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Nevada Test Site Radionuclide Inventory, 1955-1992 (U)

Scott M. Bowen, Phyllis L. Baca, Loretta F. Olivas, Carmen G. Geoffrion, Joseph L. Thompson, and Charles M. Miller, Los Alamos National Laboratory, Los Alamos, NM

Wataru Goishi, Bradley K. Esser, Jesse W. Meadows, Neil Namboodiri, David K. Smith and John F. Wild, Lawrence Livermore National Laboratory, Livermore, CA

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Nevada Test Site Radionuclide Inventory, 1955-1992 (U)

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ABSTRACT (U)

An inventory of radionuclides produced by underground nuclear testing conducted at the Nevada Test Site (NTS) by the Lawrence Livermore National Laboratory (LLNL) and the Los Alamos National Laboratory (LANL) from 1955 to 1992 includes tritium, fission products, actinides, and activation products. This inventory provides a measure of radioactivity remaining underground at the NTS after nuclear testing. The radionuclide data is reported gathered into five groups corresponding to geographic regions at NTS: Frenchman Flat, Pahute Mesa Area 19, Pahute Mesa Area 20, Rainier Mesa, and Yucca Flat, for events fired above and below the static water level. Curie activities are reported as of January 1, 1994, and January 1, 2094. This inventory does not represent the total radioactivity dissolved or potentially dissolved in the groundwater beneath the Nevada Test Site, but is strictly a compilation of the residual radionuclide inventory remaining from those underground nuclear tests conducted by LLNL and LANL from 1955 to 1992. The written report is companion to a personal computer based electronic database resident in the Nuclear Chemistry Division of the Lawrence Livermore National Laboratory and the Chemical Science and Technology Division of the Los Alamos National Laboratory. This database tabulates radionuclide totals, cavity radius, working point lithology, static water level, hole name and firing data for each event. This work has been sponsored by the U.S. Department of Energy, Nevada Operations Office, Environmental Restoration Division.

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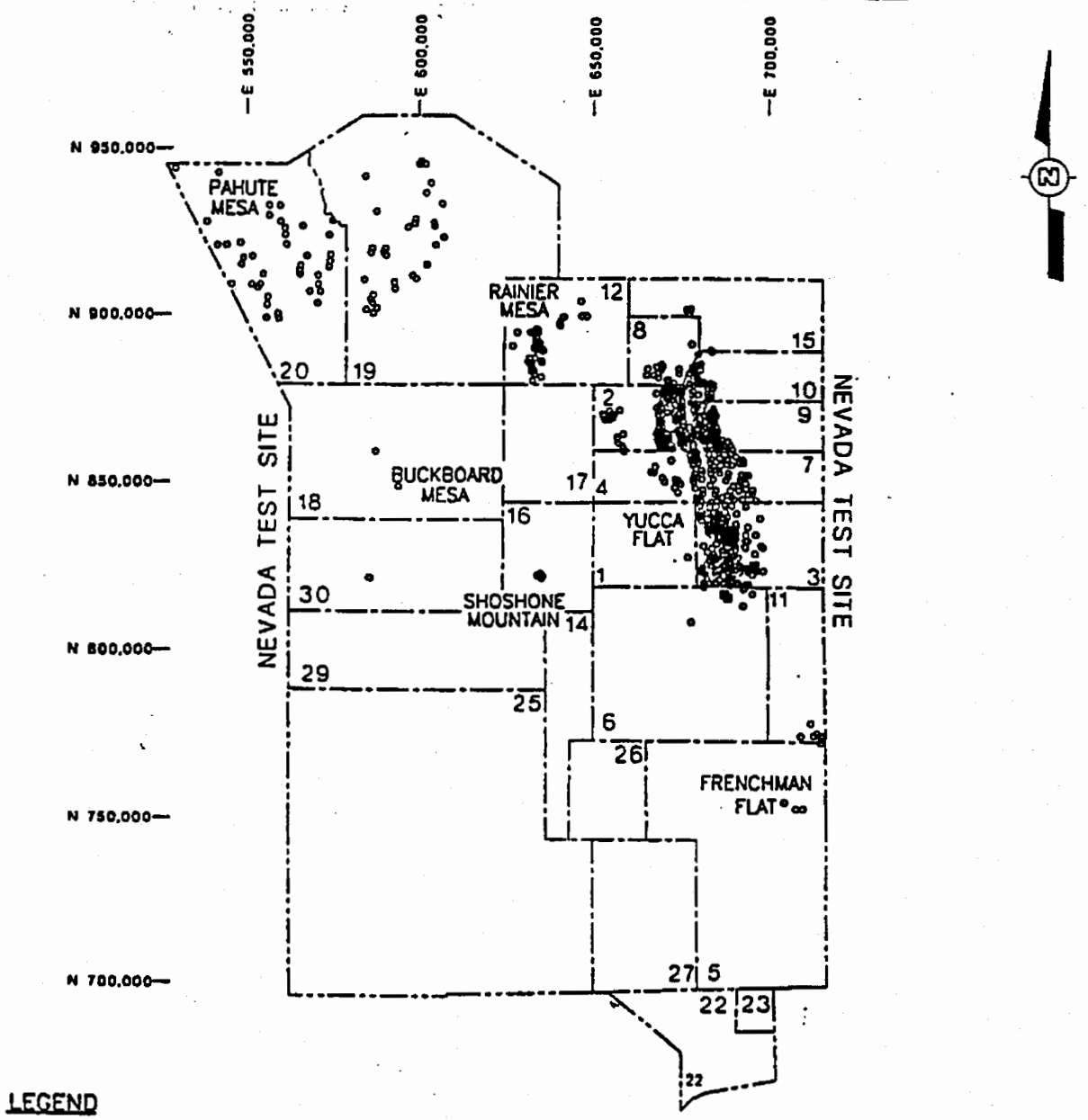

Figure 2: Map of the Nevada Test Site showing the locations of underground nuclear tests announced prior to December 1993. Over 200 previously unannounced tests were declassified by the Department of Energy in December 1993 and are omitted from this figure.

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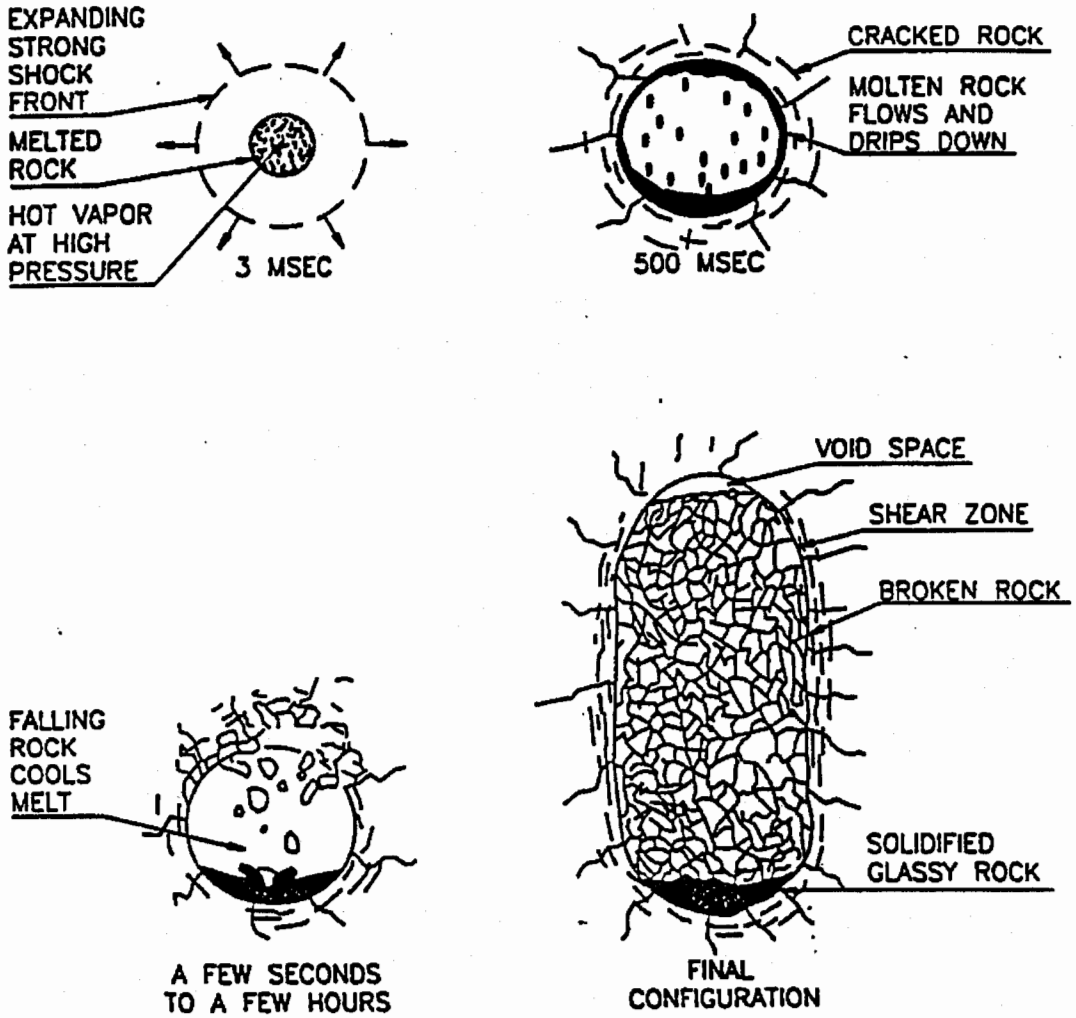

FORMATION HISTORY OF A NUCLEAR EXPLOSIVE CAVITY AND CHIMNEY

Source: Schwartz et al., 1984

Figure 3: Idealized formation history of an underground nuclear explosion in siliceous rock. Stages are in elapsed time from detonation.

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DISTRIBUTION OF RADIONUCLIDES

Phenomenology strongly influences the distribution of radionuclides within the cavity-chimney complex (Borg *et al.*, 1976). Immediately after a nuclear explosion, all of the radioactive species exist as gasses. As the cavity ceases to expand, heat is transferred to the wall rock and the cavity temperature and pressure begins to drop. The melt that flows to the floor of the cavity entrains the refractory radionuclides with higher boiling points (rare earth elements, alkaline earths, Zr and Pu). Most of these refractory species are trapped in the cooling melt; a small proportion is incorporated with the collapsed chimney rubble as splash or fine droplets entrained with escaping cavity glasses. Volatile species with lower boiling points (tritium, alkalis, Ru, U, Sb, Cl, I) circulate up cracks in the rubble chimney. Activation products are concentrated around the working point and will be largely incorporated in the melt or debris that borders the cavity. Volatile species, particularly ^{90}Kr and ^{137}Xe, are transported as gasses through the rubble and will be concentrated higher in the cavity and in the chimney relative to the refractory radionuclides. Drillback samples systematically exhibit higher volatile to refractory radionuclide ratios (Cs/Eu) for returns collected higher in the cavity and chimney. In addition, recent studies of nuclear tests fired above the water table indicate that material may be transported by prompt injection through explosion induced fractures arranged radially away from ground zero. By this mechanism, gaseous species, particularly tritium and ^{137}Cs (^{137}Cs has a gaseous ^{137}Xe precursor with a 3.8 minute half-life), may be deposited several cavity radii away from the working point. There is some evidence that refractory species may be similarly transported by prompt injection (Thompson and Gilmore, 1991).

Ultimately, the amounts and types of radionuclides resulting from a specific nuclear explosion will depend on the amount of fissile material, the fission/fusion ratio and the device yield. On an event-by-event basis, these data are classified. The present report attempts to quantify the abundance of these residual radionuclide species for five geographic test centers at the Nevada Test Site.

RADIONUCLIDE INCLUSION CRITERIA

Radionuclides to be considered for inclusion in the source term inventory are: 1) Residual fissile fuel and tracer materials, such as U isotopes, Pu isotopes, Am, and ^{244}Cm; 2) Fission products such as ^{137}Cs and ^{90}Sr, and other products of fuel burn; 3) tritium (^{3}H); 4) Activities induced by neutrons in device parts, in external hardware, and in the surrounding geologic medium (such as ^{14}C, ^{36}Cl, and ^{41}Ca). Not all of the radionuclides produced during a nuclear test are worth including in the source term inventory. Many of the nuclides have half-lives so short (microseconds to hours) that they decay to undetectable levels soon after the event. Other nuclides are produced in such low initial abundance that they never exceed levels deemed unsafe or non-permissible by regulatory agencies. Criteria were developed to exclude such unimportant nuclides from the inventory; this permits the user to focus attention on the nuclides of interest from a risk assessment point of view. Excluded were nuclides produced in such low amounts that if *all* of the amount produced during a nuclear test were dissolved into a volume of water equal to the volume of the detonation cavity for the event, 100 years from now the resulting aqueous concentration (activity) in μCi/mL (one μCi = 2.22×10^6 disintegrations per minute) would be less than one-tenth of the values for the maximum permissible concentrations (MPC's) proposed for drinking water by the Environmental Protection Agency in the Federal Register (1991). This effectively excludes almost all radionuclides with half-lives less than ten years. However, if a radionuclide exceeded these criteria for at least one event it was included for all other events for which estimates are available even if concentrations were below the 0.1 MPC criterion. The MPC of a nuclide listed in this compilation is that concentration in drinking water that will impart a dose of 4 mrem/year to a person drinking an average of 2 liters of water per day. The requirement of 100

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radionuclide amounts (reported in curies) for 57 isotopes meeting the above criteria for inclusion. Because the databases compile event-specific nuclear performance data including yield, residual nuclear fuels and thermonuclear and fission products, by necessity they are classified as Secret-Restricted Data. The databases are accessible to investigators holding an active U.S. Department of Energy Q security clearance with a valid "need-to-know."

For the purposes of summary and publication, LANL and LLNL arbitrarily subdivided the inventory into five regions corresponding to geographic test centers at the Nevada Test Site. In addition, the inventory is further subdivided by tests fired above or below the static water level. If the working point depth was below or within one cavity radius of the static water level (SWL), the test was considered to have been below the water-level. Generally, testing areas within a region have similar geologic and hydrologic properties.

TABLE II

Combination of NTS Areas into Regions

NTS region	NTS areas included
Yucca Flat	1, 2, 3, 4, 6, 7, 8, 9, 10, 15, 17
Pahute Mesa - 19	19
Pahute Mesa - 20	20
Frenchman Flat	5, 11*
Rainier Mesa	12, 16, 18, 29, 30

*Frenchman Flat also incorporates a small number of safety shots fired at NTS

The results of this compilation are presented in Tables VII and VIII, which list the aggregate activities in curies of those radionuclides that meet the selection criteria. The activities are specified at two dates (January 1, 1994, and January 1, 2094) and classified as above or below the static water level (water table) for each of the five NTS regions outlined in Table II. These tables represent the long-lived radionuclide source term for all tests conducted underground at the Nevada Test Site by the Los Alamos National Laboratory, Lawrence Livermore National Laboratory and the Defense Nuclear Agency between 1955 and 1992.

SOURCES OF RADIONUCLIDE DATA

Values for the total inventory of radionuclides were determined in two principal fashions for this compilation: measurements and calculations. Many nuclides have historically been measured from small samples recovered from the underground environment as part of the radiochemical diagnostics effort. These have been those nuclides most related to the diagnostic purposes: fission products, residual fuel species, and radiochemical detector and tracer isotopes. These measurements have also served to establish an understanding of the processes occurring in the underground environment, often reduced to computer codes used in the prediction of the production of nuclides. These codes can then be used to stand in where measurements were not made at the time of the test or where interpretation of the data for total inventory is difficult.

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METHODS OF ESTIMATING POSTSHOT RADIONUCLIDE CONTENT

Postshot radionuclides present underground at the Nevada Test Site can be conveniently grouped into four different categories: fission products, actinide elements, tritium, and activation products/natural materials. Each of these groups requires different methodologies for estimating the postshot quantities of the nuclides. In addition, historical differences in the methods employed by Los Alamos and Livermore in their radiochemical diagnostics efforts lead to slightly different techniques. The following sections describe the methods used in this study.

FISSION PRODUCTS

Most of the fission products of interest to the Underground Test Area Remedial Investigation and Feasibility Study are formed in such low yield or have such long half-lives that they were not measured radiochemically following the tests. In addition, many of these isotopes experience chemical fractionation, or preferential loss from the sampled region, and were of little or no diagnostic value.

For a large number of tests, sufficient high-yield fission products were measured in order to determine the fission yield of the device. Other fission products were also measured to characterize the fission split, the distribution of fissions among the fissile materials and neutron energy. For the remaining tests, sufficient precedent or other diagnostic measurements were available to state the fission yield and split. Thus determined, the fission yields and splits have served as the basis for the calculation of the amounts of the various fission products given in this study.

We have chosen to calculate the postshot amounts from fission yields based on the type of fissile fuel and the neutron energy spectrum. In most cases, these characteristics were available in the documentation associated with each individual test. In other cases, we reasoned by analogy. The overall, effective yield of a fission product for each event is the weighted average of the yields from that event's mix of fissions from each fissile fuel and neutron energy spectrum.

For the majority of fission products, the fission yields of England and Rider (1993) were used by both LANL and LLNL. In a few cases, values accepted by the LANL and LLNL radiochemistry groups were used (see, for example, Nethaway, 1985). Differences among these various sources reflect the different philosophies and best experience of the efforts of the two organizations; in the worst case, differences are less than 8%.

ACTINIDES

Los Alamos' Chemical Science and Technology Division and Livermore's Nuclear Chemistry Division have chosen consistent paths in the manner in which they have treated their actinide data. This is in part driven by the available data and in part by the technical biases that developed in their individual histories of weapons radiochemical diagnostics.

For many isotopes, we based our determination of the residual amounts of radionuclides from the measurements on actual core samples. These measurements were made historically in the course of the underground test program. In most cases, the postshot amount of a particular isotope or suite of isotopes were known, based on device performance. This permitted the determination of the fraction of the device present in each particular sample. Through ratio measurements to other species, the absolute abundances of the other isotopes could be calculated.

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Other isotopes were treated differently. This approach was used when measurements were not available or were compromised by underground fractionation phenomena. Based on knowledge of the device, its performance, and production/destruction characteristics, we calculated the amounts of postshot isotope, using algorithms implemented in the database structure. Some approximations were very simple: external tracer isotopes were treated as unchanged from the ingoing amount loaded with the device. Other calculations derived from many years of observations combined with explosion code information.

TRITIUM

On most U.S. nuclear tests, up to times as long as 100 to 200 years following the detonation, tritium has by far the highest activity of any radionuclide with a half-life longer than ten years. Because of this we have expended considerable effort in estimating the residual tritium on the individual nuclear tests. We have also sought to achieve agreement between Los Alamos and Livermore for this isotope. Because of their earlier start to the underground inventory effort, LLNL has provided most of the algorithms for tritium production and destruction; LANL has served to confirm these methods through independent explosion code calculations.

The amount of residual tritium from a nuclear event is the net result of the amount loaded on the device and the amounts produced and destroyed in the nuclear explosion. A wide variety of sources and reactions are involved in this production/destruction. Each unique material and device type was examined to provide methods consistent with the limited suite of tritium measurements and the results of explosion code calculations. Sources both internal and external to the device itself were considered.

ACTIVATION PRODUCTS AND NATURAL ISOTOPES

Reactions of neutrons with parts of the device other than the fuel, such as the structural materials, the stemming, and the geologic medium, produce several nuclides with half-lives longer than 10 years. There are about 50 such nuclides, but many of them can be excluded using the source term inventory criteria, because of extremely long half-life of the nuclide or very small production rate. Several nuclides do meet the selection criteria, however. The contributions of these nuclides to the source term were estimated using available data on other slow and fast neutron activation products, relevant cross section information, and neutron transport calculations.

For most of the nuclides of interest, activation of the geologic medium by reactions of slow neutrons is an important production mechanism. To estimate this, we made use of extensive data at both laboratories on the production of ^{160}Tb by ^{159}Tb(n,γ). Both the Nuclear Chemistry Division at LLNL and the Chemical Science and Technology Division at LANL have measured ^{160}Tb in debris samples from a large number of underground nuclear tests as an indicator of neutron activation of soil constituents. These measurements were used to correct other observations for such soil contributions. Empirical relationships were independently derived by the laboratories to estimate the ^{160}Tb production in cases where measurements were not made.

The amounts of various other nuclides produced by the slow neutron reactions in the soil relative to ^{160}Tb were estimated using relationships derived at LLNL using neutron transport calculations. Monte Carlo neutron transport calculations were performed using the MCNP code (Los Alamos National Laboratory, XXXX); in addition, the Monte Carlo transport calculations by Lessler and Guy (1965) were also useful. The MCNP calculations were done for different water contents of the soil in the range of 0 to 13 wt% and for soil temperatures of 0 and 100 eV. The production rates of the different isotopes relative to ^{160}Tb showed up to a factor of two variation over this

TABLE V

Estimated Accuracies for Individual Nuclides in
the Various Groups of Radionuclides in this Inventory

Fission products:	~ 10 to 30% for most important fission products
Unspent fuel materials:	~ 20% or better
Fuel activation products	~ 50% or better
Residual tritium	~ 1% to a factor of 3
Activation products	~ a factor of 10

Fission Products

For reported amounts of fission products, accuracy depends on the accuracy of the following for that event: 1) the fission yield (kilotons); 2) the fission split, and 3) the yields of the fission product (atoms per fission) from the various fissions for the event. Not counting some of the events with very low yields (<0.1 ton), the fission yields of almost all the events are known to within 5 to 30%. For those events with radiochemical measurements, the fission splits among the fissionable nuclides are usually known accurately. The accuracies for the fission product yields we quote here are from England and Rider (1993). The least accurately known yields are those for the fission products ^{94}Nb, ^{113m}Cd, ^{121m}Sn, ^{152}Eu, and ^{166m}Ho. Their yields could be in error by a factor of 2 to 100 or more. Fortunately, these are also the fission products with the lowest yields, so they are very minor contributors to the radionuclide inventory. Furthermore, the total amounts of ^{94}Nb, ^{152}Eu, and ^{166m}Ho produced as activation products are at least 1000 times larger than the amounts from fission. The accuracies of the ^{126}Sn yields are 4 to 16%. The accuracies of the yields of all the other fission products in our inventory are 0.4 to 11%. For events on which no radiochemical measurements were made, uncertainty for the fission products can exceed another factor of 2 unless the event was known to be similar in design and performance to one measured.

Actinides

Most of the tabulated amounts of unspent fission fuel and tracer materials are accurate to 1 to 10%. A few, including some of the values for events for which the device performance is known only approximately, have uncertainties of up to 20%.

The tabulated amounts of minor isotopes and activation products of fission fuel, when derived from actual measurements, are accurate to 20% or better. However, when the amounts are estimates from approximate device performance information and estimated preshot compositions, the uncertainties for some of the isotopes may rise to 50%. The total ^{232}U inventory is good only to a factory of 2 or 3 because only about 15% of it is based on actual measurements. Uranium-237 data for some of the order events is missing and still needs to be found or replaced by estimates. Because of this, our ^{237}Np inventory is probably low by a few tens of percent.

Tritium

The estimates of residual tritium have a wide range of uncertainties: from 1 - 2% to a factor of 3. The 1 or 2% applies to those events on which there was no thermonuclear yield and negligible tritium production, in which case the postshot amount of tritium equals the preshot amount. The

factor of 3 applies to devices with unusual designs and events for which performance is only known approximately.

Activation Products

Our activation product estimates are likely accurate only to a factor of 10 in most instances. This relatively large error is the result of the approximate nature of the methods we used to obtain the estimates described above, such as using data for nuclides from similar reactions and approximate Monte Carlo neutron transport models employing average elemental abundances for soil activation. The part of the europium isotope inventory that is based on direct measurements is more accurate, with uncertainties of 10 to 30%.

EXPLANATION OF RADIONUCLIDE INVENTORY TABLES

Table VII contains the summary of the radionuclide totals, broken down by area above and below the water table, as they existed on January 1, 1994. Radionuclides are arranged within each table according to their atomic number and atomic mass; amounts are listed in curies.

Table VIII contains the summary of the radionuclide totals, broken down by area above and below the water table, as it would exist on January 1, 2094.

DISCUSSION

The total underground radionuclide inventory for the Nevada Test Site provides an accurate measure of residual radioactive products present after nearly four decades of nuclear testing. The source term inventory is valuable for three reasons. First, this compilation provides a quantified and accurate radionuclide source term; in contrast, many previous compilations were qualitative only. Second, the NTS radionuclide inventory integrates in excess of 800 single radionuclide sources. Third, the inventory incorporates a significant proportion of radionuclides introduced from natural sources in addition to those expected from anthropogenic sources.

As mentioned earlier, radionuclide totals for the NTS represent an upper limit of radionuclides potentially available for transport. The radionuclide source term will never be transported in entirety; the hydrologic source term comprises those species that are dissolved in or transportable by groundwater. The mobility of radionuclides is moderated both by chemical kinetics and hydrology. Numerous experimental and field studies accompanying the nuclear test program (see Borg, 1976 and Smith, 1993) indicate, in general, long-lived radionuclides (actinides, fission products) are relatively insoluble in NTS groundwaters and are attenuated during transport by ion-exchange and surface reactions with sorptive minerals -- particularly zeolites -- characteristic of the NTS volcanic stratigraphy. With the exception of tritium, which is efficiently dispersed in groundwater as molecular HTO, the hydrologic source term will always be less than the radionuclide source term. Approximately two thirds of the devices fired at the NTS were detonated above the static water level and by probably will not substantively contribute to the hydrologic source term. Finally, the hydrology of the Nevada Test Site consists of a complex and variably transmissive stratigraphy of regional Paleozoic carbonate (transmissivities from 7 to > 10,000 m^2/day) and overlying Tertiary volcanic (transmissivities of 2.5 to 1,200 m^2/day) aquifers divided by prevailing flow directions and by discharge areas into three groundwater subbasins. While a comprehensive description of radionuclide leaching and sorption and Nevada Test Site hydrology is outside the scope of this report, radionuclide transport will not occur uniformly or pervasively

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away from the testing centers at the Nevada Test Site. Different geology and hydrologic properties will greatly affect potential radionuclide transport.

The source term integrates over 800 underground nuclear tests and provides radionuclide totals for five test areas both above and below the water table. By summing radionuclide totals for individual tests into regional areas, radionuclide production from an individual test is integrated into the sum for that region. The effect, particularly for tests with higher yield, is to minimize the contribution from any one event. Because the radionuclide source term is device dependent, local contributions will be disproportionately skewed toward the regional average. Geographic divisions were made arbitrarily and should not be treated as mutually exclusive; ideally, each test should be considered individually relative to both yield and specific radionuclide production.

The radionuclide inventory is dominated by residual radioactivity introduced by nuclear testing, but also includes a significant proportion of natural radioactivity. Volcanic tuffs and rhyolites erupted from volcanic centers over the past 15 billion years are highly evolved rocks that partition oxides of uranium, thorium and potassium -- all of which are naturally radioactive. Past undocumented studies on core samples from underground nuclear tests indicate that 700 tons of geologic media are melted per kiloton of nuclear explosive yield. (We recently confirmed this value by examining the radiochemical diagnostic data for the most device-debris-rich core sample from each of 18 LLNL tests between 1970 and 1988.) Therefore, natural ^{40}K, natural ^{232}Th and natural uranium activities are mixed into the melt created by all the events in each of the five inventory regions. The average potassium, thorium and uranium concentrations of NTS working point media are 4%, 22 ppm and 3.7 ppm by weight. Using these values, we calculate that 0.0229 Ci of ^{40}K, 0.00169 Ci of ^{232}Th, and 0.00177 Ci of uranium are incorporated into the melt per kiloton of yield. We include this contribution separately in our totals for each for the five test regions defined for the inventory. Background radioactivity from naturally occurring radionuclides constitutes an effective lower limit for monitoring or remedial levels proposed for the Nevada Test Site.

FUTURE WORK

It should be recognized that the databases represented by this work are evolving entities, subject to improvements and corrections as more sophisticated interpretations evolve and additional information is gleaned from the historic record. We expect, that with continued support, that the values of the underground nuclide inventory will improve. Large scale changes in the sums, by area or above or below the water table, are not expected.

Los Alamos and Livermore expect to maintain actively their respective databases, commensurate with the resources available. Routine communication of updates to each database will occur, ensuring that the most comprehensive and current values are available at both locations. Improvements to the database structure and implementation are envisioned to facilitate greater utility and quality assurance.

Eleven underground nuclear tests sponsored by the laboratories occurred outside the boundaries of the Nevada Test Site, within the boundaries of the United States. During fiscal year 1995, these tests will be inventoried in a fashion similar to that described here. This data will become available as a separate report.

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