

**Environmental Protection Department
Operations and Regulatory Affairs Division**

**LLNL NESHAPs
2001 Annual Report**



Lawrence Livermore National Laboratory
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**U.S. Department of Energy
Radionuclide Air Emission Annual Report
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Lawrence Livermore National Laboratory NESHAPs 2001 Annual Report

This annual report is prepared pursuant to the National Emission Standards for Hazardous Air Pollutants (NESHAPs; Title 40 Code of Federal Regulations [CFR] Part 61, Subpart H). Subpart H governs radionuclide emissions to air from Department of Energy (DOE) facilities.

SYNOPSIS

NESHAPs limits the emission of radionuclides to the ambient air from DOE facilities to levels resulting in an annual effective dose equivalent (EDE) of 10 mrem (100 μ Sv) to any member of the public. The EDEs for the Lawrence Livermore National Laboratory (LLNL) site-wide maximally exposed members of the public from operations in 2001 are summarized here.

- Livermore site: 0.017 mrem (0.17 μ Sv) (34% from point-source emissions, 66% from diffuse-source emissions). The point-source emissions include gaseous tritium modeled as tritiated water vapor as directed by EPA Region IX; the resulting dose is used for compliance purposes.
- Site 300: 0.054 mrem (0.54 μ Sv) (93% from point-source emissions, 7% from diffuse-source emissions).

The EDEs were calculated using the EPA-approved CAP88-PC air dispersion/dose-assessment model, except for doses for three diffuse sources, which were calculated from measured concentrations and dose coefficients. Site specific meteorological data, stack flow data, and emissions estimates based on radionuclide usage inventory data or continuous stack monitoring data were the specific inputs to CAP88-PC for each modeled source.

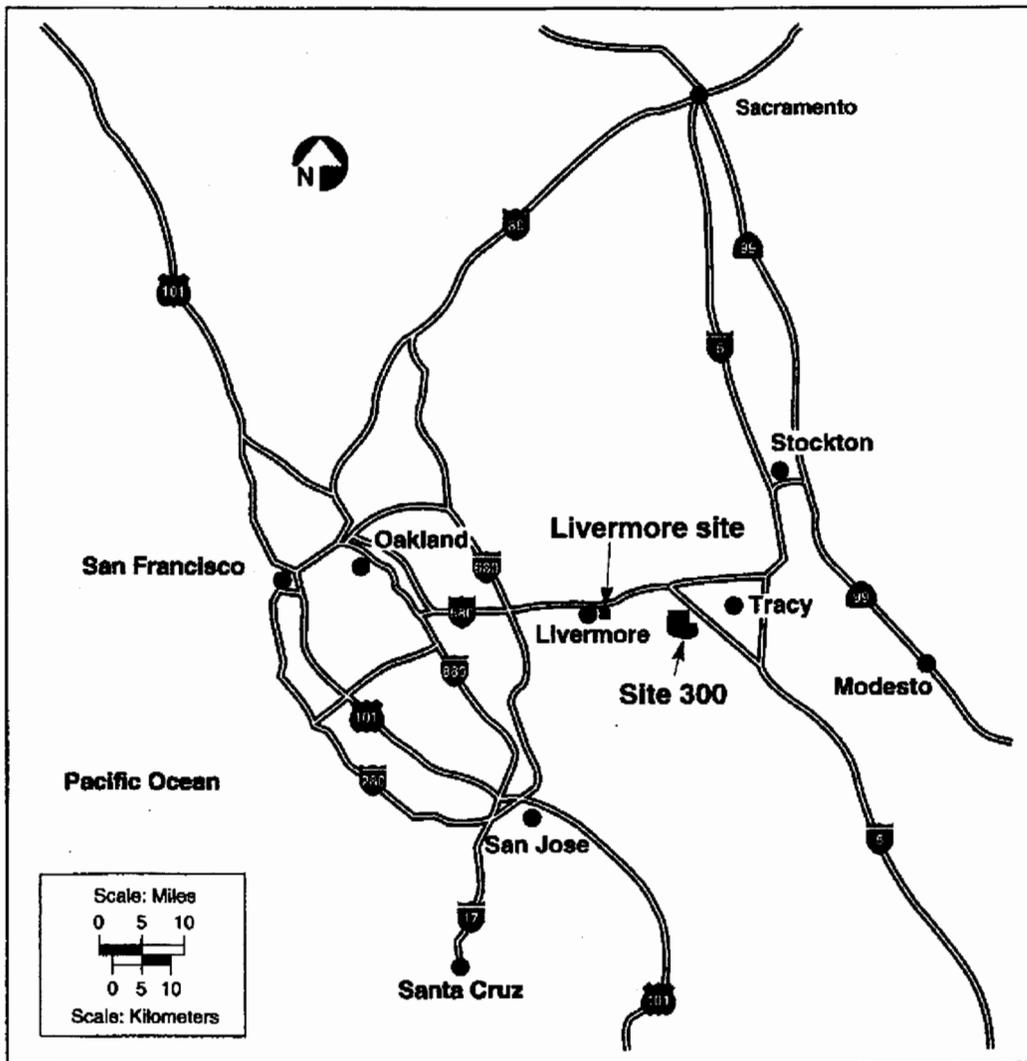


Figure 1. Locations of LLNL Livermore site and Site 300.

Site 300

Site 300, LLNL's Experimental Test Facility, is located 24 km east of the Livermore site in the Altamont Hills of the Diablo Range and occupies an area of 30.3 km². It is close to another explosives-testing facility owned and operated by SRI International. A State of California vehicular-recreation area is located nearby, and wind-turbine generators line the surrounding hills. The remainder of the surrounding area is in agricultural use, primarily pasture land for cattle and sheep. The nearest residential

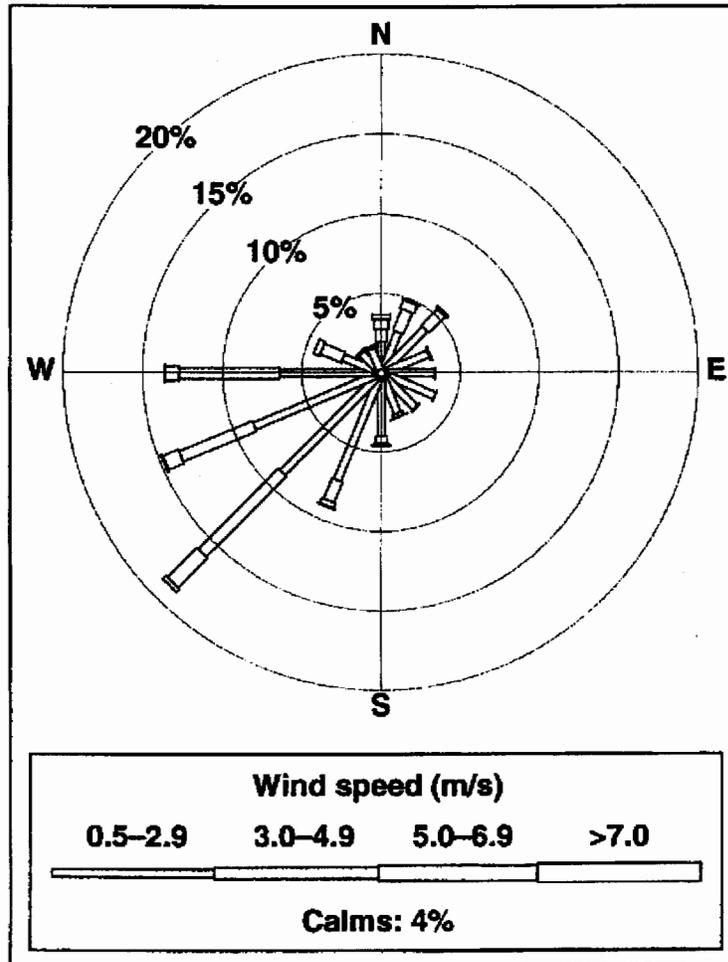


Figure 2. Wind rose showing the average annual wind speed, frequency of occurrence, and direction at the Livermore site, 2001.

area is the city of Tracy (population approximately 61,200), located 10 km to the northeast.

The topography of Site 300 is much more irregular than that of the Livermore site; it consists of a series of steep hills and ridges, which are oriented along a generally northwest/southeast trend, separated by intervening ravines. The elevation ranges from approximately 540 m in the northwestern portion of the site to 150 m at the southeast corner. The climate at Site 300 is similar to that of the Livermore site, with mild winters and dry summers. The complex topography of the site significantly influences local wind and temperature patterns, making the temperature range

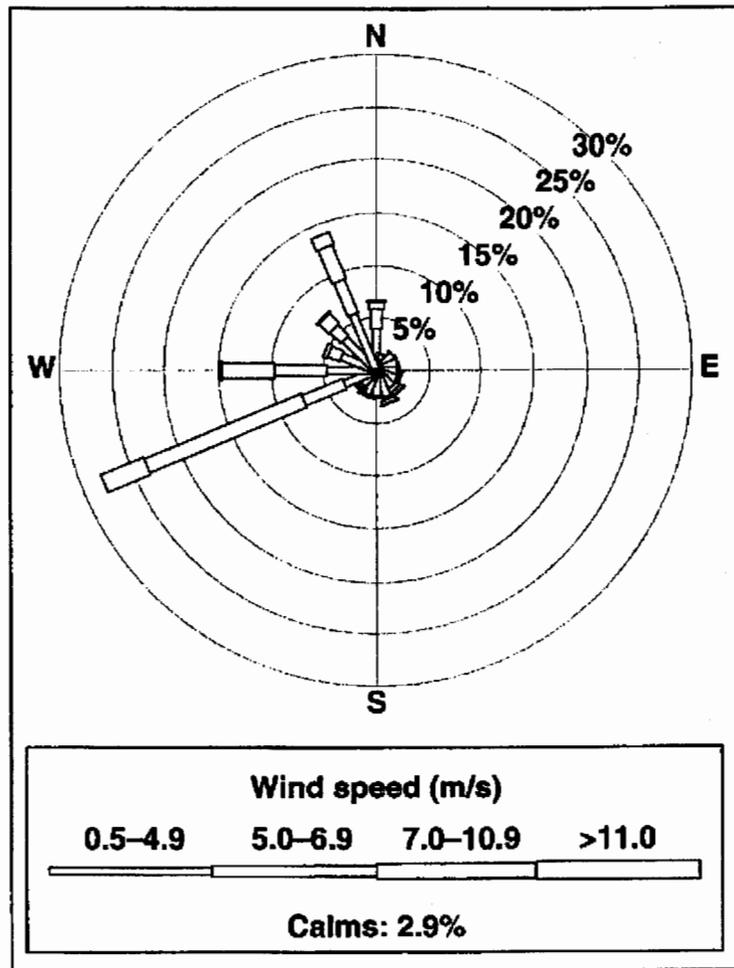


Figure 3. Wind rose showing the average annual wind speed, frequency of occurrence, and direction at Site 300, 2001.

somewhat more extreme than at the Livermore site. The 2001 annual wind data for Site 300 are displayed as a wind rose in Figure 3. Prevailing winds are from the west-southwest. As is the case at the Livermore site, precipitation is highly seasonal, with most precipitation occurring between October and April. Site 300 received 247 mm of precipitation during 2001. The mean annual temperature is about 17°C.

time tritium concentration release levels (HT, HTO, or other gaseous forms). The sieve samplers discriminate between tritiated water (HTO) vapor and molecular tritium (HT); they provide the values used for environmental reporting and are exchanged weekly. Each sieve sampler (not alarmed) is in parallel with an alarmed monitor and consists of two molecular sieves. The first sieve collects tritiated water vapor; the second sieve contains a palladium-coated catalyst that converts molecular tritium to tritiated water, which is then collected. The molecular sieve samples are submitted to the Hazards Control Analytical Laboratory where they are put into a recovery system for the bake out of tritiated water vapor and subsequent condensation and collection of the water. The retrieved tritiated water is analyzed by RML using liquid scintillation counting techniques.

Data from air particulate sampling filter and molecular sieve analyses are reviewed by Hazards Control Department Health Physicists responsible for each facility and an Environmental Protection Department Environmental Analyst.

Results of Stack Monitoring for Tritium: Operations in the Tritium Facility (Building 331) in 2001 released a total of 20 Ci (7.4×10^{11} Bq) of tritium. Of this, approximately 18.3 Ci (6.8×10^{11} Bq) were released as tritiated water (HTO). The remaining 8.5% of the tritium released, 1.7 Ci (6.4×10^{10} Bq), was elemental tritium gas (HT). The highest single weekly stack emission from the facility was 0.67 Ci (2.5×10^{10} Bq), of which 0.64 Ci (2.4×10^{10} Bq) was HTO.

Building 331 tritium emissions, as measured by stack monitoring, remained considerably lower in 2001 than emissions that occurred during the 1980s. The reduced emissions in 2001 were primarily the result of a reduction in programmatic work compared to the previous years. Over the next five years, an increasing trend in emissions may occur as research and development work is performed for new programmatic efforts. However, engineered controls designed to contain and recapture tritium leakage should maintain relatively low emissions. Figure 4 illustrates the combined HTO and HT emissions from the facility since 1981.

Stack Monitoring for Gross Alpha and Gross Beta Radiation: For most discharge points at the other facilities where continuous stack sampling is performed, the results are below the minimum detectable concentration (MDC) of the analysis; sometimes as few as 1 to 4 samples (out of 25 to 50 per year) have concentrations greater than the MDC. Generally, these few samples having results above the MDC are only marginally above the MDC. Use of zero values for this type of data can be justified based on knowledge of the facility, the use of tested, multiple stage, HEPA filters in all significant release pathways, and alpha spectroscopy based isotopic analyses of selected air sampling filters. These isotopic analyses demonstrate that detected activity on air sampling filters comes from

naturally occurring radionuclides, such as radon daughters, e.g., polonium, on the air sampling filters. In addition, because of exhaust configurations at some facilities, the monitoring systems sometimes sample air from the ambient atmosphere along with the HEPA filtered air from facility operations, giving rise to background atmospheric radioactivity being collected. Because of these considerations, the emissions from such facility operations are reported as zero. Consequently, there are no dose consequences, and doses reported for these operations are also zero. Furthermore, even if the MDC values are used in calculations of the emission estimates for these facilities, which would be an extremely conservative approach, the total dose attributable to LLNL activities is not significantly affected. None of the facilities monitored for gross alpha and beta had emissions in 2001.

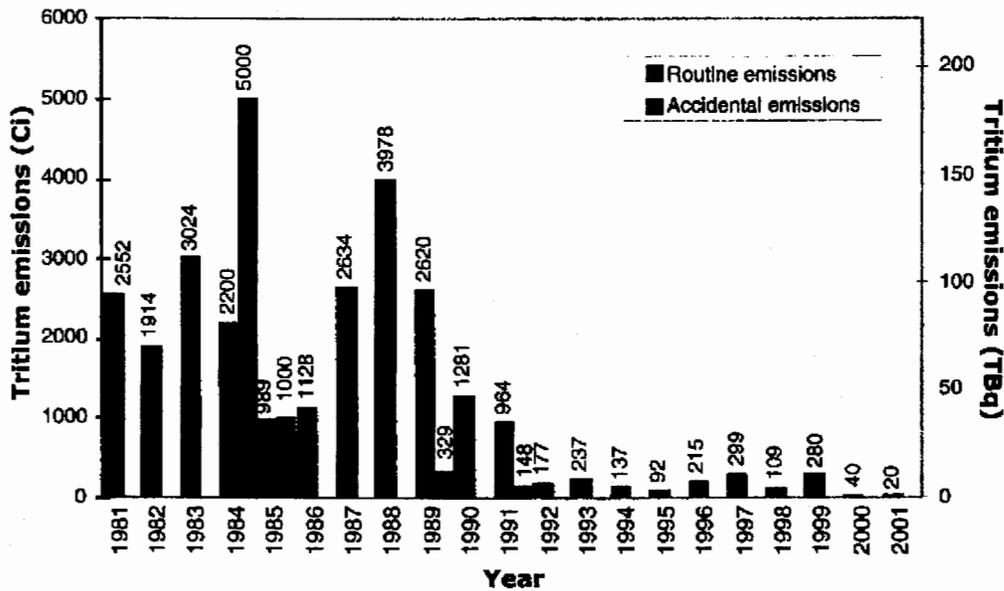


Figure 4. Combined HT and HTO emissions from the Tritium Facility, 1981–2001, distinguishing between chronic releases during normal operations (black bars) and acute accidental releases (gray bars). Accidental releases are predominantly HT gas.

Air Surveillance Monitoring for Radioactive Particles and Gases

Surveillance air monitoring for tritium and radioactive particles has been in place since the 1970s and will continue. LLNL currently maintains seven continuously operating, high volume, air particulate samplers on the Livermore site, nine in the Livermore Valley, eight at Site 300, and one in Tracy. LLNL also maintains twelve continuously operating tritiated water vapor samplers on the Livermore site, six

samplers in the Livermore Valley and one at Site 300. The samplers are positioned to ensure reasonable probability that any significant airborne concentration of particulate or tritiated water vapor effluents resulting from LLNL operations will be detected. Many of the surveillance air monitors are placed near diffuse emission sources, such as those near Buildings 292, 331, 514, and 612, as well as in and around the Southeast Quadrant of the Livermore site. As such, their results can be used to estimate and/or confirm the emissions from the associated diffuse sources. Also included is an air particulate monitor positioned at the location of the hypothetical maximally-exposed member of the public (defined in Section III) for the Livermore site. Data from air surveillance monitors provide a valuable test of predictions based on air dispersion modeling, and can help characterize unplanned releases of radioactive material.

The data from the ambient air monitoring network provide continuous measurements of the concentrations of radionuclides present in the air at the Livermore site, Site 300, and in the surrounding areas. Data from the network are presented in the LLNL Site Annual Environmental Report, which is available to the public in bound hardcopy form and on the Internet. (See, e.g., Biermann et al., *Environmental Report 2000*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-50027-00, September 2001; <http://www.llnl.gov/saer>).

Recognition of Need to Apply Correction Factors to Results of Tritium Surveillance Air Monitoring: Recently it was shown that measured tritium concentrations obtained using a method involving the extraction of water from silica gel—a method used at LLNL since 1973—are in error and require upward correction. It is important to note that this correction, while affecting the concentrations of tritium in ambient air quoted in LLNL's environmental reports, does not significantly change the doses to the public quoted in those reports. Only for the special case of a diffuse tritium source having emissions inferred from monitoring data does the correction change the inferred dose for that particular source. Doses to the public attributed to tritium emissions from the pair of 30-meter-high stacks of the Tritium Facility, in particular, are not affected.

The Environmental Monitoring Radiological Laboratory of the Analytical and Nuclear Chemistry Division at LLNL developed a correction factor that applies to all measured tritium concentrations obtained by this method (Guthrie, E.B., et al., "Isotope exchange and fractionation corrections for extraction of tritiated water in silica gel by freeze-drying techniques," LLNL draft report, Sept. 2001). The correction factor was developed based on new understanding of the properties of silica gel (Rosson, R., et al., "Isotopic exchange and the vapor pressure isotope effect in tritium oxide adsorption on silica gel," *J. Phys. Chem.* B102:10342-10346, 1998; Rosson, R., et al., (2000), "Correcting tritium concentrations in water vapor monitored with silica gel," *Health Physics* 78(1):68-73). Put simply, the

radionuclide usage data for all Site 300 explosives experiments and all significant stack and diffuse sources at both sites were included in this update.

SECTION III. Dose Assessment Methods & Concepts

Description of the Air Dispersion and Dose Model

Most estimates of individual and collective radiological doses to the public from LLNL operations were obtained using the EPA-developed computer code CAP88-PC. An LLNL-modified version of this code (designated CAP88-PC-T), which contains an improved tritium model (not yet approved by EPA for use in regulatory compliance evaluations), was also used for purposes of comparison. The four principal pathways—internal exposures from inhalation of air, ingestion of foodstuff and drinking water, external exposures through irradiation from contaminated ground, and immersion in contaminated air—are evaluated by CAP88-PC. The doses are expressed as whole-body effective dose equivalents (EDEs), in units of mrem/y (1 mrem = 10 μ Sv). Separate doses for Livermore site and Site 300 emissions are reported.

Three potential doses are emphasized: (1) The dose to the site-wide maximally exposed individual (SW-MEI), which combines the effects of all emission points, for comparison to the 10 mrem/y (100 μ Sv/y) standard; (2) the maximum dose to any member of the public (assumed to be at the LLNL fence line), in any direction, due to each unabated emission point on the site to determine the need for continuous monitoring; and (3) the collective dose to populations residing within 80 km of the two LLNL sites, adding the products of individual doses received times the number of people receiving them.

Summary of Model Input Parameters

General Model Inputs: Attachment 1 details the key identifiers and input parameters for the CAP88-PC model runs. These include building number; stack ID; isotope(s); emission rate in curies per year (1 Ci = 3.7×10^{10} Bq); and stack parameters, including height, diameter, and emission velocity.

Meteorological Data: All model runs used actual 2001 Livermore-site and Site 300 meteorological data, collected from the meteorological towers for each site. At these towers, wind speed and direction are sampled every few seconds, temperature sampled every minute, and all are averaged into quarter-hour increments, time tagged, and computer recorded. The data are converted into a CAP88-PC input wind file using EPA guidelines.

Surrogate Radionuclides: CAP88-PC contains a library of 265 radionuclides; however, it does not contain all the radionuclides in use at LLNL. As a consequence, it was necessary in a few cases to use surrogate radionuclides to estimate EDEs.

liquid, powder, or gas) are those stated in 40 CFR Part 61, Appendix D. If the material was an unconfined gas, then the factor 1.0 was used; for liquids and powders, 1.0×10^{-3} was used; and for solids, 1.0×10^{-6} was used. The U.S. EPA has granted approval for LLNL to use alternative physical state factors for elemental uranium, uranium/niobium alloy, and elemental plutonium. Table 2 provides the approved temperatures for application of the physical state factor for each material.

These factors are allowed provided that the material is not intentionally dispersed to the environment and that the processes do not alter its chemical form. The physical state dependent release fraction and the time factor are used to adjust (by multiplication) the total annual usage inventory to yield the potential annual release to air. In addition, emission control abatement factors (40 CFR 61, Appendix D), when applicable, were applied. Each HEPA filter stage was given a 0.01 abatement factor. (However, abatement factors were not used to evaluate compliance with the 0.1 mrem [1 μ Sv] standard that determines the need for continuous monitoring at a facility.) The use of actual stack effluent sampling data is much more direct, and presumably more accurate, than using assumptions based on usage inventory, time factors, release fractions, and emission control factors.

Table 2. List of materials exempted from the "treat as a gas above 100°C rule," and temperatures at which the various physical state factors apply.

Material	Solid physical state factor	Liquid physical state factor	Gas Physical state factor	Year Approved
Elemental uranium	<1100°C	Between 1100°C and 3000°C	>3000°C	1996
Uranium/niobium alloy	>3000°C	<1000°C	Between 1100°C and 3000°C	2001
Elemental plutonium	<600°	Between 600°C and 3000°C	>3000°C	2001

Site-Wide Maximally Exposed Individual: For LLNL to comply with the NESHAPs regulations, the LLNL site-wide maximally exposed individual cannot receive an EDE greater than 10 mrem/y (100 μ Sv/y). The site-wide maximally exposed individual (SW-MEI) is defined as the *hypothetical* member of the public at a single residence, school, business, or office who receives the greatest LLNL induced EDE from the combination of all radionuclide source emissions, as determined by modeling.

At the Livermore site, the SW-MEI for 2001 was found, as usual, to be located at the UNCLE Credit Union, about 10 m outside the controlled eastern fence line of the site, but about 10 m within the perimeter of the site property, as shown in Figure 5. At Site 300, the 2001 SW-MEI was again, as in the previous year, located at the boundary with the Carnegie State Vehicle Recreation Area, managed by the

Special Modeling Challenges

Among the sources at LLNL, explosives tests using depleted uranium at Site 300 and diffuse sources at both sites required special attention.

Site 300 Explosives Experiments: Some of the assemblies for Site 300 explosives experiments contain depleted uranium (DU) and possibly other radioactive materials. (The radioactive material does not contribute to the explosive energy, which is entirely chemical in origin.) The explosives assemblies are placed on an open-air firing table and detonated. Only limited data are available to characterize the initial state of the cloud of explosive decomposition products created by the detonation because properties of the cloud are not routinely measured in the experiments. Empirical scaling laws can be used, however, to define the size and height of the cloud using explosives inventories. When the assembly contains DU, the three uranium isotopes with atomic weights 238, 235, and 234 are assumed to occur in the cloud in the weight percentages 99.8, 0.2, and 5×10^{-4} . Their masses are multiplied by their specific activities to determine the total activity for each isotope in the cloud. For simplicity, it is assumed that all the uranium is dispersed as a gaseous cloud, and that the median particle size is the CAP88-PC default value of 1 μm . The assumption that all uranium is aerosolized and dispersed as a cloud results in a highly conservative off-site dose estimation—we believe a more realistic release-to-air fraction for the uranium is no greater than 0.2, but we lack sufficient data to use a value other than 1.0. CAP88-PC simulates each shot as a low level, steady state, stack-type emission occurring over one year. An alternative modeling methodology for treating these short duration explosive events, based on a “puff” code, was submitted to EPA for approval in 1992, but LLNL was directed to use the CAP88-PC code for these calculations.

Diffuse Sources: Diffuse emissions generally arise from extended-area sources external to buildings. Such sources are difficult to quantify. At present there are no EPA-mandated methods for estimation or measurement of diffuse sources; dose calculations associated with this type of source are left to the discretion of the DOE facility. Dose assessments for Livermore-site and Site 300 diffuse sources are variously derived based on radionuclide usage inventory data, environmental surveillance monitoring data, samples of contaminated materials, and other methods. The doses from principal diffuse sources in 2001 are described below in Section VIII.

Modeling Documentation

Dose assessment modeling runs were conducted for all sources (point and diffuse) meeting the criteria of the reduced accounting for 2001. The model used was EPA’s CAP88-PC code (see Section III). Files were incorporated for meteorological data (wind, precipitation, and temperature) and population data representing both sites,

along with the 2001 radionuclide usage inventory or stack effluent monitoring data. Annual dose is reported as whole-body EDE expressed in units of mrem (followed by μSv ; 1 mrem = 10 μSv). Copies of individual model runs, including input parameters and resultant calculated doses, are on file with the Terrestrial & Atmospheric Monitoring & Modeling Group (TAMM) of the Environmental Protection Department at LLNL.

SECTION IV. Results of 2001 Radiological Dose Assessment

This section summarizes the doses to the most exposed public individuals from LLNL operations in 2001, shows the temporal trends and comparison to previous years, presents the potential doses to the populations residing within 80 km of either the Livermore site or Site 300, and summarizes LLNL's compliance with 40 CFR 61, Subpart H (61.93).

Total Dose to Site-Wide Maximally Exposed Individuals

For the Livermore site, the dose calculated for the SW-MEI from diffuse emissions in 2001 totaled 0.011 mrem (0.11 μSv). The dose due to point sources was 0.0056 mrem (0.056 μSv). When combined, the total annual dose was 0.017 mrem (0.17 μSv), 66% from diffuse and 34% from point sources. The point source dose includes Tritium Facility HT emissions modeled as HTO, as directed by EPA Region IX. The SW-MEI dose calculated using NEWTRIT for tritium emissions from both point and diffuse sources at the Livermore site was 0.013 mrem (0.13 μSv).

The total dose to the Site 300 SW-MEI from operations in 2001 was 0.054 mrem (0.54 μSv). Point source emissions from firing table explosives experiments accounted for 0.050 mrem (0.50 μSv), or 93%, of this total, while 0.0037 mrem (0.037 μSv), or about 7%, was contributed by diffuse sources.

Table 3 shows the facilities or sources that accounted for more than 90% of the doses to the SW-MEI for the Livermore site and Site 300 in 2001. Although LLNL has nearly 200 sources with potential for releasing radioactive material to air according to NESHAPs prescriptions, most are very minor. Nearly the entire radiological dose to the public from LLNL operations comes from no more than a dozen sources.

A comparison of 2001 doses with those of previous years is provided by Table 4. No diffuse emissions were reported at Site 300 for years before 1993, so comparison of total Site 300 dose can only be made for 1993 and later. In addition, diffuse source doses were not reported separately from the total dose for the Livermore site for 1990 and 1991.

primarily because the stack releases from the Tritium Facility were unusually low in 2001. The corresponding collective EDE from Site 300 operations in 2001 was 9.4 person-rem (0.094 person-Sv). This value, while within the normal range seen from year to year, exceeds by almost four times the 2.5 person-rem (0.025 person-Sv) for 2000, as a result of increased firing table activity.

These population doses can be compared to the collective dose from natural background radioactivity for 6.9 million people of 2.1×10^6 person-rem (2.1×10^4 person-Sv).

Table 4. Doses (in mrem) calculated for the Site-Wide Maximally Exposed Individual (SW-MEI) for the Livermore site and Site 300, 1990 to 2001.

Year	Total Dose	Point Source Dose	Diffuse Source Dose
Livermore site			
2001	0.017 ^a	0.0057 ^a	0.011
2000	0.038 ^a	0.017 ^a	0.021
1999	0.12 ^a	0.094 ^a	0.028
1998	0.055 ^a	0.031 ^a	0.024
1997	0.097	0.078	0.019
1996	0.093	0.048	0.045
1995	0.041	0.019	0.022
1994	0.065	0.042	0.023
1993	0.066	0.040	0.026
1992	0.079	0.069	0.010
1991	0.234	— ^b	— ^b
1990	0.240	— ^b	— ^b
Site 300			
2001	0.054	0.050	0.0037
2000	0.019	0.015	0.0037
1999	0.035	0.034	0.0012
1998	0.024	0.019	0.005
1997	0.020	0.011	0.0088
1996	0.033	0.033	0.00045
1995	0.023	0.020	0.003
1994	0.081	0.049	0.032
1993	0.037	0.011	0.026
1992	0.021	0.021	— ^c
1991	0.044	0.044	— ^c
1990	0.057	0.057	— ^c

^a The dose includes HT emissions modeled as HTO as directed by EPA Region IX. EPA Region IX acknowledges that such modeling results in a conservative overestimation of the dose. This methodology is used for purposes of compliance.

^b Diffuse source doses were not reported separately from the total dose for the Livermore site for 1990 and 1991.

^c No diffuse emissions were reported at Site 300 for years before 1993.

Compliance with 40 CFR 61 Subpart H (61.93)

Calculations of effective dose equivalents for Livermore-site and Site 300 facilities having the potential to release radioactive material to the atmosphere were found to be well below the 10 mrem (100 μ Sv) NESHAPs dose standard for dose to the most-exposed individual members of the public. Tritium accounted for more than three-quarters of the Livermore-site calculated dose, while at Site 300 practically the entire calculated dose was due to the isotopes ^{238}U , ^{235}U , and ^{234}U , in depleted uranium.

In 2001, there were seven buildings (Buildings 175, 177, 235, 251, 331, 332, and 491) at the Livermore site that had radionuclide air effluent monitoring systems. These buildings are listed in Table 1, along with the number of samplers, the types of samplers, and the analytes of interest.

LLNL is committed to monitoring stack effluent air from its Tritium Facility (Building 331), Plutonium Facility (Building 332), and the seismically hardened area of its Heavy Element Facility (Building 251). In addition, other facilities are continuously monitored, as necessary, based on evaluations of potential emissions without control devices, as in the case of Building 235, or where classification or other issues prevent a usage-inventory-based evaluation.

SECTION VI. Supplemental Information on New Projects and Facilities

NESHAPs Responsibilities of LLNL Programs/Projects

Proposed facilities and significantly modified operations are assessed for NESHAPs requirements during the National Environmental Policy Act (NEPA) review process. Under NEPA, all proposed projects or actions that might involve NESHAPs issues or concerns—not just pertaining to radionuclides but to toxic air contaminants as well—are reviewed and evaluated. If the proposal includes operations that require a NESHAPs assessment, necessary modeling is conducted. If insufficient information is available for modeling at the time the NEPA documents are prepared, LLNL includes in the NEPA documents a statement that NESHAPs review, modeling, and monitoring requirements will be met. It is the responsibility of the individual project proponent to supply the specific information required for any NESHAPs modeling, analysis, and review that must be completed before operations described in the document are initiated.

Air quality compliance requirements are spelled out in the ES&H Manual, Vol. III, Part 31.1, "Air Quality Compliance." For example, Sec. 4.0 in Part 31.1 on "Radioactive Air Pollutants" states the responsibilities, directs the project to contact TAMM Group for guidance and assistance, and gives an eleven-step list under Sec. 4.3, "Process for Compliance."

Major New Facilities

Three new LLNL facilities are currently under development. All of these were assessed prior to construction for compliance with NESHAPs. Effluent sampling systems are planned for all three. These facilities are the Contained Firing Facility (CFF) at Site 300, and the Decontamination Waste Treatment Facility (DWTF) and the National Ignition Facility (NIF) at the Livermore site.

The CFF project allows containment of some explosives tests currently conducted outdoors at Site 300's Building 801. The CFF project consists of an enclosed firing chamber, a support facility, and a diagnostic equipment facility. Construction of CFF is complete, and the facility began operations with non-radiological materials in February 2002. Operations using experiments with depleted uranium commenced in March 2002. Temporary stack monitors for radioactive particulate emissions have been installed; permanent monitors should be put in place during 2002.

The DWTF is a waste handling facility that will have improved air emissions controls and will enable the handling of additional waste streams. Phase I construction (site preparation and installation of underground utilities) has been completed. Construction of the solid waste processing building, the storage building, and the

surveillance air monitor designated B624 has been placed in the Building 612 Yard to provide continuous measurements of tritium in air near this source. The median annual concentration of tritium in air for 2001 in this area was 49 pCi/m³ (1.8 Bq/m³). These data were used to calculate the total tritium emissions from the area, using a conservative approach that assumed the source to be 60 m south-southwest of the air sampler. With this assumption, a diffuse source emission of 2.0 Ci/y (7.4×10^{10} Bq/y) was required to produce the concentrations measured at the air sampler. This source term produced a calculated 2001 dose to the SW-MEI from the Building 612 Yard of 8.2×10^{-3} mrem (8.2×10^{-2} μ Sv) as calculated with CAP88-PC; a dose of 6.2×10^{-3} mrem (6.2×10^{-2} μ Sv) was calculated when the NEWTRIT model was implemented.

Southeast Quadrant

The Southeast Quadrant of the Livermore site has elevated levels of plutonium in the surface soil (from historic waste management operations) and air (from resuspension). A high volume air particulate sampler is located adjacent to the UNCLE Credit Union (the location of the SW-MEI) to monitor the plutonium levels in this area. Monitoring data from this air sampler were used as a direct measurement of potential dose via the air pathway. The median annual concentration of ²³⁹⁺²⁴⁰Pu (the analytical technique used, alpha spectroscopy, does not distinguish between ²³⁹Pu and ²⁴⁰Pu) in air was 3.41×10^{-19} μ Ci/mL (1.26×10^{-14} Bq/mL). Using the dose conversion factor of 3.08×10^5 mrem/ μ Ci (8.32×10^{-5} Sv/Bq) from Federal Guidance Report No. 11, EPA-520/1-88-020, U.S. Environmental Protection Agency (1988) for ²³⁹Pu and ²⁴⁰Pu, and the standard man breathing rates of 8400 m³/y, the dose was 8.8×10^{-4} mrem (8.8×10^{-3} μ Sv) for 2001.

Site 300 Principal Diffuse Sources

Diffuse sources at Site 300 involve primarily depleted uranium, and to a considerably lesser extent, tritium. During remediation efforts at Site 300, LLNL completed a contaminant screening to identify potential routes of migration from soil to air and other environmental media of these radionuclides and other contaminants (Final Site Wide Remedial Investigation Report; Webster-Scholten, Ed., 1994, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-AR-108131). Uranium-238 and tritium were identified as contaminants of potential concern.

Tritium Evaporation and Migration at Site 300

Tritium gas and solids containing tritium (Li³H) were components of explosives assemblies tested on the firing tables during past experiments. Most of the gaseous tritium escaped to the atmosphere during the tests, but some of the solid Li³H remained as residue in the firing table gravel. Rainwater and dust-control rinse water percolated through the gravel, causing the tritium to migrate into the subsurface soil and, in some cases, eventually to the ground water. Tritium contaminated gravel was removed from the firing tables in 1988 and disposed in the

Pit 7 landfill. Tritium in landfills, firing table soils, and ground water are potential sources of diffuse emissions of tritium to the atmosphere at Site 300. LLNL personnel maintain an air tritium sampler at a perimeter location at Site 300, and doses from diffuse tritium sources may be estimated based on the monitoring data for that sampling location. For the calendar year 2001, all measurements in ambient air at the Site 300 perimeter location were below the detection limits of the analytical method, and were consistent with natural background measurements.

Resuspension of Depleted Uranium In Soil at Site 300

Like tritium, depleted uranium has been used as a component of explosives test assemblies. It remains as a residue in surface soils, especially near the firing tables. Because surface soil is subject to resuspension by the action of wind, rain, and other environmental disturbances, the collective effects of surface soil uranium residuals on off-site doses were evaluated.

For the 1995 NESHAPs annual report, we developed calculations to separate the contribution to measured uranium activities from naturally occurring uranium (NU) (Gallegos et al., 1996, Lawrence Livermore National Laboratory, UCRL-ID-113867-96). We base our dose estimate for resuspended depleted uranium (DU) on the measured environmental surveillance monitoring total concentration in air of uranium-238, subtracting out the part contributed by NU, from the following equation:

$$\mu = \frac{0.00726 - 0.99274 \frac{M(\text{CU} - 235)}{M(\text{CU} - 238)}}{0.00526 \frac{M(\text{CU} - 235)}{M(\text{CU} - 238)} + 0.00526}$$

where μ is the fraction (by weight) of uranium contributed by operations, CU is composite uranium (both DU and NU), $M(\text{CU}-235)$ the mass of U-235 in the composite (measured) uranium, and $M(\text{CU}-238)$ the mass of U-238 in the composite (measured) uranium. (For derivation of the equation see the 1995 NESHAPs annual report, referenced above.)

As explained in last year's Site Annual Environmental Report (*Environmental Report 2000*, Biermann et al. Sept. 2001; Chapter 5, "Results" section, p. 5-7), the glass fiber filter media used in our monitors contain concentrations of ^{235}U that are too high for our approach to reliably determine the quantity of operations-contributed DU. (To correct this deficiency, in January 2002 the sampling media for airborne particulates was changed from glass fibers to cellulose filters.) In absence of a better present approach, we used an eight-year average value of the estimated SW-MEI dose based on this method. This average gave 0.0037 mrem (0.037 μSv) as the dose

attributed to resuspension of DU in soil for 2001. Coincidentally, this is the same as the previous year's (2000) value.

Modeling Dose from Tritium

To evaluate dose from tritium releases to air, we use the EPA-sanctioned CAP88-PC code. Its tritium model calculates dose from inhalation, skin absorption, and ingestion of tritium only in its tritiated water vapor form (HTO). CAP88-PC's tritium model is based on the specific activity model, which assumes that the tritium-to-hydrogen ratio in body water is the same as in air moisture. Because the specific activity model is linked in CAP88-PC with relatively high dose coefficients for HTO, the model's dose predictions generally err on the high side.

Doses from unit concentration of HT in air are a factor of 15,000 times lower than those from unit concentration of HTO in air (International Commission on Radiological Protection (ICRP), 1995, *Age dependent doses to members of the public from intake of radionuclides, Part 4, Inhalation Dose Coefficients*. Oxford: Pergamon Press; ICRP Publication 71; Ann. ICRP 25[3&4]). Thus, doses from inhaled HT can safely be ignored unless the air concentration is extremely high. A release of HT cannot be ignored, however, because HT that reaches the ground is rapidly and efficiently converted to HTO by microorganisms in soil (McFarlane, Rogers, and Bradley, *Environmental Science and Technology* 12: 590-593, 1978; Brown, Ogram, and Spencer, *Health Physics* 58:171-181, 1990) and to a lesser extent in vegetation (Sweet and Murphy, *Environmental Science and Technology*, 18:358-361, 1984).

Organically bound tritium (OBT) is formed by plants during photosynthesis and is incorporated by animals when ingested. Animals also metabolize some OBT from ingested or inhaled HTO. The ICRP dose coefficient for OBT is about 2.3 times higher than that of HTO, because the biological half-life of OBT in the body is longer than that of HTO, which is eliminated at the same rate as body water. Although doses predicted by CAP88-PC are generally high enough to account for dose from ingested OBT, nevertheless, a model that explicitly calculates dose from OBT is preferable.

A simple tritium model, NEWTRIT, has been developed that calculates ingestion dose from both HTO and OBT and accounts for conversion of HT to HTO in the environment after releases of HT (Peterson, S-R. and P.A. Davis, *Health Physics* 82(2):213-225, 2002). For this report, LLNL has used the NEWTRIT model in CAP88-PC, in addition to the default CAP88-PC code, to estimate doses from significant sources of tritium emissions; see, e.g., Table 3. A brief discussion of the NEWTRIT model was presented in Attachment 2 of last year's NESHAPs annual report (*LLNL NESHAPs 2000 Annual Report*, Gallegos et al. June 2001).

The NEWTRIT model was presented to EPA and DOE at a meeting of the Health Physics Society (Cleveland, OH, June 2001), and the paper was published in that society's journal (Peterson and Davis, 2002, Op. cit.). In October 2001, LLNL sent a letter to EPA Region IX requesting consideration of an alternative methodology for calculating doses from atmospheric releases of tritiated water vapor (HTO) and tritiated gas (HT) for use in demonstrating compliance with radionuclide NESHAPs (40 CFR 61 Subpart H). Copies of NEWTRIT, CAP88-PC-T (CAP88-PC with NEWTRIT encoded as the tritium model), and associated documentation were given to EPA and several DOE laboratories that had expressed interest. A decision has not been made as of this writing, but LLNL is hopeful that NEWTRIT, or a similar approach to modeling releases of HT and HTO for regulatory compliance, will be accepted.

Comparison of 2001 Modeling Results with Tritium Air Surveillance Monitoring Data

A comparison was made between CAP88-PC-predicted concentrations of tritium in air and ambient air monitoring data for the eleven tritiated water vapor samplers on the Livermore site (designated VIS, SALV, POOL, CAFE, MESQ, MET, COW, B331, B514, B624, and B292) and one off-site sampler (ZON7) that have been used for comparison since 1997. Monitor locations are shown in Figure 7.

Only the three most significant sources of tritium releases to air at the Livermore site were included in the model-data comparison. The largest point source is the Tritium Facility (Building 331), where the tritium is emitted from two 30-m-high, continuously monitored stacks. The Building 331 stack emissions were determined independently by stack monitoring, with the result that a total of 18.3 Ci (6.8×10^{11} Bq) of HTO was emitted in 2001. (The 1.7 Ci [6.3×10^{10} Bq] of HT emitted from the Tritium Facility stacks is not included in the comparison because the air tritium samplers only collect HTO.) These stacks make the largest contribution to the concentrations of tritium in most of the monitors, because the emissions are cast high into the air. Diffuse-source emissions are lower to the ground, primarily affecting those monitors in closest proximity. The other two principal sources in our modeling/measurement comparison are of this type: open-air diffuse emission areas associated with the Building 612 Yard and the Tritium Facility (Building 331) outside yard waste accumulation and storage areas. Emissions from the Building 612 Yard source were estimated to be 2.0 Ci (7.4×10^{10} Bq), based on calibrating CAP88PC-predictions of tritium concentrations at the tritium monitor B624 closest to it. (Thus the B624 data do not provide a test of the modeling.) Emissions from the B331 outside yard source were estimated to be 1.0 Ci (3.7×10^{10} Bq) in 2001, based on facility knowledge and environmental monitoring data (primarily the B331 monitor near this yard). While these two diffuse sources contribute significantly to tritium concentrations in all of the monitors, all other potential sources of tritiated water vapor release, such as the

it is preferable to err on the conservative side of predicting higher values. For the B292 monitor, its under-prediction is likely attributable to our neglect in the modeling of the quite small diffuse source near the building, which would noticeably contribute only to that monitor's value. In 2001, as in the past, CAP88-PC somewhat over-predicts concentrations of tritium in air at the site perimeter and offsite at ZON7.

Table 10. Comparison of measured and modeled annual-average concentrations of tritiated water vapor (HTO) in air at selected Livermore site locations, 2001.

Air monitor location (name)	Measured average concentration (pCi/m ³)	Modeled* average concentration (pCi/m ³)	Ratio of modeled-to-measured concentrations	Modeled concentration of tritium in air contributed by the indicated source (pCi/m ³)		
				B331 Stacks	B612 Yard	B331 Outside
B624	49.1	48.4	0.99	0.31	48	0.075
B331	7.67	13.1	1.7	0.051	0.84	12
B514	4.16	18.2	4.4	0.15	18	0.076
VIS	1.57	1.76	1.1	0.69	0.94	0.13
POOL	2.40	1.90	0.79	0.45	0.75	0.70
CAFE	1.12	1.69	1.5	0.39	0.91	0.39
COW	0.818	0.338	0.41	0.085	0.15	0.10
B292	1.37	0.545	0.40	0.087	0.24	0.22
SALV	0.651	2.55	3.9	0.11	2.4	0.037
MESQ	0.621	0.642	1.0	0.11	0.22	0.31
MET	0.284	0.334	1.2	0.063	0.14	0.13
ZON7	0.378	0.505	1.3	0.38	0.086	0.039
(CRED)**	—	2.39	—	0.74	1.5	0.15

* This result takes into account the three most significant tritium sources; it is the sum of the three contributions shown in the far-right column.

** The CRED location does not have a tritium surveillance air monitor, but is included since it marks the location of the SW-MEI.

Effect on Modeling/Monitoring Comparison of Corrections to Results of Tritium Air Surveillance Monitoring

As noted earlier in Section II, it has been found that LLNL's results for measured concentrations of tritium in ambient air require correction, due to new understanding of the effects of moisture retention by the silica gel prior to sampling. Comparisons between air concentrations predicted by CAP88-PC and observed air tritium concentrations have been included in LLNL's NESHAPs reports since 1997. The comparison shown in Table 10 uses the corrected measured values, but we should revise all comparisons for earlier years, replacing the original measured values by their corrected counterparts. Unfortunately, this is not possible, since there is no way to accurately correct LLNL's measured values obtained prior to January 2001. Two of

several reasons for this inability to correct old data are that (1) the correction factor is different for each batch of silica gel, which was changed from time to time in the past (most recently in May 2000), and (2) the initial dry-weights of the silica gel must be known for the correction factor determination, but these were not recorded and cannot be reconstructed. In lieu of a better alternative, a conservatively high correction factor of 2.1 was chosen to apply to air concentrations measured prior to 2001, to allow for the possibility that the silica gel used in previous years had more bound water than that used presently. (Approximately 99% of the results for 2001 had a correction factor less than or equal to 2.1.)

Using the 2.1 factor, revised predicted-to-observed (P/O) ratios of tritium concentrations in air at Livermore site perimeter locations and ZON7 are compared in Table 11 with the previously published ratios, for comparisons made each of the last four years. Without correction, only two of the thirty-two P/O ratios were less than 1.0, with the lowest being 0.84 at COW in 1997. When the observations are increased by a factor of 2.1, seven P/O ratios are below 1.0, with the lowest being 0.4 at COW in 1997.

Table 11. Previously published (upper) and revised (lower) ratios of predicted-to-observed air concentrations of tritiated water at Livermore site perimeter locations and ZON7, 1997-2000.

Monitor	1997	1998	1999	2000
CAFE	1.9	3.4	6.3	6.1
	0.89	1.6	3.0	2.9
COW	0.84	1.0	1.6	1.0
	0.40	0.49	0.77	0.50
MESQ	3.3	5.6	4.0	5.0
	1.6	2.6	1.9	2.4
MET	3.2	2.4	3.1	2.4
	1.5	1.2	1.5	1.1
POOL	0.99	2.2	3.9	4.4
	0.47	1.1	1.9	2.1
SALV	1.5	6.9	3.7	11.
	0.73	3.3	1.8	5.2
VIS	3.0	2.4	5.7	3.0
	1.4	1.2	2.7	1.4
ZON7	3.9	3.2	5.5	3.0
	1.9	1.5	2.6	1.4

It should be noted that the expected uncertainty in concentrations calculated with a Gaussian plume model (such as used by CAP88-PC) produce a range of values that bracket the dispersion in P/O ratios in Table 11. Limitations of the Gaussian plume model are discussed in general terms in Section 3.2.10 of the AIRDOS-EPA manual (Moore, R.E., et al., "AIRDOS-EPA: A computerized methodology for estimating environmental concentrations and doses to man from airborne releases of radionuclides," Oak Ridge National Laboratory; USDOE Report, ORNL-5532, NTIS; 1979). More specifically, a comparison of AIRDOS-EPA predictions of air concentrations for various radionuclides (^{234}U , ^{238}U , ^{85}Kr , and ^3H) with measurements at six different sites concluded that the 90% confidence interval for the accuracy of the CAP88-PC dispersion model ranges from a factor of 0.3 to 4.4, based on 51 samples ("Comparison of AIRDOS-EPA predictions of ground-level airborne radionuclide concentrations to measured values," Jack Faucett Associates, Bethesda, MD. 20814; JACKFAU-341/12-87; 1987).

SECTION IX. Supplemental Information on Compliance

Status of Compliance with Other Regulations

Status of compliance with 40 CFR 61 Subpart Q - National Emission Standards for Radon Emissions from Department of Energy Facilities

LLNL does not have storage and disposal facilities for radium containing materials that would be a significant source of radon.

Status of compliance with 40 CFR 61 Subpart T - National Emission Standards for Radon Emissions from the Disposal of Uranium Mill Tailings

LLNL does not have or store any uranium mill tailings.

Information on Radon-220 and Radon-222 Emissions

Radon emissions occur naturally by emanation from the earth. Radon-222 emissions that were reported in past NESHAPs annual reports from research experiments at the Livermore site did not occur in 2001.

ATTACHMENT 1. LLNL NESHAPs 2001 Annual Report Spreadsheet

Guidance for Interpreting the Data Spreadsheet

A generalized description of each facility and its operations is provided on the spreadsheet. In addition, the following information is shown for each listed emission point or stack:

- Building and room number(s)
- Specific stack identification code(s)
- Generalized description of operations in the room(s) or area(s)
- Radionuclides utilized in the operation
- Annual radionuclide usage inventory with potential for release (by isotope, in curies)
- Physical state factors (by isotope)
- Stack parameters
- Emission control devices and emission control device abatement factors
- Estimated or measured annual emissions (by isotope)
- Distance and direction to the site-wide maximally exposed individual (SW-MEI)
- Calculated EDE to the SW-MEI
- Distance and direction to the maximally exposed individual for that specific source (MEI)
- Calculated EDE to the MEI (source term not adjusted for emission controls)
- Source category

Radionuclides

The radionuclides shown in the spreadsheet are those from specific emission points where air emissions were possible. If radionuclides were present, but encapsulated or sealed for the entire year, radionuclides, annual usage inventories, and emissions are not listed.

Radionuclide Usage Inventories with Potential for Release

The annual radionuclide usage inventories for point source locations are based on data from facility experimenters and managers. For Buildings 251 (hardened area) and 332, classification issues regarding transuranic radionuclide usage inventories make use of the usage inventory/modeling approach impractical. However, all such affected emission points in these buildings are continuously monitored, and emissions are therefore directly determined.

Physical State Factors

The physical state factors listed are EPA potential release fractions from 40 CFR 61, Appendix D, whereby emissions are estimated from radionuclide usage inventories depending on their physical states for use in dispersion/dose assessment modeling. A physical state factor of 1.0×10^{-6} is used for solids, 1.0×10^{-3} is used for liquids and powders, and 1.0 is used for unconfined gases. The U.S. EPA has granted LLNL approved alternative emissions factors for elemental uranium, uranium/niobium alloy, and elemental plutonium. (See Table 2, page 14.) These factors are allowed provided that the material is not intentionally dispersed to the environment and that the processes do not alter the chemical form of the material.

Stack Parameters

Engineering surveys conducted from 1990 through 1992 form the basis for the stack physical parameters shown, which were checked and validated by facility experimenters and managers for 1994 and 1995. Stack physical parameters for sources evaluated in 2001 were updated, as necessary, by experimenters and managers for those facilities.

Emission Control Devices

High Efficiency Particulate Air (HEPA) filters are used in many LLNL facilities to control particulate emissions. For some discharge points, scrubbers and electrostatic precipitators aid the control of emissions. The operational performance of all HEPA filtration systems is routinely tested. The required efficiency of a single stage HEPA filter is 99.97%. Double staged filter systems are in place on some discharge points. Triple stage HEPA filters are used on glove box ventilation systems in the Building 332 Plutonium Facility and in the hardened portion of Building 251.

Control Device Abatement Factors

Similar to physical state factors, control device abatement factors, from Table 1 in 40 CFR 61, Appendix D, are those associated with the listed emission control devices, and are used to better estimate actual emissions for use in dispersion and dose models. By regulation, each HEPA filter stage is given a 0.01 factor (even though the required test efficiency that all LLNL HEPA filters must maintain would yield a factor of 0.0003).

Estimated Annual Emissions

For unmonitored and non-continuously monitored sources, estimated annual emissions for each radionuclide are based on the product of (1) usage inventory data, (2) time factors (discussed in "Emission Source Terms" in Section III), (3) EPA potential release fractions (physical state factors), and (4) applicable emission control device abatement factors.

Actual emission measurements are the basis for reported emissions from continuously monitored facilities. LLNL facilities that had continuous monitoring systems in 2001 were Buildings 175, 177, 235, 251, 331, 332, and 491, as noted earlier. See the discussion below under "0.1 mrem/y Monitoring Requirement" regarding the use of emissions measurements for monitored sources.

10 mrem/y Site-Wide Dose Requirement

For LLNL to comply with the NESHAPs regulations, the LLNL site-wide maximally exposed individual (SW-MEI; defined as the hypothetical member of the public at a single residence, school, business, or office who receives the greatest LLNL-induced EDE from the combination of all radionuclide source emissions) cannot receive an EDE greater than 10 mrem/y (100 μ Sv/y). (See Section II for a discussion of the SW-MEI.)

In Attachment 1, the distance and direction to the respective SW-MEI are shown for each facility at each site. Doses to the site specific SW-MEIs were evaluated for each source and then totaled for site specific evaluations against the 10 mrem/y dose standard (see "Total Dose Estimate" in Section IV).

0.1 mrem/y Monitoring Requirement

To assess compliance with the requirement for continuous monitoring (potential dose greater than 0.1 mrem/y [1.0 μ Sv/y] to the maximally-exposed public individual or MEI, discussed earlier in Section II), emissions must be individually evaluated from each point source. The location of the MEI is generally different for each emission point. The maximum dose at a location of unrestricted public access typically occurs at a point on the site perimeter. Therefore, it is often referred to as the maximum "fence line" dose, although the off-site maximum dose could occur some distance beyond the perimeter. (This could happen, e.g., when the perimeter is close to a stack; however, for nearly all emission points at the Livermore site and Site 300, calculations show that ground level concentrations of radionuclides generally decline continuously beyond LLNL boundaries.) As stipulated by the regulations, modeling for assessment of continuous monitoring requirements assumed unabated emissions (i.e., no credit was taken for emission abatement devices, such as filters), but physical state factors and time factors were applied.

The unabated EDE cannot be calculated for HEPA-filtered facilities monitored for radioactive particles. Because the monitoring equipment is placed after HEPA filtration, there is no way to obtain an estimate for what the emissions might have been had there been no filtration. It is not reasonable to apply factors for the effects of the HEPA filters on the emission rate because most of what is measured on the HEPA filters is the result of the radioactive decay of radon, which is capable of penetrating the filter. Attachment 1 gives, for each inventoried point source, the

Attachment 1 - 2001 LLNL NESHAPs Annual Report Spreadsheet

Rating	Response	Stack ID (continued)	Operation	Refrigerant	Annual Inventory with Potential for Release (lb)	Physical State	Stack Height (ft)	Stack Diameter (in)	Stack Velocity (ft/s)	Control Device(s)	Control Efficiency	Control Device Adjustment Factor	Estimated Annual Emissions (lb)	Distance to Sensitive Receptor (ft)	Direction to Sensitive Receptor	Distance to Sensitive Receptor (ft)	Direction to Sensitive Receptor	Distance to Sensitive Receptor (ft)	Direction to Sensitive Receptor	Source Category
253	17086	PHE-21	Flaring process methanols pretreatment	U-134 U-134	1.0E+00 1.0E+00 1.0E+00	1.0E+00	6.4	0.30	13.2	None	None	1	1.0E-13 1.0E-13 1.0E-13	1182	ESE	738	W	1.4E+10 1.4E+10	2	
253	1732	PHE-21	Distillation of environmental samples	Pu-239 Gross beta H-3	1.8E+13 1.2E+13 2.8E+13 1.1E+12	1.0E+00	NA	NA	NA	None	None	1	6.9E+12 5.4E+14 4.1E+13	1182	ESE	738	W	7.9E+11	2	
253	1734	PHE-21	Distillation of environmental samples	U-235 U-238	2.7E+10 4.8E+10	1.0E+00	8.1	0.10	23.0	rd99	rd99	0.01	2.7E+16 4.8E+16	1182	ESE	738	W	3.4E+19	2	
253	1734	PHE-21	Distillation of environmental samples	U-235 U-238	1.7E+11 2.8E+12	1.0E+00	10.4	0.30	12.8	None	None	1	1.7E+11 2.8E+12	1182	ESE	738	W	4.8E+10	2	
253	1734	PHE-20	Quality control samples	Pu-239 Sr-90 H-3	2.8E+12 2.8E+12 1.1E+10	1.0E+00	10.4	0.30	12.8	None	None	1	2.8E+12 2.8E+12 1.1E+10	1182	ESE	738	W	8.2E+15	2	
253	1734	PHE-11	Acid digestion for sample analysis	H-3 Gross alpha Gross beta Sr-90 Pu-239	8.8E+09 3.4E+11 2.8E+12 3.1E+12	1.0E+00	10.4	0.30	12.8	None	None	1	8.8E+09 3.4E+11 2.8E+12 3.1E+12	1182	ESE	738	W	1.4E+08 1.4E+08	2	
253	1810	PHE-02	Preparation of calibration standards	H-3 Pu-239 Pu-238	3.0E+11 1.0E+10 1.0E+10	1.0E+00	7.0	0.20	5.2	None	None	1	3.0E+11 1.0E+10 1.0E+10	1182	ESE	738	W	2.1E+14	2	
254	108	PHE-1000	Analysis of urine for radionuclides	Am-241 Am-243 Pu-239 Pu-238 H-3 Sr-90 Y-90	1.0E+17 1.0E+17 2.8E+17 1.4E+14 3.1E+14 1.0E+14	1.0E+00	8.2	1.07	8.3	None	None	1	1.0E+17 1.0E+17 2.8E+17 1.4E+14 3.1E+14 1.0E+14	1088	ESE	1070	NNE	1.4E+16 1.4E+16 1.4E+16	2	
254	108	PHE-1000	Analysis of urine for radionuclides	Am-241 Am-243 Pu-239 Pu-238 H-3 Sr-90 Y-90	1.0E+17 1.0E+17 2.8E+17 1.4E+14 3.1E+14 1.0E+14	1.0E+00	8.2	1.07	8.3	None	None	1	1.0E+17 1.0E+17 2.8E+17 1.4E+14 3.1E+14 1.0E+14	1088	ESE	1070	NNE	8.9E+19	2	
254	110	PHE-1000	Analysis of urine for radionuclides	Am-241 Am-243 Pu-239 Pu-238 H-3 Sr-90 Y-90	8.2E+18 8.2E+17 1.0E+18 1.1E+18 1.0E+18 1.0E+18	1.0E+00	8.2	1.07	8.3	None	None	1	8.2E+18 8.2E+17 1.0E+18 1.1E+18 1.0E+18 1.0E+18	1088	ESE	1070	NNE	5.0E+11	2	

Attachment 1 - 2001 LLNL NESHAPs Annual Report Spreadsheet

Building	Room/Area	Stack ID	Operation	Reference(s)	Annual Inventory for Release (lb)	Process Rate (lb/hr)	Stack Height (m)	Stack Diameter (ft)	Stack Velocity (ft/min)	Control Device(s)	Control Device Efficiency Factor	Annual Emissions (lb)	Distance to Downwind (ft)	Direction to Downwind (M)	EE (ft/min)	A. Annual Modification, Distance to NEI (ft)	Unavoidable Emissions GDE (lb/yr)	Source Category				
812	101	PNE-4	Laboratory analysis of particulate matter and velocity samples	Am-241	5.8E-06	1.0E-03	10.3	0.31	3.4	None	0.97	6.0E-10	444	NE	6.1E-04	295	9.4E-04	1				
				Am-243	4.2E-06	1.0E-03								4.2E-09								
				Ba-133	6.0E-06	1.0E-03									6.0E-09							
				Ba-137	3.8E-07	1.0E-03									3.8E-07							
				Ba-210	2.8E-07	1.0E-03									2.8E-07							
				C-14	2.0E-04	1.0E-03									2.0E-04							
				Ca-108	3.4E-10	1.0E-03									3.4E-10							
				Ca-134	2.0E-05	1.0E-03									2.0E-05							
				Ca-138	2.8E-07	1.0E-03									2.8E-07							
				Ca-144	3.4E-06	1.0E-03									3.4E-06							
				Ca-148	1.8E-07	1.0E-03									1.8E-07							
				Ca-152	5.4E-08	1.0E-03									5.4E-11							
				Ca-154	1.5E-08	1.0E-03									1.5E-09							
				Ca-156	4.8E-07	1.0E-03									4.8E-10							
				Ca-158	3.4E-08	1.0E-03									3.4E-09							
				Ca-160	1.8E-04	1.0E-03									1.8E-04							
				Ca-162	1.4E-05	1.0E-03									1.4E-05							
				Ca-164	1.0E-05	1.0E-03									1.0E-05							
				Ca-166	1.0E-05	1.0E-03									1.0E-05							
				Ca-168	1.0E-05	1.0E-03									1.0E-05							
				Ca-170	1.0E-05	1.0E-03									1.0E-05							
				Ca-172	1.0E-05	1.0E-03									1.0E-05							
				Ca-174	1.0E-05	1.0E-03									1.0E-05							
				Ca-176	1.0E-05	1.0E-03									1.0E-05							
				Ca-178	1.0E-05	1.0E-03									1.0E-05							
				Ca-180	1.0E-05	1.0E-03									1.0E-05							
				Ca-182	1.0E-05	1.0E-03									1.0E-05							
				Ca-184	1.0E-05	1.0E-03									1.0E-05							
				Ca-186	1.0E-05	1.0E-03									1.0E-05							
				Ca-188	1.0E-05	1.0E-03									1.0E-05							
				Ca-190	1.0E-05	1.0E-03									1.0E-05							
				Ca-192	1.0E-05	1.0E-03									1.0E-05							
				Ca-194	1.0E-05	1.0E-03									1.0E-05							
				Ca-196	1.0E-05	1.0E-03									1.0E-05							
				Ca-198	1.0E-05	1.0E-03									1.0E-05							
				Ca-200	1.0E-05	1.0E-03									1.0E-05							
				Ca-202	1.0E-05	1.0E-03									1.0E-05							
				Ca-204	1.0E-05	1.0E-03									1.0E-05							
				Ca-206	1.0E-05	1.0E-03									1.0E-05							
				Ca-208	1.0E-05	1.0E-03									1.0E-05							
				Ca-210	1.0E-05	1.0E-03									1.0E-05							
				Ca-212	1.0E-05	1.0E-03									1.0E-05							
				Ca-214	1.0E-05	1.0E-03									1.0E-05							
				Ca-216	1.0E-05	1.0E-03									1.0E-05							
				Ca-218	1.0E-05	1.0E-03									1.0E-05							
				Ca-220	1.0E-05	1.0E-03									1.0E-05							
				Ca-222	1.0E-05	1.0E-03									1.0E-05							
Ca-224	1.0E-05	1.0E-03									1.0E-05											
Ca-226	1.0E-05	1.0E-03									1.0E-05											
Ca-228	1.0E-05	1.0E-03									1.0E-05											
Ca-230	1.0E-05	1.0E-03									1.0E-05											
Ca-232	1.0E-05	1.0E-03									1.0E-05											
Ca-234	1.0E-05	1.0E-03									1.0E-05											
Ca-236	1.0E-05	1.0E-03									1.0E-05											
Ca-238	1.0E-05	1.0E-03									1.0E-05											
Ca-240	1.0E-05	1.0E-03									1.0E-05											
Ca-242	1.0E-05	1.0E-03									1.0E-05											
Ca-244	1.0E-05	1.0E-03									1.0E-05											
Ca-246	1.0E-05	1.0E-03									1.0E-05											
Ca-248	1.0E-05	1.0E-03									1.0E-05											
Ca-250	1.0E-05	1.0E-03									1.0E-05											
Ca-252	1.0E-05	1.0E-03									1.0E-05											
Ca-254	1.0E-05	1.0E-03									1.0E-05											
Ca-256	1.0E-05	1.0E-03									1.0E-05											
Ca-258	1.0E-05	1.0E-03									1.0E-05											
Ca-260	1.0E-05	1.0E-03									1.0E-05											
Ca-262	1.0E-05	1.0E-03									1.0E-05											
Ca-264	1.0E-05	1.0E-03									1.0E-05											
Ca-266	1.0E-05	1.0E-03									1.0E-05											
Ca-268	1.0E-05	1.0E-03									1.0E-05											
Ca-270	1.0E-05	1.0E-03									1.0E-05											
Ca-272	1.0E-05	1.0E-03									1.0E-05											
Ca-274	1.0E-05	1.0E-03									1.0E-05											
Ca-276	1.0E-05	1.0E-03									1.0E-05											
Ca-278	1.0E-05	1.0E-03									1.0E-05											
Ca-280	1.0E-05	1.0E-03									1.0E-05											
Ca-282	1.0E-05	1.0E-03									1.0E-05											
Ca-284	1.0E-05	1.0E-03									1.0E-05											
Ca-286	1.0E-05	1.0E-03									1.0E-05											
Ca-288	1.0E-05	1.0E-03									1.0E-05											
Ca-290	1.0E-05	1.0E-03									1.0E-05											
Ca-292	1.0E-05	1.0E-03									1.0E-05											
Ca-294	1.0E-05	1.0E-03									1.0E-05											
Ca-296	1.0E-05	1.0E-03									1.0E-05											
Ca-298	1.0E-05	1.0E-03									1.0E-05											
Ca-300	1.0E-05	1.0E-03									1.0E-05											
Ca-302	1.0E-05	1.0E-03									1.0E-05											
Ca-304	1.0E-05	1.0E-03									1.0E-05											
Ca-306	1.0E-05	1.0E-03									1.0E-05											
Ca-308	1.0E-05	1.0E-03									1.0E-05											
Ca-310	1.0E-05	1.0E-03									1.0E-05											

ATTACHMENT 2. Surrogate Radionuclides List

The need for selection of a surrogate isotope occurs when an isotope used in operations (isotope of interest) is not contained in the limited nuclide library in the NESHAPs dose compliance model CAP88-PC. The selection of a suitable surrogate is based upon several criteria. If possible, a surrogate isotope is chosen from the CAP88-PC radionuclide library that has a metabolically similar behavior to the isotope of interest. Following an acute inhalation exposure, the metabolically similar surrogate would concentrate in the same specific organs and tissues as the isotope of interest. In most cases the surrogate selected possesses similar modes of decay and decay energies of the radiation type of the isotope of interest. Thus, the surrogate models the behavior of the isotope with similar relative biological effect due to deposition energy.

According to present knowledge, the daughter nuclides produced following physical decay are assumed to remain organ site specific and follow the translocation pathway of the parent. Therefore, when a surrogate of similar metabolic behavior is not available or has a greatly dissimilar half-life, the surrogate chosen is a daughter nuclide of the isotope of interest that will remain organ site specific and follow the translocation pathway of the parent.

Once a surrogate has been selected, the equivalent source term is adjusted by the product of the initial inventory of the isotope of interest and the ratio of the effective dose equivalent of the surrogate to that of the isotope of interest. For determining the dose ratio, the primary exposure pathway is assumed to be that of inhalation and inhalation dose conversion factors (International Commission on Radiological Protection Publication No. 71, "Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 4 Inhalation Dose Coefficients," Elsevier Science Ltd., 1996) are used for determination of the effective dose equivalents.

In addition, isotopic analysis of mixtures of radionuclides are not always available, and radionuclide usage inventories are stated as "gross alpha," "gross beta," "gross gamma," or "mixed fission products" (MFP). In these cases, ^{239}Pu is used as the surrogate for gross alpha, ^{137}Cs is used as the surrogate for gross gamma, and ^{90}Sr is used as the surrogate for gross beta and mixed fission products to provide conservative dose estimates.

Table 2-1 provides a list of radionuclides not in the CAP88-PC library and their respective surrogates.

Table 2-1. List of surrogate radionuclides.

Isotope	Half-Life	Lung Class ^a	ALI (inh) μCi	DAC (inh) $\mu\text{Ci}/\text{m}^3$	Surrogate	Half-Life	Lung Class ^a	ALI (inh) μCi	DAC (inh) $\mu\text{Ci}/\text{m}^3$
Ca-106m	127 y	Y	2.0×10^1	1.0×10^{-8}	Co-60	5.271 y	Y	3.0×10^1	1.0×10^{-8}
Bi-207	38 y	W	4.0×10^2	1.0×10^{-7}	Bi-214	19.9 min	W	9.0×10^2	4.0×10^{-7}
Ca-45	163 d	W	8.0×10^2	4.0×10^{-7}	Sr-90	29.12 y	D	2.0×10^1	8.0×10^{-9}
Cd-109	464 d	Y	1.0×10^2	5.0×10^{-8}	Co-60	5.271 y	Y	3.0×10^1	1.0×10^{-8}
Cf-249	350.6 y	Y	1.0×10^{-2}	4.0×10^{-12}	Cm-245	8500 y	W	6.0×10^{-3}	3.0×10^{-12}
Cf-250	13.1 y	W	9.0×10^{-3}	4.0×10^{-12}	Am-241	432.2 y	W	6.0×10^{-3}	3.0×10^{-12}
Cl-36	3.01×10^5 y	W	2.0×10^2	1.0×10^{-7}	Cs-137	30 y	D	2.0×10^2	6.0×10^{-8}
Es-254	275.7 d	W	7.0×10^{-2}	3.0×10^{-11}	Pu-239	24065 y	Y	2.0×10^{-2}	7.0×10^{-12}
Eu-149	93.1 d	W	3.0×10^3	1.0×10^{-6}	Pm-151	28.4 hr	Y	3.0×10^3	1.0×10^{-6}
Gd-148	93 y	D	8.0×10^{-3}	3.0×10^{-12}	La-140	40.272 h	W	1.0×10^3	5.0×10^{-7}
Os-185	94 d	D	5.0×10^2	2.0×10^{-7}	Mo-99	66 h	Y	1.0×10^3	6.0×10^{-7}
P-33	25.4 d	W	3.0×10^3	1.0×10^{-6}	P-32	14.29 d	D	9.0×10^2	4.0×10^{-7}
Re-184	38 d	W	1.0×10^3	6.0×10^{-7}	Mo-99	66 h	Y	1.0×10^3	6.0×10^{-7}
Se-75	119.8 d	W	6.0×10^2	3.0×10^{-7}	As-76	26.32 h	W	1.0×10^3	6.0×10^{-7}
Sr-85	64.8 d	D	3.0×10^3	1.0×10^{-6}	Sr-90	29.12 y	D	2.0×10^1	8.0×10^{-9}
Ta-182	115 d	Y	1.0×10^2	6.0×10^{-8}	Hf-181	42.4 d	W	4.0×10^2	2.0×10^{-7}
Tb-157	110 y	W	3.0×10^2	1.0×10^{-7}	La-140	40.272 h	W	1.0×10^3	5.0×10^{-7}
Tb-158	180 y	W	2.0×10^1	8.0×10^{-9}	La-140	40.272 h	W	1.0×10^3	5.0×10^{-7}
Tl-204	3.78 y	D	2.0×10^3	9.0×10^{-7}	Pb-214	26.8 min	D	8.0×10^2	3.0×10^{-7}
Tm-168	93.1 d	W	2.0×10^3	8.0×10^{-7}	La-140	40.272 h	W	1.0×10^3	5.0×10^{-7}
Tm-171	1.92 y	Y	3.0×10^2	1.0×10^{-7}	La-140	40.272 h	W	1.0×10^3	5.0×10^{-7}
Y-88	106.64 d	Y	2.0×10^2	1.0×10^{-7}	Y-90	64 h	Y	6.0×10^2	3.0×10^{-7}
Am-244	10.1 h	W	2.0×10^2	8.0×10^{-8}	Cm-244	18.11 y	W	1.0×10^{-2}	5.0×10^{-12}
Au-195	183 d	Y	4.0×10^2	2.0×10^{-7}	Ba-133	10.74 y	D	7.0×10^2	3.0×10^{-7}
Co-56	78.76 d	Y	2.0×10^2	8.0×10^{-8}	Co-60	5.271 y	Y	3.0×10^1	1.0×10^{-8}
Gd-146	48.3 d	W	3.0×10^2	1.0×10^{-7}	Sm-147	1.06×10^{11} y	W	4.0×10^{-2}	2.0×10^{-11}
Kr-85	10.72 y	Gas	See Note	1.0×10^{-4}					
Rh-102	2.9 y	Y	6.0×10^1	2.0×10^{-8}	Rh-106m	29.9 s	Y	4.0×10^4	1.0×10^{-5}
U-239	23.54 min	Y	2.0×10^5	6.0×10^{-5}	U-240	14.1 h	Y	2.0×10^3	1.0×10^{-6}
Zr-90	809 ms	W	N/A	N/A	Y-90	64 h	Y	6.0×10^2	3.0×10^{-7}
Po-209 ^b	102 y	N/A	N/A	N/A	Pu-239	24065 y	Y	2.0×10^{-2}	7.0×10^{-12}

Note: The DAC for Kr-85 also has been relaxed considerably since its beta emission only irradiates the skin. The DAC is based on limitation of non-stochastic effects in the skin; the MPC was derived assuming that the beta particles of energy greater than 0.1 MeV contributed to the whole body dose.

^a D = days, W = weeks, Y = years.

^b No ALI or DAC information available. Pu-239 used to provide a conservative alpha-emitter dose.

Source: Limiting Values of Radionuclide Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion and Ingestion, Federal Guidance Report No. 11, EPA-520/1-88-020, U.S. Environmental Protection Agency, 1988.

Mr. Jack Broadbent, Director, Air Division
U.S. Environmental Protection Agency, Region IX

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SUBJECT: Graded Approach to Compliance Demonstration for Radionuclide NESHAPs

Table 1. Comparison of graded approaches by emission category for calendar year 2000.

Emission category	Current graded approach				Proposed graded approach			
	Frequency of evaluation	Dose level (mrem)	Risk level	Number of sources	Frequency of evaluation	Dose level (mrem)	Risk level	Number of sources
Monitored stack emissions	Annual	Any	Any	46	Same	Same	Same	Same
Site 300 Shots	Annual	Any	Any	1	Same	Same	Same	Same
Monitored diffuse sources	Annual	Annual	Annual	4	Same	Same	Same	Same
inventoried stack emissions, room air, diffuse sources	Annual	Approx. 0.001 (New sources and top 90% contributors to dose)	Approx. $\geq 2 \times 10^{-8}$	5	Same	$\geq 5 \times 10^{-6}$	$\geq 1 \times 10^{-10}$	18
Inventoried stack emissions, room air, diffuse sources	Triennial update	Approx. <0.001	Approx. $< 2 \times 10^{-8}$	135	Pre-operational (NEPA/IWS) review as operations are proposed or changed (to be documented internally)	$> 5 \times 10^{-6}$	$< 1 \times 10^{-10}$	122

We believe this administrative change from our current practice is justifiable based on nearly ten years of reporting data for very small sources. In addition, LLNL has made an institutional commitment to Integrated Safety Management, which has added another venue for surfacing NESHAPs Subpart H issues in planned and existing projects.

The requested reporting method is also consistent with other radionuclide NESHAPs regulations. Facilities having radionuclide emissions, but regulated by 40 CFR 61, Subpart I, are exempt from reporting requirements if the dose consequences are less than 10% of the 10 mrem standard (i.e., less than 1 mrem); they must, however, keep internal records of the determination of dose. The total LLNL dose consequences are now less than 1% of the standard, and the sources that would not be reported, but documented internally, under the new graded approach represent doses less than 0.01% of the standard.

Finally, we believe this proposed change is consistent with the U.S. EPA's position taken with respect to reporting exemptions for releases of naturally occurring radionuclides

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