

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

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**LLNL NESHAPs**  
**2005 Annual Report**



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**Lawrence Livermore National Laboratory**  
University of California  
Livermore, CA 94551

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# **LLNL NESHAPs 2005 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 2005**

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# Lawrence Livermore National Laboratory NESHAPs 2005 Annual Report

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

## SYNOPSIS

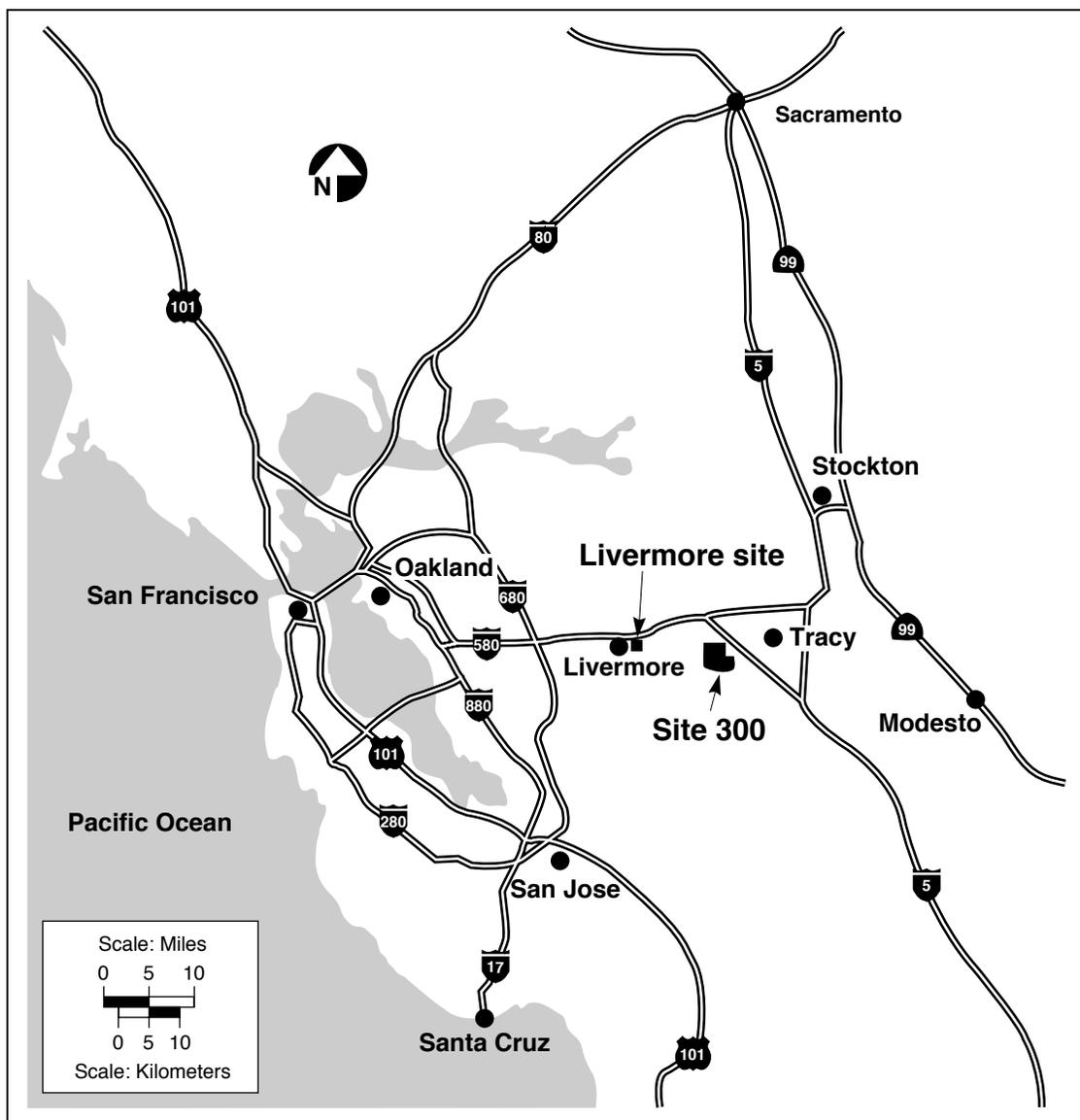
NESHAPs limits the emission of radionuclides to the ambient air from DOE facilities to levels resulting in an annual effective dose equivalent (EDE) of 10 mrem (100  $\mu$ Sv) to any member of the public. The EDEs for the Lawrence Livermore National Laboratory (LLNL) site-wide maximally exposed members of the public from operations in 2005 are summarized here.

- Livermore site: 0.0065 mrem (0.065  $\mu$ Sv) (41% from point source emissions, 59% from diffuse source emissions). The point source emissions include gaseous tritium modeled as tritiated water vapor as directed by EPA Region IX; the resulting dose is used for compliance purposes.
- Site 300: 0.018 mrem (0.18  $\mu$ Sv) (48% from point source emissions, 52% from diffuse source emissions).

The EDEs were calculated using the EPA-approved CAP88-PC air dispersion/dose-assessment model, except for doses for two diffuse sources that were estimated using measured radionuclide concentrations and dose coefficients. Specific inputs to CAP88-PC for the modeled sources included site-specific meteorological data and source emissions data, the latter variously based on continuous stack effluent monitoring data, stack flow or other release-rate information, ambient air monitoring data, and facility knowledge.

## SECTION I. Site Description

LLNL, a U.S. DOE facility operated by the University of California, was established in 1952 to conduct nuclear weapons research and development. The Laboratory serves as a national resource in science, engineering, and technology. LLNL's primary mission focuses on nuclear weapons and national security, including stockpile stewardship. Its mission is dynamic and has been broadened over the years to include areas such as strategic defense, nonproliferation, homeland security, energy, the environment, bioscience and biotechnology, and science and mathematics education. LLNL comprises 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.



**Figure 1.** Locations of LLNL's Livermore site and Site 300.

## **Livermore Site**

LLNL's Livermore site occupies an area of 3.3 km<sup>2</sup> located about 60 km east of San Francisco, California, adjacent to the City of Livermore in the eastern part of Alameda County. In round numbers, 7 million people live within 80 km of the Livermore site; about 80,000 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 above sea level at the eastern end to approximately 90 m above sea level at the southwest corner.

The climate of the Livermore Valley is characterized by mild, rainy winters and warm, dry summers. The mean daily maximum, minimum, and average temperatures for the Livermore site in 2005 were 22.0°C, 8.0°C, and 15.0°C, respectively, typical for the site. Temperatures typically range from -5°C during some pre-dawn hours in the winter, to 40°C on a few summer afternoons. The 2005 annual wind data for the Livermore site are displayed as a wind rose in **Figure 2**. In the wind rose, the length of each spoke is proportional to the frequency at which the wind blows from the indicated direction; different line widths of each spoke represent wind speed classes. These data show that 49% of the time the winds blew from the south-southwest through west directions. However, during the winter, the wind often blew from the northeast. The average wind speed in 2005 at the Livermore site was 2.3 m/s (5.2 mph). Most precipitation occurs as rain between October and April with very little rainfall during the summer months. In 2005, the Livermore site received 45.5 cm of precipitation.

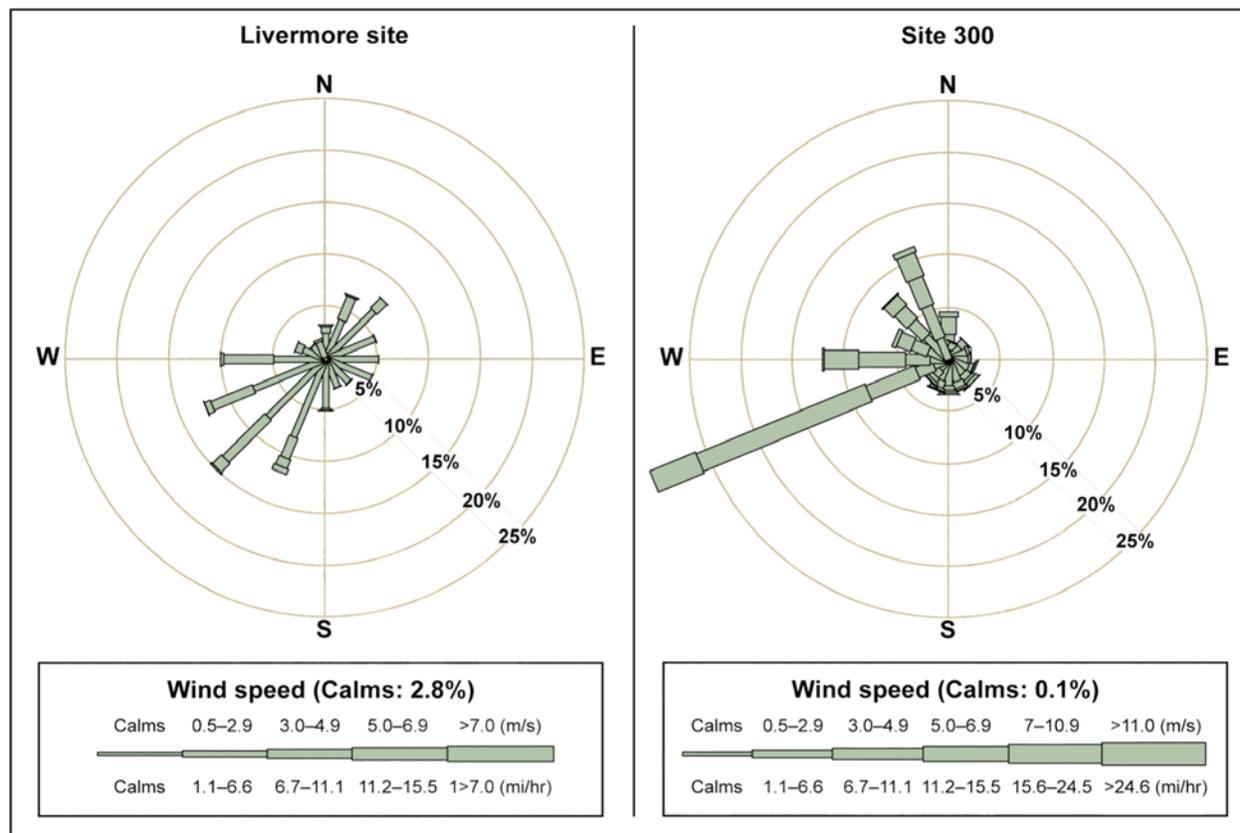
## **Site 300**

Site 300, LLNL's Experimental Test Facility, is located 24 km east of the Livermore site in the Altamont Hills of the Diablo Range and occupies an area of 30.3 km<sup>2</sup>. SRI International operates a testing site located approximately 1 km south of Site 300. Property immediately to the east of Site 300 is owned by Fireworks America which uses it for packaging and storing fireworks displays. The Carnegie State Vehicular Recreation Area is located south of the western portion of Site 300, and wind-turbine generators line the hills to the northwest. The remainder of the surrounding area is in agricultural use, primarily grazing land for cattle and sheep. The nearest residential area is the city of Tracy (population of over 80,000), located 10 km to the northeast. About 6.2 million people live within 80 km of Site 300. Ninety-five percent live more than 32 km from Site 300 in such distant metropolitan areas as Oakland, San Jose, and Stockton.

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. The stronger

winds occurring at the higher elevations of Site 300 results in warmer nights and slightly cooler days than at the Livermore site.

The 2005 annual wind data for Site 300 are displayed as a wind rose on the right side of **Figure 2**. Winds from the west-southwest through west occurred 43% of the time during 2005. As is the case at the Livermore site, Site 300 precipitation is highly seasonal, with most precipitation occurring between October and April. Site 300 received 32.5 cm of precipitation during 2005 and had mean daily maximum, minimum, and average temperatures of 21.1°C, 12.6°C, and 16.9°C, respectively. The average wind speed at the site was 5.5 m/s (12.3 mph).



Note: The length of each spoke is proportional to the frequency at which the wind blows from the indicated direction. Different line widths of each spoke represent wind speed classes. The average wind speed in 2005 at the Livermore site was 2.3 m/s (5.2 mph); at Site 300 it was 5.5 m/s (12.3 mph).

**Figure 2.** Wind roses, showing wind speed, direction, and frequency of occurrence at the Livermore site and Site 300 during 2005.

## SECTION II. Air Emission Sources and Data

### Sources

Approximately eighty different radioisotopes were available for use at LLNL in 2005 for research purposes, including biomedical tracers, tritium, mixed fission products, transuranic isotopes, and others—see **Table 1**. Radioisotope handling procedures and work enclosures are determined for each project or activity, depending on the isotopes, the quantities being used, and the types of operations being performed. Work enclosures include gloveboxes, exhaust hoods, and laboratory bench tops. Exhaust paths to the atmosphere include High Efficiency Particulate Air (HEPA) filtered ventilation systems, roof vents and stacks lacking abatement devices, direct open-air dispersal of depleted uranium during explosives testing at Site 300, and releases to ambient air from a variety of diffuse area sources.

**Table 1.** Radionuclides at LLNL during 2005.

Hydrogen-3	Iron-55	Silver-110m	Bismuth-207	Uranium-233	Americium-242m
Nitrogen-13	Cobalt-57	Iodine-125	Polonium-208	Uranium-234	Plutonium-242
Carbon-14	Nickel-59	Iodine-131	Polonium-209	Uranium-235	Americium-243
Oxygen-15	Cobalt-60	Barium-133	Polonium-210	Neptunium-236	Curium-243
Sodium-22	Nickel-63	Cesium-134	Lead-210	Plutonium-236	Curium-244
Phosphorus-32	Zinc-65	Cesium-137	Radium-226	Uranium-236	Plutonium-244
Phosphorus-33	Selenium-75	Cerium-144	Actinium-227	Neptunium-237	Californium-249
Sulfur-35	Krypton-85	Promethium-147	Radium-228	Uranium-237	Californium-250
Chlorine-36	Strontium-85	Europium-152	Thorium-228	Plutonium-238	Californium-252
Potassium-40	Yttrium-88	Europium-154	Thorium-229	Uranium-238	
Calcium-41	Strontium-89	Europium-155	Thorium-230	Plutonium-239	
Argon-41	Strontium-90	Rhenium-187	Protactinium-231	Plutonium-240	
Chromium-51	Technetium-99	Mercury-203	Thorium-232	Americium-241	
Manganese-54	Cadmium-109	Thallium-204	Uranium-232	Plutonium-241	

Sources of radioactive material emissions to air at LLNL are divided into two categories for purposes of evaluating NESHAPs compliance: point sources and diffuse area sources. The former includes stacks, roof vents, and explosive experiments conducted on Site 300's firing tables; the latter are for the most part dedicated waste accumulation areas and other areas of known contamination, generally external to buildings.

### Air Monitoring in 2005

Continuous stack-effluent sampling systems at selected LLNL facilities and ambient air monitors in place at numerous locations on and off LLNL sites are described in this section.

#### *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 2005, there were seven buildings (Buildings 235, 251, 331, 332, 491, and 695/696; the last two share a common stack) at the Livermore site and one building (the Contained Firing Facility, Building 801A) at Site 300 that had radionuclide air

effluent monitoring systems. These buildings are listed in **Table 2**, along with the number of samplers, the types of samplers, and the analytes of interest.

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 gross beta activity on a weekly or bi-weekly frequency depending on the facility. In most cases, passive filter 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 an unplanned 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. The Radiological Measurements Laboratory (RML) in LLNL's Hazards Control Department (HCD) performs the analyses.

Each stack of the Tritium Facility (Building 331) is monitored for tritium release by both an alarmed continuous monitoring system and by molecular sieve continuous samplers. The alarmed monitors provide real time tritium concentration release levels (HT, HTO, or other gaseous forms). The sieve samplers discriminate between tritiated water (HTO) vapor and molecular tritium (HT); they provide the values used for environmental reporting and are exchanged weekly. Each sieve sampler (not alarmed) is in parallel with an alarmed monitor and consists of two molecular sieves. The first sieve collects tritiated water vapor; the second sieve contains a palladium-coated catalyst that converts molecular tritium to tritiated water, which is then collected. The molecular sieve samples are submitted to the Hazards Control Analytical Laboratory where they are put into a recovery system for the bake out of tritiated water vapor and subsequent condensation and collection of the water. The retrieved tritiated water is analyzed by RML using liquid scintillation counting techniques.

In addition to particulate monitoring for gross alpha and gross beta, the Decontamination and Waste Treatment Facility (DWTF) stack effluent has continuous monitoring for tritium with the use of a glycol bubbler. The bubbler uses a two-stage glycol impinging process. Stack air to be sampled enters the instrument and flows through two impingers in series capturing the HTO present. Next, the sampled air is directed through a palladium catalyst where oxidation of any HT in the sample takes place, converting HT to HTO. Then the HTO is collected in two more bottle impingers in series. The impingers are analyzed by the RML using liquid scintillation analysis. This type of sampling quantifies the amount of tritium for both species HT and HTO.

Environmental Protection Department (EPD) environmental analysts review data from air particulate sampling filters, molecular sieves, and the glycol bubbler.

**Table 2.** Air effluent sampling systems and locations.

Building	Facility	Analytes	Sample type	Number of samplers
235	Chemistry and Materials Science	Gross $\alpha$ , $\beta$ on particles	Filter	1
251	Heavy Elements			
	Unhardened <sup>a</sup> area	Gross $\alpha$ , $\beta$ on particles	Filters	24
	Hardened <sup>a</sup> area	Gross $\alpha$ , $\beta$ on particles	Filters	4
		Gross $\alpha$ , $\beta$ on particles	CAM <sup>b</sup>	2
331	Tritium	Tritium	Ionization Chamber <sup>b</sup>	4
		Gaseous tritium/tritiated water vapor	Molecular sieves	4
332	Plutonium	Gross $\alpha$ , $\beta$ on particles	CAM <sup>b</sup>	12
		Gross $\alpha$ , $\beta$ on particles	Filters	15
491	Isotope Separation <sup>c</sup>	Gross $\alpha$ , $\beta$ on particles	Filter	1
695/696	Decontamination and Waste Treatment Facility	Gross $\alpha$ , $\beta$ on particles	Filter	1
		Gaseous tritium/tritiated water vapor	Glycol Bubbler	1
695 Yard	TRU Mover <sup>d</sup>	Gross $\alpha$ , $\beta$ on particles	Filter	1
801A	Contained Firing Facility	Gross $\alpha$ , $\beta$ on particles	Filter	1

Note: "CAM" denotes Eberline continuous air monitors.

<sup>a</sup> Hardening refers to seismic reinforcement.

<sup>b</sup> Alarmed systems.

<sup>c</sup> Isotope separation operations were discontinued; area now used for storage of contaminated parts.

<sup>d</sup> In January 2006, with the completion of TRU Mover activities, air effluent sampling was discontinued.

### Results of Stack Monitoring for Tritium

Operations in the Tritium Facility (Building 331) in 2005 released a total of 32 Ci (1.2 TBq) of tritium. Of this, approximately 30 Ci (1.1 TBq) were released as HTO. The remaining tritium released, 2.0 Ci ( $7.4 \times 10^{-2}$  TBq), was tritiated hydrogen gas (HT). The highest single weekly stack emission from the facility was 4.9 Ci (0.18 TBq), of which greater than 99% was HTO.

This 2005 level of tritium emissions continues to be low in comparison to those typically seen in the 1980's and 1990's, indicative of a reduced level of operations in the Tritium Facility.

**Table 3** displays the combined HTO and HT emissions from the Tritium Facility since 1981.

Continuously monitored tritium releases from the stack of DWTF began in February of 2005. A total of 2.3 Ci ( $8.5 \times 10^{-2}$  TBq) of measured tritium was released with 2.2 Ci ( $8.1 \times 10^{-2}$  TBq) as HTO and 0.088 Ci ( $3.3 \times 10^{-3}$  TBq) as HT. Since monitoring did not begin at the first of the year, an estimate emission of 0.47 Ci ( $1.7 \times 10^{-2}$  TBq), was calculated by taking an average of measured emissions and applying it to the time period when monitoring was not in place. The

total emission for 2005 (measured and estimate emissions combined) was 2.7 Ci ( $1.0 \times 10^{-1}$  TBq), of which 0.11 Ci ( $4.0 \times 10^{-3}$  TBq) was HT. The tritium emissions from Building 695 are more than a hundred times below the level of regulatory requirement for monitoring; monitoring is in place as part of a best management practice.

**Table 3.** Combined HT and HTO emissions from the Tritium Facility, 1981–2005.

Year	Tritium emissions <sup>a</sup> (Ci)	Year	Tritium emissions <sup>a</sup> (Ci)
2005	32	1992	177
2004	17	1991	964 (148)
2003	110	1990	1281
2002	36	1989	2620 (329)
2001	20	1988	3978
2000	40	1987	2634
1999	280	1986	1128
1998	109	1985	989 (1000)
1997	299	1984	2200 (5000)
1996	215	1983	3024
1995	92	1982	1914
1994	137	1981	2552
1993	237		

<sup>a</sup> Chronic releases from normal operations are distinguished from acute accidental releases by showing the latter in parentheses. Accidental releases were predominately HT gas.

### 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 26 to 52 per year) have concentrations greater than the MDC. Generally, these few samples having results above the MDC are only marginally above it. 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-spectrometry-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. As a result, there are no dose consequences, and doses reported for these operations are zero. Furthermore, even if the MDC values were used in calculations of the emission estimates for these facilities, which would be an extremely conservative approach, the total dose attributable to LLNL activities would not be significantly affected.

In 2005, seven samples collected throughout the year from the release emission point at Building 801A (Site 300) yielded gross alpha results greater than the MDC. Gross alpha is used as the primary indicator of potential emissions for operations that involve the use of uranium or transuranic materials, such as those at Building 801A. The gross alpha and gross beta activity

emissions for Building 801A were  $4.2 \times 10^{-7}$  Ci/y ( $1.6 \times 10^4$  Bq/y) and  $1.6 \times 10^{-6}$  Ci/y ( $5.9 \times 10^4$  Bq/y). Because more than ten percent of the weekly samples had values above the MDC, we have taken a conservative approach and are reporting gross alpha and gross beta measurements as actual emissions.

The resulting radiological dose to the maximally exposed individual (MEI) member of the public determined with CAP88-PC modeling was  $1.1 \times 10^{-5}$  mrem/y ( $1.1 \times 10^{-6}$   $\mu$ Sv/y); doses are discussed in Sections III and IV and in Attachment 1.

Among the facilities monitored for particulate gross alpha and gross beta in 2005, only Building 801A (Site 300) showed emissions.

### ***Air Surveillance Monitoring for Radioactive Particles and Gases***

Surveillance air monitoring for tritium and radioactive particles has been in place since the early 1970s. LLNL currently maintains seven continuously operating, high volume, air particulate samplers on the Livermore site, nine in the Livermore Valley, eight at Site 300, and one in Tracy. LLNL also maintains eleven continuously operating tritiated water vapor samplers on the Livermore site, six in the Livermore Valley and one at Site 300. The samplers are positioned to provide reasonable probability that any significant airborne concentration of particulate or tritiated water vapor effluents resulting from LLNL operations will be detected. Several surveillance air monitors are placed near diffuse emission sources, such as those near Building 331 and in the Building 612 Yard, as well as in and around the Southeast Quadrant of the Livermore site. Their results can be used to estimate and/or confirm emissions from associated diffuse sources. Both an air particulate monitor and an ambient air tritium sampler are positioned at the location of the hypothetical maximally exposed member of the public (defined in Section III) for the Livermore site. Data from air surveillance monitors provide a valuable test of predictions based on air dispersion modeling and can help characterize unplanned releases of radioactive material.

Summary data are provided in **Table 4** and in Section VII, "Comparison of 2005 Modeling Results with Tritium Surveillance Air Monitoring Data." Detailed data from the surveillance air-monitoring network are presented annually in the LLNL Site Annual Environmental Report (SAER), which is available to the public in hardcopy form, on CD, and on the Internet at the address <http://www.llnl.gov/saer>.

### ***Compliance Demonstration for Minor Radiological Sources***

With the Environmental Protection Agency's (EPA) Region IX approval, LLNL demonstrates compliance for minor emissions sources (which are primarily non-monitored stack sources) through the use of existing ambient air monitoring data. The method entails comparing measured ambient air concentrations at the location of the site-wide maximally exposed individual (SW-MEI), defined in Section III, to concentration limits set by the U.S. EPA in its Table 2 of Appendix E to 40 CFR 61. The radionuclides for which the comparison is made are tritium and plutonium-239+240 for the Livermore SW-MEI and uranium-238 for the Site 300 SW-MEI. At the Livermore site, all 2005 monitoring results from the Discovery Center (VIS) and the UNCLE Credit Union (CRED) sampling locations (shown in **Figure 5** in Section VII) that are greater than zero are averaged to represent the SW-MEI for the purposes of this minor

source comparison. At Site 300, wind-driven resuspension of soil contaminated with depleted uranium is of greatest interest in the minor source category. Because this is a diffuse source covering a wide area, the average of the results for all monitoring locations at the site is used to represent the SW-MEI.

The measured concentrations at the SW-MEI are presented in **Table 4**. The measured concentration of plutonium-239 is about two times greater than the result for 2004. This slight increase reflects a change in the way the average was calculated: unlike previous years in which all of the monitoring results were averaged, including negative and zero values, the 2005 is based on the more conservative averaging of all results greater than zero. Also shown in **Table 4** are EPA's standards from Table 2 of Appendix E to 40 CFR 61. As demonstrated by the calculation of the fraction of the standard, LLNL measured concentrations in air for tritium, plutonium-239+240, and uranium-238 are a fraction 0.005 or less of the standard for these radionuclides.

**Table 4.** Mean concentrations of radionuclides of concern at the location of the SW-MEI in 2005 compared to EPA's concentration standard.

Location	Nuclide	EPA's Table 2 concentration standard	Mean measured concentration	Measured concentration as a fraction of the standard	Detection limit
Livermore site SW-MEI	Tritium	$1.5 \times 10^{-9}$ Ci/m <sup>3</sup>	$1.3 \times 10^{-12}$ Ci/m <sup>3*</sup>	$8.7 \times 10^{-4}$	$1 \times 10^{-12}$ Ci/m <sup>3</sup>
Livermore site SW-MEI	Plutonium-239	$2.0 \times 10^{-15}$ Ci/m <sup>3</sup>	$2.4 \times 10^{-19}$ Ci/m <sup>3**</sup>	$1.2 \times 10^{-4}$	$5 \times 10^{-19}$ Ci/m <sup>3</sup>
Site 300 SW-MEI	Uranium-238	$8.3 \times 10^{-15}$ Ci/m <sup>3</sup>	$1.9 \times 10^{-17}$ Ci/m <sup>3***</sup>	$2.3 \times 10^{-3}$	$3 \times 10^{-20}$ Ci/m <sup>3</sup>

\* The measured tritium value includes contributions from all tritium sources, i.e., the Tritium Facility, Building 612 Yard, the DWTF stack, the Building 331 Outside Yard, etc.; there is no way to distinguish tritium by its source of emission.

\*\* Note that the mean measured concentration for plutonium is less than the detection limit; only 3 of the 24 values comprising the mean were measured detections. Only values greater than zero are used in the calculation of the mean.

\*\*\* The ratio for the mean uranium-238 and uranium-235 concentrations for 2005 is 0.005, which is less than 0.00726, the ratio of these isotopes for naturally occurring uranium. This results in approximately 57% of the resuspension being attributable to natural occurring uranium and 43% to depleted uranium.

The LLNL radiological facilities included in the "minor sources" classification in 2005 are listed in **Table 5**.

**Table 5.** Buildings with minor radiological emissions (by directorate) for 2005.<sup>a</sup>

<b>C&amp;MS</b>	<b>P&amp;AT</b>	<b>SEP</b>	<b>E&amp;E</b>	<b>Eng.</b>	<b>Biosci.</b>	<b>DNT</b>	<b>NIF</b>	<b>Institut.</b>
B 132	B 194	B 253	B 281	B 131	B 361	B 801	B 298	B 212
B 151	B 282	B 254	B 292	B 231	B 362	B 804		(vacant)
B 235	B 341	B 255	B 378	B 321	B 363			
B 241				B 321A	B 364			
B 810A				B 321B	B 365			
B 810B				B 321C	B 366			
				B 322				
				B 327				

<sup>a</sup> Directorate abbreviations refer to Chemistry and Materials Science (C&MS), Physics and Advanced Technologies (P&AT), Safety and Environmental Protection (SEP), Energy and Environment (E&E), Engineering (Eng.), Biosciences (Biosci.), Defense and Nuclear Technologies (DNT), National Ignition Facility Programs (NIF), and Institutional (Director's Office).

### **Radionuclide Usage Inventories**

Radionuclide usage inventories were utilized in 2005 to calculate public dose impacts only for the open-air explosives experiments at Site 300 (see Attachment 1) and for pre-start evaluations for various other radiological activities/experiments that commenced operations in 2005.

Radionuclide usage inventory documentation and pre-start evaluations are archived in the NESHAPs data library maintained by the Terrestrial and Atmospheric Monitoring and Modeling (TAMM) Group in the Operations and Regulatory Affairs Division of the Environmental Protection Department.

## SECTION III. Dose Assessment Methods & Concepts

### Description of the Air Dispersion and Dose Model

Most estimates of individual and collective radiological doses to the public from LLNL operations were obtained using the EPA's computer code, CAP88-PC. The four principal pathways—internal exposures from inhalation of air, ingestion of drinking water (for tritium only) and foodstuff, external exposures through irradiation from contaminated ground, and immersion in contaminated air—are evaluated by CAP88-PC. The doses are expressed as whole-body effective dose equivalents (EDEs), in units of mrem/y (1 mrem = 10  $\mu$ Sv). Separate doses for Livermore site and Site 300 emissions are reported. For purposes of comparison, tritium doses from inhalation and ingestion were also calculated with an improved tritium model, NEWTRIT (see “Modeling Dose from Tritium” in Section VII); NEWTRIT is not yet approved by EPA for use in regulatory compliance evaluations.

Three potential doses are emphasized: (1) The dose to the site-wide maximally exposed individual (SW-MEI), which combines the contributions of all evaluated emission points to dose at a publicly-accessible facility for comparison to the 10 mrem/y (100  $\mu$ Sv/y) standard; (2) the maximum dose to any member of the public, in any direction attributed to each unabated emission point on the site to determine the need for continuous monitoring; and (3) the collective dose to populations residing within 80 km of the two LLNL sites, summing the products of individual doses received and number of people receiving them.

### Summary of Model Input Parameters

#### General Model Inputs

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

#### Meteorological Data

All model runs used actual 2005 Livermore site and Site 300 meteorological data collected from the meteorological towers for each site. At these towers, wind speed and direction and temperature are sampled every one or two seconds and 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

Even though CAP88-PC contains a library of 265 radionuclides, it does not contain all radionuclides available for use at LLNL. As a consequence, use of surrogate radionuclides to estimate EDEs is sometimes necessary. The selection of a suitable surrogate is based upon several criteria, including metabolically similar behavior and similar modes of decay and decay energies of the radiation type of the isotope of interest. Once a surrogate is selected, the equivalent source term is adjusted by the product of the initial inventory of the isotope of interest and the ratio of the effective dose equivalent of the surrogate to that of the isotope of interest. In some cases, isotopic analyses of mixtures of radionuclides are not available and the radionuclides used are identified simply as “gross alpha,” “gross beta,” “gross gamma,” or “mixed fission products” (MFP). In these cases, for compliance modeling purposes  $^{239}\text{Pu}$  is used as the

surrogate for gross alpha,  $^{137}\text{Cs}$  for gross gamma, and  $^{90}\text{Sr}$  for gross beta and mixed fission products to provide conservative dose estimates. For a list of surrogate radionuclides, see Table 2-1 in the 2003 NESHAPs annual report (Harrach et al. *LLNL NESHAPs 2003 Annual Report*, UCRL-ID-11367-04, June 2004).

### Population Inputs

The population distributions centered on the two LLNL sites are based on the LandScan Global Population 2001 Database (Dobson, J. E., E. A. Bright, P. R. Coleman, R.C. Durfee, B. A. Worley, LandScan: A Global Population Database for Estimating Populations at Risk, *Photogrammetric Engineering & Remote Sensing* Vol. 66, No. 7, July 2000, pp. 849-857; see also the Website <http://www.ornl.gov/sci/landscan>). The population distributions were developed using the geographic information system software, ArcView©, to construct five equidistant radial sectors in each of the 16 wind directions required by CAP88-PC. The population for each sector segment was determined by running code developed in the LandScan project and distributed with the LandScan Database. Key population centers affected by LLNL emissions are the 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 DOE, there are 7.1 million residents included for the Livermore site collective dose determination, and 6.2 million for Site 300.

### Land Use and Agricultural Inputs

For agricultural parameters in CAP88-PC, LLNL used mean values for California based on data from the California Department of Food and Agriculture (CDFA 2002; California Department of Food and Agriculture Resource Directory 2002; [http://www.cdffa.ca.gov/card/pdfs/cdfa\\_rd02.pdf](http://www.cdffa.ca.gov/card/pdfs/cdfa_rd02.pdf)). The mean values are shown in **Table 6**.

**Table 6.** CAP88PC's agricultural parameter values representing LLNL.

Parameter	Value
Beef cattle density (# cows/km <sup>2</sup> )	1.9
Milk cattle density (# cows/km <sup>2</sup> )	4.0
Land fraction cultivated for vegetable crops	0.046

For individual dose from ingestion, it was assumed that 25% of the vegetables and meat are home-grown, while the remaining 75% of vegetables and meat and 100% of the milk is imported (i.e., free from LLNL-generated radioactivity). For collective dose, the urban default choice in CAP88-PC was used (in which 7.6% of vegetables, 0% of milk, and 0.8% of meat are home-grown, with the balances obtained from the assessment area exposed to the released radioactivity).

### Emission Source Terms

The source term for each emission source in the calculations was determined by one of three methods. For continuously monitored sources, the sampling data (curies released per unit time) for each radionuclide were used directly. For sources qualifying as "minor sources," ambient air

monitoring data were used to gauge the maximum dose to the public from their emissions (see the subsection on “Compliance Demonstration for Minor Sources” in Section II). For unmonitored facilities or activities that do not fall in the category of minor sources, or that were new operations in the year covered by the report, potential emissions to air were estimated based on radionuclide usage inventories and facility knowledge, or the combined use of surveillance air monitoring and air dispersion modeling. Generally, model runs for sources characterized by inventory data utilize “time factors” and EPA-specified physical state factors. Time factors adjust for the fact that a 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 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. However, the U.S. EPA has granted approval for LLNL to use alternative physical state factors based on actual physical form for elemental uranium, various uranium compounds/alloys, and elemental plutonium. **Table 7** provides the approved temperatures for application of the physical state factor for each of these materials. Otherwise, if the material was an unconfined gas, or any material heated above 100°C (with exceptions noted in **Table 7**), then the factor 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.

**Table 7.** List of materials exempted from the “treat as a gas above 100°C” rule and temperatures at which the various physical state factors apply.

Material	Solid physical state factor	Liquid physical state factor	Gas physical state factor	Year approved
Elemental uranium	<1100°C	Between 1100°C and 3000°C	>3000°C	1996
Uranium/niobium alloy	<1000°C	Between 1000°C and 3000°C	>3000°C	2001
Uranium oxide	<2000°C	Between 2000°C and 2500°C	>2500°C	2004
Uranium nitride	<2000°C	Between 2000°C and 2500°C	>2500°C	2004
Uranium carbide	<2000°C	Between 2000°C and 2500°C	>2500°C	2004
Elemental plutonium	<600°C	Between 600°C and 3000°C	>3000°C	2001

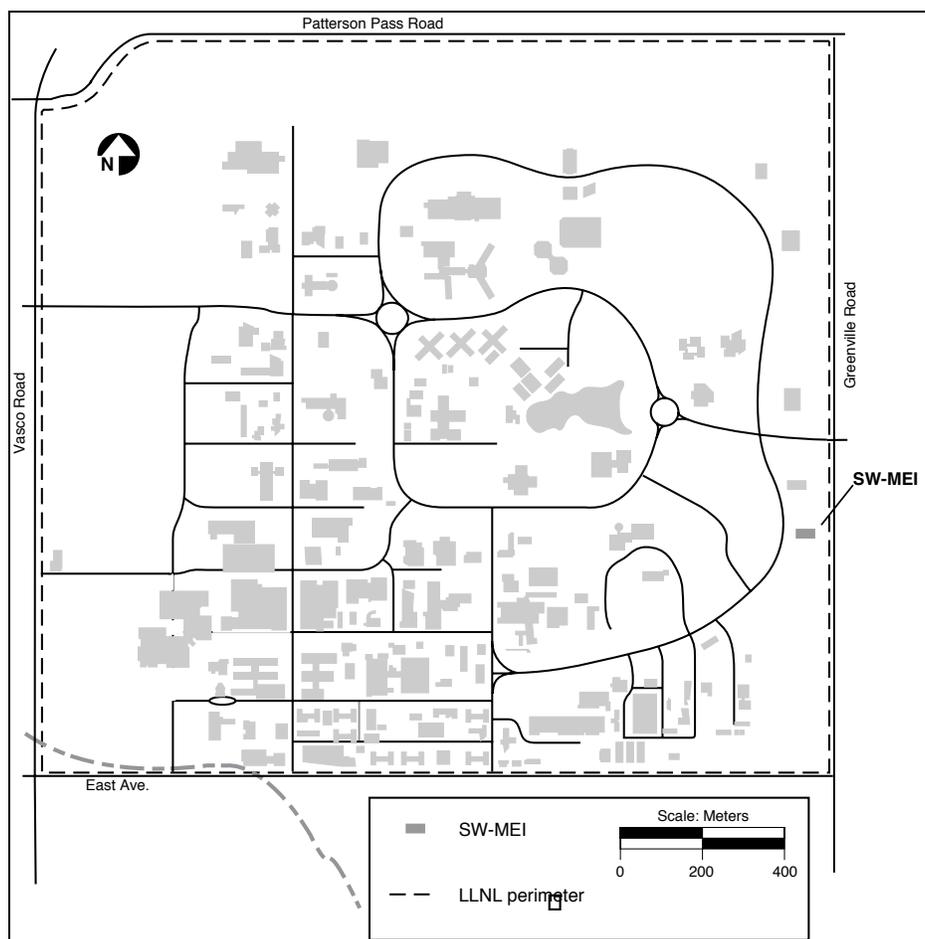
In addition to physical state factors, emission control abatement factors (40 CFR 61, Appendix D) were used when applicable. Each HEPA filter stage was given a 0.01 abatement factor. (However, abatement factors were not used to evaluate compliance with the 0.1 mrem [ $1 \mu\text{Sv}$ ] standard that determines the need for continuous monitoring at a facility.)

#### ***Site-Wide Maximally Exposed Individual***

For LLNL to comply with the NESHAPs regulations, the LLNL site-wide maximally exposed individual cannot receive an EDE greater than 10 mrem/y ( $100 \mu\text{Sv/y}$ ). The site-wide maximally exposed individual (SW-MEI) is defined as the *hypothetical* member of the public at a single residence, school, business, church, or other such facility who receives the greatest LLNL induced EDE from the combination of all evaluated radionuclide source emissions, as determined by modeling.

At the Livermore site, the SW-MEI for 2005 was found, as usual, to be located at the UNCLE Credit Union, about 10 m outside the controlled eastern fence line of the site, but about 10 m within the perimeter of the site property, as shown in **Figure 3**. At Site 300, the 2005 SW-MEI was located, as in the past several years, at the boundary with the Carnegie State Vehicle Recreation Area, managed by the California Department of Parks and Recreation, approximately 3.2 km south-southeast of the firing table at Building 851, as shown in **Figure 4**.

Doses to the SW-MEIs were evaluated for each source and then totaled for site-specific evaluations against the 10 mrem/y (100  $\mu$ Sv) dose standard (see “Total Dose to Site-Wide Maximally Exposed Individuals” in Section IV).

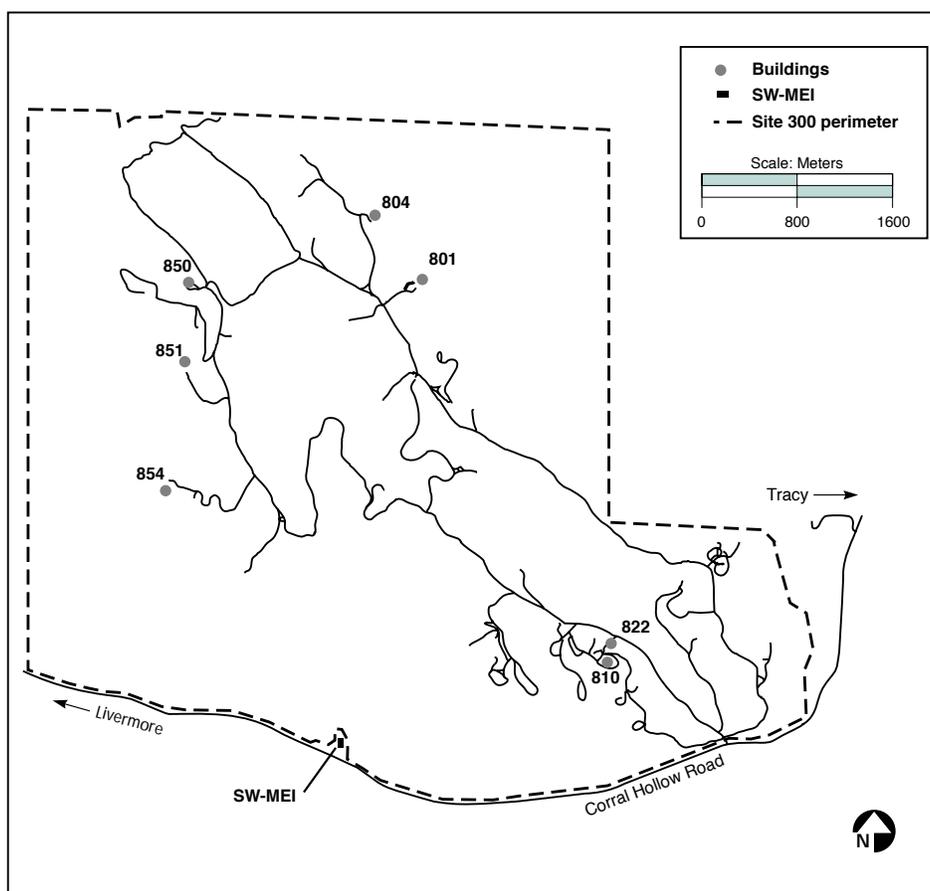


**Figure 3.** Location of Site-Wide Maximally Exposed Individual (SW-MEI) at the Livermore site, 2005.

### *Maximally Exposed Public Individual*

To assess compliance with the EPA requirement for continuous monitoring of a release point (potential dose greater than 0.1 mrem/y [1.0  $\mu$ Sv/y]), emissions must be individually evaluated from each point source to determine the dose to the maximally exposed individual (MEI) member of the public. The location of the MEI is generally different for each emission point, and must occur at a location of unrestricted public access. Typically, this location is a point on

the site perimeter, prompting the MEI dose to be referred to as the maximum “fence line” dose. However the off-site maximum dose can occur some distance beyond the perimeter, e.g., when a facility stack is close to the perimeter. Modeling calculations show that ground level concentrations of radionuclides can be expected to reach maximum values beyond the LLNL boundaries for releases from the DWTF stack on the Livermore site and dispersals from open-air explosives experiments conducted at Site 300. As stipulated by the regulations in 40 CFR Section 61.93 (b)(4)(ii), modeling for evaluation of the need for continuous monitoring must assume unabated emissions (i.e., no credit can be taken for emission abatement devices, such as filters). Model run documentation typically includes evaluation of the dose to the MEI, specification of emission abatement factors (in place but not credited for the required monitoring evaluation), and the distance and direction to the LLNL fence line point where (or beyond which) the MEI is located; see Attachment 1.



**Figure 4.** Location of Site-Wide Maximally Exposed Individual (SW-MEI) at Site 300, 2005.

## SECTION IV. Results of 2005 Radiological Dose Assessment

This section summarizes the doses to the most exposed public individuals from LLNL operations in 2005, shows the comparison to previous years, presents the potential doses to the populations residing within 80 km of either the Livermore site or Site 300, and summarizes LLNL's compliance with 40 CFR 61, Subpart H (61.93).

### Total Dose to Site-Wide Maximally Exposed Individuals

The total dose to the Livermore site SW-MEI from operations in 2005 was 0.0065 mrem (0.065  $\mu$ Sv). Of this, 0.0027 mrem (0.027  $\mu$ Sv), or 41%, was contributed by point sources, while diffuse emissions accounted for 0.0038 mrem (0.038  $\mu$ Sv), or 59%, of the total. The point source dose includes Tritium Facility HT emissions modeled as HTO, as directed by EPA Region IX. (See "Modeling Dose from Tritium" in Section VII for changes [decreases] in the dose from tritium when this assumption is not used.)

This SW-MEI dose is the lowest reported for the Livermore site since 1990, when NESHAPs reporting commenced. There were no significant changes in LLNL operations or changes to modeling assumptions in 2005, and so this dose is comparable to the value reported for 2004. The most significant factors leading to this and the 2004 low dose were the relatively low level of operations and emissions from the Tritium Facility (**Table 3**) and a readjustment of the food intake assumptions (see "Land Use and Agricultural Inputs" in Section III, *LLNL NESHAPs 2004 Annual Report*, Harrach et al., UCRL-ID-113867-05, June 2005). The reorganization of operations of the Radioactive and Hazardous Waste Management Division (see "Changes in Operations and Facilities" in Section VI) was also a contributing factor, although a far less significant one.

The total dose to the Site 300 SW-MEI from operations in 2005 was 0.018 mrem (0.18  $\mu$ Sv). Point source emissions from firing table explosives experiments accounted for about 48% of this total, while 52% was contributed by diffuse sources. The total dose to the Site 300 SW-MEI was comparable to historical values, although the percent contributions from point and diffuse sources did not reflect the typical values reported over the last decade. The reason for the change is the coincident occurrences at Site 300 of decreased Building 851 Firing Table activities and increased soil resuspension, attributable to an extensive wildfire in July.

**Table 8** shows the facilities or sources that collectively accounted for 98% or more of the doses to the SW-MEI for the Livermore site and Site 300 in 2005. Although LLNL has more than 150 sources with potential for releasing radioactive material to air according to NESHAPs prescriptions, most are very minor. Each year, nearly the entire radiological dose to the public from LLNL operations comes from no more than a dozen sources.

**Table 8.** Ranked list of facilities or sources whose emissions collectively accounted for nearly 100% of the SW-MEI doses for the Livermore site and Site 300 in 2005.

Facility (Source Category)	CAP88-PC Dose in mrem/y	CAP88-PC Percentage Contribution to Total Dose
<b>Livermore site</b>		
Building 331 stacks (point source)	0.0026	40%
Building 612 Yard (diffuse source)	0.0020	31%
Building 331 outside (diffuse source)	0.0012	18%
Southeast Quadrant (diffuse source)	0.00061	9%
<b>Site 300</b>		
Soil resuspension (diffuse source)	0.0094	52%
Building 851 Firing Table (point source)	0.0088	48%

**Table 9** compares 2005 doses with those of previous years. Diffuse source doses were not reported for the Livermore site for 1990 and 1991. In addition, no diffuse emissions were reported at Site 300 for years before 1993, so a comparison of the total Site 300 dose can only be made for 1993 and later.

**Table 9.** Doses (in mrem) calculated for the Site-Wide Maximally Exposed Individual (SW-MEI) for the Livermore site and Site 300, 1990 to 2005.

Year	Total Dose	Point Source Dose	Diffuse Source Dose
<b>Livermore site</b>			
2005	0.0065 <sup>a</sup>	0.0027 <sup>a</sup>	0.0038
2004	0.0079 <sup>a</sup>	0.0021 <sup>a</sup>	0.0058
2003	0.044 <sup>a</sup>	0.024 <sup>a</sup>	0.020
2002	0.023 <sup>a</sup>	0.010 <sup>a</sup>	0.013
2001	0.017 <sup>a</sup>	0.0057 <sup>a</sup>	0.011
2000	0.038 <sup>a</sup>	0.017 <sup>a</sup>	0.021
1999	0.12 <sup>a</sup>	0.094 <sup>a</sup>	0.028
1998	0.055 <sup>a</sup>	0.031 <sup>a</sup>	0.024
1997	0.097	0.078	0.019
1996	0.093	0.048	0.045
1995	0.041	0.019	0.022
1994	0.065	0.042	0.023
1993	0.066	0.040	0.026
1992	0.079	0.069	0.010
1991	0.234	— <sup>b</sup>	— <sup>b</sup>
1990	0.240	— <sup>b</sup>	— <sup>b</sup>

**Table 9. Continued**

Year	Total Dose	Point Source Dose	Diffuse Source Dose
<b>Site 300</b>			
2005	0.018	0.0088	0.0094
2004	0.026	0.025	0.00086
2003	0.017	0.017	0.00034
2002	0.021	0.018	0.0033
2001	0.054	0.050	0.0037
2000	0.019	0.015	0.0037
1999	0.035	0.034	0.0012
1998	0.024	0.019	0.005
1997	0.020	0.011	0.0088
1996	0.033	0.033	0.00045
1995	0.023	0.020	0.003
1994	0.081	0.049	0.032
1993	0.037	0.011	0.026
1992	0.021	0.021	— <sup>c</sup>
1991	0.044	0.044	— <sup>c</sup>
1990	0.057	0.057	— <sup>c</sup>

<sup>a</sup> The dose includes HT emissions modeled as HTO. Modeling HT emissions as such results in an overestimation of the dose. This methodology is used for purposes of compliance, as directed by EPA Region IX.

<sup>b</sup> Diffuse source doses were not reported for the Livermore site for 1990 and 1991.

<sup>c</sup> No diffuse emissions were evaluated at Site 300 for years before 1993.

### Doses from Unplanned Releases

There were no unplanned atmospheric releases of radionuclides at the Livermore site or Site 300 in 2005.

### Population Doses

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. This air dispersion and dose assessment model evaluates the four principal exposure pathways: ingestion through water (for tritium only) and food consumption, inhalation, air immersion, and irradiation by contaminated ground surface.

The CAP88-PC result for potential collective dose attributed to 2005 Livermore site operations was 1.2 person-rem (0.012 person-Sv); the corresponding collective EDE from Site 300 operations was 1.7 person-rem (0.017 person-Sv). For the Livermore site, this population dose is attributable to tritium, and for Site 300, the isotopes in depleted uranium (<sup>238</sup>U, <sup>235</sup>U, and <sup>234</sup>U). The value for the Livermore site collective dose from tritium was slightly higher than in 2004 as anticipated from the higher release rate of tritium from the Tritium Facility in 2005. These potential collective dose values are both quite small and within the normal range of variation seen from year to year. By way of comparison, the collective dose to the roughly 7 million people within 80 km of LLNL's two sites from exposure to the average level of natural background radioactivity in the United States is two million person-rem (twenty thousand person-Sv).

The collective doses from LLNL are high relative to many other DOE facilities because of the large populations lying within 80 km of the Livermore site and Site 300. Although the collective doses may be the same, a large dose to a small number of people is not equivalent to a small dose to many people. A better way to present the collective doses from LLNL operations is to disaggregate them into categories of individual dose, which demonstrates the tiny doses received by all of the population.

For the Livermore site, population doses from stack and area releases of tritium may be broken down as shown in **Table 10**. It can be seen in the table that the individuals that make up more than 99% of the population received less than 0.001 mrem/y (0.01  $\mu$ Sv/y).

**Table 10.** Disaggregations of collective dose for the Livermore site, 2005.

Individual dose mrem/y	Collective dose person-rem/y	Percent total collective dose
0.001 to 0.01	0.005	0.4%
0.0001 to 0.001	0.72	61.0%
0.00001 to 0.0001	0.34	28.9%
0.000001 to 0.00001	0.11	9.7%
Total*	1.2	100%

\* 0.05% of the individuals in the population received a dose of less than 0.000001 mrem/y.

Collective doses can be broken down similarly for the shots from the Building 851 Firing Table and the Contained Firing Facility at Site 300, as shown in **Table 11**. In this case individuals that make up about 94% of the population receive less than 0.001 mrem/y (0.01  $\mu$ Sv/y).

**Table 11.** Disaggregations of collective dose for Site 300, 2005.

Individual dose mrem/y	Collective dose person-rem/y	Percent total collective dose
0.001 to 0.01	0.11	6.3%
0.0001 to 0.001	1.06	62.0%
0.00001 to 0.0001	0.51	29.6%
0.000001 to 0.00001	0.03	2.0%
0.000000001 to 0.000001	0.003	0.2%
Total	1.7	100%

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

Calculations of effective dose equivalents for Livermore site and Site 300 facilities having the potential to release or releasing radioactive material to the atmosphere were found to be well below the 10 mrem (100  $\mu$ Sv) NESHAPs dose standard for dose to the maximally exposed individual members of the public. Tritium accounted for 91% of the Livermore site calculated dose, while at Site 300 the entire calculated dose was due to the isotopes  $^{238}\text{U}$ ,  $^{235}\text{U}$ , and  $^{234}\text{U}$ , in depleted uranium.

In 2005, there were seven buildings (Buildings 235, 251, 331, 332, 491, 695, and 696) at the Livermore site and one (Building 801A, the Contained Firing Facility) at Site 300 that had radionuclide air effluent monitoring systems. (Buildings 695 and 696 in the DWTF complex vent through a common stack.) These buildings are listed, along with the number of samplers, the types of samplers, and the analytes of interest in **Table 2** of Section II.

LLNL remains committed to monitoring stack effluent air from its Tritium Facility (Building 331), Plutonium Facility (Building 332), Decontamination and Waste Treatment Facility (Buildings 695 and 696), Contained Firing Facility (Building 801A), and the seismically hardened area of its Heavy Element Facility (Building 251). In addition, other facilities are continuously monitored, as necessary, based on evaluations of potential emissions without control devices, as in the case of Building 235, or where classification or other issues prevent a usage-inventory-based evaluation.

## SECTION V. Certification

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 personnel 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.

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**Signature:** William A. Bookless **Date:** 6/23/06  
William A. Bookless

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. See 18 U.S.C. 1001.

**Name:** Phillip Hill  
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**Signature:** Phillip Hill **Date:** 6/29/06  
Phillip Hill

## **SECTION VI. Supplemental Information on NESHAPs Compliance and QA/QC Activities**

### **Periodic Confirmatory Measurements**

Results of NESHAPs periodic confirmatory measurements (PCM) are intended to support or confirm two objectives: (1) that those operations not continuously monitored do not, in fact, need to be continuously monitored and (2) that radionuclide usage-inventory-based estimates of emissions and their corresponding doses are conservative.

For sources evaluated to have a potential to result in a dose less than the regulatory value of 0.1 mrem/y that requires continuous monitoring under Subpart H, LLNL achieves the PCM objectives by fulfilling the requirements stated in 40 CFR 61.93 paragraph (e) with its ambient air monitoring program. The ambient air monitoring effort includes thirty sampling locations with more than forty samplers placed in strategic areas (see the Air Monitoring Programs section in the LLNL Site Annual Environmental Report [<http://www.llnl.gov/saer>] for a description of LLNL's ambient air radiological monitoring).

### **NESHAPs Quality Assurance (QA) Program**

The LLNL NESHAPs quality assurance program is a multi-organizational effort. Its major components are the LLNL facilities/programs that have continuous stack effluent monitoring systems; the Radiological Measurements Laboratory (RML) and the Hazards Control Analytical Laboratory (HCAL), both in the Hazards Control Department (HCD); and the Environmental Protection Department (EPD). To coordinate the activities of these organizations, NESHAPs Agreement of Roles and Responsibilities (NARRs) documents are in place between EPD and the facilities and/or programs and HCD. NARRs formalize responsibilities and obligations of the organizations regarding many tasks for the air effluent sampling network. Tasks that are addressed in the NARRs include air sampler design and installation, procedures and their implementation, sampling, sample analysis and tracking, maintenance and repair of sampling systems, guidance on regulatory requirements, documentation of the sampling network, reporting, and the archiving of records.

The NESHAPs quality assurance project plan is included in the "NESHAPs Compliance Guidance Document and Quality Assurance Project Plan (G. Gallegos, EMP-NS-S, 2006). This document recites the key elements of the NESHAPs Quality Assurance Project Plan (QAPP) as specifically prescribed by 40 CFR 61, App. B, Method 114. Because LLNL's NESHAPs quality assurance activities are conducted by two LLNL departments, EPD and HCD, the documentation for the elements of a complete quality assurance project plan are independently maintained by these organizations. The NESHAPs QAPP presents a cross-walk between the requirements of a complete QAPP, the documents that meet those requirements, and the responsible organizations.

A general overview of these requirements and the responsible organizations is as follows. EPD is responsible for an annual assessment and demonstration of LLNL's compliance with NESHAPs, as documented in the present report. EPD's Terrestrial and Atmospheric Monitoring and Modeling (TAMM) Group is responsible for environmental monitoring; calibration, inspection, and maintenance of all stack sampling activities; air dispersion and dose assessment

modeling; assessment (in cooperation with Laboratory Program personnel) of usage of radioactive materials and their potential releases to air in operations throughout the Laboratory; record keeping; and reporting to EPA and DOE to demonstrate the Laboratory's compliance with NESHAPs. HCD is responsible for conducting the stack sampling and radiological analyses. HCD is also responsible for assuring the quality of the samples, sample tracking, and analytical quality control. The LLNL Assurance Review Office periodically audits EPD and HCD activities.

### **Evaluation of New Radiological Projects**

The TAMM Group is informed of proposed new operations and modified operations where significant changes in radiological usage inventories occur by several mechanisms. These include reviews of National Environmental Policy Act (NEPA) documentation, Integration Work Sheets, Occupational Safety Plans (describing facility-specific safety procedures and plans), and knowledge derived from participation on EPD's Environmental Support Teams (ESTs). In the NESHAPs context, the EST representatives from the TAMM Group and the Environmental Operations Group (EOG) have primary responsibilities. Written communications between NESHAPs analysts and project principal investigators, including records of model runs carried out to evaluate the need for monitoring of radiological releases and the need to obtain permission from EPA to start up operations, are retained in the TAMM Group for at least the period of time specified in 40 CFR 61 Subpart H.

### **Quality Control (QC) for 2005 Air Dispersion and Dose Assessment Model Runs**

The only radiological facilities or projects providing an accounting by means of radionuclide inventories were ones commencing operation in 2005 or unmonitored point source releases that contributed significantly in 2005 to the dose to the public. The former underwent NESHAPs evaluation in which NEPA or other documents such as Integration Work Sheets and Occupational Safety Plans were examined prior to start-up of operations, and CAP88-PC model runs were performed to determine the maximum potential doses to the public from the activities. The latter were six explosives experiments conducted in 2005 at Firing Table 851 at Site 300. Both the input data and model runs for all six explosives experiments were independently checked and validated.

Model runs were performed for about one dozen sources in the 2005 assessment, including the activities mentioned above and two stack-monitored facilities that released tritium to air (the Tritium Facility and Decontamination and Waste Treatment Facility [DWTF]). More than half of all model runs were recalculated independently. Facility personnel reviewed and concurred with source term data inferred by the NESHAPs analysts for the Building 331 Outside Yard.

Copies of individual model runs, including input parameters and resultant calculated doses, are archived in the records kept by the TAMM Group.

Based on these QC efforts, the data, results, and conclusions presented in this report meet applicable quality assurance objectives.

## **SECTION VII. Supplementary Information on Radiological Dose Assessment for 2005**

### **Livermore Site Principal Diffuse Sources**

The dose evaluations for diffuse sources at the Livermore site in 2005 required several different modeling approaches. Building 331 WAA, Building 612 Yard, and DWTF Transportainer Storage emissions estimates were based on back-calculations in which the CAP88-PC air concentration for unit source strength in model runs was used to convert the concentration determined from environmental surveillance air monitoring data into a source term. The dose in each of these cases was calculated using CAP88-PC. Air surveillance monitoring data for plutonium from two ambient air monitors at the location of the SW-MEI and at the Discovery Center were used directly to evaluate the dose from historical plutonium contamination in the Southeast Quadrant.

#### ***Building 331 Outside Yard***

As the Tritium Facility (Building 331) conducts operations, tritium-contaminated equipment and material slated for disposal are packaged in a storage area, removed from the building to an outside storage container, and finally sent to Radioactive and Hazardous Waste Management Division (RHWM) facilities. During 2005, outgassing from such waste released an estimated 4.8 Ci ( $1.8 \times 10^{11}$  Bq) of tritium to the atmosphere outside Building 331. This amount was derived from a combination of environmental surveillance monitoring data and air dispersion back-calculation, and agreed with estimates based on process and facility knowledge. Its release was modeled in CAP88-PC as a  $1 \text{ m}^2$  area source, leading to a calculated 2005 dose to the SW-MEI of  $1.2 \times 10^{-3}$  mrem ( $1.2 \times 10^{-2}$   $\mu\text{Sv}$ ). A dose 0.89 times this amount was calculated using the NEWTRIT model with air concentrations calculated by CAP88-PC (see "Modeling Dose from Tritium" later in this section).

#### ***Building 612 Yard***

The Building 612 Yard is a potential source of diffuse emissions of tritium. This area is dedicated to hazardous waste, radioactive waste, and mixed waste management activities. The yard consists of several areas where waste containers are stacked outdoors. Several of these containers outgas tritium. A surveillance air monitor designated B624 has been placed in the Building 612 Yard to provide continuous measurements of tritium in air near this source. The mean annual concentration of tritium in air for 2005 in this area was  $39.7 \text{ pCi/m}^3$  ( $1.5 \text{ Bq/m}^3$ ). This value was used to calculate the total tritium emissions from the area using a conservative approach that assumed the source to be 60 m south-southwest of the air sampler. With this assumption, a diffuse source emission of 1.5 Ci/y ( $5.6 \times 10^{10}$  Bq/y) was required to produce the concentrations measured at the air sampler. This source term produced a CAP88-PC calculated 2005 dose to the SW-MEI from the Building 612 Yard of  $2.0 \times 10^{-3}$  mrem ( $2.0 \times 10^{-2}$   $\mu\text{Sv}$ ). As in the preceding section, a dose 0.89 times this amount was calculated using the NEWTRIT model with air concentrations calculated by CAP88-PC. (Under LLNL's presently used ingestion assumptions, the ratio of dose predicted by NEWTRIT to that by CAP88-PC is always 0.89 for a source releasing only HTO.)

### ***DWTF Transportainer Storage***

The Decontamination and Waste Treatment Facility is comprised of five buildings and, in addition, as part of the operations of this facility, transportainers are stored outside. In 2005, one transportainer stored east of Building 693 contained some tritium. Using back-calculation from the DWTF ambient air sampler to this diffuse source, it is estimated that 0.21 Ci ( $7.77 \times 10^{10}$  Bq) was released during 2005 from the transportainer. The dose contribution from this source to the SW-MEI is negligible ( $3.7 \times 10^{-5}$  mrem/y [ $1.7 \times 10^{-4}$   $\mu$ Sv/y]). The dose to the MEI on the perimeter of the site is bigger ( $2.9 \times 10^{-3}$  mrem/y [ $1.9 \times 10^{-2}$   $\mu$ Sv/y]) because the source is diffuse and in close proximity to the perimeter of the site. The dose calculated using NEWTRIT was 0.89 times these amounts.

### ***Southeast Quadrant***

The Southeast Quadrant of the Livermore site has elevated levels of plutonium in the surface soil (from historic waste management operations) and air (from resuspension). A high volume air particulate sampler is located adjacent to the UNCLE Credit Union (the location of the SW-MEI) and a second sampler is located next to the Discovery Center to monitor the plutonium levels in this area. Monitoring data from these air samplers were used as a direct measurement of potential dose via the air pathway. The 2005 mean annual concentration in air of  $^{239+240}$ Pu (alpha spectroscopy does not distinguish between  $^{239}$ Pu and  $^{240}$ Pu) for all results greater than zero was  $2.4 \times 10^{-19}$  Ci/m<sup>3</sup> ( $8.7 \times 10^{-9}$  Bq/m<sup>3</sup>). Using the dose conversion factor of  $3.08 \times 10^5$  mrem/ $\mu$ Ci ( $8.32 \times 10^{-5}$  Sv/Bq) from Federal Guidance Report No. 11, EPA-520/1-88-020, U.S. Environmental Protection Agency (1988) for  $^{239}$ Pu and  $^{240}$ Pu, and the reference man breathing rate of 8400 m<sup>3</sup>/y (International Commission on Radiological Protection [ICRP], 1975, *Reference Man: Anatomical Physiological and Metabolic Characteristics*. Oxford: Pergamon Press; ICRP Publication 23), the dose was determined to be  $6.1 \times 10^{-4}$  mrem ( $6.1 \times 10^{-3}$   $\mu$ Sv) for 2005.

### **Site 300 Principal Diffuse Sources**

Diffuse sources at Site 300 predominantly feature the radioisotopes in depleted uranium, with trace amounts of tritium being the only other radiological component of concern as having potential for release to air.

#### ***Tritium Evaporation and Migration at Site 300***

Tritium gas and solids containing tritium ( $\text{Li}^3\text{H}$ ) were components of explosives assemblies tested on the firing tables during experiments in years past. Most of the gaseous tritium escaped to the atmosphere during the tests, but some of the solid  $\text{Li}^3\text{H}$  remained as residue in the firing table gravel. Rainwater and dust-control rinse water percolated through the gravel, causing the tritium to migrate into the subsurface soil and, in some cases, eventually to the ground water. Tritium contaminated gravel was removed from the firing tables in 1988 and disposed in the Pit 7 landfill. Tritium in landfills, firing table soils, and ground water are potential sources of diffuse emissions of tritium to the atmosphere at Site 300. LLNL personnel maintain an air tritium sampler at a perimeter location at Site 300, and doses from diffuse tritium sources may be estimated based on the monitoring data for that sampling location. For the calendar year 2005, all measurements in ambient air at the Site 300 perimeter location were at or near the minimum detection limit of the analytical method (about 0.65 pCi [ $25$  mBq]/m<sup>3</sup>).

### ***Resuspension of Depleted Uranium in Soil at Site 300***

Depleted uranium has been used as a component of explosives test assemblies over many years. 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.

A model was developed to distinguish between the contribution to measured uranium activities arising from naturally occurring uranium (NU) and that from depleted uranium (DU) contributed by LLNL operations. (A derivation of the model was presented in Gallegos et al., *LLNL NESHAPs 1995 Annual Report*, UCRL-ID-113867-96, June 1996.) We base our dose estimate for resuspended 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  $\mu$  is the fraction (by weight) of uranium contributed by operations, CU is composite uranium (both DU and NU),  $M(CU-235)$  is the mass of U-235 in the composite (measured) uranium, and  $M(CU-238)$  is the mass of U-238 in the composite (measured) uranium.

For 2005, all eight air-particulate monitors at Site 300 were used to determine the annual-average concentrations of isotopes U-238 and U-235. These site-average values gave an estimate of  $9.4 \times 10^{-3}$  mrem ( $9.4 \times 10^{-2}$   $\mu$ Sv) for the SW-MEI dose resulting from resuspension of DU in soil for 2005, the highest diffuse source dose reported in the last decade (see **Table 9**). The elevated dose is attributed to increased resuspension of particulate from, by far, the most aggressive wildfire to burn at Site 300 in the past ten years. This July 2005 wildfire consumed more than 6,200 acres, approximately one-third of which burned at Site 300, causing elevated concentrations of DU at two air particulate sampling locations downwind from the fire and near its perimeter. (For more information on the sampling data, see the "Air Monitoring Programs" chapter in LLNL's Site Annual Environmental Report for 2005.)

### **Radioactive and Hazardous Waste Management Operations and Facilities**

A state-of-the-art integrated facility for storing and processing hazardous, radioactive, and mixed wastes, LLNL's DWTF, opened for operation in September 2003. Five buildings comprise the facility: Building 695, Building 696, and three others (see the report "Recent Advances in the Environmental Protection Department," UCRL-BR-208053, Dec. 2004, pp. 15-17, for a description of the facility). Building 695 and Building 696 share a complex ventilation system that connects to the atmosphere through DWTF's HEPA-filtered, continuously monitored (for radioactive particles) 30-m stack.

Building 695 houses most of the higher-dose activities attributed to RHWM in recent years. Regarding maximum potential dose to the public, these activities in 2002 and 2003 contributed approximately 0.003 mrem/y (0.03  $\mu$ Sv) to the SW-MEI dose. In 2004, dose to the SW-MEI was determined by back-calculation using ambient air tritium data from the DWTF air tritium

sampler and facility knowledge (as opposed to the previous inventory based determinations). The result was a decrease in dose by approximately a factor of four compared to 2002 and 2003 for these operations, to a level of 0.0007 mrem (0.007  $\mu$ Sv). In 2005, this number was determined using continuous stack monitoring data and the dose was 0.00008 mrem (0.0008  $\mu$ Sv), even lower than the result for 2004 by nearly a factor of nine. The decrease in the contribution from these operations reflects improvements in RHW's infrastructure, relocation of these operations relative to the location of the SW-MEI, and the different methodologies used for calculating the potential dose.

### **Modeling Dose from Tritium**

To evaluate dose from tritium releases to air, we use the EPA-sanctioned CAP88-PC code. Its tritium model calculates dose from inhalation, skin absorption, and ingestion of tritium only in its tritiated water vapor form (HTO). Doses from releases of tritiated gas (HT) or organically bound tritium (OBT) are not calculated. CAP88-PC's tritium model is based on specific activity and 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.

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

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

A simple tritium model, NEWTRIT, has been developed that calculates ingestion dose from both HTO and OBT and accounts for conversion of HT to HTO in the environment following releases of HT (Peterson, S-R. and P.A. Davis, *Health Physics* 82(2): 213-225, 2002). A discussion of the NEWTRIT model was presented in Attachment 2 of the 2000 NESHAPs annual report (Gallegos et al., *LLNL NESHAPs 2000 Annual Report*, UCRL-ID-113867-01, June 2001).

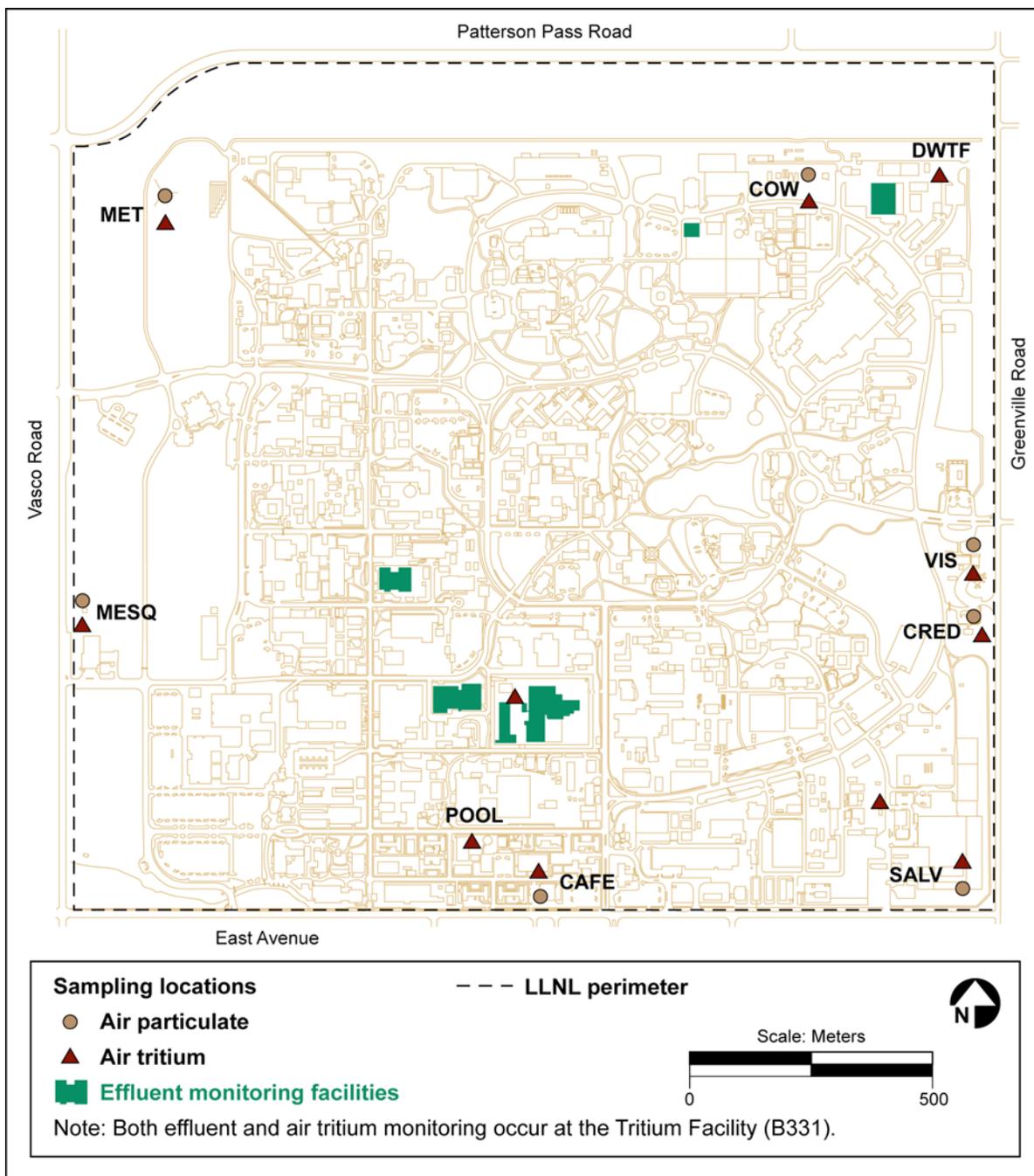
Tritium doses from 2005 Livermore site operations were calculated using NEWTRIT and compared to those obtained by our standard procedure using CAP88-PC (the latter are presented in Section IV). NEWTRIT does not model dispersion, so tritium concentrations in air calculated

by CAP88-PC are used as input. For the principal comparison of the total tritium contribution to the Livermore site SW-MEI dose in 2005 calculated using NEWTRIT instead of CAP88-PC, the result was 0.0051 mrem (0.051  $\mu$ Sv), about 15% lower than the CAP88-PC value of 0.0059 mrem (0.059  $\mu$ Sv). Both NEWTRIT and CAP88-PC doses for each significant source of tritium are presented in the data spreadsheet (columns 16 and 19) in Attachment 1.

In October 2001, LLNL sent a letter to EPA Region IX requesting consideration of NEWTRIT as an alternative methodology for calculating doses from atmospheric releases of tritiated water vapor (HTO) and tritiated gas (HT) for use in demonstrating compliance with radionuclide NESHAPs (40 CFR 61 Subpart H). In late 2003, the EPA had NEWTRIT coded into GENII-NESHAPs, a version of GENII that the EPA plans to approve as a regulatory model for evaluating radionuclide NESHAPs compliance (B.A. Napier, et al., GENII - The Hanford Environmental Radiation Dosimetry Software System. Richland, WA: Pacific Northwest Laboratory, PNL-6584 Vol. UC-60; 1988). At this writing, GENII-NESHAPs is undergoing peer review and should be approved soon. As well, DOE surveyed users of CAP88-PC (currently the code most-used for compliance) and determined that there is support to have NEWTRIT incorporated into EPA's CAP88-PC. However, NEWTRIT was not coded into CAP88-Pc Version 3.0, approved February 2006.

### **Comparison of 2005 Modeling Results with Tritium Surveillance Air Monitoring Data**

A comparison was made between CAP88-PC-predicted concentrations of tritium in air and ambient air monitoring data for nine tritiated water vapor samplers on the Livermore site (designated POOL, CAFE, MET, CRED, VIS, DWTF, COW, B331, and B624). Other locations (MESQ, SALV, and ZON7) that have been used for the comparison in past years were sampled normally, but more than half of their biweekly concentrations were below the lower limits of analytical detection, making any comparison with predicted results meaningless. **Figure 5** shows the locations of the tritium air surveillance monitors. Modeled predictions have been compared with tritium monitoring data since 1997.



**Figure 5.** Radiological air monitoring at the Livermore site showing locations for air surveillance monitoring of tritiated water vapor (triangles) and radioactive particles (circles) and stack air effluent monitoring (indicated by building number and icon).

Only the four most significant sources of tritium releases to air at the Livermore site (B331 stacks, B331 Outside Yard, B612 Yard, and DWTF Stack) were included in the initial model-data comparison. The release of HTO from the two 30-m-high, continuously monitored stacks at the Tritium Facility (Building 331) was determined from stack monitoring data and emission reconstruction to be 30.2 Ci ( $1.1 \times 10^{12}$  Bq) in 2005. (An estimated 1.57 Ci [ $5.8 \times 10^{10}$  Bq] of

HT emitted from the Tritium Facility stacks is not included in the comparison calculation because the tritium air surveillance monitors only absorb HTO.) Stack monitoring of the Decontamination and Waste Treatment Facility determined a release rate of 2.6 Ci HTO; the 0.1 Ci of HT released was ignored for this comparison. The other two principal sources in our initial modeling/measurement comparison were open-air diffuse emission areas associated with the Building 612 Yard and the Tritium Facility's outside yard storage area. Emissions from the Building 612 Yard source were estimated to be 1.5 Ci ( $5.6 \times 10^{10}$  Bq) based on back-calculating a source term from observed tritium concentrations at the tritium monitor B624. (Thus the B624 data do not provide a test of the dispersion modeling.) The release rate for the B331 area source was determined by back-calculation to be 2.3 Ci ( $8.5 \times 10^{10}$  Bq) for fifty weeks in 2005. This value differs from that used to calculate the annual dose to the SW-MEI from this source (4.8 Ci;  $1.8 \times 10^{11}$  Bq) because it does not include an extremely high concentration (640 pCi/m<sup>3</sup>; 23.7 Bq/m<sup>3</sup>) from the December 1-15 sampling period. This very high sample contaminated the Analytical Laboratory and the other samples being prepared at the same time. Thus all other samples for that sampling period were rejected and their values could not be included in the calculated mean annual concentrations used in this comparison. While these two diffuse sources contribute significantly to tritium concentrations at all monitoring locations, other potential sources of tritiated water vapor release were thought to be too minor to influence the overall model-data comparison.

Annual mean concentrations of HTO in air (pCi/m<sup>3</sup>) at the nine air tritium samplers were initially modeled for each of the four principal sources and the sum of the four contributions was compared to the measured annual mean concentrations. The mean observed concentration was underestimated at the DWTF sampler by a factor of 2.5 which indicated a contribution from another tritium source. The additional source was a transportainer containing tritium located in the DWTF yard (see Section VII, "DWTF Transportainer Storage"). The tritium estimated to have been released from the transportainer in 2005 was then included in the model/data comparison. The results, displayed in **Table 12**, show that fairly good agreement is obtained between predictions and observations for all air tritium monitors.

Air concentrations were overestimated by CAP88-PC at six of the eight locations at which the model was tested. The under-predictions seen at COW and MET are probably due to outgassing of tritium from a transportainer containing tritiated wastes that was removed from a building undergoing renovations. The shift from under- to over-prediction for DWTF emphasizes the importance of even small sources when they are diffuse and close to a receptor. These model predictions are consistent with other tests of CAP88-PC (S-R. Peterson, "Testing CAP88-PC's Predicted Air Concentrations Against Historical Air Tritium Monitoring Data, 1986-2001, at Lawrence Livermore National Laboratory," Health Physics 87(6):583-595. 2004; Jack Faucett Associates, Report JACKFAU-341/12-87; 1987).

**Table 12.** Comparison of measured and modeled annual mean concentrations of tritiated water vapor (HTO) in air at selected Livermore site locations, 2005.

Air monitor (name)	Mean measured concentration (pCi/m <sup>3</sup> )	Modeled* average concentration (pCi/m <sup>3</sup> )	Ratio of modeled-to-measured concentrations	Modeled concentration of tritium in air contributed by the indicated source (pCi/m <sup>3</sup> )				
				B331 Stacks	B612 Yard	B331 WAA	DWTF Stack	DWTF Outside
B331	21.7	22.9	1.1	0.041	0.75	22	0.058	0.019
B624	40.5	42.5	1.0**	1.2	40.5	0.26	0.027	0.0095
DWTF	3.75	5.29	1.4	0.95	0.14	0.17	0.23	3.8
POOL	3.37	4.78	1.4	1.5	0.63	2.6	0.039	0.014
COW	2.87	2.13	0.74	1.05	0.14	0.24	0.014	0.690
MET	1.30	0.802	0.62	0.43	0.088	0.21	0.044	0.03
CRED	1.29	2.71	2.1	1.10	1.3	0.26	0.031	0.016
VIS	1.27	2.21	1.7	1	0.94	0.22	0.029	0.022
CAFÉ	1.12	1.91	1.7	0.64	0.59	0.64	0.030	0.010

\* This result takes into account the four most significant tritium sources and a minor source near the DWTF; it is the annual-mean concentration comprising the sum of the five contributions shown in the far right columns.

\*\* This agreement was obtained by calibration.

## **SECTION VIII. Supplemental Information on Other Compliance**

### **Status of Compliance with Other Regulations**

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

LLNL does not have storage and disposal facilities for radium containing materials that would be a significant source of radon. Emissions of radon from LLNL research experiments did not occur in 2005.

#### ***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.

## **ATTACHMENT 1. LLNL NESHAPs 2005 Annual Report Guidance and Spreadsheet**

### **Guidance for Interpreting the Data Spreadsheet**

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

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

#### ***Radionuclides***

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

#### ***Radionuclide Usage Inventories***

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

#### ***Physical State Factors***

The physical state factors listed are EPA potential release fractions from 40 CFR 61, Appendix D, whereby emissions are estimated from radionuclide usage inventories depending on their physical states for use in dispersion/dose assessment modeling. A physical state factor of  $1.0 \times 10^{-6}$  is used for solids,  $1.0 \times 10^{-3}$  is used for liquids and powders, and 1.0 is used for unconfined gases and substances heated above 100°C. Regarding the latter, U.S. EPA has granted LLNL approved alternative emissions factors for selected radionuclides (see **Table 7** in Section III.) These factors are allowed provided that the material is not intentionally dispersed to the environment and that the processes do not alter the chemical form of the material.

### ***Stack Parameters***

Stack physical parameters for sources evaluated in 2005 were updated, as necessary, by experimenters and managers for those facilities. The TAMM Group annually measures the stack velocity of each monitored stack.

### ***Emission Control Devices***

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

### ***Control Device Abatement Factors***

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

### ***Estimated Annual Emissions***

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

Actual emission measurements are the basis for reported emissions from continuously monitored facilities. LLNL facilities that had continuous monitoring systems in 2005 were Buildings 235, 251, 331, 332, 491, and 695/696 at the Livermore site, and Building 801A (the Contained Firing Facility) at Site 300, as noted earlier in the subsection on "Compliance with 40 CFR Subpart H (61.93)" in Section IV. See also the discussion below under "0.1 mrem/y Monitoring Requirement" regarding the use of emissions measurements for monitored sources.

### ***10 mrem/y Site-Wide Dose Requirement***

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

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

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

### ***Source Categories***

LLNL radionuclide air emission sources have been classified into seven source categories, indicated by the number in the last column of the following spreadsheet: (1) unmonitored or non-continuously monitored Livermore site facilities that have had a radionuclide usage inventory update for 2005, (2) unmonitored or non-continuously monitored Livermore site facilities with a previous radionuclide usage inventory update, (3) continuously monitored Livermore site and Site 300 facilities, (4) Site 300 explosives experiments, (5) diffuse sources where emissions and subsequent doses were estimated using inventory processes, (6) diffuse sources where emission and dose estimates were supported by environmental surveillance measurements, and (7) sources whose emissions estimates and subsequent doses were estimated by confirmatory air sampling rather than continuous sampling.

Attachment 1 - 2005 LLNL NESHAPs Annual Report Spreadsheet

Building	Room/Area	Stack ID	Operation	Radionuclides	Annual Inventory with Potential for Release (Ci)	Physical State Factor	Stack Height (m)	Stack Diameter (m)	Stack Velocity (m/s)	Control Device(s)	Control Device Abatement Factor	Estimated Annual Emissions (Ci)	0.05 mrem/y Site-Wide Dose Requirement Distance to SWMEI (m)	Direction to SWMEI	EDE (mrem)	0.1 mrem/y Monitoring Requirement Distance to MEI (m)	Direction to MEI	Unabated EDE (mrem)	Source Category
<b>LIVERMORE SITE POINT SOURCES</b>																			
Building 235 is part of the Chemistry and Materials Sciences Directorate. Operations in the facility include examination of material structure, surface, and subsurface; precision cutting, ion implanting, and metallurgical studies.																			
235	1130	FHE-1A/1B, FHE2A/2B, and FGBE-1A/1B through FHE-1000/2002	Preparation of plutonium samples for diamond anvil studies	Gross alpha Gross beta	<sup>a</sup> <sup>a</sup>	NA NA	10.7	0.30	7.2	Double HEPA	0.0001	0.0E+00 0.0E+00	1065	ENE	0.0E+00	b	b	b	c
Building 251, the Heavy Element Facility, is managed by the Safety and Environmental Protection Directorate for the Institution as a non-operational facility in which transuranic isotopes remain until they can be disposed. One area of the facility has been "hardened" to resist damage from earthquakes. Room exhausts from this hardened area are double HEPA filtered; glove box exhausts are triple HEPA filtered. Exhausts from the unhardened area, also HEPA filtered, are continuously sampled by simple filter systems.																			
Unhardened Area <sup>c</sup>																			
251	1003	FHE-5	General chemistry	Gross alpha	<sup>a</sup>	NA	4.3	0.26	7.6	HEPA	0.01	0.0E+00	1188	E	6.8E-07	b	b	b	c
	1003	FHE-4		Gross beta			4.3	0.27	7.6			0.0E+00							
	1142	FHE-8					4.3	0.32	3.6										
	1142	FHE-9					4.3	0.26	4.7										
	1142	FHE-10					4.3	0.28	5.1										
	1150	FGBE-33,34					8.0	0.15	2.4										
	1150	FFE-15					4.3	0.31	4.8										
	1165	FGBE-31,32					5.5	0.87	2.8										
	1211	FHE-6					6.4	0.25	6.9										
	1211	FHE-7					6.4	0.25	4.5										
	1212	FGBE-15,16					5.5	0.10	7.4										
	1232	FGBE-38,39					7.2	0.15	3.2										
	1234	FFE-9					4.3	0.19	2.8										
	1235	FFE-12					4.3	0.25	7.4										
	1235	FGBE-29,30					5.5	0.13	9.3										
	1363	FHE-12					4.3	0.32	9.3										
	1363	FHE-13					6.4	0.28	7.9										
	1364	FFE-23					4.3	0.34	11.9										
	1364	FGBE-35,36					4.3	0.13	6.4										
	1314, 1354	FGBE-44,45					10.2	0.15	2.6										
	Hot cells	FGBE-40,41					5.5	0.23	3.4										
	Hot cells	FGBE-42,43					5.5	0.36	9.6										
	1150	FFE-13					5.5	0.28	4.8										
Hardened Area																			
251	Glove Boxes <sup>c</sup>	FGBE-1000 FGBE-2000	Previous transuranic research	Gross alpha Gross beta	<sup>a</sup>	NA	7.8 7.8	0.30 0.30	5.5 6.5	Triple HEPA	0.000001	0.0E+00 0.0E+00	1188	E	0.0E+00	b	b	b	c
	Room Exhaust <sup>c</sup>	FFE-1000 FFE-2000		Gross alpha Gross beta	<sup>a</sup>	NA	7.8 7.8	0.50 0.50	11.2 10.8	Double HEPA	0.0001	0.0E+00 0.0E+00	1188	E	0.0E+00	b	b	b	c
Building 331 is operated by the Defense and Nuclear Technologies Directorate. The building houses the tritium research facility and associated laboratories.																			
331	All <sup>f</sup>	Stack 1 Stack 2	Tritium research and development Decontamination of parts	H-3 H-3	<sup>d</sup> <sup>d</sup>	1.0E+00 1.0E+00	30.0 30.0	1.22 1.22	5.4 7.4	None None	1 1	5.2E-01 3.1E+01	957	ENE	2.6E-03 2.2E-03 <sup>g</sup>	573	SW	2.9E-03 2.4E-03 <sup>g</sup>	c
Building 332 is operated by the Defense Sciences Program for plutonium research. Exhausts from glove box operations and the workplace are triple filtered by high efficiency particulate air (HEPA) filters. Exhausts are monitored with both continuous filter sampling (PAMs) and plutonium-specific, continuous real-time monitors (CAMs).																			
332	Increment 1 Rooms	FHE-1000/2000	Plutonium research	Transuranics	<sup>a,j</sup>	NA	8.8	0.8x1.1	16.7	Double HEPA	0.0001	0.0E+00	912	ENE	0.0E+00	b	b	b	c
332	Increment 1 Glove boxes	FGBE-1000/2000	Plutonium research	Transuranics	<sup>a,j</sup>	NA	11	0.3	5.2	Triple HEPA	0.000001	0.0E+00	912	ENE	0.0E+00	b	b	b	c
332	Loft	FE-4,5W FE-4,5E	Loft exhaust Loft exhaust	Transuranics Transuranics	<sup>a,j</sup> <sup>a,j</sup>	NA	11 11	0.6x0.9 0.6x0.9	3.8 3.3	HEPA HEPA	0.01 0.01	0.0E+00 0.0E+00	912 912	ENE ENE	0.0E+00 0.0E+00	b b	b b	b b	c
332	Increment 1 Glove boxes	FGBE-3000/4000	Plutonium research	Transuranics	<sup>a,j</sup>	NA	11	0.3	5.4	Triple HEPA	0.000001	0.0E+00	912	ENE	0.0E+00	b	b	b	c
332	Increment 3 Glove boxes	FFE-1000/2000 FGBE-7000/8000	Plutonium research Plutonium research	Transuranics Transuranics	<sup>a,j</sup> <sup>a,j</sup>	NA	10.1 10.1	0.9 0.27	10.4 2.8	com—Double HEPA ve Box—Triple HE	0.0001 0.000001	0.0E+00 0.0E+00	912	ENE	0.0E+00 0.0E+00	b b	b b	b b	c
Building 491 is operated by the Space Action Team as an area for the storage of contaminated parts. Isotope separation activities that previously occurred in this building have been discontinued. Stack sampling is continuous. The facility operates with two in-series high efficiency particulate (HEPA) filter banks to control emissions.																			
491	All	FFE-1	Storage	Gross alpha Gross beta	<sup>g</sup> <sup>g</sup>	NA	9.1	0.9	6.1	Double HEPA	0.0001	0.0E+00 0.0E+00	1000	SSE	0.0E+00	b	b	b	c
Building 695/696 is the Decontamination Waste Treatment Facility operated by Radiological and Hazardous Waste Management Division. All operations are HEPA filtered and have pre-filters in place; some operations have additional HEPA filtration.																			
695/696	DWTF	FHE 1000/2000/3000	Waste treatment	Gross alpha Gross beta	<sup>a</sup> <sup>a</sup>	NA	20.0	1.98	10.5	HEPA Pre-filter	0.01 0.1	0.0E+00 0.0E+00	953	S	8.4E-05 7.2E-05 <sup>g</sup>	198	ENE	0.00074 <sup>h</sup> 8.7E-04 <sup>g</sup>	c

Attachment 1 - 2005 LLNL NESHAPs Annual Report Spreadsheet

Building	Room/Area	Stack ID	Operation	Radionuclides	Annual Inventory with Potential for Release (Ci) <sup>d</sup>	Physical State Factor	Stack Height (m)	Stack Diameter (m)	Stack Velocity (m/s)	Control Device(s)	Control Device Abatement Factor	Estimated Annual Emissions (Ci)	0.1 mrem/y Site-Wide Dose Requirement Distance to SWMEI (m)	Direction to SWMEI	EDE (mrem)	0.1 mrem/y Monitoring Requirement Distance to MEI (m)	Direction to MEI	Unabated EDE (mrem)	Soil Category
				Tritium		NA						2.7E+00							
<b>SITE 300 POINT SOURCES</b>																			
Building 801 is the Contained Firing Facility, where explosives tests are conducted. This facility and the 851 Firing Table are operated by the Defense and Nuclear Technologies Directorate.																			
801	Contained Firing Facility	FEFH-1, FE-2	Explosive tests	U-238 U-235 U-234	<sup>a</sup> <sup>a</sup> <sup>a</sup>	NA NA NA	16.8	1.60	7.8	HEPA Pre-filter	0.01 0.1	4.2E-07 8.4E-10 2.1E-12	3770	S	7.9E-07				
Explosives tests in which radionuclides may be present are conducted on open-air firing tables located at Bunker 851. These tests have depleted uranium material as part of the material inventory. There are multiple tests per year.																			
851	Firing Table	None	Explosive tests	U-238 U-235 U-234	8.9E-03 1.1E-04 8.3E-04	1 1 1	NA	NA	NA	None	1	8.9E-03 1.1E-04 8.3E-04	3170	SSE	8.8E-03	1396	WSW	8.9E-03	4
<b>LIVERMORE SITE DIFFUSE SOURCES</b>																			
Building 331 - Contaminated equipment outside the facility is awaiting transport and storage by Radioactive and Hazardous Waste Management.																			
331	Outside	None	Storage of contaminated parts	Tritium	NA	1	NA	NA	NA	None	1	4.8E+00	957	ENE	1.2E-03 1.0E-03 <sup>f</sup>	441	SSW	5.0E-03 4.5E-03 <sup>f</sup>	6
The Building 612 Yard is operated by the Radioactive and Hazardous Waste Management Division. The Yard consists of several areas where containers having radioactive wastes are stacked outdoors. The containers, which are not air tight, can outgas tritium.																			
612	Yard	Area Source	Storage of low level waste	Tritium	NA	NA	NA	NA	NA	None	1	1.5E+00	444	NE	2.0E-03 1.8E-03 <sup>f</sup>	212	SSW	6.1E-03 5.4E-03 <sup>f</sup>	6
Building 695/696 is the Decontamination Waste Treatment Facility operated by Radiological and Hazardous Waste Management Division.																			
695	Yard	Transportainer	Temporary storage of waste	Tritium	NA	1	NA	NA	NA	None	1	2.1E-01	876	S	3.7E-05 3.3E-05 <sup>f</sup>	101	ENE	2.9E-03 2.5E-03 <sup>f</sup>	6
The Southeast Quadrant of the Livermore Site has slightly elevated levels of Pu-239 in the surface soil and air. The source of the Pu-239 was past waste management operations.																			
Southeast Quadrant	Area Source		Resuspension	Pu-239	NA	NA	NA	NA	NA	None	1	NA	NA	NA	6.1E-04	NA	NA	NA	6
<b>SITE 300 DIFFUSE SOURCES</b>																			
Diffuse sources consist of resuspension of depleted uranium from historical explosive tests.																			
Site 300	All	Area Source	Soil resuspension	U-238 U-235 U-234	NA NA NA	NA NA NA	NA	NA	NA	None	1	NA NA NA	NA	NA	9.4E-03	NA	NA	NA	6
<b>EMISSION SOURCES THAT ACCOUNT FOR NEARLY 100% OF THE POTENTIAL EFFECTIVE DOSE EQUIVALENT AT EACH SITE.</b>																			
<b>LIVERMORE SITE SOURCES</b>																			
331	All <sup>f</sup>	Stack 1	tritium research and development	H-3	<sup>d</sup>	1.0E+00	30.0	1.22	5.4	None	1	5.2E-01	957	ENE	2.6E-03	573	SW	2.9E-03	3
		Stack 2	Decontamination of parts	H-3	<sup>d</sup>	1.0E+00	30.0	1.22	7.4	None	1	3.1E+01			2.2E-03 <sup>g</sup>			2.4E-03 <sup>g</sup>	3
612	Yard	Area Source	Storage of low level waste	Tritium	NA	NA	NA	NA	NA	None	1	1.5E+00	444	NE	2.0E-03 1.8E-03 <sup>f</sup>	212	SSW	6.1E-03 5.4E-03 <sup>f</sup>	6
331	Outside	None	Storage of contaminated parts	Tritium	NA	1	NA	NA	NA	None	1	4.8E+00	957	ENE	1.2E-03 1.0E-03 <sup>f</sup>	441	SSW	5.0E-03 4.5E-03 <sup>f</sup>	6
Southeast Quadrant	Area Source		Resuspension	Pu-239	NA	NA	NA	NA	NA	None	1	NA	NA	NA	6.1E-04	NA	NA	NA	6
<b>SITE 300 SOURCES</b>																			
Site 300	All	Area Source	Soil resuspension	U-238 U-235 U-234	NA NA NA	NA NA NA	NA	NA	NA	None	1	NA NA NA	NA	NA	9.4E-03	NA	NA	NA	6
851	Firing Table	None	Explosive tests	U-238 U-235 U-234	8.9E-03 1.1E-04 8.3E-04	1 1 1	NA	NA	NA	None	1	8.9E-03 1.1E-04 8.3E-04	3170	SSE	8.8E-03	1396	WSW	8.9E-03	4
NOTE: To convert curies to becquerels use 1 Ci=3.7E+10 Bq and to convert millirem to sieverts use 1 Sv=1.0E+05 mrem.																			
<sup>a</sup> Gross alpha and Gross beta emissions are continuously monitored at the stack.																			
<sup>b</sup> Because monitoring takes place after HEPA filtration, an unabated EDE cannot be determined (see discussion in Section II, subsection "Stack Monitoring for Gross Alpha and Gross Beta Radiation.")																			
<sup>c</sup> Stack emissions have been combined as permitted by the EPA/DOE Memorandum of Understanding.																			
<sup>d</sup> Tritium HT and HTO emissions from the stack are continuously monitored.																			
<sup>e</sup> The dose from HT and HTO emissions calculated using the NEWTRIT model; see discussion in Section VII, "Modeling Dose"																			
<sup>f</sup> The air monitoring data for all emission points show no detectable released alpha activity, i.e., the measurements are at or below the limit of sensitivity of the analytical method.																			
<sup>g</sup> Air emissions are continuously sampled at the post-HEPA-filter atmospheric discharge points, although potential emissions are low enough that stack monitoring is not required per the NESHAPs 40 CFR 61.101.																			
<sup>h</sup> The unabated EDE shown is only for the tritium source term.																			
Except for high-bay exhaust that is not HEPA-filtered, monitoring takes place after HEPA filtration, and an unabated EDE cannot be determined (see discussion in Section II, "Stack Monitoring for Gross Alpha and Gross Beta Radiation.")																			
The dose from HTO emissions calculated using the NEWTRIT model; see discussion in Section VII, "Modeling Dose from Tritium"																			

## ATTACHMENT 2. ERRATA for the NESHAPs 2004 Annual Report

In the *LLNL NESHAPs 2004 Annual Report* (UCRL-TR-113867-05, dated June 2005), two tables require correction, as follows:

- The 80 radionuclides listed in **Table 1** on page 5 include one that is misidentified. The entry in row 2, column 5 that reads “Palladium-231” should be removed. This elimination changes the number of radionuclides listed in **Table 1** from 80 to 79.
- **Table 11** on page 22 contains several errors and should be replaced by the following corrected version:

**Table 11.** Disaggregations of collective dose for Site 300, 2004.

Individual dose mrem/y	Collective dose person-rem/y	Percent total collective dose
0.001 to 0.01	0.0753	1.96%
0.0001 to 0.001	1.39	36.2%
0.00001 to 0.0001	2.38	61.8%
0.000001 to 0.00001	0	0%
Total	3.85	100%