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Radiological Dose Assessment

Robert J. Harrach
Gretchen M. Gallegos
S. Ring Peterson

Introduction

Radiological doses to the public result from both natural and man-made radiation. The doses received by individuals and populations can be determined by measurements and calculations. This chapter describes Lawrence Livermore National Laboratory's radiological dose assessments, which are made to determine the impact of LLNL operations on the public and the environment. It includes a discussion of the analyses performed to demonstrate LLNL's compliance with the radiological *National Emission Standards for Hazardous Air Pollutants* (NESHAPs; Title 40 Code of Federal Regulations [CFR], Part 61, Subpart H).

Background Information

Because this chapter is written for a diverse readership, ranging from scientists and regulators to interested citizens with limited scientific training, a description is given of concepts, methods, tools, and other basic material in the first few sections as well as in Appendix D. Part D-1, "Radiation Basics," covers the different sources and types of radiation and the units used to quantify radiation, and it provides perspective on the wide range of radiation levels that people commonly encounter. Part D-2, "Radiation Control Measures at LLNL," sketches the standard operating procedures used to protect employees, the public, and the environment from uncontrolled releases and unsafe levels of radiation.

A discussion of sources, principal public receptors, and other aspects of modeling and monitoring follows the introductory material in the main text, leading to a presentation of key results on dose impacts from operations conducted in 2001. Readers desiring to go directly to these principal new results can turn to the section "Results of 2001 Radiological Dose Assessment".

Marie Curie

Releases of Radioactivity to Air

Releases of radioactive material to air (for example, in the form of air effluent dispersed from stacks or wind-driven resuspension of contaminated soil) are by far the major source of public radiological exposures from LLNL operations.

In contrast, releases to groundwater, surface water, and sewerable water are not sources of direct public exposures because these waters are not directly consumed or used by the public. Water releases can cause indirect exposures, which are analyzed as special cases. A recent case of this type concerned the potential dose to the public from inhalation and ingestion of soil that had been contaminated by sewer effluent containing radioactivity (U.S. Department of Health and Human Services 1999). Apart from such unusual occurrences, measurements and modeling of radiological releases to air determine LLNL's dose to the public.

Data supporting LLNL's radiological dose assessment are gathered by three principal means: continuous monitoring of stack effluent at selected facilities at the Livermore site (described in Chapter 4); routine surveillance air monitoring for radioactive particles and gases, both on and off Laboratory property (described in Chapter 5); and radioactive material usage inventories (described in LLNL's NESHAPs annual reports). The inventories cover noncontinuously monitored or unmonitored facilities housing radioactive materials management areas, and the explosive experiments conducted at Site 300.

Despite this emphasis on air monitoring, it should be noted that LLNL's extensive environmental monitoring program encompasses a variety of media and a wide range of potential contaminants; it is not limited to radioactive ones. In addition to ambient and effluent air monitoring and the three categories of water monitoring already mentioned, the Laboratory samples soil, vegetation, and wine, and measures environmental (gamma) radiation.

Monitoring has been described extensively since 1971 in LLNL's environmental reports (e.g., Biermann et al. 2001; see also Chapters 4 through 12 in the present report) and in LLNL's triennially updated *Environmental Monitoring Plan* (Tate et al. 1999) and its companion volume on procedures and guidance documents.

Air Dispersion and Dose Models

Theoretical/calculational models are needed to describe the transport and dispersion in air of contaminants and the doses received by exposed persons. Various factors dictate this need for modeling: (1) the amounts of LLNL-generated radioactive material dispersed into the atmosphere cause doses thousands of times smaller than those caused by natural background radiation (arising from irradiation by cosmic rays, inhalation of radon gas, exposure to radioactive materials in soil and rock, and ingestion of naturally occurring radionuclides present in our food and water; see Appendix D, Part D-1), so it is difficult to demonstrate compliance with standards through physical measurements alone; (2) all potentially significant exposure pathways need to be taken into account when estimating dose impacts; and (3) the U.S. Department of Energy (DOE) and the U.S. Environmental Protection Agency (EPA) sanction the use of specific computer codes that implement their approved dosimetry and dispersion models for evaluating potential doses to the public from both routine and unplanned releases. Beyond its role in dose assessment for regulatory compliance, advantages of a well-developed modeling capability include its utility in source design and optimization by estimating effects of hypothetical and/or dangerous sources and in interpreting past events through dose reconstruction.

The computer programs used at LLNL to model air releases and their impacts feature idealized, Gaussian-shaped plumes and can be run on personal computers. The CAP88-PC code incorporates dosimetric and health effects data and equations that are mandated by EPA to be used in compliance assessments (Parks 1992). Furthermore, CAP88-PC accommodates site-specific input data files to characterize meteorological conditions and population distributions for both individual and collective dose evaluations, and the code is relatively easy to use and understand. For these reasons, CAP88-PC has been the "work-horse" modeling tool for LLNL's regulatory compliance assessments since its availability in March 1992, particularly as applied to chronic releases of radioactivity to air occurring in the course of routine operations.

Radiation Protection Standards

The release of radionuclides from operations at LLNL and the resultant radiological impact to the public are regulated by both DOE and EPA.

DOE environmental radiation protection standards, provided under the authority of the Atomic Energy Act of 1954 and the DOE Organization Act of 1977 (both as amended), are defined in DOE Order 5400.5, *Radiation Protection of the Public and the Environment*. The standards for controlling exposures to the public from operations at DOE facilities that are incorporated in this order are based on recommendations by the International Commission on Radiological Protection (ICRP). The radiological impact to the public is assessed in accordance with the applicable portions of DOE Order 5400.1, *General Environmental Protection*.

The primary DOE radiation standards for protection of the public are 1 millisievert per year (1 mSv/y) or 100 millirem per year (100 mrem/y) whole-body effective dose equivalent (EDE) for prolonged exposure of a maximally exposed individual in an uncontrolled area and 5 mSv/y (500 mrem/y) EDE for occasional exposure of this individual. (EDEs and other technical terms are discussed in Appendix D, Part D-1 and defined in the glossary of this report.) These limits pertain to the sum of the EDE from external radiation and the committed 50-year EDE from radioactive materials ingested or inhaled during a particular year that may remain in the body for many years.

Radionuclide emissions to the atmosphere from DOE facilities are further regulated by the EPA, under the authority of Section 112 of the Clean Air Act. Subpart H of NESHAPs, under 40 CFR 61, referenced earlier, sets standards for public exposure to airborne radioactive materials (other than radon) released by DOE facilities; radon is regulated by Subparts Q and T. NESHAPs implements the dosimetry system recommended by the ICRP in Publication 26 (ICRP 1977).

The EPA's radiation dose standard, which applies only to air emissions, limits the EDE to members of the public caused by operations at a DOE facility to 100 µSv/y (10 mrem/y). EPA regulations specify not only the allowed levels, but also the approved methods by which airborne emissions and their impacts must be evaluated. With respect to all new or modified projects, NESHAPs compliance obligations define the requirements to install continuous air-effluent monitoring and to obtain EPA approval before the startup of new operations. NESHAPs regulations require that any operation with the potential to produce an annual-averaged off-site dose greater than or equal to 1 µSv/y (0.1 mrem/y), taking full credit for emission-abatement devices such as high-efficiency particulate air (HEPA) filters, must obtain EPA approval prior to the startup of operations. This same calculation, but without taking any credit for emission abatement devices, determines whether or not

continuous monitoring of emissions to air from this project is required. These requirements are spelled out in LLNL's online *Environment, Safety, and Health (ES&H) Manual* in Document 31.1, "Air Quality Compliance," which can be found at the following Internet address: http://www.llnl.gov/es_and_h/hsm/doc_31.01/doc31-01.html.

Air Emission Sources and Data

Sources

More than a hundred different radioisotopes are used at LLNL for research purposes, including biomedical tracers, tritium, mixed fission products, transuranic isotopes, and others. 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-filtered ventilation systems, to roof vents and stacks lacking abatement devices, to direct dispersal of depleted uranium during explosives testing at Site 300's open-air firing tables, to a variety of diffuse area sources.

Sources of radioactive material emissions to air at LLNL are divided into two categories for purposes of evaluating compliance: point sources (including stacks, roof vents, and Site 300's explosive experiments), and diffuse area sources (including dedicated waste accumulation areas and other areas of known contamination). Sources external to buildings, such as Hazardous Waste Management's "Tank Farm" operations at Building 514 and waste storage at the Building 612 Yard at the Livermore site, are treated as diffuse area sources. Detailed information on releases of radioactivity from LLNL's operations during 2001 is given in *LLNL NESHAPs 2001 Annual Report* (Harrach et al. 2002).

2001 Air Monitoring

This section briefly describes continuous stack-effluent sampling systems at selected LLNL facilities and ambient air monitors in place at numerous locations on and off LLNL sites. More complete information is provided in Chapters 4 and 5.

Continuous Stack Air Effluent Monitoring

Actual measurements of radioactivity in air and effluent flow are the basis for reported emissions from continuously monitored sources. In 2001, Buildings 175, 177, 235, 251, 331, 332, and 491 at the Livermore site had radionuclide air effluent monitoring systems. The number of samplers, the types of samplers, and the analytes of interest in these buildings are described in Chapter 4. All but Building 331 employed filter-type samplers to monitor gross alpha and beta radiation on particles.

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. In the pair of 30-meter stacks of the Tritium Facility (Building 331), the analytes being monitored are elemental gaseous tritium, tritiated water vapor, and total tritium; the sampling utilizes an ionization chamber and molecular sieves (see Chapter 4). Both the Tritium Facility and Plutonium Facility (Building 322) feature alarmed monitoring systems.

Results of Stack Monitoring for Tritium: Operations in the Tritium Facility in 2001 released a total of 7.4×10^{11} Bq (20 Ci) of tritium. Of this, approximately 6.8×10^{11} Bq (18.3 Ci) were released as tritiated water (HTO). The remaining

8.5% of the tritium released, 6.4×10^{10} Bq (1.7 Ci), was elemental tritium gas (HT). The highest single weekly stack emission from the facility was 2.5×10^{10} Bq (0.67 Ci), of which 2.4×10^{10} Bq (0.64 Ci) was HTO.

Building 331 tritium emissions, as measured by stack monitoring, remained considerably lower in 2001 than emissions that occurred during the 1980s. (Figure 4-2 illustrates the combined HTO and HT emissions from the facility since 1981.) The reduced emissions in 2001 were primarily the result of a reduction in programmatic work compared to 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 from this effort should maintain relatively low emissions.

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, the few samples with results above the MDC are only marginally above. 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 like 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. As a result, 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. On this basis, none of the facilities monitored for gross alpha and beta had emissions in 2001.

Air Surveillance Monitoring for Radioactive Particles and Gases

Surveillance air monitoring for tritium and radioactive particles has been in place since the 1970s. The data from this 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. As described in Chapter 5, LLNL currently maintains 7 continuously operating, high volume, air particulate samplers on the Livermore site, 9 in the Livermore Valley, 8 at Site 300, and 1 in Tracy, and maintains 12 continuously operating tritiated water vapor samplers on the Livermore site, 6 samplers in the Livermore Valley and 1 at Site 300. The samplers are positioned to ensure reasonable probability that any significant airborne concentration of particulate and 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 or confirm the emissions from the associated diffuse sources. Also included are air particulate and tritiated water vapor monitors positioned at or near the location of the hypothetical maximally

exposed member of the public (defined later in the subsection "Identification of Key Receptors") 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.

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 significantly affecting the concentrations of tritium in ambient air quoted in LLNL's environmental reports, results in negligible changes in the radiological doses to the public documented in those reports. Only for the special case of a diffuse tritium source having emissions inferred from monitoring data does the correction apply and change the inferred dose for that particular source. In particular, doses to the public attributed to tritium emissions from the pair of 30-meter-high stacks of the Tritium Facility 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 et al. 2001). The correction factor was developed based on new understanding of the properties of silica gel (Rosson et al. 1998; Rosson et al. 2000).

Put simply, the concentration of tritium measured in water extracted from the silica gel is lower than the concentration of the air moisture absorbed by the silica gel. This phenomenon occurs because tritium from ambient air exchanges with water bound in the silica gel that cannot be removed by the drying process. The bound water fraction is about 5% or 6% by weight, depending upon the type of silica gel. The magnitude of the correction depends upon the amount of water collected compared with the amount of exchangeable water bound in the silica gel and is specific to the silica gel used by LLNL. For 2001, the average correction factor was 1.6 (range of 1.3 to 2.3, with 99% of the correction factors being less than 2.1). The correction factor was applied to each sample based upon the amount of water collected and the initial weight of the dry silica gel.

An illustration of the quantitative effect produced annually by these corrections over the period 1997–2001 is given in the section "Results of 2001 Radiological Dose Assessment". Results of computer modeling are compared to measured concentrations of tritium in air at a dozen surveillance air monitoring locations on or near the Livermore site over that five-year period.

Radionuclide Usage Inventory Update

A partial accounting of LLNL's radiological emission sources was made in 2001 in accordance with the allowance by EPA that a 100% accounting need be made only every third year. The previous year, when reviewing and reporting on operations conducted in 2000, a 100% accounting was made.

The partial accounting focused on sources in four categories: (1) the group of sources that collectively (in a ranked list) accounted for at least 90% of the dose to the maximally exposed public individual from both the Livermore site and Site 300 in the year 2000 assessment; (2) all "new" sources that commenced emissions in 2001, or sources that showed significantly elevated releases over 2000 levels; (3) all monitored sources; and (4) all sources in the major LLNL waste stream dealt with by Hazardous Waste Management (HWM).

Radionuclide usage inventory forms, with guidance for completing them, were sent to all assurance managers, facility managers, and project-responsible persons connected with activities meeting these criteria for our partial accounting. The forms were completed by experimenters and certified by facility managers. Radionuclide usage data for all Site 300 explosives experiments and all significant stack and diffuse sources at both sites were included in this update.

Dose Assessment Methods and Concepts

Principal Modeling Approaches

Most estimates of individual and collective radiological doses to the public from LLNL operations were obtained using the EPA-developed computer code, CAP88-PC, as noted in the "Introduction". An LLNL-modified version of this code called CAP88-PC-T, which contains an improved tritium model (submitted to but not yet approved by EPA for use in regulatory compliance evaluations), was also used for purposes of comparison.

The user's guide for CAP88-PC (Parks 1992) provides useful information, including discussions of the basic equations and key input and output files. Additional information about LLNL-site-specific data files and several important caveats on use of the code can be found in the LLNL radiological dose assessment guidance document (Harrach 1998). The four principal pathways of exposure from air releases — 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 EDEs in units of mrem/y (1 mrem = 10 μSv). Separate doses for Livermore site and Site 300 emissions are evaluated and reported.

Other codes, such as EPA's INPUFF code (Peterson and Lavdas 1986) or LLNL's HOTSPOT code (Homann 1994), can be used as needed to address unplanned releases or transient releases from normal operations or accidents. In 2000, the EPA granted regulatory "guideline model" status to two codes—the AERMOD and CALPUFF codes— which are of considerably greater complexity than CAP88-PC, INPUFF, and HOTSPOT. Many other Gaussian-plume-type computer models are available for modeling specific types of releases; see, for example, the annotated lists in *Atmospheric Dispersion Modeling Resources* (Oak Ridge 1995) and *Supplement B to the Guideline on Air Quality Models (Revised)* (U.S. EPA 1993).

A complementary approach to deriving EDEs using the built-in dosimetry model in CAP88-PC or other codes is to explicitly calculate EDEs using mathematical formulas from the Nuclear Regulatory Commission's Regulatory Guide 1.109 (U.S. NRC 1977), which incorporate dose conversion factors consistent with those in the International Commission on Radiation Protection's Publication 30 (ICRP 1980). This approach, outlined in Appendix A of this report, has been used at LLNL since 1979 and can be used to evaluate annual doses to the public inferred from sampling of local environmental media (air, water, vegetation, and wine).

Identification of Key Receptors

When assessing probable off-site impacts, LLNL pays particular attention to doses received by three hypothetical receptors. First is the dose to the site-wide maximally exposed individual member of the public (SW-MEI; defined below). Second is the dose to the maximally exposed individual (MEI) member of the public from a given source point.

Third is the collective or "population" dose received by people residing within 80 km of either of the two LLNL sites.

The SW-MEI is defined as the hypothetical member of the public at a single, publicly accessible location (where members of the public reside or abide) who receives the greatest LLNL-induced EDE from all sources at a site. For LLNL to comply with the NESHAPs regulations, the LLNL SW-MEI cannot receive an EDE greater than 10 mrem/y (100 μSv/y) from releases of radioactive material to air. Public facilities that could be the location of the SW-MEI include schools, churches, businesses, and residences. This hypothetical person is assumed to remain at this location 24 hours per day, 365 days per year, continuously breathing air having the ground-level radionuclide concentration, and consuming a specified fraction of food and drinking water that is affected by the releases of radioactivity from the site. Thus, the SW-MEI dose is not received by any actual individual and is used as a conservative estimate of the highest possible dose to any member of the public. The location of the SW-MEI is sensitive to the frequency distribution of wind speeds and directions and locations of key sources in a given year and can change from one year to the next.

At the Livermore site, evaluation showed that the SW-MEI in 2001 was located at the UNCLE Credit Union, about 10 m outside the controlled eastern perimeter of the site. This location lies 948 m from the Tritium Facility (Building 331), in an east-northeast direction (the typical prevailing wind direction).

At Site 300, the SW-MEI occupied a position on the south-central boundary of the site bordering the Carnegie State Vehicular Recreation Area, approximately 3.2 km south-southeast of the firing table at Building 851. These SW-MEI locations are depicted in **Figure 13-1** and **Figure 13-2**.

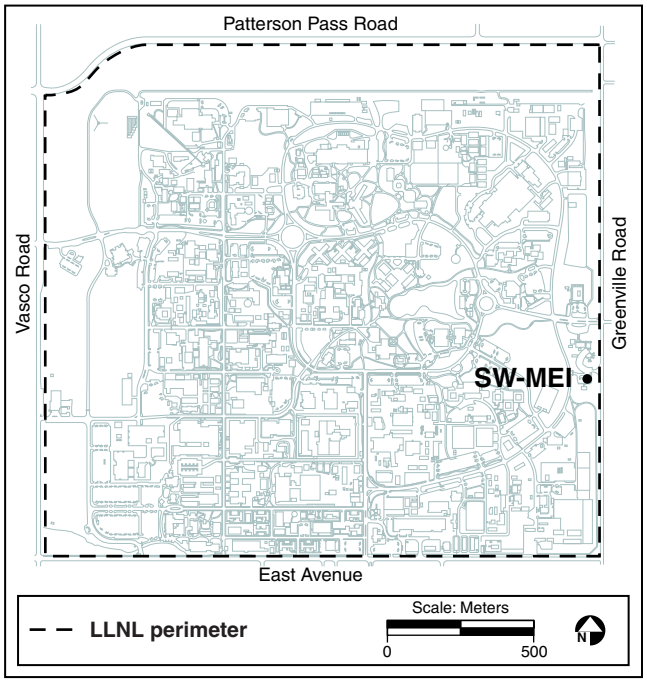

Figure 13-1. Location of the sitewide maximally exposed individual (SW-MEI) at the Livermore site, 2001

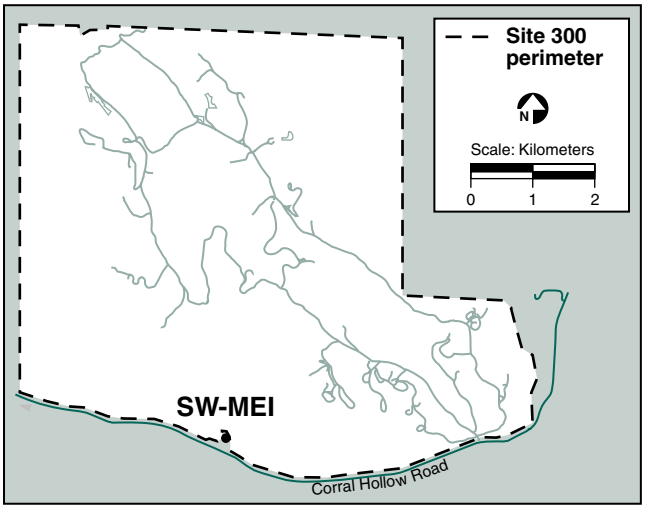

Figure 13-2. Location of the sitewide maximally exposed individual (SW-MEI) at Site 300, 2001

While the SW-MEI location is determined collectively by all sources at a site and coincides with an actual publicly accessible facility, the location of the MEI is any point of unrestricted public access receiving the largest potential dose from a given source and is generally different for each emission point. Such a point typically occurs at the site perimeter, and is often referred to as the maximum "fence line" dose. However, the off-site maximum dose could occur some distance beyond the perimeter (e.g., when a stack is close to the perimeter).

All new or modified LLNL projects in which releases of radioactivity to the environment may occur are reviewed for joint compliance with NESHAPs and the National Environmental Policy Act (NEPA). Dose to the MEI is used to evaluate whether continuous monitoring of the emissions from a given project is required, and whether it is necessary to petition the EPA for permission to start up the activity.

Summary of Input Parameters to CAP88-PC

General Model Inputs

Basic input parameters for running the CAP88-PC model include the specification of radionuclides, their emission rates in curies per year (1 Ci = 3.7×10^{10} Bq), and data on the nature of the emissions (e.g., stack parameters, including height, diameter, and emission velocity). A complete listing of required input data is given in the *User's Guide for CAP88-PC* (Parks 1992).

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 is 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 derive surrogate radionuclides to estimate EDEs. Attachment 2 in *LLNL NESHAPs 2001 Annual Report* (Harrach et al. 2002) shows the surrogate radionuclides used by LLNL in CAP88-PC over the years.

Population Inputs

Population distributions centered on the two LLNL sites were compiled from the LandScan Global Population 1998 Database developed by Dr. Jerome Dobson at Oak Ridge National Laboratory. The population data files (distribution of population with distance and direction) used in the 2001 modeling effort are the same as those described in *LLNL NESHAPs 2000 Annual Report* (Gallegos et al. 2001).

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. The "user entered" option was again selected for the CAP88-PC modeling effort for 2001. 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 for individual dose assessment. The assumption that all milk comes from local cows is not supported by the agricultural activities conducted in the area. For population dose assessments, all food is considered to be grown within an 80 km radius about the site; default densities of agricultural products in California are used.

Source Specification

The source term for 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 usage inventories, together with time factors and EPA-specified physical state factors, were used to estimate the potential annual emissions to air from a source. The time factors are used to adjust for the fact that the radionuclide may not always be in the same facility all year or may be encapsulated or enclosed for a substantial part of the year. The time factors are chosen to allow a reasonable estimate of the amount of radioactive material that may potentially be released into the atmosphere. The EPA-specified factors for potential release to air of materials in different physical states (solid, 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; see Table 4 in *LLNL NESHAPs 2001 Annual Report* (Harrach et al. 2002). 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. Abatement factors are taken into account in an evaluation for start up of operations, but are not included in the evaluation of need to install continuous monitoring of emissions.

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 in *LLNL NESHAPs 2001 Annual Report* (Harrach et al. 2002).

Modeling Dose from Tritium

Tritium (^{3}H) emissions account for the major dose from operations at the Livermore site. These emissions exist in two major chemical forms: tritium oxide or tritiated water vapor (HTO) and tritium gas (HT). The CAP88-PC code's tritium model calculates dose from inhalation, skin absorption, and ingestion of tritium, but only in its HTO form. 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 (ICRP 1995). 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 1990) and to a lesser extent in vegetation (Sweet and Murphy 1984).

A third important form of tritium to consider is organically bound tritium (OBT), which is formed by plants during photosynthesis and 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.

A new, simple tritium model developed at LLNL, called NEWTRIT, calculates ingestion dose from both HTO and OBT and accounts for conversion of HT to HTO in the environment after releases of HT (Peterson and Davis 2002). Both for this and last year's report, LLNL has used the NEWTRIT model incorporated into CAP88-PC, (called CAP88-PC-T) in addition to the default CAP88-PC code, to estimate doses from significant sources of tritium emissions. A brief discussion of the NEWTRIT model was presented in last year's NESHAPs annual report (Gallegos et al. 2001).

The NEWTRIT model was presented to EPA and DOE at a meeting of the Health Physics Society (Cleveland, Ohio, June 2001), and the associated paper was published in that society's journal (Peterson and Davis 2002).

In October 2001, LLNL sent a letter to EPA Region IX requesting consideration of an alternative methodology for calculating doses from atmospheric releases of HTO and HT for use in demonstrating compliance with radionuclide NESHAPs (40 CFR 61 Subpart H). Copies of NEWTRIT, CAP88-PC-T 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.

Reporting the Contribution of Tritium to Total Dose

Prior to the *Environmental Report 1998,* LLNL considered only the contribution to tritium dose of HTO releases. In April 1999, EPA mandated that LLNL use a more conservative approach when calculating dose to the public for NESHAPs compliance purposes, by treating all HT released as though it were HTO, rather than treating the dose from HT as negligible.

The introduction of NEWTRIT gives a third version of the contribution of tritium releases to total dose from LLNL operations. Starting with the present report, only the results derived using the latter two approaches will be quoted: (1) the CAP88-PC result inputting all curies of HTO released, plus an additional number of curies of HTO equal to the number of curies of HT released, and (2) the result from CAP88-PC-T (i.e., using the NEWTRIT model for tritium), inputting separately the number of curies of HTO and HT released. It should be noted that this tritium dose problem is important only for the Livermore site; at Site 300, tritium makes a negligible contribution to the public dose.

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 by comparison to previous years, presents the potential doses to the populations residing within 80 km of either the Livermore site or Site 300, and places the potential doses from LLNL operations in perspective with doses from other sources. Comments are provided on dose to biota and an illustration is given of the possible effect of silica gel correction factors on comparisons between modeling and monitoring.

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 0.11 µSv (0.011 mrem). The dose due to point sources was 0.057 µSv (0.0057 mrem). When combined, the total annual dose was 0.17 µSv (0.017 mrem), 66% from diffuse and 34% from point sources.

The 0.17 µSv (0.017 mrem) total 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 0.13 µSv (0.013 mrem).

The total dose to the Site 300 SW-MEI from operations in 2001 was 0.54 µSv (0.054 mrem). Point source emissions from firing table explosives experiments accounted for 0.50 µSv (0.050 mrem), or 93%, of this total, while 0.037 µSv (0.0037 mrem), or about 7%, was contributed by a diffuse source representing resuspension by wind of soil throughout the site containing low levels of depleted uranium.

Tritium accounted for more than three-quarters of the Livermore site's calculated dose, while at Site 300 practically the entire calculated dose was due to the isotopes uranium-238, uranium-235, and uranium-234 in depleted uranium. Regarding pathways, the relative significance of inhalation and ingestion depends on the assumptions made about the origin of food consumed. The assumption when assessing individual LLNL doses that milk is

imported while the remainder of the food is produced locally results in ingestion dose exceeding inhalation dose in the case of tritium, approximately in the percentages 80% to 20%. For uranium, these numbers are nearly reversed: 17% by the ingestion pathway versus 83% via inhalation. LLNL doses from air immersion and ground irradiation are negligible for both tritium and uranium.

Table 13-1 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 releasing radioactive material to air, most are very minor; nearly the entire radiological dose to the public comes from fewer than a dozen sources.

The trends in dose to the SW-MEI from emissions at the Livermore site and Site 300 over the last 12 years are shown in **Table 13-2**. The general pattern, particularly over the last decade, shows year-to-year fluctuations around a quite low dose level, staying at or below about 1% of the federal standard.

The SW-MEI dose estimates are intentionally conservative, predicting potential doses that are generally higher than would actually be experienced by any member of the public. Potential doses from Site 300 firing table operations are especially so, as explained in the section "Special Modeling Challenges."

Table 13-3 shows the Site 300 SW-MEI dose values attributed to firing table experiments for 1990 through 2001 exhibited along with the total amounts of depleted uranium and the total quantity of high explosives used each year in the experiments. (Only explosives experiments that included depleted uranium are considered here; most have none.) The 2001 total was indicative of increased firing table activity compared to the previous year but quite typical of levels in the past decade (see also the "Point source dose" column for Site 300 in **Table 13-2**).

Table 13-1. List of facilities or sources whose emissions accounted for more than 90% of the SW-MEI doses for the Livermore site and Site 300 in 2001

Facility (source category)	CAP88-PC dose in μSv/y	CAP88-PC percentage contribution to total dose
Livermore site		
Building 612 Yard (diffuse source)	0.082[a]	48
Building 331 stacks (point source)	0.043[a]	25
Building 514 Tank Farm (diffuse source)	0.013	8
Southeast Quadrant (diffuse source)	0.0088	5
Building 612, R102 (point source)	0.0062	4
Building 514 Evaporator (point source)	0.0058	3
Site 300		
Building 851 Firing Table (point source)	0.50	93
Soil resuspension (diffuse source)	0.037	7

a When LLNL's NEWTRIT model is used in CAP88-PC in place of CAP88-PC's default tritium model, the doses for Building 612 yard and Building 331stacks become 0.0061 mrem and 0.0031 mrem, respectively, and their percentages of the total dose from Livermore site operations each drop by 1%.

Table 13-2. Doses (in μSv) calculated for the sitewide 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	0.17[a]	0.057 [a]	0.11
2000	0.38[a]	0.17[a]	0.21
1999	1.2[a]	0.94[a]	0.28
1998	0.55[a]	0.31[a]	0.24
1997	0.97	0.78	0.19
1996	0.93	0.48	0.45
1995	0.41	0.19	0.22
1994	0.65	0.42	0.23
1993	0.66	0.40	0.26
1992	0.79	0.69	0.10
1991	2.34	—[b]	—[b]
1990	2.40	—[b]	—[b]
Site 300			
2001	0.54	0.50	0.037
2000	0.19	0.15	0.037
1999	0.35	0.34	0.012
1998	0.24	0.19	0.050
1997	0.20	0.11	0.088
1996	0.33	0.33	0.0045
1995	0.23	0.20	0.030
1994	0.81	0.49	0.32
1993	0.37	0.11	0.26
1992	0.21	0.21	—[c]
1991	0.44	0.44	—[c]
1990	0.57	0.57	—[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 before 1993.

Table 13-3. Annual dose to the SW-MEI from explosives experiments on firing tables at Site 300, 1990–2001, related to the total quantity of depleted uranium used in the experiments and the total quantity of high explosives driving the detonations

Year	Annual dose to SW-MEI		Total depleted uranium used in experiments (kg)	Total HE [a] used in depleted uranium experiments (kg)
	μSv	mrem		
2001	0.50	0.050	187	104
2000	0.15	0.015	43	34
1999	0.34	0.034	216	168
1998	0.19	0.019	230	192
1997	0.11	0.011	163	122
1996	0.33	0.033)	272	112
1995	0.20	0.020	165	199
1994	0.49	0.049	230	134
1993	0.11	0.011	99	74
1992	0.21	0.021	151	360
1991	0.44	0.044	221	330
1990	0.57	0.057	340	170

a HE = high explosives

Doses from Unplanned Releases

There were no unplanned atmospheric releases of radionuclides at the Livermore site or Site 300 in 2001.

Population Doses

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

Population centers affected by LLNL emissions include the relatively nearby communities of Livermore and Tracy; the more distant metropolitan areas of Oakland, San Francisco, and San Jose; and the San Joaquin Valley communities of Modesto and Stockton. Within the 80 km outer distance specified by DOE, there are 6.9 million residents included for the Livermore site population dose determination, and 6.0 million for Site 300. Population data files (distribution of population with distance and direction) used for the present report were the same as in the previous year; see Tables 7 and 8 in *LLNL NESHAPs 2000 Annual Report* (Gallegos et al. 2001).

The CAP88-PC result for potential population dose attributed to 2001 Livermore site operations was 0.0016 person-Sv (0.16 person-rem). This amount is less than typical, 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 0.094 person-Sv (9.4 person-rem). This value, while within the normal range seen from year to year, exceeds the 0.025 person-Sv (2.5 person-rem) for 2000 as a result of increased firing table activity.

Effect of Silica Gel Correction Factors on Modeling vs. Monitoring Comparison

LLNL's results for measured concentrations of tritium in ambient air require correction in light of new understanding of the effects of chemically bound water in "dry" silica gel (see Data Supplement, Chapter 5). LLNL's reported doses are negligibly affected by these corrections, since modeling, not monitoring, is used to determine dose to the public. These corrections do influence LLNL's opinions regarding the margin of conservatism represented in LLNL's modeling.

Comparisons between air concentrations predicted by CAP88-PC and measured air tritium concentrations have been included in LLNL's NESHAPs reports for the past five years. All of these comparisons now need revision, replacing the original measured values by their corrected counterparts.

Unfortunately, this is not possible for years prior to 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 multiplicative 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 a correction factor of 2.1 for years 1997 through 2000 and the actual (measured) sample-to-sample correction factors for 2001, revised predicted-to-observed (P/O) ratios of tritium concentrations in air at Livermore site perimeter locations and ZON7 were obtained. These were compared with the ratios using uncorrected measured values, with results as shown in **Table 13-4**. Without correction, 35 of the 40 P/O ratios were greater than 1.0, with the lowest being 0.69 at COW in 2001 and the highest 11 at SALV in 2000. When the observations are increased by applying correction factors, 30 of the P/O ratios are greater than 1.0, with the lowest being 0.40 at COW in 1997 and the highest 5.1 at SALV in 2000.

The diffferences between the sets of P/O ratios with and without correction are not regarded as significant. Considering the uncertainty in the numerator "P" alone, differences of this same general magnitude are expected to arise from use of a Gaussian plume dispersion model (such as used by CAP88-PC). For example, a comparison of AIRDOS-EPA predictions of air concentrations for various radionuclides (uranium-234, uranium-238, krypton-85, and tritium) 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 (Jack Faucett Assoc. 1987).

Doses to the Public Placed in Perspective

These levels of variation in population and SW-MEI doses from one year to the next are within the expected range of operations-driven fluctuations in small radiation quantities. A frame of reference to gauge the magnitude of these LLNL doses is

Table 13-4. Uncorrected (upper) and corrected (lower) ratios of predicted-to-observed air concentrations of tritiated water at Livermore site perimeter locations and ZON7, 1997–2001

Monitor	1997	1998	1999	2000	2001
CAFE	1.9 0.89	3.4 1.6	6.3 3.0	6.1 2.9	2.0 1.5
COW	0.84 0.40	1.0 0.49	1.6 0.77	1.1 0.50	0.58 0.41
MESQ	3.3 1.6	5.6 2.6	4.0 1.9	5.0 2.4	1.5 1.0
MET	3.2 1.5	2.4 1.2	3.1 1.5	2.4 1.1	1.6 1.2
POOL	0.99 0.47	2.2 1.1	3.9 1.9	4.4 2.1	1.0 0.79
SALV	1.5 0.73	6.9 3.3	3.7 1.8	11.0 5.1	3.8 3.9
VIS	3.0 1.4	2.4 1.2	5.7 2.7	3.0 1.4	1.5 1.1
ZON7	3.9 1.9	3.2 1.5	5.5 2.6	3.0 1.4	2.1 1.3

provided in **Table 13-5**. The table compares the conservatively estimated population doses and doses to the maximally exposed public individuals caused by LLNL operations against average doses received in the United States from exposure to natural background radiation and medical treatments. The population doses attributed to LLNL operations in 2001 are about 200,000 times smaller than ones from natural background radiation; the estimated maximum potential doses to individual members of the public from operations at the two LLNL sites in 2001 are more than 5,500 times smaller than ones received from background radiation in the natural environment.

Estimate of Dose to Biota

In recent years, it has been recognized that a past principle of radiological protection—that by protecting man, other living things are also protected—is not adequate. In 2000, DOE presented its standards for protection of the natural environment from the effects of ionizing radiation in its detailed guidance document "DOE Standard (Proposed): A Graded Approach for Evaluating Radiation Doses to Aquatic and Terrestrial Biota" (U.S. DOE 2002). DOE sites are requested to calculate dose to biota based upon this guidance. The guidance includes a manual, spreadsheets, and a database giving biota concentration guides (BCGs). Cases where human access to an area of exposure is restricted or exposure pathways favor biota exposure are especially important to consider. The effort required to show compliance is minimized by several features of the guidance: its use of a graded approach; its allowance of use of existing generic and site-specific data (not requiring new monitoring programs tailored to biota); and the fact that current and proposed standards are not very restrictive. Regarding the latter, the limit on

Table 13-5. Comparison of background (natural and man-made) and LLNL radiation doses, 2001

Location/source	Individual dose[a]		Population dose[b]	
	(μSv)	(mrem)	(person-Sv)	(person-rem)
Livermore site sources				
Atmospheric emissions	0.17	0.017	0.0016	0.16
Site 300 sources				
Atmospheric emissions	0.54	0.054	0.094	9.4
Other sources[c]				
Natural radioactivity[d,e]				
Cosmic radiation	300	30	1900	190,000
Terrestrial radiation	300	30	1900	190,000
Internal (food consumption)	400	40	2500	250,000
Radon	2000	200	12,500	1,250,000
Medical radiation (diagnostic procedures)[e]	530	53	3,300	330,000
Weapons test fallout[e]	11	1.1	68	6800
Nuclear fuel cycle	4	0.4	25	2500

a For LLNL sources, this dose represents that experienced by the SW-MEI member of the public.

b The population dose is the collective (combined) dose for all individuals residing within an 80-km radius of LLNL (approximately 6.9 million people for the Livermore site and 6.0 million for Site 300), calculated with respect to distance and direction from each site.

c From National Council on Radiation Protection and Measurements (NCRP 1987a, b)

d These values vary with location.

e This dose is an average over the U.S. population.

absorbed dose is 10 mGy/d (1 rad/d) for aquatic animals and terrestrial plants, and 1 mGy/d (0.1 rad/d) for terrestrial animals. (See Appendix D, Part D-1, Radiation Basics, and the Glossary for a discussion of radiation units.)

Screening calculations for LLNL impacts were performed in 2001 using the electronic spreadsheet provided with the guidance. Each radionuclide in each medium (soil, sediment, surface water) is assigned a derived concentration limit in the guidance. For each measured maximum concentration input to the spreadsheet, a fraction of the derived concentration limit for that radionuclide is automatically calculated, and the fractions are summed for each medium.

For aquatic biota, the sum of the fractions for water exposure are added to the sum of the fractions for sediment exposure. Similarly, the fractions for water and soil are summed for terrestrial biota. If the sums for the aquatic and terrestrial biota are both less than 1.0 mGy/d (0.1 rad/d), the site has passed the screening analysis, and the biota are assumed to be protected without further analysis.

In the LLNL assessment, the maximum concentration of each radionuclide measured in soils, sediments, and surface waters during 2001, whether measured on the Livermore site, off site in the Livermore Valley, or at Site 300, was entered into the screening calculation. Measurements of storm water runoff were used, although it is questionable

whether biota would be exposed to this concentration for more than a very short while. Principal measured radionuclides were americium-241 (non-detects), cesium-137, cobalt-60 (non-detects), tritium, plutonium-239, thorium-228, uranium-234, uranium-235 and uranium-238. Natural background levels of beryllium-7, potassium-40, radium-226 and radium-228 were also measured but not used as input to the spreadsheet. For LLNL, the sum of the fractions for aquatic biota was 0.21, and the sum for terrestrial biota was 0.016. Both are indicative of doses to aquatic and terrestrial biota from LLNL operations that are well below allowable dose limits.

Summary and Conclusion

The annual radiological dose from all emissions at the Livermore site and Site 300 in 2001 was found to be well below the applicable standards for radiation protection of the public, in particular the NESHAPs standard. This standard limits to 100 μSv/y (10 mrem/y) the EDE to any member of the public, arising as a result of releases of radioactive material to air from DOE facilities. Using EPA-mandated computer models and actual LLNL meteorology appropriate to the two sites, the potential doses to the LLNL SW-MEI members of the public from operations in 2001 were evaluated, with the following results:

- Livermore site: 0.17 μSv (0.017 mrem)—34% from point-source emissions, 66% from diffuse-source emissions—calculated by modeling releases of elemental gaseous tritium as tritiated water vapor, for compliance purposes as directed by EPA Region IX.

- Site 300: 0.54 μSv (0.054 mrem)—93% from explosive experiments, which are classified as point-sources, 7% from diffuse-source emissions.

The major radionuclides accounting for the doses were tritium at the Livermore site and the three isotopes in depleted uranium (uranium-234, uranium-235, and uranium-238) at Site 300. The only significant exposure pathway was release of radioactive material to air, leading to doses by inhalation and ingestion.

The collective EDE or population dose attributable to LLNL operations in 2001 was estimated to be 0.0016 person-Sv (0.16 person-rem) for the Livermore site and 0.094 person-Sv (9.4 person-rem) for Site 300. These doses include potentially exposed populations of 6.9 million people for the Livermore site and 6.0 million people for Site 300 living within a distance of 80 km from the site centers.

The doses to the SW-MEI members of the public resulting from Livermore site and Site 300 operations in 2001 were below 0.6% of the federal standard and were more than 5500 times smaller than the dose from background radiation. The population doses from LLNL operations in 2001 were about 200,000 times smaller than those caused by natural radioactivity in the environment (see **Table 13-5**).

Potential doses to aquatic and terrestrial biota from LLNL operations were assessed and found to be well below DOE allowable dose limits.

We conclude that the potential radiological doses from LLNL operations were well below regulatory standards and were very small compared with doses normally received by these populations from natural background radiation sources, even though highly conservative assumptions were used in the determinations of LLNL doses. These maximum credible doses to the public indicate that LLNL's use of radionuclides had no significant impact on public health during 2001.