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Lawrence Livermore National Laboratory
NESHAPs 2006 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 U.S. 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 μSv) 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 2006 are summarized here.

- Livermore site: 0.0045 mrem (0.045 μSv) (36% from point source emissions, 64% from diffuse source emissions). The point source emissions include gaseous tritium modeled as tritiated water vapor as directed by the U.S. Environmental Protection Agency (EPA) Region IX; the resulting dose is used for compliance purposes.
- Site 300: 0.016 mrem (0.16 μSv) (87.5% from point source emissions, 12.5% 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 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 km² 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 topographic and structural depression oriented east-west within the Diablo Range. The valley is approximately 22.6 km long and generally varies in width between 4 and 11.3 km. The valley floor is at its highest elevation of 220 m above sea level along the eastern margin and gradually dips to 92 m at the southwest corner.

The climate of the Livermore Valley is characterized by mild, rainy winters and warm-to-hot, dry summers. The mean daily maximum, minimum, and average temperatures for the Livermore site in 2006 were 22.1 °C, 7.5 °C, and 14.8 °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 2006 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 winds blew from the south-southwest through west-southwest about 45% of the time and more frequently during the summer. During the winter, winds from the northeast were more common. The average wind speed in 2006 at the Livermore site was 2.3 m/s (5.1 mph). Eighty percent of the precipitation occurs as rain between November and March with very little rainfall during the summer months. In 2006, the Livermore site received 38.7 cm of rain.

Site 300
Site 300, LLNL’s Experimental Test Facility, is located 24 km east of the Livermore site in the Altamont Hills of the Diablo Range and occupies an area of 30.3 km². 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 packages and stores fireworks at that location. 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 above sea level in the northwestern portion of the site to 150 m above sea level at the southeast corner. The climate at Site 300 is similar to that of the
Livermore site, with mild winters and warm-to-hot 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 2006 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 42% of the time during 2006. As is the case at the Livermore site, Site 300 precipitation is highly seasonal, with eighty percent of precipitation occurring between November and March. Site 300 received 32.2 cm of rain during 2006 and had mean daily maximum, minimum, and average temperatures of 21.2 °C, 12.5 °C, and 16.9 °C, respectively. The average wind speed at the site was 5.4 m/s (12.1 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 2006 at the Livermore site was 2.3 m/s (5.1 mph); at Site 300 it was 5.4 m/s (12.1 mph).

**Figure 2.** Wind roses, showing wind speed, direction, and frequency of occurrence at the Livermore site and Site 300 during 2006.
SECTION II. Air Emission Sources and Data

Sources
Approximately a hundred different radioisotopes were available for use at LLNL in 2006 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 2006.

<table>
<thead>
<tr>
<th>Hydrogen-3</th>
<th>Selenium-75</th>
<th>Iodine-129</th>
<th>Europium-155</th>
<th>Actinium-227</th>
<th>Plutonium-239</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beryllium-7</td>
<td>Krypton-85</td>
<td>Tellurium-129m</td>
<td>Europium-156</td>
<td>Actinium-228</td>
<td>Plutonium-240</td>
</tr>
<tr>
<td>Beryllium-10</td>
<td>Strontium-85</td>
<td>Iodine-131</td>
<td>Holmium-166m</td>
<td>Radium-228</td>
<td>Americium-241</td>
</tr>
<tr>
<td>Carbon-14</td>
<td>Yttrium-88</td>
<td>Barium-133</td>
<td>Rhenium-187</td>
<td>Thorium-228</td>
<td>Plutonium-241</td>
</tr>
<tr>
<td>Sodium-22</td>
<td>Strontium-90</td>
<td>Cesium-134</td>
<td>Iridium-192</td>
<td>Thorium-229</td>
<td>Americium-242m</td>
</tr>
<tr>
<td>Phosphorus-32</td>
<td>Niobium-95</td>
<td>Cesium-137</td>
<td>Gold-195</td>
<td>Protactinium-231</td>
<td>Americium-243</td>
</tr>
<tr>
<td>Phosphorus-33</td>
<td>Zirconium-95</td>
<td>Cerium-139</td>
<td>Mercury-203</td>
<td>Thorium-232</td>
<td>Curium-243</td>
</tr>
<tr>
<td>Sulfur-35</td>
<td>Technetium-99</td>
<td>Barium-140</td>
<td>Thallium-204</td>
<td>Uranium-232</td>
<td>Curium-244</td>
</tr>
<tr>
<td>Chlorine-36</td>
<td>Technetium-99m</td>
<td>Lanthanum-140</td>
<td>Bismuth-205</td>
<td>Uranium-233</td>
<td>Plutonium-244</td>
</tr>
<tr>
<td>Potassium-40</td>
<td>Rhodium-102</td>
<td>Cerium-141</td>
<td>Bismuth-207</td>
<td>Thorium-234</td>
<td>Curium-245</td>
</tr>
<tr>
<td>Manganese-54</td>
<td>Ruthenium-103</td>
<td>Cerium-144</td>
<td>Polonium-208</td>
<td>Uranium-234</td>
<td>Curium-248</td>
</tr>
<tr>
<td>Cobalt-57</td>
<td>Silver-108m</td>
<td>Neodymium-147</td>
<td>Polonium-210</td>
<td>Uranium-236</td>
<td>Californium-250</td>
</tr>
<tr>
<td>Cobalt-58</td>
<td>Cadmium-109</td>
<td>Promethium-147</td>
<td>Lead-210</td>
<td>Neptunium-237</td>
<td>Californium-252</td>
</tr>
<tr>
<td>Nickel-59</td>
<td>Tin-113</td>
<td>Gadolinium-148</td>
<td>Bismuth-212</td>
<td>Plutonium-238</td>
<td></td>
</tr>
<tr>
<td>Cobalt-60</td>
<td>Antimony-125</td>
<td>Europium-152</td>
<td>Bismuth-214</td>
<td>Uranium-238</td>
<td></td>
</tr>
<tr>
<td>Nickel-63</td>
<td>Iodine-125</td>
<td>Europium-154</td>
<td>Radium-226</td>
<td>Neptunium-239</td>
<td></td>
</tr>
</tbody>
</table>

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 2006
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 2006, 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.

Table 2. Air effluent sampling systems and locations.

<table>
<thead>
<tr>
<th>Building</th>
<th>Facility</th>
<th>Analytes</th>
<th>Sample type</th>
<th>Number of samplers</th>
</tr>
</thead>
<tbody>
<tr>
<td>235</td>
<td>Chemistry, Materials, and Life Sciences</td>
<td>Gross $\alpha, \beta$ on particles</td>
<td>Filter</td>
<td>1</td>
</tr>
<tr>
<td>251</td>
<td>Heavy Elements Unhardened\textsuperscript a area</td>
<td>Gross $\alpha, \beta$ particles</td>
<td>Filters</td>
<td>24</td>
</tr>
<tr>
<td></td>
<td>Hardened\textsuperscript a area</td>
<td>Gross $\alpha, \beta$ on particles</td>
<td>Filters</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Gross $\alpha, \beta$ on particles</td>
<td>CAM\textsuperscript b</td>
<td>2</td>
</tr>
<tr>
<td>331</td>
<td>Tritium</td>
<td>Tritium</td>
<td>Ionization Chamber\textsuperscript b</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Gaseous tritium/tritiated water vapor</td>
<td>Molecular Sieves</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Gaseous tritium/tritiated water vapor</td>
<td>Glycol Bubblers</td>
<td>2</td>
</tr>
<tr>
<td>332</td>
<td>Plutonium</td>
<td>Gross $\alpha, \beta$ on particles</td>
<td>Filters</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Gross $\alpha, \beta$ on particles</td>
<td>CAM\textsuperscript b</td>
<td>12</td>
</tr>
<tr>
<td>491</td>
<td>Isotope Separation\textsuperscript c</td>
<td>Gross $\alpha, \beta$ on particles</td>
<td>Filter</td>
<td>1</td>
</tr>
<tr>
<td>695/696</td>
<td>Decontamination and Waste Treatment</td>
<td>Gross $\alpha, \beta$ on particles</td>
<td>Filter</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Gaseous tritium/tritiated water vapor</td>
<td>Glycol Bubblers</td>
<td>1</td>
</tr>
<tr>
<td>801A</td>
<td>Contained Firing</td>
<td>Gross $\alpha, \beta$ on particles</td>
<td>Filter</td>
<td>1</td>
</tr>
</tbody>
</table>

Note: “CAM” denotes Eberline continuous air monitors.
\textsuperscript a Hardening refers to seismic reinforcement.
\textsuperscript b Alarmed systems, not used for NESHAPs compliance demonstration.
\textsuperscript c Isotope separation operations are discontinued; area now used for storage of contaminated parts.
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 all cases, continuous passive filter aerosol collection systems are used. At some facilities, alpha continuous air monitors (CAMs) are also deployed for sampling. CAMs have an alarm capability for the facility in the event of an unplanned release of alpha activity. CAMs are used for facility personnel safety; they are not used for NESHAPs compliance demonstration.

Detection of gross alpha and gross 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 short-lived 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.

In 2006, each stack of the Tritium Facility (Building 331) was monitored for tritium release by the use of ion chambers, molecular sieve samplers, and glycol bubblers. The release of tritium is either in the form of tritiated water vapor (HTO) or tritiated hydrogen gas (HT). All of the stack samplers monitor continuously. The alarmed ion chamber monitors provide real-time tritium concentration release levels (HT, HTO, or other gaseous forms). Similar to the CAMs for particulate systems, the ion chambers are used for facility personnel safety; they are not used for NESHAPs compliance demonstration.

The molecular sieve sampling at the Tritium Facility was discontinued in September of 2006 and replaced with glycol bubblers after a nine-week comparative study showed the bubblers out-performed the sieves in tritium capture. Each bubbler (not alarmed) is in parallel with an alarmed monitor. The bubblers use 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, which is then collected in the final two impingers (also 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.

In addition to particulate monitoring for gross alpha and gross beta, the Building 695/696 Decontamination and Waste Treatment Facility (DWTF) stack effluent was continuously monitored in 2006 for tritium with the use of a glycol bubbler. However, the glycol bubbler became disabled in November 2006 and was returned to service in June 2007. Sampling for tritium at the DWTF was not required in 2006, but occurred as a best management practice.
Environmental Protection Department (EPD) environmental analysts review data from air particulate sampling filters, ion chambers, molecular sieves, and glycol bubblers.

**Results of Stack Monitoring for Tritium**

Operations in the Tritium Facility (Building 331) in 2006 released a total of 18 Ci (0.67 TBq) of tritium. Of this, approximately 11 Ci (0.41 TBq) were released as HTO. The remaining tritium released, 7.1 Ci (0.26 TBq), was HT. The highest stack emission occurred over a two-week sample interval and was 4.2 Ci (0.16 TBq), of which approximately 85% was HTO.

This 2006 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 HT and HTO emissions from the Tritium Facility since 1981.

**Table 3.** Combined HT and HTO emissions from the Tritium Facility, 1981–2006.

<table>
<thead>
<tr>
<th>Year</th>
<th>Tritium emissions a (Ci)</th>
<th>Year</th>
<th>Tritium emissions a (Ci)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2006</td>
<td>18</td>
<td>1993</td>
<td>237</td>
</tr>
<tr>
<td>2005</td>
<td>32</td>
<td>1992</td>
<td>177</td>
</tr>
<tr>
<td>2004</td>
<td>17</td>
<td>1991</td>
<td>964 (148)</td>
</tr>
<tr>
<td>2003</td>
<td>110</td>
<td>1990</td>
<td>1281</td>
</tr>
<tr>
<td>2002</td>
<td>36</td>
<td>1989</td>
<td>2620 (329)</td>
</tr>
<tr>
<td>2001</td>
<td>20</td>
<td>1988</td>
<td>3978</td>
</tr>
<tr>
<td>2000</td>
<td>40</td>
<td>1987</td>
<td>2634</td>
</tr>
<tr>
<td>1999</td>
<td>280</td>
<td>1986</td>
<td>1128</td>
</tr>
<tr>
<td>1998</td>
<td>109</td>
<td>1985</td>
<td>989 (1000)</td>
</tr>
<tr>
<td>1997</td>
<td>299</td>
<td>1984</td>
<td>2200 (5000)</td>
</tr>
<tr>
<td>1996</td>
<td>215</td>
<td>1983</td>
<td>3024</td>
</tr>
<tr>
<td>1995</td>
<td>92</td>
<td>1982</td>
<td>1914</td>
</tr>
<tr>
<td>1994</td>
<td>137</td>
<td>1981</td>
<td>2552</td>
</tr>
</tbody>
</table>

a Chronic releases from normal operations are distinguished from acute accidental releases by showing the latter in parentheses. Accidental releases were predominately HT gas. Total emissions for the year are the sum of both chronic and accidental releases.

Continuously monitored tritium releases from the stack of DWTF (Building 695/696) were measured in 2006. From January through November of 2006, a total of 2.8 mCi (1.0 x 10^-4 TBq) of tritium was released as HT, and there were no measured HTO emissions from the DWTF. The measured tritium emissions from the DWTF, which were approximately one thousand times lower than the previous year, were more than one hundred times below the level of regulatory requirement for monitoring. The monitoring is currently in place as part of a best management practice, a practice which includes anticipating an increase in the quantity of tritium waste treated by the DWTF as a result of waste generated by the National Ignition Facility Project in future activities.
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; comparability of stack sampling results to results from upwind samplers; 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. As based on the criteria discussed above, if appropriate, 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.

None of the facilities monitored for gross alpha and gross beta had emissions in 2006.

Ambient Air Surveillance Monitoring for Radioactive Particles and Gases  
Surveillance monitoring of ambient air for tritium and radioactive particles has been in place since the early 1970s. In 2006, LLNL maintained seven continuously operating, high volume, air particulate samplers on the Livermore site, eight at Site 300, one in Tracy, and, with the addition of a new sampler in 2006 strategically positioned to monitor future National Ignition Facility Project activities, a total of ten in the Livermore Valley. LLNL also maintained eleven continuously operating tritiated water vapor samplers on the Livermore site, six in the Livermore Valley and one at Site 300; a seventh Livermore Valley sampler was added in October at the same location as the new particulate air sampler. 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 tritium surveillance monitors provide a valuable test of predictions based on air dispersion modeling, and all surveillance monitors can help characterize unplanned releases of radioactive material.
Summary ambient air data are provided in Table 4 and in Section VII, “Comparison of 2006 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.

Table 4. Mean concentrations of radionuclides of concern at the location of the SW-MEI in 2006 compared to EPA’s concentration standard.

<table>
<thead>
<tr>
<th>Location</th>
<th>Nuclide</th>
<th>EPA’s Table 2 concentration standard</th>
<th>Mean measured concentration</th>
<th>Measured concentration as a fraction of the standard</th>
<th>Detection limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Livermore site SW-MEI</td>
<td>Tritium</td>
<td>1.5 x 10^{-9} Ci/m^3</td>
<td>7.7 x 10^{-13} Ci/m^3*</td>
<td>5.1 x 10^{-4}</td>
<td>1 x 10^{-12} Ci/m^3</td>
</tr>
<tr>
<td>Livermore site SW-MEI</td>
<td>Plutonium-239</td>
<td>2.0 x 10^{-15} Ci/m^3</td>
<td>1.8 x 10^{-19} Ci/m^3**</td>
<td>9.0 x 10^{-5}</td>
<td>5 x 10^{-19} Ci/m^3</td>
</tr>
<tr>
<td>Site 300 SW-MEI</td>
<td>Uranium-238</td>
<td>8.3 x 10^{-15} Ci/m^3</td>
<td>1.3 x 10^{-17} Ci/m^3***</td>
<td>1.5 x 10^{-3}</td>
<td>3 x 10^{-20} Ci/m^3</td>
</tr>
</tbody>
</table>

* The measured tritium value includes contributions from all minor sources (including the Building 612 Yard and the Building 331 Outside Yard), Tritium Facility, and DWTF; it is not possible to differentiate the contributions of the Tritium Facility and DWTF from those of the minor sources.

** Note that the mean measured concentration for plutonium is less than the detection limit; only 1 of the 24 values comprising the mean was a measured detection. 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 2006 is 0.0065, which is less than 0.00725, the ratio of these isotopes for naturally occurring uranium. This results in approximately 86% of the resuspension being attributable to natural occurring uranium and 14% to depleted uranium.

Compliance Demonstration for Minor Radiological Sources

With the EPA’s Region IX approval, LLNL demonstrates compliance for minor emissions sources (both non-monitored stack and area sources) through the use of 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 comparisons are 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 2006 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. 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’s measured concentrations in air for tritium, plutonium-239+240, and uranium-238 are a fraction 0.002 or less of the standard for these radionuclides.

The LLNL radiological facilities included in the “minor sources” classification in 2006 are listed in Table 5. In addition, out-gassing tritiated wastes that are stored in transportainers at various locations on-site are also categorized as minor sources.

Table 5. Buildings with minor radiological emissions (by directorate) for 2006.\(^a\)

<table>
<thead>
<tr>
<th>CMLS</th>
<th>P&amp;AT</th>
<th>SEP</th>
<th>E&amp;E</th>
<th>Eng.</th>
<th>DNT</th>
<th>NIF</th>
<th>Institution</th>
</tr>
</thead>
<tbody>
<tr>
<td>B132</td>
<td>B194</td>
<td>B253</td>
<td>B281</td>
<td>B131</td>
<td>B801</td>
<td>B298</td>
<td>B212</td>
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<tr>
<td>B151</td>
<td>B282</td>
<td>B254</td>
<td>B292</td>
<td>B231</td>
<td>B804</td>
<td>(vacant)</td>
<td></td>
</tr>
<tr>
<td>B235</td>
<td>B341</td>
<td>B255</td>
<td>B378</td>
<td>B321</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>B241</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>B321A</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B361</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>B321B</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B362</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>B321C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B363</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>B322</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B364</td>
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<td></td>
<td></td>
<td></td>
<td>B327</td>
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<tr>
<td>B365</td>
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<tr>
<td>B366</td>
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<td></td>
</tr>
<tr>
<td>B810A</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>B810B</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^a\) Directorate abbreviations refer to Chemistry, Materials, and Life Sciences (CMLS), Physics and Advanced Technologies (P&AT), Safety and Environmental Protection (SEP), Energy and Environment (E&E), Engineering (Eng.), Defense and Nuclear Technologies (DNT), National Ignition Facility Programs (NIF), and Director’s Office (Institution).

Radionuclide Usage Inventories

Radionuclide usage inventories were utilized in 2006 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 2006.

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 μSv). 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).

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 μSv/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 x 10¹⁰ Bq), and stack parameters, including height, diameter, and emission velocity.

Meteorological Data
All model runs used actual 2006 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 second and are averaged into quarter-hour increments, time tagged, and computer recorded. Stability is estimated in real-time using the Solar Radiation/Delta method as suggested by the EPA. The wind speed and direction 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 $^{239}$Pu is used as the surrogate for gross alpha, $^{137}$Cs for gross gamma, and $^{90}$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. 2000. "LandScan: A Global Population Database for Estimating Populations at Risk," Photogrammetric Engineering & Remote Sensing Vol. 66, No. 7, July 2000, pp. 849-857. Available at [http://www.ornl.gov/sci/landscan](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**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beef cattle density (# cows/km$^2$)</td>
<td>1.9</td>
</tr>
<tr>
<td>Milk cattle density (# cows/km$^2$)</td>
<td>4.0</td>
</tr>
<tr>
<td>Land fraction cultivated for vegetable crops</td>
<td>0.046</td>
</tr>
</tbody>
</table>

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 stack sources, the sampling data (curies released per unit time) for each radionuclide were used directly. For minor sources such as unmonitored facilities or activities, 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 other minor sources, such as diffuse area 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.

<table>
<thead>
<tr>
<th>Material</th>
<th>Solid physical state factor</th>
<th>Liquid physical state factor</th>
<th>Gas physical state factor</th>
<th>Year approved</th>
</tr>
</thead>
<tbody>
<tr>
<td>Elemental uranium</td>
<td>&lt;1100 °C</td>
<td>Between 1100 °C and 3000 °C</td>
<td>&gt;3000 °C</td>
<td>1996</td>
</tr>
<tr>
<td>Uranium/niobium alloy</td>
<td>&lt;1000 °C</td>
<td>Between 1000 °C and 3000 °C</td>
<td>&gt;3000 °C</td>
<td>2001</td>
</tr>
<tr>
<td>Uranium oxide</td>
<td>&lt;2000 °C</td>
<td>Between 2000 °C and 2500 °C</td>
<td>&gt;2500 °C</td>
<td>2004</td>
</tr>
<tr>
<td>Uranium nitride</td>
<td>&lt;2000 °C</td>
<td>Between 2000 °C and 2500 °C</td>
<td>&gt;2500 °C</td>
<td>2004</td>
</tr>
<tr>
<td>Uranium carbide</td>
<td>&lt;2000 °C</td>
<td>Between 2000 °C and 2500 °C</td>
<td>&gt;2500 °C</td>
<td>2004</td>
</tr>
<tr>
<td>Elemental plutonium</td>
<td>&lt;600 °C</td>
<td>Between 600 °C and 3000 °C</td>
<td>&gt;3000 °C</td>
<td>2001</td>
</tr>
</tbody>
</table>

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 μSv] 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 (SW-MEI) cannot receive an EDE greater than 10 mrem/y (100 μSv/y). The 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 2006 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 2006 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.

Figure 3. Location of Site-Wide Maximally Exposed Individual (SW-MEI) at the Livermore site, 2006.
**Figure 4.** Location of Site-Wide Maximally Exposed Individual (SW-MEI) at Site 300, 2006.

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

**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 μSv/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. 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.
SECTION IV. Results of 2006 Radiological Dose Assessment

This section summarizes the doses to the most exposed public individuals from LLNL operations in 2006, shows the comparison to previous years, and summarizes LLNL’s compliance with 40 CFR 61, Subpart H (61.93). Also included in this section are potential doses to the populations residing within 80 km of either the Livermore site or Site 300 and unplanned releases, as requested in supplementary guidance for NESHAPs reporting issued in 1992 by DOE Headquarters, Office of Environment, Safety and Health.

Total Dose to Site-Wide Maximally Exposed Individuals

The total dose to the Livermore site SW-MEI from operations in 2006 was 0.0045 mrem (0.045 μSv). Of this, 0.0016 mrem (0.016 μSv), or 36%, was contributed by point sources, while diffuse emissions accounted for 0.0029 mrem (0.029 μSv), or 64%, 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 2006, and so this dose is comparable to the historically low value also reported for 2005. The most significant factor leading to this low dose was a continued low level of operations and emissions from the Tritium Facility (see Table 3 in Section II).

The total dose to the Site 300 SW-MEI from operations in 2006 was 0.016 mrem (0.16 μSv). Point source emissions from firing table explosives experiments accounted for about 87.5% of this total, while 12.5% was contributed by diffuse sources. Although neither the point source nor diffuse source contributions are individually the lowest values ever reported for these source types, together they result in the lowest potential dose ever determined for the Site 300 SW-MEI.

Table 8 shows the facilities or sources that collectively accounted for 99% or more of the doses to the SW-MEI for the Livermore site and Site 300 in 2006. 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 2006.

<table>
<thead>
<tr>
<th>Facility (Source Category)</th>
<th>CAP88-PC Dose in mrem/y</th>
<th>CAP88-PC Percentage Contribution to Total Dose</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Livermore site</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Building 331 stacks (point source)</td>
<td>0.0016</td>
<td>36%</td>
</tr>
<tr>
<td>Building 612 Yard (diffuse source)</td>
<td>0.0013</td>
<td>29%</td>
</tr>
<tr>
<td>Building 331 outside (diffuse source)</td>
<td>0.0011</td>
<td>25%</td>
</tr>
<tr>
<td>Southeast Quadrant (diffuse source)</td>
<td>0.00061</td>
<td>10%</td>
</tr>
<tr>
<td><strong>Site 300</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Building 851 Firing Table (point source)</td>
<td>0.014</td>
<td>87.5%</td>
</tr>
<tr>
<td>Soil resuspension (diffuse source)</td>
<td>0.0020</td>
<td>12.5%</td>
</tr>
</tbody>
</table>

Table 9 compares 2006 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 2006.

<table>
<thead>
<tr>
<th>Year</th>
<th>Total Dose</th>
<th>Point Source Dose</th>
<th>Diffuse Source Dose</th>
</tr>
</thead>
<tbody>
<tr>
<td>Livermore site</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2006</td>
<td>0.0045</td>
<td>0.0016</td>
<td>0.0029</td>
</tr>
<tr>
<td>2005</td>
<td>0.0065</td>
<td>0.0027</td>
<td>0.0038</td>
</tr>
<tr>
<td>2004</td>
<td>0.0079</td>
<td>0.0021</td>
<td>0.0058</td>
</tr>
<tr>
<td>2003</td>
<td>0.044</td>
<td>0.024</td>
<td>0.020</td>
</tr>
<tr>
<td>2002</td>
<td>0.023</td>
<td>0.010</td>
<td>0.013</td>
</tr>
<tr>
<td>2001</td>
<td>0.017</td>
<td>0.0057</td>
<td>0.011</td>
</tr>
<tr>
<td>2000</td>
<td>0.038</td>
<td>0.017</td>
<td>0.021</td>
</tr>
<tr>
<td>1999</td>
<td>0.12</td>
<td>0.094</td>
<td>0.028</td>
</tr>
<tr>
<td>1998</td>
<td>0.055</td>
<td>0.031</td>
<td>0.024</td>
</tr>
<tr>
<td>1997</td>
<td>0.097</td>
<td>0.078</td>
<td>0.019</td>
</tr>
<tr>
<td>1996</td>
<td>0.093</td>
<td>0.048</td>
<td>0.045</td>
</tr>
<tr>
<td>1995</td>
<td>0.041</td>
<td>0.019</td>
<td>0.022</td>
</tr>
<tr>
<td>1994</td>
<td>0.065</td>
<td>0.042</td>
<td>0.023</td>
</tr>
<tr>
<td>1993</td>
<td>0.066</td>
<td>0.040</td>
<td>0.026</td>
</tr>
<tr>
<td>1992</td>
<td>0.079</td>
<td>0.069</td>
<td>0.010</td>
</tr>
<tr>
<td>1991</td>
<td>0.234</td>
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<tr>
<td>1990</td>
<td>0.240</td>
<td>—</td>
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</table>
Table 9. Continued

<table>
<thead>
<tr>
<th>Year</th>
<th>Total Dose</th>
<th>Point Source Dose</th>
<th>Diffuse Source Dose</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Site 300</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2006</td>
<td>0.016</td>
<td>0.014</td>
<td>0.0020</td>
</tr>
<tr>
<td>2005</td>
<td>0.018</td>
<td>0.0088</td>
<td>0.0094</td>
</tr>
<tr>
<td>2004</td>
<td>0.026</td>
<td>0.025</td>
<td>0.00086</td>
</tr>
<tr>
<td>2003</td>
<td>0.017</td>
<td>0.017</td>
<td>0.00034</td>
</tr>
<tr>
<td>2002</td>
<td>0.021</td>
<td>0.018</td>
<td>0.0033</td>
</tr>
<tr>
<td>2001</td>
<td>0.054</td>
<td>0.050</td>
<td>0.0037</td>
</tr>
<tr>
<td>2000</td>
<td>0.019</td>
<td>0.015</td>
<td>0.0037</td>
</tr>
<tr>
<td>1999</td>
<td>0.035</td>
<td>0.034</td>
<td>0.0012</td>
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<tr>
<td>1998</td>
<td>0.024</td>
<td>0.019</td>
<td>0.005</td>
</tr>
<tr>
<td>1997</td>
<td>0.020</td>
<td>0.011</td>
<td>0.0088</td>
</tr>
<tr>
<td>1996</td>
<td>0.033</td>
<td>0.033</td>
<td>0.00045</td>
</tr>
<tr>
<td>1995</td>
<td>0.023</td>
<td>0.020</td>
<td>0.003</td>
</tr>
<tr>
<td>1994</td>
<td>0.081</td>
<td>0.049</td>
<td>0.032</td>
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<tr>
<td>1993</td>
<td>0.037</td>
<td>0.011</td>
<td>0.026</td>
</tr>
<tr>
<td>1992</td>
<td>0.021</td>
<td>0.021</td>
<td>— c</td>
</tr>
<tr>
<td>1991</td>
<td>0.044</td>
<td>0.044</td>
<td>— c</td>
</tr>
<tr>
<td>1990</td>
<td>0.057</td>
<td>0.057</td>
<td>— c</td>
</tr>
</tbody>
</table>

a 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.
b Point and diffuse source doses were not reported separately from the total dose for the Livermore site for 1990 and 1991.
c No diffuse emissions were evaluated at Site 300 for years before 1993.

Doses from Unplanned Releases
In June of 2006, a solid titanium tritide source was transferred from one Livermore site building to another for potential use as a check source. Subsequently, after routine radiation swipes identified tritium contamination in both buildings, it was determined that this legacy source had leaked tritiated particulate matter. During the transfer, the source was wrapped, but tritium contamination was inadvertently spread to the environment via personnel contact with the particulate matter. Contamination measured above the DOE’s release limit for tritium contamination was remediated. The bioassays performed for personnel handling the source or working in the rooms impacted by the incident indicated that there was either no tritium intake or else there was none attributable to the incident. Because the greatest potential dose would have been to these personnel, rather than a member of the public, any potential dose to a member of the public from this incident would have been completely negligible.

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 center of each site 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 2006 Livermore site operations was 0.8 person-rem (0.008 person-Sv); the corresponding collective EDE from Site 300 operations was 3.3 person-rem (0.033 person-Sv). For the Livermore site, this population dose is attributable to tritium, and for Site 300, the isotopes in depleted uranium ($^{238}\text{U}$, $^{235}\text{U}$, and $^{234}\text{U}$). The value for the Livermore site collective dose from tritium was lower than in 2005 primarily due to lower tritium releases from the Tritium Facility. 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 μSv/y) and the vast majority received a dose less than 0.0001 mrem/y (0.001 μSv/y).

Table 10. Disaggregations of collective dose for the Livermore site, 2006.

<table>
<thead>
<tr>
<th>Individual dose mrem/y</th>
<th>Collective dose person-rem/y</th>
<th>Percent total collective dose</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.001 to 0.01</td>
<td>0.003</td>
<td>0.4%</td>
</tr>
<tr>
<td>0.0001 to 0.001</td>
<td>0.047</td>
<td>6.3%</td>
</tr>
<tr>
<td>0.00001 to 0.0001</td>
<td>0.666</td>
<td>89.1%</td>
</tr>
<tr>
<td>0.000001 to 0.00001</td>
<td>0.032</td>
<td>4.3%</td>
</tr>
<tr>
<td>Total</td>
<td>0.75</td>
<td>100%</td>
</tr>
</tbody>
</table>

Collective doses can be broken down similarly for the shots from the Building 851 Firing Table, as shown in Table 11. In this case, individuals that make up more than 91% of the population receive less than 0.001 mrem/y (0.01 μSv/y).

Table 11. Disaggregations of collective dose for Site 300, 2006.

<table>
<thead>
<tr>
<th>Individual dose mrem/y</th>
<th>Collective dose person-rem/y</th>
<th>Percent total collective dose</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.001 to 0.01</td>
<td>0.29</td>
<td>8.8%</td>
</tr>
<tr>
<td>0.0001 to 0.001</td>
<td>2.2</td>
<td>67%</td>
</tr>
<tr>
<td>0.00001 to 0.0001</td>
<td>0.76</td>
<td>23%</td>
</tr>
<tr>
<td>0.000001 to 0.00001</td>
<td>0.051</td>
<td>1.5%</td>
</tr>
<tr>
<td>Total</td>
<td>3.3</td>
<td>100%</td>
</tr>
</tbody>
</table>
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 µSv) NESHAPs dose standard for dose to the maximally exposed individual members of the public. Tritium accounted for 90% 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 2006, 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.

Name: William A. Bookless
Associate Director
Safety and Environmental Protection
Lawrence Livermore National Laboratory
7000 East Avenue, L-668
Livermore, CA 94550

Signature: [Signature]
William A. Bookless
Date: 6/24/07

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
Technical Deputy
Safety and Environmental Programs
U.S. Department of Energy
7000 East Avenue, L-293
Livermore, CA 94550

Signature: [Signature]
Phillip Hill
Date: 6/27/07
SECTION VI. Supplemental Information on NESHAPs Compliance and Quality Assurance/Quality Control 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-one 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 Program
The LLNL NESHAPs quality assurance (QA) 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.

LLNL’s QA project plan for NESHAPs 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 QA activities are conducted by two LLNL departments, EPD and HCD, the documentation for the elements of a complete quality assurance project plan is independently maintained by these organizations. The LLNL NESHAPs QAPP presents a cross-walk between the requirements of a complete quality assurance project plan, 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.

Based on the key elements addressed by the LLNL QA program as presented in LLNL’s NESHAPs QAPP, LLNL has met the requirements prescribed by 40 CFR 61, App. B. Method 114 to: 1) identify organizational structure, functional responsibilities, levels of authority, and lines of communication; 2) establish administrative controls; 3) describe sample collection and analyses procedures; 4) document objectives of the QA program; 5) establish a quality control program; 6) establish a sample tracking system; 7) perform maintenance, calibration, and field checks; 8) perform audits; 9) establish a corrective action program; 10) prepare periodic reports; and 11) document the QA program.

**Evaluation of New Radiological Projects**

The TAMM Group is informed by several mechanisms of proposed new operations and modified operations where significant changes in radiological usage inventories occur. 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 TAMMM Group for at least the period of time specified in 40 CFR 61, Subpart H.

**Quality Control for 2006 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 2006 or unmonitored point source releases that contributed significantly in 2006 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 seven explosives experiments conducted in 2006 at Firing Table 851 at Site 300. Both the input data and
model runs for all seven explosives experiments were independently checked and validated.

Model runs were performed for about one dozen sources in the 2006 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 quality control efforts, the data, results, and conclusions presented in this report meet applicable quality assurance objectives.
SECTION VII. Supplementary Information on Radiological Dose Assessment for 2006

Livermore Site Principal Diffuse Sources
The dose evaluations for diffuse sources at the Livermore site in 2006 required several different modeling approaches. The Building 331 Outside Yard and the Building 612 Yard 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. A back-calculation was also done for the outside transportainer storage and the portable tank berm at the DWTF, where small quantities of tritium are handled. This calculation indicated that 0.11 Ci of tritium was emitted. Since the dose contribution from this quantity of tritium was too small to impact the total dose at the SW-MEI, a discussion of this source is not included.

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 2006, outgassing from such waste released an estimated 3.6 Ci (1.4 x 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 leading to a calculated 2006 dose to the SW-MEI of 1.1 x 10\(^{-3}\) mrem (1.1 x 10\(^{-2}\) 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 2006 in this area was 23.8 pCi/m\(^3\) (0.88 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 0.71 Ci/y (2.6 x 10\(^{10}\) Bq/y) was required to produce the concentrations measured at the air sampler. This source term produced a
CAP88-PC calculated 2006 dose to the SW-MEI from the Building 612 Yard of 1.3 x 10^{-3} mrem (1.3 x 10^{-2} \mu 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.)

**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 2006 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 $1.8 \times 10^{-19}$ Ci/m$^3$ ($6.6 \times 10^{-9}$ Bq/m$^3$). Using the dose conversion factor of 3.08 $\times$ 10$^{5}$ mrem/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$^3$/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 4.6 $\times$ 10$^{-4}$ mrem (4.6 $\times$ 10$^{-3}$ \mu Sv) for 2006.

**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 (Li$^3$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 Li$^3$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 2006, 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$^3$).
**Resuspension of Depleted Uranium in Soil at Site 300**

Depleted uranium is currently used and 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.

The contribution to measured uranium activities arising from naturally occurring uranium (NU) can be distinguished from depleted uranium (DU) contributed by LLNL operations. (A derivation of the arithmetic calculation used for this purpose 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 2006, 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 \( 2.0 \times 10^{-3} \) mrem (\( 2.0 \times 10^{-2} \) Sv) for the SW-MEI dose resulting from resuspension of DU in soil for 2006. (For more information on the sampling data, see the “Air Monitoring Programs” chapter in LLNL’s Site Annual Environmental Report for 2006, available at [http://www.llnl.gov/saer](http://www.llnl.gov/saer).)

**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 ingestion of 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;
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).

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.


Tritium doses from 2006 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 2006, calculated using NEWTRIT instead of CAP88-PC, the result was 0.0030 mrem (0.030 μSv), about 25% lower than the CAP88-PC value of 0.0040 mrem (0.040 μSv).

Comparison of 2006 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 the eleven tritiated water vapor samplers on the Livermore site (designated B331, B624, CAFE, COW, CRED, DWTF, MESQ, MET, POOL, SALV, and VIS) and one location off-site and downwind (ZON7). Figure 5 shows the locations of the tritium air surveillance monitors on the Livermore site. 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 darkened building icons).

Because the air tritium monitors only absorb HTO, only releases of HTO from stack and area sources were modeled. The release rate of HTO from the two 30-m-high, continuously monitored stacks at the Tritium Facility (Building 331) was determined from stack monitoring data to be 10.9 Ci (4.0 x 10^{11} Bq) in 2006. Stack monitoring of the DWTF determined a release of only HT (2.8 mCi), so no release from the DWTF was
included in the model test. Emissions from the Building 612 Yard source were estimated to be 0.71 Ci (2.6 x 10^{10} Bq) based on back-calculating a source term from observed tritium concentrations at the tritium monitor B624. The release rate for the B331 area source was determined by back-calculation to be 3.6 Ci (1.33 x 10^{11} Bq) in 2006. (Because the B331 and B624 release rates are calculated, rather than measured directly, these locations cannot be used to test the dispersion modeling.) While these two diffuse sources have historically contributed significantly to tritium concentrations at all monitoring locations, other potential sources of tritiated water vapor release (e.g., the area outside the DWTF) are too minor to influence the overall model-data comparison.

Annual mean concentrations of HTO in air (pCi/m^3) at the twelve air tritium samplers were modeled for each of the three principal sources, and the sum of the three contributions was compared to the measured annual mean concentrations. The results, displayed in Table 12, show that all air concentrations predicted by CAP88-PC were within a factor of three of the measured values, as is expected for a Gaussian dispersion model, and all predictions are consistent with other tests of CAP88-PC (Peterson, S-R. “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). Because of the high frequency of samples below the detection limit, only the results for DWTF, POOL, and COW are truly meaningful. Underestimation at DWTF, and to a lesser extent at COW, would be largely accounted for by including the localized contribution of the tritium released from the minor source outside the DWTF (estimated to 0.11 Ci (4.1 x 10^9 Bq).
Table 12. Comparison of measured and modeled annual mean concentrations of tritiated water vapor (HTO) in air at selected Livermore site locations and one off-site location, 2006.

<table>
<thead>
<tr>
<th>Air monitor (name)</th>
<th>Mean measured concentration (pCi/m³)</th>
<th>Modeled* average concentration (pCi/m³)</th>
<th>Ratio of modeled-to-measured concentrations</th>
<th>Modeled concentration of tritium in air contributed by the indicated source (pCi/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>B331 Stacks</td>
</tr>
<tr>
<td>B331</td>
<td>140</td>
<td>140</td>
<td>1.0**</td>
<td>0.021</td>
</tr>
<tr>
<td>B624</td>
<td>23.8</td>
<td>25</td>
<td>1.0**</td>
<td>0.33</td>
</tr>
<tr>
<td>DWTF</td>
<td>2.44</td>
<td>0.80</td>
<td>0.33</td>
<td>0.36</td>
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<tr>
<td>POOL</td>
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<td>5.6</td>
<td>2.3</td>
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<td>COW</td>
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<td>0.41</td>
<td>0.39</td>
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<tr>
<td>SALV</td>
<td>0.43</td>
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<td>-----</td>
<td>0.12</td>
</tr>
<tr>
<td>ZON7</td>
<td>0.32</td>
<td>0.29</td>
<td>-----</td>
<td>0.15</td>
</tr>
</tbody>
</table>

* This result takes into account the three significant tritium sources; it is the annual-mean concentration comprising the sum of the three contributions shown in the far right columns.
** This agreement was obtained by calibration.

Note: When half or more of the measurements were below the detection limit, predicted-to-observed (P/O) ratios are given in brackets. When no P/O ratio is shown, the mean measured concentration was below the detection limit of about 0.7 pCi/m³.
SECTION VIII. Supplemental Information on Other Compliance

Status of Compliance with Other Regulations

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 2006.

LLNL does not have or store any uranium mill tailings.
ATTACHMENT 1. LLNL NESHAPs 2006 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 effective dose equivalent (EDE) to the SW-MEI
- Distance and direction to the maximally exposed individual (MEI) for that specific source
- 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, the 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 are updated, as necessary, by experimenters and managers for those facilities. The Tamm Group annually measures the stack velocity and sampler flow and calibrates mass flow sensors for 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.

**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 2006 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 61, 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 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 μSv/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 μSv/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 2006, 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.
## Tritium Activities

### Operations

<table>
<thead>
<tr>
<th>Area</th>
<th>1150</th>
<th>331</th>
<th>1150</th>
<th>1003</th>
<th>1128</th>
<th>1128</th>
<th>1150</th>
<th>1128</th>
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<td>Gravimetric</td>
<td>General Chemistry</td>
<td>Gas Chromatography</td>
<td>Preparative purification</td>
<td>General Chemistry</td>
<td>Gas Chromatography</td>
<td>Preparative purification</td>
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<tr>
<td>Mass Spectrometer</td>
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<td>Gravimetric</td>
<td>General Chemistry</td>
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<td>General Chemistry</td>
<td>Gas Chromatography</td>
<td>Preparative purification</td>
<td></td>
</tr>
</tbody>
</table>

### Filters

- HEPA filters are used to filter air entering the facility. HEPA filters are used to filter air exiting the facility. HEPA filters are used to filter air entering the facility. HEPA filters are used to filter air exiting the facility.

### Area Activities

- **Gross alpha**:
  - 1150: 0.0E+00, 0.0E+00
  - 331: 4.3, 0.26, 7.6
  - 1003: 6.8, 0.32, 7.6
  - HEPA: 0.00001

- **Gross beta**:
  - 1150: 1.22, 0.0E+00, 0.0E+00
  - 331: 7.8, 0.30, 6.5
  - 1003: 7.8, 0.30, 6.5
  - HEPA: 0.00001

- **Triplet HEPA**:
  - 1150: 9.1, 0.17, 9.5
  - 331: 9.1, 0.17, 9.5
  - 1003: 9.1, 0.17, 9.5
  - HEPA: 0.00001

- **Double HEPA**:
  - 1150: 9.1, 0.17, 9.5
  - 331: 9.1, 0.17, 9.5
  - 1003: 9.1, 0.17, 9.5
  - HEPA: 0.00001

### Environmental Activities

- **Gloves Boxes**
  - 1150: 0.0E+00, 0.0E+00
  - 331: 4.3, 0.26, 7.6
  - 1003: 6.8, 0.32, 7.6

### Radiation Monitoring

- **Continuous Real-Time Monitors**
  - 1150: 0.0E+00, 0.0E+00
  - 331: 4.3, 0.26, 7.6
  - 1003: 6.8, 0.32, 7.6

### Area Status

- **Hardened Area**
  - 1150: 0.1 1.1 6.4
  - 331: 0.1 1.1 6.4
  - 1003: 0.1 1.1 6.4

- **Unhardened Area**
  - 1150: 0.1 1.1 6.4
  - 331: 0.1 1.1 6.4
  - 1003: 0.1 1.1 6.4

### Additional Information

- **Gross alpha**:
  - 1150: 0.0E+00, 0.0E+00
  - 331: 4.3, 0.26, 7.6
  - 1003: 6.8, 0.32, 7.6
  - HEPA: 0.00001

- **Gross beta**:
  - 1150: 1.22, 0.0E+00, 0.0E+00
  - 331: 7.8, 0.30, 6.5
  - 1003: 7.8, 0.30, 6.5
  - HEPA: 0.00001

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  - 1003: 9.1, 0.17, 9.5
  - HEPA: 0.00001

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  - 1150: 9.1, 0.17, 9.5
  - 331: 9.1, 0.17, 9.5
  - 1003: 9.1, 0.17, 9.5
  - HEPA: 0.00001
### SITE 100 Stack Sources

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<thead>
<tr>
<th>Site Name</th>
<th>Source Type</th>
<th>Source ID</th>
<th>Area</th>
<th>Source Type</th>
<th>Activity</th>
<th>Contaminant</th>
<th>Location</th>
<th>Emission Rate</th>
<th>Control</th>
<th>Containment</th>
<th>Fraction</th>
<th>Backward Dispersion</th>
<th>Total Emissions</th>
<th>Toxicity</th>
<th>Local Emission</th>
<th>Site-Wide Emission</th>
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<td>Tritium</td>
<td>NA</td>
<td>1.0E-02</td>
<td>NA</td>
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<td>1.0E-02</td>
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### SITE 100 Diffuse Sources

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<th>Source Type</th>
<th>Activity</th>
<th>Contaminant</th>
<th>Location</th>
<th>Emission Rate</th>
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<th>Containment</th>
<th>Fraction</th>
<th>Backward Dispersion</th>
<th>Total Emissions</th>
<th>Toxicity</th>
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</tbody>
</table>

### SITE 100 Stack Sources

- **Dose from Tritium:** The dose from HTO emissions calculated using the NEWTRIT model; see discussion in Section VII, "Modeling, and an unabated EDE cannot be determined from the monitoring data. There are no Tritium emissions from historical explosive tests.
- Air emissions are continuously monitored as part of the Radioactive and Hazardous Waste Management Division.
- The dose from Tritium is not required by the U.S. Environmental Protection Agency, but is monitored for historical explosive tests.
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- The dose from Tritium is calculated for historical explosive tests.
ATTACHMENT 2. ERRATA for the NESHAPs 2005 Annual Report

In the LLNL NESHAPs 2005 Annual Report (UCRL-TR-113867-06, dated June 2006), two sections correction, as follows:

- In the section titled “Maximally Exposed Public Individual” on page 16, the text states that “modeling calculations show that ground level concentrations of radionuclides can be expected to reach maximum values beyond the LLNL boundaries for...dispersals from open-air explosive experiments conducted at Site 300;” this statement is incorrect. Maximum values are reached closest to the source, well within site boundaries. This correction also applies to the text in the NESHAPs annual reports for calendar years 2003 and 2004.

- In the section titled “DWTF Transportainer Storage” on page 26, the dose contribution from this source to the SW-MEI should be 3.7 x 10^-4 μSv/y (rather than 1.7 x 10^-4 μSv/y) and to the MEI, it should be 2.9 x 10^-2 μSv/y (rather than 1.9 x 10^-2 μSv/y).