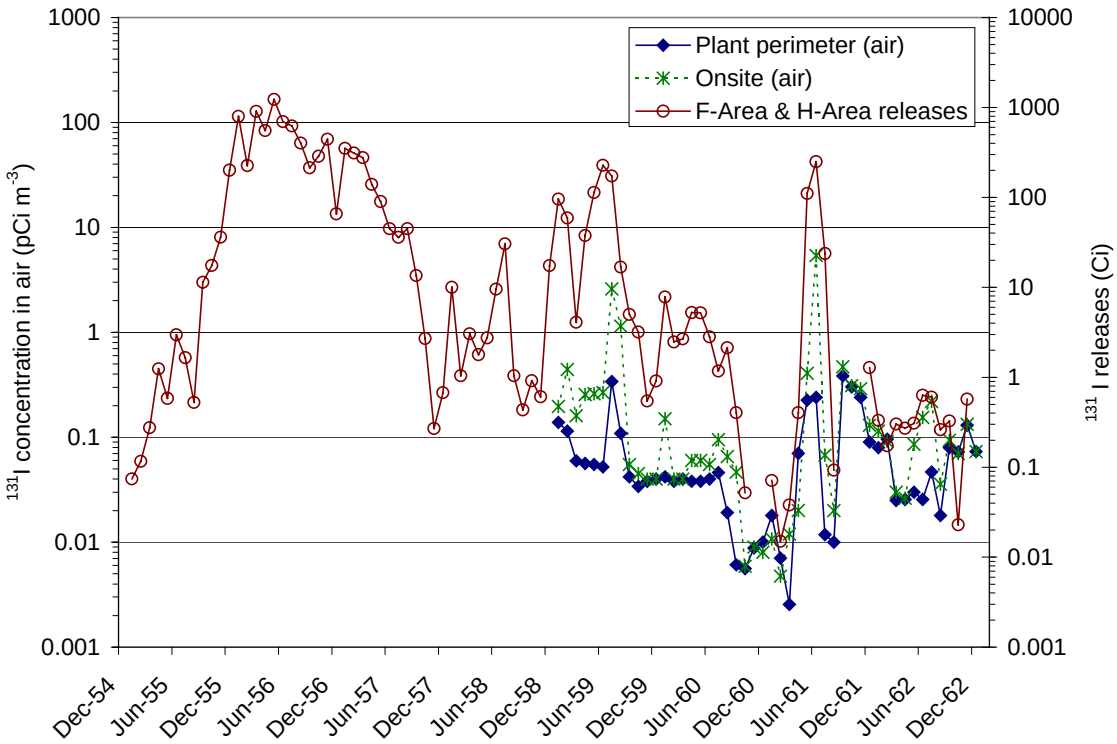

Figure K-5. Elemental ^{131}I release estimates from F-Area and H-Area and monthly average air concentrations measured at the plant perimeter and onsite (F-Area and H-Area) locations. Link to tabulated data.

Figure K-6 shows total annual atmospheric tritium release estimates (Chapter 4.1) from the reactor and separations areas from 1955 through 1991 compared to annual average tritium concentrations measured in vegetation and rainwater (Chapters 9 and 8). The vegetation concentrations depicted here before 1974 represent estimates made based on the ratio of atmospheric releases to vegetation concentrations measured from 1974 through 1991. This ratio was then used to estimate vegetation concentrations before 1974, based on atmospheric tritium releases between 1955 and 1973. Because vegetation concentrations and release estimates are well correlated between 1974 and 1991, this likely represents a good approximation of average vegetation concentrations that may have occurred before 1974.

Concentrations measured in rainwater and vegetation are well correlated at both onsite and plant perimeter locations from 1974 through 1991; however, data are limited for onsite rainwater concentrations. Additionally, estimated vegetation concentrations between 1955 and 1973 are generally well correlated with rainwater concentrations measured and reported for that time period.

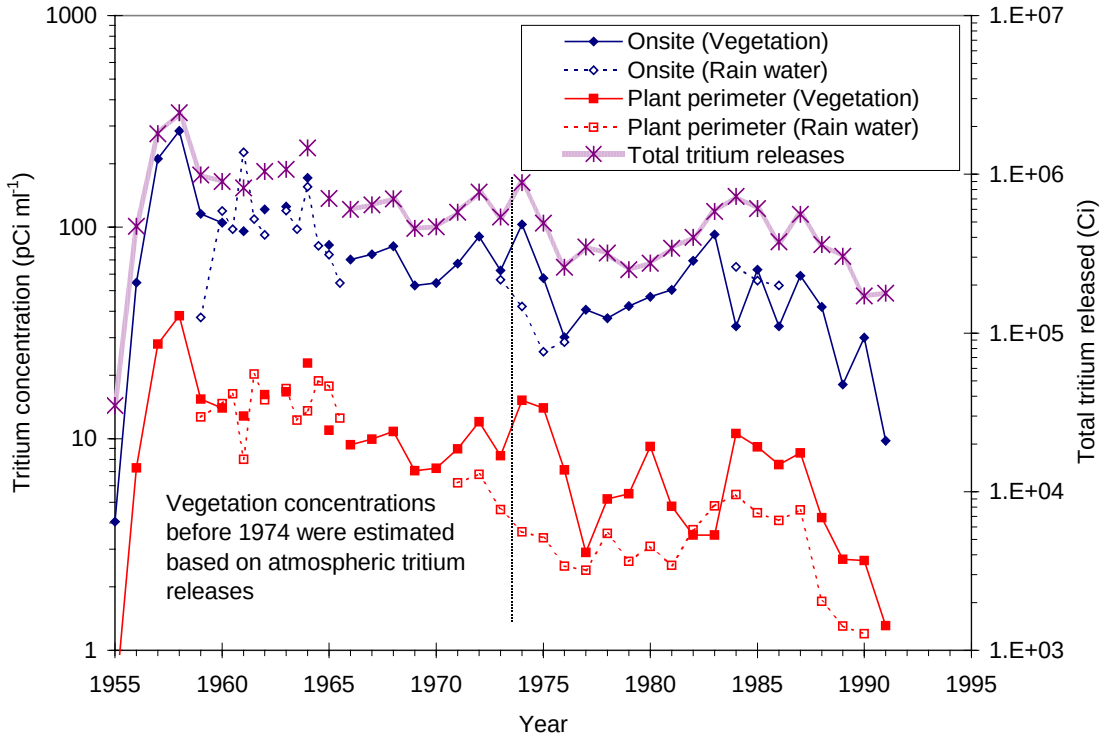

Figure K-6. Atmospheric tritium releases and annual average tritium concentrations measured in onsite and plant perimeter vegetation and rainwater. Tritium concentrations were not consistently reported for vegetation before 1974, and the concentrations shown here are estimated based on atmospheric tritium release estimates. Link to tabulated data.

Figure K-7 shows ^{137}Cs concentrations measured in water collected from Steel Creek at Road A from 1959 through 1976, which are the basis for surface water release estimates to Steel Creek (Chapter 5). Also shown are ^{137}Cs concentrations measured in fish collected from Steel Creek at Road A and at a location 2 mi downstream from Road A (Chapter 14). The concentrations reported for fish at both locations appear well correlated with water concentrations and, therefore, release estimates during this time period.

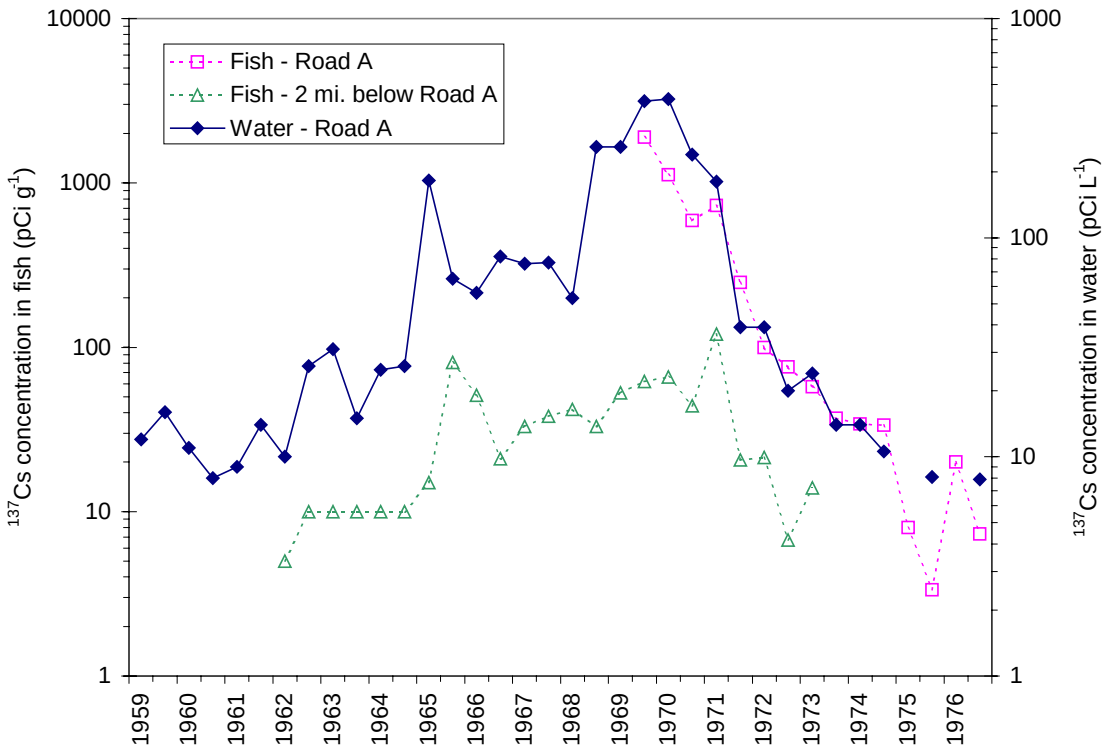

Figure K-7. Semiannual average ^{137}Cs concentrations measured in water and fish from Steel Creek at Road A and in fish from Steel Creek 2 mi below Road A. Link to tabulated data.

COMPARING CONCENTRATIONS MEASURED IN DIFFERENT MEDIA

Comparing concentrations measured in different environmental media can assist with filling existing data gaps or holes and help verify reported values, which provide confidence in the data. These comparisons are often difficult to interpret because of varying degrees of temporal and spatial resolution. However, correlation is evident for concentrations measured in different media. Figure K-6 demonstrates the correlation between vegetation and rainwater, and Figure K-7 demonstrates the correlation between fish and surface water.

Figure K-8 illustrates the general agreement between ^{131}I concentrations measured between 1959 and 1966 in air and vegetation samples collected at the plant perimeter. The air and vegetation concentrations show the same general trends, with peaks in concentration evident during July 1959 and June 1961 that are likely related to elevated Site releases (see Figure K-5). As previously discussed, the higher concentrations measured during the latter part of 1961 are likely the result of increased weapons testing during that time period. Vegetation concentrations were reported on a wet weight basis before 1961; however, beginning in 1961, concentrations were reported on a dry weight basis. This helps account for the relatively lower vegetation concentrations observed before 1961.

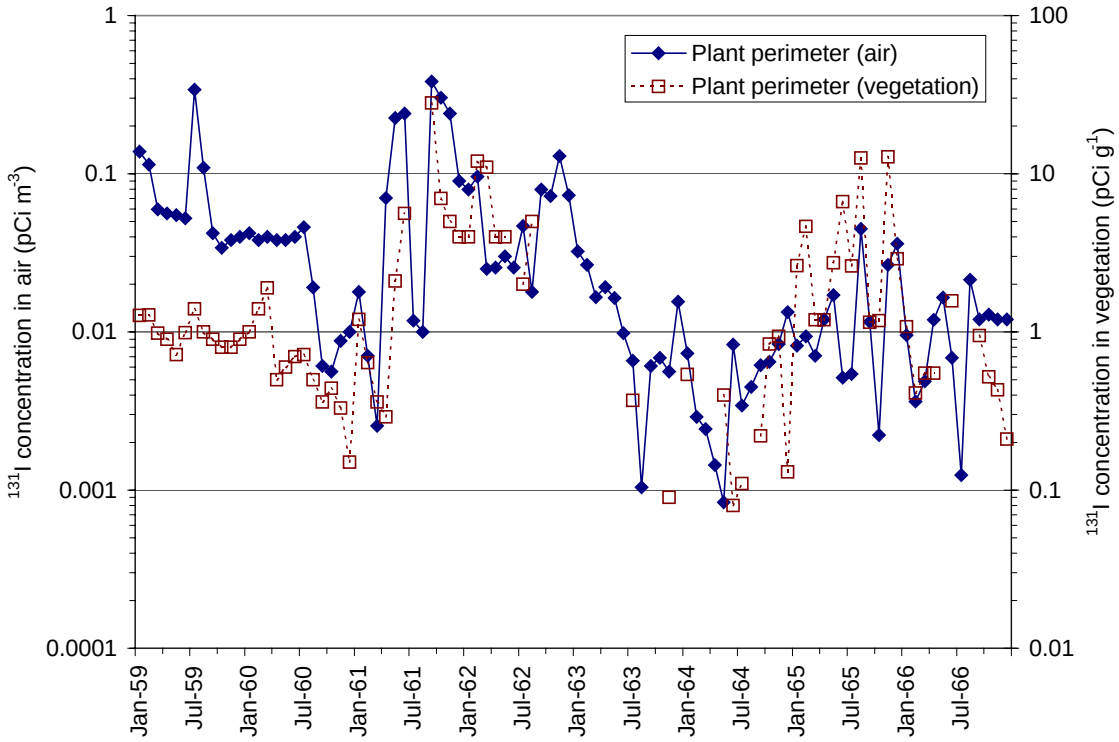

Figure K-8. Monthly average ^{131}I concentrations measured in vegetation and air at the plant perimeter locations from 1959 through 1966. Link to tabulated data.

Figure K-9 compares monthly average ^{131}I concentrations measured in vegetation (Chapter 9) and air (Chapter 8) at the plant perimeter and in milk (Chapter 10) from the Aiken, South Carolina, dairy during 1961 and 1962. Similar trends are evident for the three media, with peaks in concentration during May and June 1961 likely the result of increased separations area releases following the inadvertent reprocessing of short-cooled fuel.

The figures in this appendix are examples of the ways in which the environmental monitoring data may be useful during subsequent phases of this dose reconstruction project. Although they do not necessarily tell the whole story, they are important pieces of the puzzle and can often help identify the source of contamination and assist with quantifying potential offsite impacts related to Site releases. However, inadequate temporal or spatial resolution may limit the usefulness of the various data, particularly during time periods before 1959.

Using the data for model validation is not yet possible and will be part of subsequent phases of the dose reconstruction project following selection of appropriate dispersion models. It may also be possible to use the environmental monitoring data to develop site-specific model parameters. For example, concentration ratios can be calculated using fish and water concentrations. Calculation and selection of appropriate concentration ratios are discussed in greater detail in Chapter 14. Some of the data may also be useful for direct exposure assessment. This is likely possible for wild game and fish data, which are discussed in detail in Chapters 11 and 14, respectively.

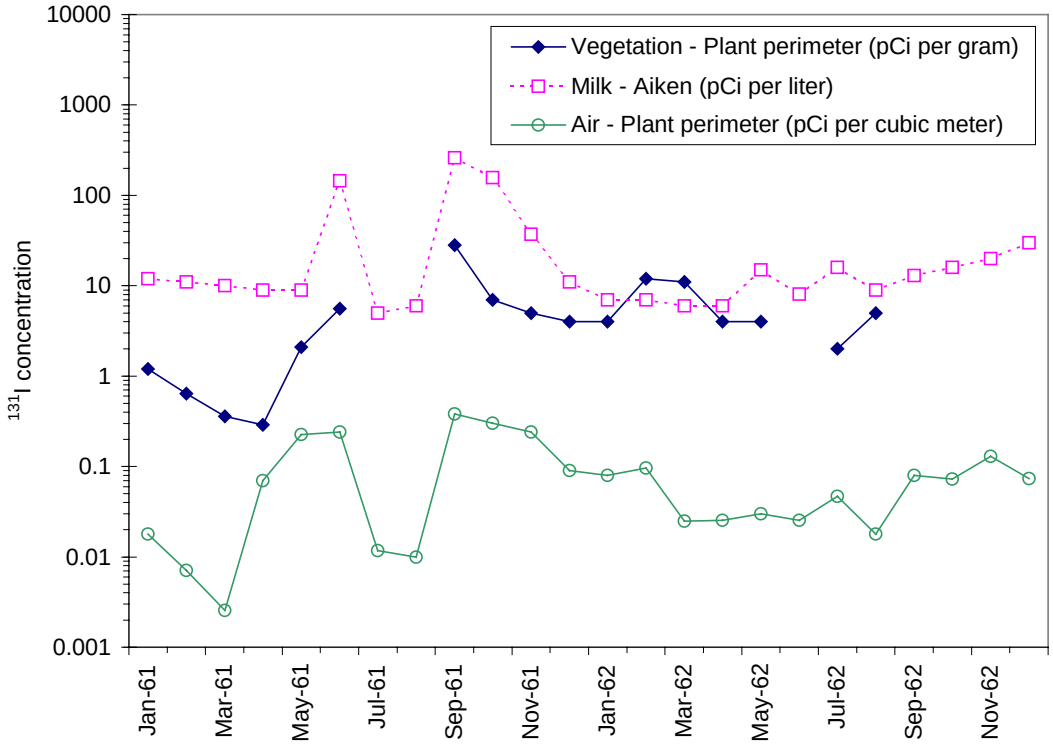

Figure K-9. Monthly average ^{131}I concentrations measured in vegetation and air collected at plant perimeter locations and in milk collected at the Aiken, South Carolina, dairy. Link to tabulated data.

The data compiled for developing the figures in this appendix are described in detail and provided in various other chapters of this report, identified separately for each figure in this appendix. The tabular data used to produce the figures depicted in this appendix can be accessed directly by readers of the electronic version of this document by clicking on the following hyperlink: AppK-Figure_data.xls.