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# **LLNL NESHAPS 1996 Annual Report**

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## **U.S. Department of Energy Radionuclide Air Emission Annual Report (under Subpart H of 40 CFR Part 61) Calendar Year 1996**

**Site Name: Lawrence Livermore National Laboratory**

## **ODe** ● **rations OffIce Information**

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**Address: 1301 Clay Street Oakland, CA 94612**



### **Site Information**

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**June 1997**

## Table of Contents













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# List of Figures and Tables





## **Lawrence Livermore National Laboratory NESHAPS 1996 Annual Report**

**This amual report is prepared pursuant to the National Emissions Standards for Hazardous Air Pollutants (NESHAPS) 40 CFR Part 61, Subpart H; Subpart H governs radionuclide emissions to air from Department of Energy (DOE) facilities.**

## **SYNOPSIS**

**1**

**NESHAPS limits the emission of radionuclides to the ambient air from DOE facilities to levels resulting in an animal effective dose equivalent (EDE) of 10 mrem (100 pSv) to any member of the public. The EDEs for the Lawrence Livermore National Laboratory (LLNL) site-wide maximally exposed members of the public from 1996 operations were**

- **Livermore** site: 0.093 mrem (0.93  $\mu$ Sv) (52% from point-source **emissions, 48% from diffuse-source emissions);**
- **Site 300:0.033 rnrem (0.33 @v) (9970 from point-source, 170 from diffuse-source emissions).**

**The EDEs were generally calculated using the EPA-approved CAP88-PC airdispersion/dose-assessment model. Site-specific meteorological data, stack flow data, and emissions estimates based on radionuclide inventory data or continuous-monitoring systems data were the specific input to CAP88-PC for each modeled source.**

## **SECTION 1. Facilities Information**

### **Site Description**

**The University of California operates LLNL for DOE. LLNL was established in 1952 to conduct weapons research and development. LLNL's mission is to serve as a national resource in science and engineering, with a special responsibility for nuclear weapons. Laboratory activities focus on global security, energy, global ecology, biomedicine, economic competitiveness, and science and mathematics education. The Laboratory's mission is dynamic and has been broadened over the years to meet new national needs. LLNL consists of two sites-the main laboratory site located in Livermore, California (Livermore site), and the Experimental Test Facility (Site 300) located near Tracy, California. Figure 1 shows the locations of the sites.**

**LRermore site: LLNL's Livermore site occupies an area of 3.3 kmz located about 60 km east of San Francisco, California, adjacent to the City of Livermore in the eastern part of Alameda County. More than 6 million people live within 80 km of LLNL; approximately 65,000 of them live in the City of Livermore.**

**The Livermore site is located in the southeastern portion of the Livermore Valley, a topographical and structural depression oriented east-west within the Diablo Range of the California Coast Range Province. The Livermore Valley forms an irregularly shaped lowland area approximately 26 km long and an average of 11 km wide. The floor of the valley slopes from an elevation of approximately 200 m at the eastern end to approximately 90 m at the southwest corner.**

**The climate of the Livermore Valley is characterized by mild, rainy winters and warm, dry summers. The mean annual temperature is about 15"C. Temperatures typically range from –5°C during some pre-dawn hours during the winter, to 40"C on a few summer afternoons. The 1996 annual wind data for the Livermore site are shown in Table 1 and displayed as a wind rose in Figure 2. Although winds are variable, the prevailing wind direction is from the southwest, especially during the summer. However, during the winter, the wind often blows from the northeast. Most preapitation occurs as rain between October and April with very little rainfall during the summer months. In 1996, the Llvermore site received 527 mm of preapitation.**

**Site 300 Site 300, LLNL's Experimental Test Site, is located 24 km east of the Livermore site in the Altamont Hills of the Diablo Range and occupies an**



Figure 1. **Locations of LLNL Liverrnoresite and Site 300.**

**area of 30.3 kmz. It is close to two other explosives-testing facilities; one operated by Primex Physics International, the other by SRI Intermtional. A State of California vehicular-recreation area is located nearby, and windturbine generators line the surrounding hills. The remainder of the**



**Table 1. Wind rose for LLNL's Livermore site at the 10-m level for 1996. Values are frequency of occurrence (in percent). Columns and rows may not exactly sum to the listed totals due to rounding.**

**Table 2. Wind rose for LLNL's Site 300 at the 10-m level for 1996. Values are frequency of occurrence (in percent). Columns and rows may not exactly sum to the listed totals due to rounding.**





**Figure** 2. **Wind rose showing the average annual wind speed, frequency of occurrence, and direction at the Livermoresite, 1996.**

**surrounding area is in agricultural use, primarily pasture land for cattle and sheep. The nearest residential area is the City of Tracy (population approximately 45,000), 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 somewhat **more extreme than at the**

**LLNLNESHAPSReport 1996**



Figure 3. Wind **rose showing the average annual wind speed, frequency of occurrence, and directionat Site 300, 1996.**

**Livermore site. The 1996 amual wind data for Site 300 are shown in Table 2 and displayed as a wind rose in Figure 3. Prevailing winds are from the westsouthwest. As is the case at the Livermore site, precipitation is highly seasonal, with most precipitation occurring between October and April. The average annual rainfall over the past 20 years was 257 mm; Site 300 received 362 mm of precipitation during 1996.**

## **Source Description**

**Many different radioisotopes are used at LLNL for research purposes, including transuranics, biomedical tracers, tritium, mixed fission products, and others (Table 3). Radioisotope-handling procedures and work enclosures**

are determined for each project, depending on the isotopes, the quantities being used, and the types of operations being performed. Radioisotope handling and working environments include glove boxes, exhaust hoods, and laboratory bench tops. Exhaust paths to the atmosphere range from triple-HEPA (High-Efficiency-Particulate-Air)-filtered stacks, to roof vents and stacks lacking abatement devices, to direct dispersal of depleted uranium during explosives testing at Site 300, to a variety of diffuse-area sources.



Table 3. Radionuclides used at LLNL during 1996.

## **SECTION Il. Air-Emission Data**

### **Sources**

**At LLNL, areas where radioactive materials are used or stored, or where activation products occur, are called Radioactive Materials Management Areas (RMMAs ). Detailed information is given in Attachment 1 for pointsource emissions from the Livermore-site RMMAs in which radiological operations took place during 1996. Building 514 and five other Livermore-site sources external to buildings (including the RMMA at the Building 612 Hazardous Waste Management Yard) are treated as diffuse-area sources.**

**Similarly, detailed information is given in Attachment 1 for experiments at two Site 300 explosives-testing facilities (Buildings 801 and 851 and their associated fining tables). Six Site 300 sources, including the two firing tables where surface and subsurface contamination exists, are treated as diffuse-area sources.**

## **1996 Inventory Update and Effective Dose Equivalent (EDE) Calculations**

**For this year's report, covering activities in 1996, we updated the radionuclide inventories in our key facilities, defined as those that accounted for 90% of the 1995 Livermore site radiological dose to members of the public. We also inventoried all RMMAs that began operations in 1996. Radionuclide inventory forms, with detailed guidance for completing them, were sent to the unmonitored facilities that contributed to 90% of the dose in 1995 and to new unmonitored facilities having the potential for radionuclide emissions to the air. The forms were completed by experimenters, and certified by facility managers. Radionuclide inventories for all Site 300 explosives experiments and assessments of source terms for known diffuse sources at both sites were also updated.**

**Dose-assessment modeling runs were conducted for all diffuse sources and for all point sources using actual radionuclide releases to air, or potential releases based on radionuclide inventory data. The model used was CAP88-PC (see Section III); we incorporated 1996 on-site meteorological data (wind, precipitation**, and **temperature**) along with the 1996 radionuclide inventory **or monitoring data. Annual dose is reported as whole-body EDE expressed in units of mrem (followed by p.%). When reasonable to do so, modeling runs were combined by buildin~ rather than a separate model run for each stack**

**or room. This is permitted by the 1995 Memorandum of Understanding between the U.S. EPA and the DOE concerning radionuclide NESHAPS.**

**A generalized description of each facility and its operations is provided in Attachment 1. The following information is shown for each listed emission point or stack:**

- **Building** and **room number(s)**
- **Specific stack identification code(s)**
- e **Generalized operations in the room(s) or area(s)**
- e **Radionuclides utilized during 1996**
- o **Annual radionuclide inventory with potential for release (by isotope, in curies)**
- 0 **Physical-state factors (by isotope)**
- e **Stack parameters**
- o **Emission-control devices and emission-control-device abatement factors**
- 0 **Estimated or measured amual emissions (by isotope)**
- @ **Distance and direction to the site-wide maximally exposed individual (SW-MEI)**
- e **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**
- **Below Appendix E Quantity (Y or N)**

**A more complete description of these terms is provided in the introductory material to the attachment.**

**The radionuclides shown in the attachment are those from specific emission points where there was a potential for air emissions. If radionuclides were present, but encapsulated or sealed for the entire year, radionuclides, amual inventories, and emissions are not listed.**

**Actual measurements of air radioactivity and effluent flow are the basis for reported emissions from continuously monitored sources. LLNL facilities that have continuously monitored discharge points are Buildings 166, 175, 231-vault, 251,331,332,419,490, and 491. For most of the discharge points, sample results are below the minimum detectable concentration (MIX) 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 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. Furthermore, even if the MDC values are used in calculations of the emission estimates for these facilities, an extremely conservative approach, the total dose attributable to LLNL activities is not significantly affected.**

**In 1996, samples from 8 emission points at three facilities, three in Building 175, three in Building 251 (the unhardened area) and two in Building 419, yielded gross alpha results greater than the MDC on 15% or more of the samples collected throughout the year. We use gross alpha as the primary indicator of potential emissions for operations, such as those at Buildings 175 which involve the use uranium, and Buildings 251 and 419, that involve the use of uranium and transuranic materials. Because of the number of samples with values above the MDC, we have taken a conservative approach and reported gross alpha and gross beta measurements as actual emissions. The gross alpha and gross beta emissions for Building 175 were determined to be**  $1.0 \times 10^{-7}$  Ci/y  $(3.8 \times 10^3$  Bq/y) and  $1.1 \times 10^{-6}$  Ci/y  $(3.9 \times 10^4$  Bq/y); for Building  $251,4.9 \times 10^{-7}$  Ci/y  $(1.8 \times 10^{4}$  Bq/y) and  $7.9 \times 10^{-6}$  Ci/y  $(2.9 \times 10^{5}$  Bq/y); and for **Building 419,**  $1.6 \times 10^{-7}$  **Ci/y**  $(5.9 \times 10^{3}$  **Bq/y) and**  $2.5 \times 10^{-6}$  **Ci/y**  $(9.2 \times 10^{4}$  **Bq/y).** Modeling these emissions resulted the following doses:  $2.3 \times 10^{-4}$  mrem  $(2.3 \times 10^{-3} \,\mu\text{Sv})$  for Building 175, 7.7  $\times 10^{-5}$  mrem  $(7.7 \times 10^{-4} \,\mu\text{Sv})$  for **Building 251, and 1.0**  $\times 10^{-4}$  mrem (2.3  $\times 10^{-3}$  µSv) for Building 419.

**We have looked into possible causes of the emissions being reported from Building 419 operations. We found that, because of the physical configuration of the sampling system and faulty seals in the samplers, some air from the workplace decontamimtion and decommissioning operations was being sampled by the continuous air samplers. New samplers were installed in October, and since that time, no gross alpha or gross beta analyses reported from the new samplers have indicated concentrations above the MDC.**

**Therefore, the estimated emissions listed in Attachment 1 are not indicative of emissions from the facility. Actual emissions are likely to be zero. Similarly, the emissions reported for Buildings 175 and 251 have not been confirmed to be emissions from facility operations. As in the case of Building 419, further investigation into the reported emissions is continuing and will likely include isotopic amlyses of selected samples and special air sampling. So it is possible that these emissions from Buildings 175 and 251 are due to naturally occurring, or background, radioactivity, or the facility exhaust configuration as previously mentioned. In any case, assessment of the emissions being reported for these facilities indicates the radiological dose is far less than the dose due to other facility emissions at the Livermore site.**

## **SECTION Ill. Dose Assessment**

## **Description of Dose Model**

**Estimates of individual and collective radiological doses to the public from all point sources and most diffuse sources at LLNL were obtained using the EPAdeveloped computer code CAP88-PC. The four principal pathways-internal exposures from inhalation of air and ingestion of foodstuffs and drinking water, and external exposures through irradiation from contamimted 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**  $m$ **rem/y** (1 mrem = 10  $\mu$ Sv). Separate doses for the Livermore site and **Site 300 from point-source emissions (i.e., stack emissions) and diffuse-source emissions at the two sites 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  $\mu$ 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 the 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  $(1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq})$ ; and **stack parameters, including height, diameter, and emission velocity.**

**Meteorological Data: All model runs used actual 1996 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 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: Because the EPA-mandated model CAP88-PC does not contain all the radionuclides in use at LLNL, it was necessary in a few cases to use surrogate radionuclides to estimate EDEs. Attachment 2**

**shows the surrogate radionuclide lists for CAP88-PC. In selecting the surrogates, the most-restrictive lung class (whether clearance from the lungs takes place in days, weeks, or years) was used. When possible, a surrogate radionuclide with similar chemistry and similar values for "amual limits of intake via inhalation and derived air concentration," as specified in the EPA's Federal Guidance Report No. 11 was used. CA.P88-PC contains a library of 265 radionuclides. In some cases, experimenters did not have isotopic analyses of mixtures of radionuclides and could only identify their radionuclide inventory as "gross alpha" or "gross beta. " In these cases, 23%?u was used as the surrogate for gross alpha and \$%r was used as the surrogate for gross beta in modeling efforts designed to provide conservative dose estimates.**

**Population Inputs: Population distributions centered on the two LLNL sites were compiled from 1990 census data. The population data files (distribution of population with distance and direction) used in the 1996 modeling effort are described in Section VI under "Collective Effective Dose Equivalent. "**

**Land-Use and Agricultural Inputs: Options for model inputs regarding agricultural characteristics and land use are established by the EPA, and the particular designation selected can strongly influence the ingestion dose received by the population being evaluated. Following our investigation in 1995 into the use of the various options, the "user entered" option was selected for the CAP88-PC modeling effort for 1996. The values entered corresponded to the "local agriculture" option (i.e., everything is home produced), with one exception —all milk consumed was assumed to be imported. The assumption that all milk comes from local cows is not supported by the agricultural activities conducted in the area. A detailed discussion of how the dose from tritium is calculated by CAP88-PC is presented in the LLNL NESHAPS 1995 Annual report (Gallegos et al., 1996, Lawrence Livermore National Laboratory, UCRL-ID-113867-96).**

**Emission Source Terms: The source term(s) from each emission point in the calculations was determined by one of two methods: For continuously monitored sources, the sampling data (curies released per unit time) for each radionuclide were used directly. For unmonitored facilities, the radionuclide inventories, together with the EPA-speafied fractions for potential release to air of materials in different physical states (solid, liquid, powder, or gas), in accordance with 40 CFR Part 61, Appendix D were used. The state-dependent release fraction was used to adjust (by multiplication) the total amual inventory to yield the potential amual release to air. If the material was an unconfined gas, then the fraction 1.0 was used; for liquids and powders,**

 $1.0 \times 10^{-3}$  was used; and for solids,  $1.0 \times 10^{-6}$  was used. In addition, emission**control abatement factors (40 CFR 61, Appendix D), when applicable, were applied. Each EIEPA filter stage was given a 0.01 factor, electrostatic precipitators, as well as venturi scrubbers, were each given a 0.05 factor, and each activated-charcoal filter was given a 0.1 factor. (However, abatement factors were not used to evaluate compliance with the 0.1 mrem standard that determines the need for continuous monitoring at a facility.) The use of actual monitoring data is much more direct, and presumably more accurate, than using assumptions based on inventory, release fractions, and emissioncontrol factors.**

**Site-Wide Maximally Exposed Individual: For LLNL to comply with the NESHAPS regulations, the LLNL site-wide maximally exposed individual camot receive an EDE greater than 10 mrem/y (100 I.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.**

**To determine the location of the 1996 SW-MEI, CAP88-PC results from multiple sources were combined. Sources were selected to include those expected to give significant contributions to the EDE. These included Building 331 point and area sources, Building 514 Tank Farm, and Building 612 area source. Because EDE results from CAP88-PC are relative to the location of the specified source, direct summing of results from multiple sources can only be accomplished using an interpolation method. To do this, the location of each selected source relative to a common location (the Liverrn.ore-site center) and a set of receptor locations (where the combined EDEs from the selected sources were to be evaluated), also relative to the site center, were specified in the modeling efforts that supported determimtion of the SW-MEI. The receptor locations included 48 equally spaced directions from the site center and 4 additional receptor locations along the eastern and southern Livermore-site boundaries. The interpolation method was used to calculate the EDEs for the desired set of receptor locations for each source. These resulting interpolated EDEs for each source, now for the same set of locations, were then summed, and the SW-MEI determined.**



Figure 4. Location of Site-Wide Maximally Exposed Individual (SW-MEI) at the Livermore site, 1996.

At the Livermore site, the SW-MEI for 1996 was located at the UNCLE Credit Union, about 10 m outside the controlled eastern perimeter of the site, as shown in Figure 4.



**Figure 5. Locationof Site-Wide Maximally Exposed Individual(SW-MEI) at Site 300, 1996.**

**At Site 300, the 1996 SW-MEI was located in an experimental area termed "Bunker 2" operated by Primex Physics International. Bunker 2 lies about 300 m outsid~ the east-\_central boundary of Ske 300, as shown in Figure 5. This bunker is 2.4 km east-southeast of the principal firing table at Building 801.**

**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).**

**Maximally Exposed Public Individual: To assess compliance with the requirement for continuous monitoring (potential dose greater than 0.1 rnrem/y {1.0 jLSv/y}), emissions must be individually evaluated from each point source; the location of the maximally exposed public individual (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 all emission points at the Livermore site and Site 300, calculations show that ground-level concentrations of radionuclides decline monotonically beyond LLNL boundaries.) As stipulated by the regulations in 40 CFR Section 61.93 (b)(4) (ii), 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 were applied. Attachment 1 provides, for each point source, the dose to the MEI and the distance and direction to the LLNL fence line where the MEI is located.**

**Special Modeling Challenges: Among the sources at LLNL, explosives tests using depleted uranium at Site 300 and difiuse sources at the two sites required special attention.**

**Site 300 Explosives Experiments: During Site 300 explosives experiments, the device containing depleted uranium is placed on an openair 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 cloud using the radionuclide and explosives inventories. Isotopic ratios for depleted uranium are used; the tluee uranium isotopes with atomic weights 238,235, and 234 occur in the weight percentages 99.8,0.2, and 5 x Id, respectively. Their masses are multiplied by their respective specific activities to determine the total number of curies for each isotope in the cloud. It is assumed that all the uranium is dispersed into the cloud, and the median particle size is assumed to be the CAP88-PC default value of 1 pm. 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-t~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 was submitted for approval in 1992, but LLNL was directed by EPA to use the CAP88-PC code for these calculations.**

**Diffuse Sources: Diffuse emissions are generally area sources external to buildings, as discussed in Section IV, below. The dose assessments for diffuse sources can be derived from modeling based on radionuclideinventory data, or can be determined from environmental-surveillance monitoring data.**

**Modeling Documentation: 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.**

## **Point Source Summary**

**The 1996 calculated EDE to the SW=MEI from Livermore-site point sources was 0.048 mrem (0.48 pSv). Emissions from the two 30-meter stacks at the LLNL Tritium Facility (Building 331) accounted for 0.045 mrem (0.45 @v). In 1995, emissions from the Tritium Facility resulted in a modeled dose of 0.017 mrem (0.17 I.&v). The relative increase in 1996 in emissions and dose occurred primarily as a result of glovebox decontamination and decommissioning activities.**

**The calculated EDE to the SW-MEI at Site 300 was calculated to be 0.033 mrem (0.33 p%) from point-source emissions. All of this EDE resulted from Building 801 and Building 851 firing-table emissions in the course of explosives experiments—55%** from the former and 45% from the latter. This **is an increase over the 0.020 mrem (0.20 p%) dose modeled for 1995; the increase is the result of an increase in the amount of depleted uranium used in experiments at the site.**

**All the dose evaluations from point-source emissions, and those from most diffuse sources discussed below, were made using the EPA-mandated CAP88- PC dispersion model. They result in levels of public exposure well below the EPA standard, which limits the whole-body EDE to members of the public from DOE activities to 10 rnrem/ y (100 @v/y). Discussion of the contribution to EDE to members of the public from diffuse sources is presented in Section IV.**

## **SECTION IV. Additional Information**

## **Construction and Modifications**

**During 1996, no construction projects or modifications were completed for which approval to construct or modify was required or waived under 40 CFR 61.96. Only maintenance, repair, and replacement activities, as well as those considered normal or routine, were conducted. Proposed facilities and significantly modified operations are assessed for NESHAI?S requirements during the National Environmental Policy Act (NEI?A) process. Under NEI?A, all proposed projects or actions that might involve NESHAPS issues or concerns-not just pertaining to radionuclides but to air toxics 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.**

### **Unplanned Releases**

**There was one unplanned atmospheric radionuclide release from the Livermore site in 1996. On April 15, 1996, approximately 1.5 L of contaminated oil leaked from a 55-gallon drum in the 514 yard. The leaked material contained about 6 nCi of depleted uranium and was dispersed over a 0.37 mz area.** The emission resulted in a calculated  $4.9 \times 10^{-9}$  mrem  $(4.9 \times 10^{-8} \text{ }\mu\text{Sv})$  dose **to the site-wide maximally exposed individual (estimated using CAP88-PC). There were no unplamed atmospheric releases at Site 300 in 1996.**

## **Diffuse Source Dose Assessments**

**Diffuse, or non-point, sources are difficult to quantify. There are no EPAmandated methods for estimation or measurement, although LLNL did review a second draft of EPA guidance on this topic during 1994. At this time, however, dose calculations associated with this type of source remain left to the discretion of the DOE facility. Livermore-site and Site 300 diffuse sources are described separately.**

### **Livermore-Site Diffuse Sources**

**The dose calculations from 1996 diffuse sources at the Livermore site required three different modeling approaches. Building 331, Building 292 and Building 612 Yard needed faality persomel knowledge and envirorunentalsurveillance data to estimate emissions; Building 514 required radiologicalinventory data and CAP88-PC modeling ,techniques; and in the Southeast Quadrant, data from ambient-air monitoring were used to calculate the dose. The unplamed release at Building 514, discussed previously, was also a diffuse source release.**

**Building 292: Elevated tritium concentrations in soil moisture near Building 292 resulted from a historic leak in an underground retention tank. This contamination has resulted in diffuse tritium emissions from evaporation of soil moisture and transpiration from vegetation. A surveillance air monitor has been placed near Building 292 to provide continuous measurements of tritiuin near this source. The median annual** concentration of tritium in air for 1996 in this area was 0.0039 pCi/L  $(1.4 \times 10^{-4})$ **Bq/L). These data were used to calculate the total tritium emissions from the area, using a conservative approach that assumed the source to be 10 m east of** the air sampler. With this assumption, a diffuse source emission of  $1.4 \times 10^{-3}$  $Ci/y$  **(5.2**  $\times$  10<sup>6</sup>  $Bq/y$ ) would have been required to produce the concentrations **measured at the air sampler. This source term produced a calculated 1996 dose to** the SW-MEI from the Building 292 area of  $3.6 \times 10^{-7}$  mrem  $(3.6 \times 10^{-6} \mu Sv)$ .

**Building 331: As the Tritium Facility (Building 331) undergoes both decommissioning/decontamination and redirection of its research and development efforts, tritium-contaminated equipment slated for disposal is removed from the building, packaged in a waste-accumulation area, and sent to Hazardous Waste Management Division (HWM) facilities. During 1996, outgassing from such waste processing released approximately 3 Ci (1.1 x 1011 Bq) of tritiurn to the atmosphere outside Building 331. The estimated releases were derived from measurements of surface contamination on the material, process and faality knowledge, and environmental-surveillance measurements. The estimated 3 Ci (1.1x 1011 Bq) release was modeled in CAP88-PC as a 1 m2 area source, leading to a calculated 1996 dose to the**  $SW-MEI$  of  $3.1 \times 10^{-3}$  mrem  $(3.1 \times 10^{-2} \,\mu Sv)$ .

**Building 514: bother potential source of diffuse emissions of a variety of radionuclides was HWM waste-storage and treatment operations. Building 514 houses the HWM "tank farm; consisting of six 7,170-liter tanks with**

**ancillary equipment such as pumps, mixers, probes, and a bulking station. The tanks are used to store and treat liquid and solid radioactive and/or mixed wastes. Treatment is performed on a batch basis. Chemicals and waste are added to the tanks to achieve the desired treatment objectives. A 1996 radionuclide inventory was conducted for the facility to determine the diffuse source term (Attachment 1). During 1996, hazardous waste operations increased treatment of legacy waste, including materials containing higher levels of cesium-137, thorium-228, uranium-238 and plutonium-239 than had been treated in 1995. CAP88-PC modeling gave a 1996 EDE for the Tank Farm to** the SW-MEI of  $1.6 \times 10^{-2}$  mrem  $(1.6 \times 10^{-1} \,\mu\text{Sv})$ .

**Building 612 Yard: The Building 612 Yard is a potential source of diffuse emissions of tritium. This area is dedicated to hazardous-waste-, radioactivewaste-, and mixed-waste-management activities. The yard consists of several areas where waste containers are stacked outdoors. Many of these containers are not air tight and outgas tritium. A surveillance air monitor has been placed in the Building 612 Yard to provide continuous measurements of tritium near this source. The median amual concentration of tritium in air for 1996** in this area was  $0.169$   $pCi/L$   $(6.3 \times 10^{-3}$   $Bq/L)$ . 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. The assumption that the source is 60 m from the sampler was changed from 120 m because waste repackaging was being conducted in the yard within 60 m of the sampler. Using 60 m yielded a reasonable prediction** of the concentration of tritium at another nearby tritium sampler  $(3.0 \times 10^{-3})$ **pCi/L predicted versus 2.1 x l(ha pCi/L measured at SALV monitoring location). With this assumption, a diffuse source emission of 3.0 Ci/ y (1.1 x 1011 Bq/y) was required to produce the concentrations measured at the air sampler. This source term produced a calculated 1996 dose to the SW-MEI from** the Building 612 Yard of  $2.5 \times 10^{-2}$  mrem  $(2.5 \times 10^{-1} \,\text{\ensuremath{\mu}Sv})$ .

**Southeast Quadrant: The Southeast Quadrant of the Livermore site has elevated levels of 239Pu in the surface soil (from historic waste-management operations) and air (presumably from resuspension). A high-volume airparticulate sampler is located adjacent to the UNCLE Credit Union (the location of the SW-MEI) to monitor the 239Pu levels in this area. Monitoring data from this air sampler were used as a direct measurement of potential dose via the air pathway. The mean amual concentration of ~9Pu in air of**  $3.0 \times 10^{-19} \mu$ Ci/mL  $(1.1 \times 10^{-14}$  Bq/mL), the dose-conversion factor of  $3.08 \times 10^5$ **mrem/pCi (8.33x 10\_s Sv/Bq) from Federal Guidance Report No. 11, EPA-520/l-88-020, U.S. Environmental Protection Agency (1988) for 239Pu,**

**and the standard-man breathing rates of 8.4 x 109 mL/y were used to calculate the estimated EDE** of  $7.8 \times 10^{-4}$  **mrem**  $(7.8 \times 10^{-3} \text{ uSv})$  for 1996.

## **Site 300 Diffuse Sources**

**Diffuse sources at Site 300 involve tritium and uranium. 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. Information provided in the Final Site Wide Remedial Investigation Report (Webster-Scholten, Ed., 1994, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-AR-108131) was used in the diffuse-source evaluations. In the course of the remedial investigation, the rate of intermedia migration and the exposure-point concentrations of contaminants were evaluated. Tritium and Z3SU were identified as contaminants of potential concern at six locations.**

**Tritium contamination is well characterized at Site 300. Five diffuse tritium sources are discussed individually. Uranium, on the other hand, is not as well characterized. Diffuse uranium sources were treated collectively in a resuspension calculation, presented following the individual tritium discussions below.**

**Tritium gas and solid tritium (Li3H) were components of explosives assemblies tested on the firing tables during past experiments. Most of the gaseous tritiurn escaped to the atmosphere during the tests, but some of the solid LisH remained as residue in the firing table gravel. Rainwater and dustcontrol 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 source terms for diffuse emissions of tritium to the atmosphere at Site 300.**

**Pit 7 Complex: The Pit 7 Complex is an area where four landfills were established. All the pits contain gravel and debris generated from explosives tests conducted at the Building 850 and 851 firing tables. Tritiurn is a known residue in this waste, and tritiurn contamination in both subsurface soils and ground water in the area has been characterized. Tritium in subsurface moisture can evaporate to the atmosphere. The affected area is estimated at 18,000 m2. Tritiurn flux was calculated from tritium activity data obtained from subsurface soil samples collected at depths from 0.15 to 3 m, and was**

**estimated to be**  $8.3 \times 10^{-1}$  **Ci**  $(3.1 \times 10^{10}$  Bq) for 1996. A correction (decrease) in **source term from the time the samples were taken accounts for both radioactive decay and loss of the original tritium activities in the soil due to evapotranspiration. In addition, well purge water (water collected from wells and left to evaporate to the atmosphere before ground water sampling) in this area often contains elevated levels of tritiurn. During 1996, ground water monitoring operations involved purging three wells with tritium levels above 20,000 pCi/L (740 Bq/L). The evaporation of this water to the atmosphere represents another component of the Pit 7 diffuse emission source term;** it was estimated **to contribute**  $1.3 \times 10^8$  pCi  $(4.8 \times 10^6$  Bq) during **1996. This emission estimate is based on the total volume of water purged during monitoring activities and the detection levels reported in the 1996 LLNL Site 300 Compliance Monitoring Program Report (Chris tofferson and MacQueen, 1997, Lawrence Livermore National Laboratory, UCAR-10191-96-4). The 1996 calculated EDE to the SW-MEI from the** combined **tritium** emissions at the Pit 7 Complex was  $3.0 \times 10^{-5}$  mrem  $(3.0 \times 10^{-4} \,\text{uSv}).$ 

**Well 8 Spring: Tritium released to the soils, and eventually to the ground water, near the Building 850 firing table has been transported to areas where ground water flows near the surface and can evaporate to the atmosphere. Such is the case at the Well 8 Spring, where ground water is very shallow. To estimate tritium flux from this spring, tritium activity data obtained from water samples collected at the spring were used. These data were corrected for radioactive decay, but not for removal by evapotranspiration because the spring was assumed to have a continuous source of tritiated water for the period in question. The affected area of the spring was estimated at 9.3 m% and the 1996 source term was estimated to be 2.1** *x 10–3 Ci* **(7.8** *x 107***Bq). The 1996 calculated EDE to the SW-MEI from tritium emissions at the Well 8 Spring was**  $1.3 \times 10^{-7}$  mrem  $(1.3 \times 10^{-6} \,\text{\upmu Sv})$ .

**Building 802: Tritium in the subsurface soils near the Building 802 firing table may evaporate to the atmosphere. The affected area was estimated to be 900 mz. Tritium flux was calculated from tritium activity data obtained from subsurface soil samples collected at depths from 0.15 to 3 m. The tritium emission rate from subsurface soils to air was the product of the spatialaverage tritium flux, the natural flux of water, the fraction of tritium in the water, and the affected surface area. The 1996 tritium emissions from this source** were estimated to be  $5.0 \times 10^{-4}$  Ci  $(1.9 \times 10^{7}$  Bq). The 1996 calculated EDE **to** the SW-MEI from **tritium** emissions at Building 802 was  $5.4 \times 10^{-8}$  mrem  $(5.4 \times 10^{-7} \,\mu\text{Sv})$ .

**Building 850:** Approximately  $2.1 \times 10^4$  Ci  $(7.8 \times 10^{14}$  Bq) of tritium was **expended in explosives tests at the Building 850 firing table in the past. Although a significant source of tritium (firing-table gravel) was removed from the area during 1988, tritiurn remains in subsurface soils beneath the Building 850 firing table, sand pile area, and lower corporation yard. Tritium in the subsurface soils in the vicinity can evaporate to the atmosphere. The affected area was estimated to be 20,000 mz. The tritium flux and tritium emission rate from subsurface soil to air were calculated as in the Building 802 case. The 1996 tritium emissions from this source were estimated to be**  $1.0 \times 10^{-1}$  Ci (3.7  $\times$  10<sup>9</sup> Bq). The 1996 calculated EDE to the SW-MEI from **tritium** emissions at Building 850 was  $5.7 \times 10^{-6}$  mrem  $(5.7 \times 10^{-5} \mu Sv)$ .

**Building 851:** About  $1.0 \times 10^3$  Ci  $(3.7 \times 10^{13}$  Bq) of tritium were expended **during past explosives research conducted at the Building 851 firing table. Although gravel was removed routinely from the area, subsurface soil below the firing table contains residual tritium in soil moisture that can evaporate to the atmosphere. The affected area was estimated to be 470 mz. The tritium flux and tritium emission rate from subsurface soil to air were calculated as in the Building 802 case. The 1996 tritium emissions from this source were estimated to be**  $2.9 \times 10^{-4}$ Ci  $(1.1 \times 10^{7}$  Bq). The 1996 calculated EDE to the **SW-MEI** from **tritium** emissions at Building 850 was  $1.8 \times 10^{-8}$  mrem  $(1.8 \times 10^{-7} \,\mu\text{Sv})$ .

**Resuspension of Depleted Uranium 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 (IVU) (Gallegos et al., 1996, Lawrence Liverrnore 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**

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

**where y is the fraction (by weight) of uranium contributed by operations, CU is composite uranium (both DU and NV), M(CU-235) the mass of U-235 in the composite (measured) uranium, and M(CU-238) the mass of U-238 in the composite (measured) uranium. (For derivation of the equation see the 1995 NESHAI?S amual report, referenced above.) This equation is used for those months in which explosives shots were not conducted.**

**Using these calculations to apportion the M(CU) for 1996, and excluding the appropriate months, we obtain an annual average concentration of DU in air from resuspension** of  $1.13 \times 10^{-12}$  g/m<sup>3</sup>. Using the fractions 0.998,0.002, and **0.000005 to represent the amounts of 238U, 235U, and 2UU; specific activities of**  $3.32 \times 10^{-7}$ ,  $2.13 \times 10^{-6}$ , and  $6.16 \times 10^{-3}$  Ci/g for  $238$ U,  $235$ U, and  $234$ U; a yearly **inhalation rate of 8400 m3/ y, and dose conversion factors from EPA Regulatory** Guide 11 of  $1.18 \times 10^{-11}$ ,  $1.23 \times 10^{-11}$ , and  $1.23 \times 10^{-11}$  mrem/Ci; we **obtain a total dose for resuspended DU of 4.1 x lfi mrem for 1996.**

## **Total Dose Estimate and Comparison with Previous Years' Data**

**For the Livermore site, the dose calculated for the SW-MEI from diffuse emissions in 1996 was 0.045 mrem (0.45 @v). When point and diffuse** sources were combined, the total annual dose was 0.093 mrem (0.93  $\mu Sv$ ). **Therefore, the relative contributions to the total were 52% from diffuse sources and 48% from point source emissions. The total dose to the Site 300 SW-MEI from Site 300 operations in 1996 was 0.033 mrem (0.33 @v). Pointsource emissions from firing-table explosives experiments accounted for 0.033 rnrem (0.33 @v), or 9970, of this total, while 0.00045 mrem (0.0045 p%), or 1%, was contributed by diffuse sources. Table 4 presents the facilities or sources that account for 90% or more of the doses for the Livermore site or Site** *300* **SW-MEI.**

**Comparison of the 1996 total dose estimate with that of previous years can be made by reviewing the information presented in Table 5. No diffuse emissions were reported at !Me 300 for years before 1993, so comparison for total dose can only be made with the values for 1993,1994, and 1995; in addition, diffuse source doses were not reported separately from the total dose for the Livermore site for 1990 and 1991. The increased point source**

**contribution to dose for the Livermore site for 1996 compared to 1995 is attributed to glovebox decontamination and decommissioning operations at Building 331. The increased diffuse source contribution to dose is attributed to increased treatment of legacy waste at the Building 514 Tank Farm.**

**Table 4. List of facilities or sources whose emissions account for 90% or more of the doses for the Livermore site and Site 300 SW-MEI**



**Table 5. Doses (in mrem) calculated for the Site-Wide Maximally Exposed Individual for the Livermore site and Site 300, 1990 to 1996.**



<sup>a</sup>Diffuse source doses were not reported separately from the total dose for the Livermore site for 1990and 1991.

b<sub>No</sub> diffuse emissions were reported at Site 300 for years before 1993.

## **SECTION V. Certification**

**I certify under penalty of law that I have personally examined and am familiar with the information submitted herein, and based on my inquiry of those individuals immediately responsible for obtaining the information, I believe that the submitted information is true, accurate, and complete. I am aware that there are significant penalties for submitting false information, including the possibility of fine and imprisonment.**



**I certify under penalty of law that this document and all attachments were prepared under my direction or supervision in accordance with a system designed to assure that qualified persomel properly gather and evaluate the information submitted. Based on my inquiry of the person or persons who manage the system, or those persons directly responsible for gathering the information, the information submitted is, to the best of my knowledge and belief, true, accurate, and complete. I am aware that there are significant penalties for submitting false information, including the possibility of fine and imprisonment for knowing violations.**

**Name: Dennis K. Fisher Assoaate Deputy Director for Operations Lawrence Livermore National Laboratory 7000 East Avenue Livermore, CA 94551**

**Signature: Date:** 6/13/97 **Dennis K. Fisher**

## **SECTION V1. Supplemental Information**

## **Collective Effective Dose Equivalent**

**Population doses, or collective EDEs, for both LLNL sites were calculated out to a distance of 80 km in all directions from the site-centers using CAP88-PC. As noted earlier, CAP88-PC evaluates the four principal exposure pathways: ingestion through food and water corqumption, inhalation, air immersion, and irradiation by contaminated ground surface.**

**In 1996, we reconstructed the population distributions centered on the two LLNL sites. These population distributions, as were the previous distributions, are based on 1990 census data. However, the new distributions were developed using commercially available, computer-map-based population data and the geographic information system software, ArcView@. The population for each sector segment was determined by selecting census block level data for that segment. Key population centers affected by LLNL emissions are the relatively nearby communities of Livermore and Tracy, and the more distant metropolitan areas of Oakland, San Francisco, and San Jose, as well as the San Joaquin Valley communities of Modesto and Stockton. Within the 80 km outer distance specified by the EPA, there are 6.3 million residents included for the Livermore site collective dose determination, and 5.2 million for Site 300. Our population data files (distribution of population with distance and direction) are shown in Tables 6 and 7 for the Livermore site and Site 300, respectively.**

**For the evaluation of the population dose, as distinct from the individual dose, all food (and in particular milk) was assumed to be produced locally. This decision was made because, although there are no commercial dairy animals within the distances used to evaluate individual doses, many dairy animals live within 80 km of the Livermore site and Site 300.**

**The collective EDE, which is the sum of the individual doses to all 6.3 million people within 80 km of the Livermore site, due to 1996 Livermore-site operations was 1.1 person-rem (0.01 1 person-Sv). This number can be compared to the collective EDE from natural background radioactivity for 6.3 million people of 1.88 x 106 person-rem (1.88 x 104 person-Sv). The 1996 collective EDE value is greater than the 1995 value of 0.59 person-rem (0.0059 person-Sv). The reason for the increase in the collective EDE is greater stack releases in 1996 than in 1995. Stacks release effluents at considerable speed high above the ground, allowing contaminants to be more readily transported toward population centers downwind.**



**Table 6. Population distribution for LLNL's Livermore site, based on 1990 census information. Values are population in sector segment\$ bounded by the indicated inner and outer radii, for each of sixteen 22.50-sector directions.**

**Table 7. Population distribution for LLNL's Site 300, based on 1990 census information. Values are population in sector segments bounded by the indicated inner and outer radii, for each of sixteen 22.50-sector directions.**



**The corresponding collective EDE from Site 300 operations in 1996, 10.0 person-rem (0.010 person-Sv), was due to point-source emissions. The total collective EDE value is very similar to the 1995 value of 7.7 person-rem (0.77 person-Sv). These differences are the result of differences in the amounts of high explosives and depleted uranium used each year in explosives experiments.**

**The larger value for Site 300 compared to the Livermore site is traceable primarily to the highly conservative assumptions about the Site 300 explosives experiments, especially regarding the fraction of radioactive material that is aerosolized and the height and trajectory of the explosivedebris cloud. This conservative modeling methodology over-predicts the quantity of radionuclides released to air by at least a factor of five, we believe, and over-estimates the long-range dispersal of material in these experiments. In 1992, we submitted to EPA a modeling protocol designed to treat the transient expl~sive experiments more realistically than does CAP88-PC, but this protocol was not accepted.**

## **Compliance with 40 CFR 61 Subpart H (61.93)**

**Calculations of effective dose equivalents for ail Livermore-site and Site 300 facilities having the potential to release radionuclides to the atmosphere have been completed. Annual doses from actual total emissions of all facilities during 1996 were found to be well below the 10 mrem (100 p%) NESHAPS dose standard. Tritium accounted for most of the Livermore-site calculated dose, while at Site 300 practically the entire calculated dose was due to the isotopes 238U, 235U, and 2~U, in depleted uranium.**

**Based on potential emissions without control devices and EPA agreement, 22 emission points in three facilities at the Livermore site will maintain continuous monitoring systems in compliance with NESHAPS requirements. Continuous monitoring will be maintained in Building 332 and the seismically hardened area of Building 251 instead of a modeling or measurement effort to demonstrate the actual need for monitoring. Continuous monitoring at Building 166 will be maintained, based on EDEs determined from modeling of the building radionuclide inventory. Continuous monitoring is being continued at Building 331 even though the EDEs that result from measured emissions do not require monitoring under 40 CFR 61.93(b).**

**Several other Livermore-site facilities (Buildings 175,231,251 unhardened, 419, 490, and 491) also will maintain continuous-monitoring systems; however, calculations using umbated potential emissions resulted in EDEs of less than**

**0.1 mrem/ y (1 @v/y) for the emissions from each of these facilities. While this monitoring also will be continued, it is not required under NESHAPS.**

**For facilities having discharge points without continuous monitoring, the requirement for continuous monitoring was individual y evaluated. The evaluation was based on unabated emissions, even if emission-control systems existed. No additional facilities at either LLNL site were found to require continuous monitoring.**

## **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 amual reports from research experiments at the Livermore site did not occur in 1996.**

## **Site Periodic Confirmatory Measurements**

**LLNL uses a graded approach to determine the required level of periodic confirmatory measurements. The greater the calculated EDE, the more intensive the measurements will be. LLNL invokes a four-tier approach: (1) continuous monitoring at selected facilities, (2) annual effluent sampling, (3) general surveillance monitoring, and (4) site-specific surveillance monitoring, as described below.**

**Continuous Monitoring: There are currently nine buildings (Buildings 166, 175,231,251,331,332, 419,490, and 491) at the LLNL site that have radionuclide air-monitoring systems. These buildings are listed in Table 8, along with the number of samplers, the types of samplers, the analytes of interest, and the number of monitored discharge points at the building. In all,** **there are 103 samplers operating continuously. Many would operate from emergency power systems if normal power were lost.**





Note: "CAM" denotes Eberline continuous air monitors.

aAlternate**blower system measured by the same sampler.**

**bAlarmed systems.**

**Air samples for particulate emissions are extracted downstream of HEPA filters and prior to the discharge point to the atmosphere. Particles are collected on membrane filters. The sample filters are removed and analyzed for gross alpha and beta activity on a weekly or hi-weekly frequency depending on the facility. In most cases, simple filter-type aerosol collection systems are used. However, in some facilities, alpha continuous-air monitors**

**(CAMS) are used for sampling. In addition to collecting a sample of particles, the CAM units provide an alarm capability for the facility in the event of a release of alpha activity.**

**Detection of gross alpha and beta activity resulting from particles collected on the air filters is accomplished using gas-flow-proportional counters. Analysis is delayed for at least four days from the end of sample collection to allow for the decay of naturally occurring radon daughters. For verification of the operation of the counting system, calibration sources, as well as background samples, are intermixed with the sample filters for analysis. Analysis is performed by the Radiological Measurements Laboratory (RML) in the Hazards Control Department (HCD).**

**Each stack of the Tritium Facility (Building 331) is monitored for tritium release by both a continuous-monitoring alarm system and continuous molecular-sieve samplers. The alarmed samplers, Overhoff ion chambers, provide real-time tritium concentration release levels (HT and HTO). The sieve samplers, which can discriminate between tritiated-water (HTO) vapor and molecular tritium (HT), provide the values used for environmental reporting and are exchanged weekly or hi-weekly depending on the rate of tritium releases expected from planned work. Each sieve sampler (unalarmed) is in parallel with an alarmed monitor and consists of two molecular sieves. The first sieve collects tritiated water vapor; then a palladium-coated catalyst converts molecular tritium to tritiated water, which is then collected on a second sieve. The molecular sieve samples are submitted to the Hazards Control Analytical Laboratory where they are installed 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 the Hazards Control Department Health Physicist responsible for each facility and an Environmental Protection Department Environmental Analyst.**

**Annual Effluent Sampling: For point sources where the fence line EDE is between 0.1% and 1% of the NESHAPS emission standard of 10 rnrem/y (100 pSv/y) (between 0.01 and 0.1 rnrem/y or 0.1 and 1.0 pSv/y), and no existing monitoring system is in place, LLNL strives to perform amual confirmatory sampling. Measurements of the effluent from such sources are plamed for the year following the annual dose assessment. These measurements are planned to be taken downstream of any emission control**

**devices and when operations are being performed. In 1995, no point sources that were not already subject to continuous monitoring fell into these criteria for sampling. We, nonetheless, conducted three evaluations for NESHAPS periodic confirmatory measurements in 1996. These include confirmatory sampling of discharge points at Buildings 177 and 490, and a comparison of the inventory approach with workplace sampling at Building 298.**

**Building 177: At Building 177, we performed air sampling of a stack exhaust that vents a uranium dissolution process. Three samples were taken over a 12-day period while dissolution operations were being performed. To obtain an appropriate background, one sample was also taken while dissolution operations were not being performed. Samples of particulate emissions were taken by single-probe, isokinetic sampling of the exhaust using 47-mm-diameter, cellulose membrane filters. The filters were analyzed for gross alpha and gross beta activity as well as total uranium by induced coupled plasma mass spectroscopy (ICPMS). The average measured concentration of uranium in the exhaust air as determined by ICPMS results was 5.7 x 10-10 g/ins. Assuming the operations were performed continuously the entire year**, the estimated **emission** is  $1.9 \times 10^{-2}$  g of **uranium**. Modeling of **this source was performed with CAP88-PC. The resulting dose to the MEI and** SW-MEI is 2.1  $\times$  10<sup>-6</sup> and 1.3  $\times$  10<sup>-7</sup> mrem respectively. Thus, this source is **not a significant contributor to the Livermore site dose nor is continuous monitoring of the stack emissions required by regulation.**

**Further, the estimated emission based on the above sampling results was compared to the inventory approach for NESHAPS source evaluation. This was done using the sampling results and knowledge of the amount of uranium processed during the sampling period. A release fraction of**  $5.7 \times 10^{-5}$  to  $5.7 \times 10^{-6}$  was estimated. These fractions are 17 to 170 times **lower than 0.001, the release fraction for a liquid that would be used by the approach given in Appendix D of 40 CFR 61 Subpart H. Therefore, this sampling effort indicates that the inventory-based approach is conservative for this type of operation.**

**Building 298: Building 298 houses operations involving the use of tritium. In 1996, estimates of emissions were made using continuous samplers that are in place for workplace concentration measurements and compared to the inventory approach. The average concentration as measured by** the workplace samplers is  $3.5 \times 10^{-3} \mu\text{Ci/m}^3$ . Using estimates of ventilation **rates for the rooms, the estimated tritiurn release was calculated to be 0.044 Ci for the year. This quantity is less than 0.05 Ci which was provided by the**

**inventory-based approach. CAP88-PC modeling of the workplace-based source term as an area-type release resulted in an MEI dose of 8.3 x 10-4 mrem and a SW-MEI dose of 1.9 x 10-5 mrem. Therefore, no continuous sampling of these operations is required.**

**Building 490: At Building 490, we performed periodic confirmatory measurements of a vacuum pump exhaust from an operation using uranium. Since this type of exhaust is intermittent depending on the operation of the process, a special, HEl?A-filtered sampling train was placed on the exhaust to facilitate continuous sampling. The sampling train provided filtered air for the continuous sampler during times when there was little or no flow from the pump exhaust. Four samples were taken over a 2-week period while experimental operations in the facility were being conducted. Background measurements taken of the sampling train without the vacuum pump exhaust comected were also made. Samples of particulate emissions were collected on either 47-mm diameter cellulose membrane or glass fiber filters. The filters were analyzed for gross alpha and gross beta activity and for total uranium by ICPMS. The average measured concentration as determined by ICPMS results was not distinguishable, or statistically different, from the background measurements. (As for continuously monitored stacks, for which measured emissions are not significantly higher than background, thesq emissions are considered to be zero.) Since the background concentration measurements have a lower limit of sensitivityy that would result in a MEI dose far less than the 0.1 mrem requirement for continuous sampling, the exhaust from this operation does not require continuous sampling. Similarly, there is no significant contribution to the SW-MEI dose.**

**General Surveillance Monitoring: Surveillance air monitoring for tritium and radioactive particles has been in place since the 1970s and will continue. LLNL currently maintains eight continuously operating, highvolume, air-particulate samplers on the Livermore site, nine in the Livermore Valley, eight at Site 300, and one in Tracy. LLNL also maintains eleven continuous y operating airborne-tritium samplers on the Livermore site and six samplers in the Livermore Valley, The samplers are positioned to ensure reasonable probability that any significant airborne concentration of particulate and tritium effluents resulting from LLNLoperations will be detected. The data from this 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. This network allows for direct measurements of the overall impact of LLNL operations. Data horn**

**this network are presented in the LLNL Environmental Report, which is prepared annually and available to the public. (Harrach et al., Environmental Report for 1996, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-50027-96, to be published in October 1997.)**

**Site-Specific Surveillance Monitoring: Surveillance air monitors are placed near diffuse emission sources, such as those associated with Buildings 292,331,514, and 612, as well as in and around the Southeast Quadrant of the Livermore site. The data from these monitoring networks provide continuous measurements of the concentrations of specific radionuclides present in the air near these sources and allow a direct and accurate determination of their environmental impact. This practice will continue at these locations. It has been determined that the use of site-specific surveillance monitoring for Site 300 diffuse sources of tritium is umecessary because of the low emissions and resultant dose values displayed in Attachment 1.**

## **Status of the NESHAPS QA Program**

**The LLNL NESHAPS Quality Assurance (QA) Program is a multiorganizational effort that relies on the Quality Assurance/Quality Control programs that are in place at the LLNL facilities with continuous airmonitoring systems, the Radiological Measurements Laboratory (RML) and the Analytical Laboratory of the Hazards Control Department, and the Environmental Protection Department (EPD).**

**Facility Safety Procedures (FSPS), Safety Analysis Reports (SARs) and QA Manuals for monitored facilities describe their organizational structures, responsibilities for sampling locations used for continuous air monitoring, and the procedures to be followed in the case of unplamed radionuclide releases. For example, the FSP for the Plutonium Facility (Building 332) describes in detail the procedure for responding to detection of radioactive materials in a release from the stacks. These documents also describe the sample-collection systems for both real-time and passive (i.e., not alarmed) air-monitoring systems, and procedures to be used for measuring flow rates, sampling, and calibration.**

**The RML Quality Assurance Program describes laboratory-analysis procedures, precision, accuracy and completeness objectives, sample-tracking procedures, quality-control (QC) sampling, sample handling, and data reporting. For example, the Gross Alpha-Beta Procedures Manual of the RML describes operational procedures for analyzing the air sampler filters for radioactivity.**

**EPD, which is responsible for NESHAPS modeling and reporting, also operates under a Quality Assurance Management Plan and associated procedures. Detailed records are kept of all measurements, CAP88-PC model runs, and calculations, and selected model runs are validated. The Terrestrial and Atmospheric Monitoring and Modeling Group (TAMM) of EPD is responsible for modeling and reporting radionuclide emissions for NESHAPS compliance. TAMM members continue to refine mechanisms that ensure they are informed whenever new operations are proposed, significant changes in radionuclide inventories occur, or existing operations are modified so that NESHAPS modeling can be performed and appropriate action taken. All NESHAPS calculations are archived with the supporting information used to make the calculations.**

## **Quality Control (QC) for 1996 Radiological Inventory Update and Modeling**

**Radiological Inventory Update QC: Approximately 15% of the 61 Livermore-site facilities that completed radiological-inventory updates in 1996 were randomly selected for validation. For this QC check, radiological inventories from eleven potential emission points were selected for validation five from Building 151, two from Building 222, two from Building 177, and one each from Building 446 and the Building 514 Tank Farm. An EPD Environmental Analyst contacted the responsible party who signed the NESHAPS Inventory Forms and physically visited and inspected the facilities to verify inventory data. The responsible party was asked to demonstrate how he/she arrived at the data submitted on the original inventory form. Stack parameters also were verified. The QC data were compared to the original data. The accuracy of the inventory data was confirmed.**

**Modeling QC: Fifteen percent of the CAI?88-PC modeling runs were selected for validation by a second analyst using a different computer and copy of CAP88-PC. The analyst performing this QC effort ran the model following independent gathering of radionuclide inventories and stack data from the NESHAPS Inventory Forms and pertinent distances from site maps. The QC modeling verified the values from the original CAI?88-PC modeling runs. The data that are presented in the attached spreadsheet are as accurate as possible, demonstrating that quality objectives are being met.**

## **EPA Compliance Evaluation investigation**

**On May 28,1996, the U.S. EPA, Region IX conducted a Compliance Evaluation Investigation at Buildings 332,255,253, and 331. LLNL persomel made a**

**number of presentations during the course of the inspection including summaries of stack monitoring systems, the HEPA falter testing program, the Hazards Control Radiological Measurements Laboratory operations, Building 332 operations overview and faality tour, and NESHAPS compliance overview. LLNL was found to be in compliance with 40 CFR 61 Subpart H and no additional compliance activities were required.**

**In December 1996, LLNL held an informational meeting with U.S. EPA Region IX staff to discuss plamed construction of the Decontamination Waste Treatment Facility at the Livermore site and Contained Firing Facility at Site 300. Potential NESHAPS issues and plans for monitoring at the facilities were discussed. Additional subjects covered at the meeting included periodic confirmatory measurements, the potential for the use of de nzinirnis values in determining NESHAI?S compliance, and the status of the delegation of NESHAPS regulatory oversight to the State of California.**

### **Uranium Physical State Exemption**

**In discussions between LLNL and U.S. EPA staff, LLNL persomel pointed out the burden of assuming, as required by 40 CFR Part 61 Subpart H Appendix D, that all materials heated in excess of 100"C are hi a gaseous physical state. Such an assumption is quite unrealistic for uranium and other refractory metals. Uranium has a melting point of 1132°C and a boiling point of 3818"C. The effect of the assumption that all materials are gaseous when at temperatures above 100"C is to apply a physical state factor of 1, rather than**  $1 \times 10^{-3}$  for liquids and  $1 \times 10^{-6}$  for solids. Evaluation of a new source which **involved heating of uranium, and using the required physical state factor, could lead to a dose estimate that requires continuous monitoring of the source, whereas using a physical state factor based on the actual physical state of the materials would not. On July 25, 1996, LLNL requested an exemption from the temperature-based physical state assumptions for uranium. U.S. EPA granted approved alternative emissions factors for elemental uranium as follows: an emission factor of 1 x lb can be used for elemental uranium heated** at **temperatures** below 1100°C, an emission factor of  $1 \times 10^{-3}$  can be **used for elemental uranium heated at temperatures below 3000"C, and an emission factor of 1 shall be used for temperatures greater than 3000°C. These factors are allowed provided that the uranium is not intentionally dispersed to the environment and that the processes do not alter the chemical form of the uranium. We are working towards similar exemptions for uranium compounds.**

### **Guidance for Interpreting Attachment f**

**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:**

- $\bullet$ **Building and room number(s)**
- 0 **Specific stack identification code(s)**
- $\bullet$ **Generalized, operations in the room(s) or area(s)**
- **e Radionuclides utilized during 1996**
- **o Annual radionuclide inventory with potential for release (by isotope, in curies)**
- **\* Physical-state factors (by isotope)**
- **Stack parameters**
- **o Emission-control devices and emission-control-device abatement factors**
- **e Estimated or measured amual emissions (by isotope)**
- **Distance and direction to the site-wide maximally exposed individual (SW-MEI)**
- **o Calculated EDE to the SW-MEI**
- **e Distance and direction to the maximally exposed individual for that specific source (MEI)**
- **e Calculated EDE to the MEI (source term not adjusted for emission controls)**
- **Source category**
- **e Below Appendix E Quantity (Y or N)**

**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, amual inventories, and emissions are not listed.**

**Radionuclide Inventories with Potential for Release: The annual radionuclide inventories for point-source locations are based on data from facility experimenters and mamgers. For Buildings 251 (hardened area) and 332, classification issues regarding transuranic-radionuclide inventories make use of the inventory/modeling approach impractical. However, all such affected emission points in these buildings are continuously monitored, and emissions are therefore directly determined. LLNL conducted a complete radionuclide-inventory update in 1994. Because of the magnitude of effort**

**required to complete a site-wide inventory, the 1996 inventory was conducted for all new sources and for those sources that cumulatively contributed to 90?40or more of the dose for 1995.**

**Physical-State Factors: The physical-state factors listed are EPA potentialrelease fractions from 40 CFR 61, Appendix D, whereby emissions are estimated from radionuclide inventories depending on their physical states for use in dispersion/dose assessment modeling. A physical-state factor of**  $1.0 \times 10^{-6}$  is used for solids,  $1.0 \times 10^{-3}$  is used for liquids and powders, and 1.0 is **used for unconfined gases.**

**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 new sources in 1996 were provided 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°/0. 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), venturi scrubbers and electrostatic precipitators are each given a 0.05 factor, and each activated-charcoal filter is given a 0.1 factor.**

**Estimated Annual Emissions: For unmonitored and non-continuously monitored sources, estimated amual emissions for each radionuclide are based on the product of (1) inventory data, (2) EPA potential-release fractions (physical-state factors), and (3) applicable emission-contiol-device abatement factors.**

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**Actual emission measurements are the basis for reported emissions from continuously monitored facilities. LLNL facilities that have continuous monitoring systems are Buildings 166, 175, 231-vault, 251,331, 332, 419, 490, and 491. See pages 9-10 for a discussion of 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  $10 \text{ mrem/y}$   $(100 \mu\text{Sv/y})$ .

**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 pSv/ y]), emissions must be individually evaluated from each point source; the location of the maximally exposed public individual (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 all emission points at the Livermore site and Site 300, calculations show that ground-level concentrations of radionuclides decline monotonically 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 were applied.**

**The unabated EDE camot be calculated for monitored facilities. 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 dose to**

**the MEI and the distance and direction to the LLNL fence line where the MEI is located. However, for monitored sources, no value is shown.**

**Source Categories: LLNL radionuclide air-emission sources have been classtiled into six source categories, indicated by the number in the next to last column of the spreadsheet: (1) Unmonitored or non-continuously monitored Livermore-site facilities that have had a radionuclide-inventory update for 1996; (2) Unmonitored or non-continuously monitored Livermore site facilities with a previous (1994 or 1995) radionuclide-inventory update; (3) Continuously monitored Livermore-site facilities; (4) Site 300 explosives experiments; (5) Diffuse sources where emissions and subsequent doses were estimated using inventory processes; and (6) Diffuse sources where emission and dose estimates were supported by environmental-surveillance measurements.**

**Below Appendix E Quantity: In 1995, DOE and EPA entered into a memorandum of understanding that, among other things, made the contents of 40 CFR 61, Appendix E acceptable "other procedures" for DOE facilities to establish compliance with Section 61.93(a) of Subpart H. Part of Appendix E is a list of "Annual Possession Quantities for Environmental Compliance. " Facilities having less than these quantities of radionuclides need not report to EPA under NESHAPS. A letter "Y" in this column denotes those inventoried sources at LLNL facilities that contain radionuclides in amounts below the annual possession quantities listed in Appendix E.**

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## Attachment 2. Surrogate Radionuclides List

Although CAP88-PC supports calculations for many radionuclides, there are some in use at LLNL that are not included in CAP88-PC. Consequently, this list of surrogate radionuclides has been developed to account for the contribution of those radionuclides.



Table 2-1. List of surrogate radionuclides.

 $\mathbf a$  $D = days$ ,  $W = weeks$ ,  $Y = years$ .

The annual inventory is multiplied by the correction factor, and a resulting surrogate b equivalency is used for the modeling calculation.

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.

<u>57.</u>