

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

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**LLNL NESHAPs  
2003 Annual Report**



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

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# LLNL NESHAPs 2003 Annual Report

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**U.S. Department of Energy  
Radionuclide Air Emission Annual Report  
(under Subpart H of 40 CFR Part 61)  
Calendar Year 2003**

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# Lawrence Livermore National Laboratory NESHAPs 2003 Annual Report

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

## SYNOPSIS

NESHAPs limits the emission of radionuclides to the ambient air from DOE facilities to levels resulting in an annual effective dose equivalent (EDE) of 10 mrem (100  $\mu$ Sv) to any member of the public. The EDEs for the Lawrence Livermore National Laboratory (LLNL) site-wide maximally exposed members of the public from operations in 2003 are summarized here.

- Livermore site: 0.044 mrem (0.44  $\mu$ Sv) (55% from point-source emissions, 45% from diffuse-source emissions). The point-source emissions include gaseous tritium modeled as tritiated water vapor as directed by EPA Region IX; the resulting dose is used for compliance purposes.
- Site 300: 0.017 mrem (0.17  $\mu$ Sv) (98% from point-source emissions, 2% 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 concentrations and dose coefficients. Site specific meteorological data, stack flow data, and emissions estimates based on radionuclide usage inventory data or continuous stack monitoring data were the specific inputs to CAP88-PC for each modeled source.

## SECTION I. Site Description

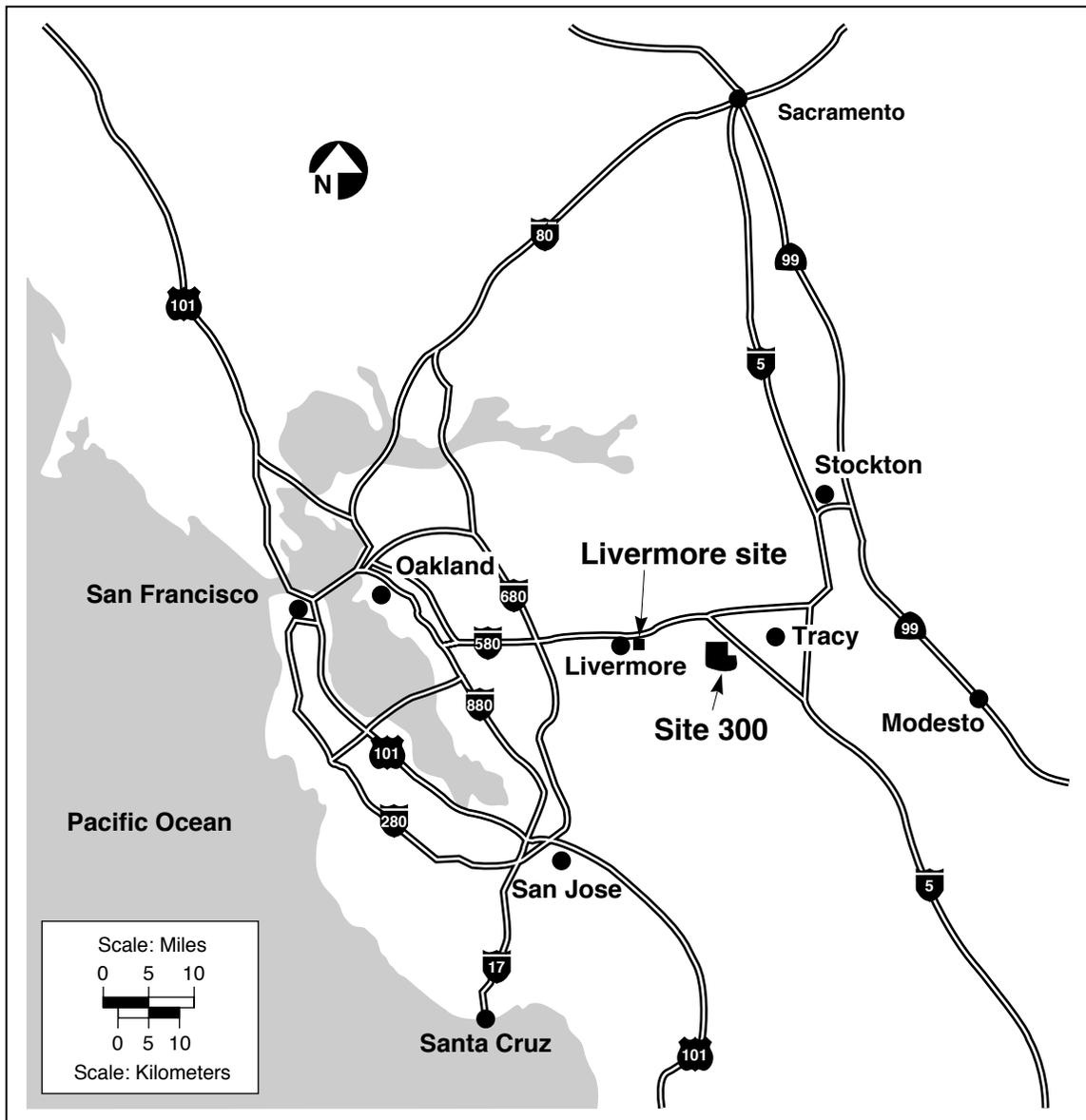
LLNL was established in 1952 to conduct nuclear weapons research and development. The Laboratory's mission is dynamic and has been broadened over the years to meet new national needs. LLNL serves as a national resource in science and engineering; its activities focus on global security, energy, global ecology, biomedicine, economic competitiveness, 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. The University of California operates LLNL for DOE.

### Livermore Site

LLNL's Livermore site occupies an area of 3.3 km<sup>2</sup> located about 60 km east of San Francisco, California, adjacent to the City of Livermore in the eastern part of Alameda County. In round numbers, 7 million people live within 80 km of the Livermore site; about 77,000 of them live in the City of Livermore.

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

The climate of the Livermore Valley is characterized by mild, rainy winters and warm, dry summers. The mean annual temperature was 15.2°C in 2003, 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 2003 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 over 50% of the time the winds blow from the south-southwest through west directions. However, during the winter, the wind often blows from the northeast. The average wind speed in 2003 at the Livermore site was 2.4 m/s (5.3 mph). Most precipitation occurs as rain between October and April with very little rainfall during the summer months. In 2003, the Livermore site received 23.9 cm of precipitation.



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

### Site 300

Site 300, LLNL’s Experimental Test Facility, is located 24 km east of the Livermore site in the Altamont Hills of the Diablo Range and occupies an area of 30.3 km<sup>2</sup>. A State of California vehicular-recreation area is located nearby, and wind-turbine generators line the surrounding hills. The remainder of the surrounding area is in agricultural use, primarily pasture land for cattle and sheep. The nearest residential area is the city of Tracy (population approximately 66,000), located 10 km to the northeast.



## SECTION II. Air Emission Sources and Data

### Sources

Nearly sixty different radioisotopes were used at LLNL in 2003 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, depending on the isotopes, the quantities being used, and the types of operations being performed. Work places include gloveboxes, exhaust hoods, and laboratory bench tops. Exhaust paths to the atmosphere include triple HEPA (High Efficiency Particulate Air) 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 used at LLNL during 2003.

Hydrogen-3	Argon-41	Strontium-90	Rhenium-187	Uranium-233	Plutonium-240
Nitrogen-13	Chromium-51	Technetium-99	Thallium-204	Uranium-234	Americium-241
Carbon-14	Manganese-54	Iodine-125	Lead-210	Uranium-235	Plutonium-241
Oxygen-15	Iron-55	Iodine-131	Radium-226	Plutonium-236	Americium-242m
Sodium-22	Cobalt-57	Barium-133	Radium-228	Uranium-236	Plutonium-242
Phosphorus-32	Cobalt-60	Cesium-134	Thorium-228	Neptunium-237	Americium-243
Phosphorus-33	Nickel-63	Cesium-137	Thorium-229	Uranium-237	Californium-249
Sulfur-35	Zinc-65	Cerium-144	Palladium-231	Plutonium-238	Californium-252
Chlorine-36	Selenium-75	Promethium-147	Thorium-232	Uranium-238	
Potassium-40	Strontium-89	Europium-152	Uranium-232	Plutonium-239	

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 2003

In this section we describe continuous stack-effluent sampling systems at selected LLNL facilities and ambient air monitors in place at numerous locations on and off LLNL sites.

#### **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 2003, there were seven buildings (Buildings 175, 235, 251, 331, 332, 491, and 695) 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. Many samplers would operate from emergency power systems if normal power were lost.

Air samples for particulate emissions are extracted downstream of HEPA filters and prior to the discharge point to the atmosphere. Particles are collected on membrane filters. The sample filters are removed and analyzed for gross alpha and beta activity on a weekly or bi-weekly frequency depending on the facility. In most cases, simple filter aerosol collection systems are used. However, in some facilities, alpha continuous air monitors (CAMs) are used for sampling. In addition to collecting a sample of particles, the CAM units provide an alarm capability for the facility in the event of an unplanned release of alpha activity.

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

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

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

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

Building	Facility	Analytes	Sample type	Number of samplers
175	MARS <sup>a</sup>	Gross $\alpha$ , $\beta$ on particles	Filter	6
235	Chemistry and Materials Science	Gross $\alpha$ , $\beta$ on particles	Filter	1
251	Heavy Elements Unhardened area	Gross $\alpha$ , $\beta$ on particles	Filters	23
	Hardened area	Gross $\alpha$ , $\beta$ on particles	Filters	4
331	Tritium	Tritium	Ionization Chamber <sup>b</sup>	4
		Gaseous tritium/ tritiated water vapor	Molecular sieves	4
332	Plutonium	Gross $\alpha$ , $\beta$ on particles	CAM <sup>b</sup>	12
		Gross $\alpha$ , $\beta$ on particles	Filters	15
491	Isotope Separation <sup>a</sup>	Gross $\alpha$ , $\beta$ on particles	Filter	1
695	Decontamination and Waste Treatment Facility	Gross $\alpha$ , $\beta$ on particles	Filter	1
801A	Contained Firing Facility	Gross $\alpha$ , $\beta$ on particles	Filter	1

Note: "CAM" denotes Eberline continuous air monitors.

<sup>a</sup> Operations discontinued, however, air effluent sampling systems at this building continued to operate as part of the maintenance and surveillance shutdown plan for the facilities. The sampling system in Building 175 was removed from service in May 2003; the building no longer contained an inventory of radioactive materials.

<sup>b</sup> Alarmed systems.

**Results of Stack Monitoring for Tritium:** The stack effluent monitoring equipment at the Tritium Facility (Building 331) began functioning improperly in July 2003. Repairs of the sampling systems were completed in late October 2003, and measured emissions returned to normal, giving results in the expected range. For the July through October time period, emissions were reconstructed using data from LLNL ambient air tritium monitors. The estimated tritium emission from the stacks during the nearly four-month period of faulty performance was estimated to be 41 Ci ( $1.5 \times 10^{12}$  Bq) (HTO and HT combined). The measured emission during months in 2003 when stack sampling was behaving normally and considered accurate was 69 Ci ( $2.6 \times 10^{12}$  Bq). Combining the two periods resulted in a total tritium release from the Tritium Facility stacks in 2003 of 110 Ci ( $4.1 \times 10^{12}$  Bq). Of this, approximately 104 Ci

( $3.8 \times 10^{12}$  Bq) were released as tritiated water (HTO) and 6 Ci ( $2.2 \times 10^{11}$  Bq) as elemental tritium gas (HT). The highest single weekly stack emission from the facility was 10.2 Ci ( $3.8 \times 10^{11}$  Bq), of which more than 97% was HTO.

This 2003 level of tritium emissions was comparable to those in recent years; **Table 3** displays the combined HTO and HT emissions from the Tritium Facility since 1981. We anticipate that emissions over the next five years will exceed the 2000–2003 levels, as research and development work is performed for new programmatic efforts. However, engineered controls designed to contain and recapture tritium leakage should maintain relatively low emissions.

**Stack Monitoring for Gross Alpha and Gross Beta Radiation:** For most discharge points at the other facilities where continuous stack sampling is performed, the results are below the minimum detectable concentration (MDC) of the analysis; sometimes as few as 1 to 4 samples (out of 25 to 50 per year) have concentrations greater than the MDC. Generally, these few samples having results above the MDC are only marginally above it. Use of zero values for this type of data can be justified based on knowledge of the facility, the use of tested, multiple stage, HEPA filters in all significant release pathways, and alpha spectroscopy based isotopic analyses of selected air sampling filters. These isotopic analyses demonstrate that detected activity on air sampling filters comes from naturally occurring radionuclides, such as radon daughters, e.g., polonium, on the air sampling filters. In addition, because of exhaust configurations at some facilities, the monitoring systems sometimes sample air from the ambient atmosphere along with the HEPA filtered air from facility operations, giving rise to background atmospheric radioactivity being collected. Because of these considerations, the emissions from such facility operations are reported as zero. 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.

An effluent sampling system was installed at the Contained Firing Facility (CFF, Building 801A) at Site 300 in early 2002. Although all facility operations are HEPA filtered, this building has a large high bay room that exhausts to the stack without HEPA filtration. Consequently, some of the air sampled by the effluent sampling system is essentially outside, ambient air. In order to determine if any releases actually occurred from this facility, the sampling results must be compared to ambient air. In 2003, ten samples out of 48 had concentrations greater than the MDC. The median concentration of CFF stack detections,  $6.6 \times 10^{-15}$  Ci/m<sup>3</sup> ( $2.4 \times 10^{-4}$  Bq/m<sup>3</sup>), was slightly higher than the median concentrations of the detections from two offsite sampling locations that are used to establish background levels of gross

**Table 3.** Combined HT and HTO emissions from the Tritium Facility, 1981–2003.

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

<sup>a</sup> The doses calculated from these emissions include HT releases modeled as HTO, as directed by EPA Region IX. EPA Region IX acknowledges that such modeling results in an overestimation of the tritium dose. This methodology is used for purposes of evaluating NESHAPs compliance.

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

alpha and beta activity for direct comparison to results from the air effluent samplers. The median of all 48 of the CFF samples,  $7.4 \times 10^{-16}$  Ci/m<sup>3</sup> ( $2.8 \times 10^{-5}$  Bq/m<sup>3</sup>), was approximately three times lower than the median of all of the offsite sampling location samples. Because the median concentration of the detectable CFF gross alpha samples exceeded the median concentration of the current background locations, we take a conservative approach and report gross alpha measurements as actual emissions. The gross alpha emissions for CFF were determined to be  $5.1 \times 10^7$  Ci/y ( $1.9 \times 10^4$  Bq/y). The resulting radiological dose determined with CAP88-PC modeling was  $1.3 \times 10^{-6}$  mrem ( $1.3 \times 10^{-5}$   $\mu$ Sv); doses are discussed in Section III and Attachment 1.

Among the facilities monitored for gross alpha and beta in 2003, only the CFF showed emissions.

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

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

reasonable probability that any significant airborne concentration of particulate or tritiated water vapor effluents resulting from LLNL operations will be detected. Many of the surveillance air monitors are placed near diffuse emission sources, such as those near Buildings 331 and 614, as well as in and around the Southeast Quadrant of the Livermore site. As such, their results can be used to estimate and/or confirm the emissions from the associated diffuse sources. Also included is an air particulate monitor positioned at the location of the hypothetical maximally-exposed member of the public (defined in Section III) for the Livermore site. Data from air surveillance monitors provide a valuable test of predictions based on air dispersion modeling, and can help characterize unplanned releases of radioactive material.

Data from the surveillance air-monitoring network are presented in the LLNL Site Annual Environmental Report (SAER), which is available to the public in hardcopy form, on CD, and on the Internet. See, e.g., Sanchez et al., *Environmental Report 2002*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-50027-02, September 2003; <http://www.llnl.gov/saer>.

### ***Characterizing Minor Radiological Sources By Ambient Air Monitoring***

Since 1991, LLNL has demonstrated compliance for minor sources (which are primarily non-monitored stack sources) through a labor-intensive inventory and modeling process. The dose consequences to the public for these sources were 8 to 20 orders of magnitude below the regulatory standard of 10 mrem/y and never affected LLNL's reported dose. To better allocate resources, LLNL made a request, pursuant to the NESHAPs regulations, to use existing ambient air monitoring to demonstrate compliance for minor emissions sources. This request was made in March 2003 and granted in April 2003; see Attachment 3 in last year's NESHAPs annual report (Harrach et al. *LLNL NESHAPs 2002 Annual Report*, UCRL-ID-113867-03, June 2003). For the present compliance report, covering LLNL operations in 2003, LLNL is for the first time demonstrating NESHAPs compliance for minor sources using this new method.

Basically the method entails comparing measured ambient air concentrations at the location of the SW-MEI to concentrations limits set by the U.S. EPA in its Table 2 Appendix E to 40 CFR 61. The radionuclides for which the comparison is made are tritium and plutonium-239+240 for the Livermore SW-MEI and uranium-238 for the Site 300 SW-MEI. At the Livermore site, the average of the monitoring results for locations L-VIS and L-CRED (shown in **Figure 6** in Section VII) 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 air particulate monitoring locations at the site were used to represent the concentration at the SW-MEI location.

EPA's Table 2 Appendix E to 40 CFR 61 standards and the 2003 measured concentrations at the location representing the SW-MEI are shown in **Table 4**. As demonstrated by the calculation of the fraction of the standard, LLNL measured concentrations for tritium and plutonium-239+240, and uranium-238 in air are a fraction 0.003 or less of the standard for these radionuclides.

**Table 4.** Mean concentrations of radionuclides of concern at the location of the SW-MEI in 2003.

Location	Nuclide	EPA Table 2 concentration standard (Ci/m <sup>3</sup> )	Mean measured concentration (Ci/m <sup>3</sup> )	Measured concentration as a fraction of the std.	Detection limit (approx.) (Ci/m <sup>3</sup> )
Livermore site SW-MEI	Tritium	$1.5 \times 10^{-9}$	$5.0 \times 10^{-12} *$	$3.3 \times 10^{-3}$	$1 \times 10^{-12}$
Livermore site SW-MEI	Plutonium-239	$2.0 \times 10^{-15}$	$1.3 \times 10^{-19} **$	$6.5 \times 10^{-5}$	$5 \times 10^{-19}$
Site 300 SW-MEI	Uranium -238	$8.3 \times 10^{-15}$	$7.0 \times 10^{-18} ***$	$8.4 \times 10^{-4}$	$3 \times 10^{-20}$

\* The tritium value includes contribution of emissions from the Tritium Facility, estimated at  $3.8 \times 10^{-12}$  Ci/m<sup>3</sup>

\*\*Note that the mean measured concentration for plutonium is less than the detection limit; only 3 of the 24 values comprising the mean were measured detections.

\*\*\*The mean ratio for uranium-235/uranium-238 for 2003 is 0.00708, which is only slightly less than 0.00726, the ratio of these isotopes for naturally occurring uranium. This indicates that approximately 96% of the measured quantities of uranium-238 were caused by resuspension of soil containing naturally occurring uranium.

The LLNL radiological facilities included in the "minor sources" classification in 2003 are listed in **Table 5**.

## Radionuclide Usage Inventories

Reliance upon radionuclide usage inventory forms was much reduced in 2003 due to implementation of the new emissions accounting method for minor sources. Inventories were utilized to calculate public dose impacts only for the 5 principal operations of the Radioactive and Hazardous Waste Management (RHWM) Division at the Livermore site, and the open-air explosives experiments at Site 300 (see Attachment 1). Other inventory forms were provided for 2003 operations of the

National Ignition Facility (NIF) and various other activities/experiments having the potential for radiological releases to air; all fell into the category of minor sources.

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

**Table 5.** Buildings with minor radiological emissions (by directorate), for 2003.<sup>a</sup>

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

<sup>a</sup> Directorate abbreviations refer to Chemistry and Materials Science, Physics and Advanced Technologies, Safety and Environmental Protection, Energy and Environment, Engineering, Biology and Biotechnology Research, Defense and Nuclear Technologies, National Ignition Facility, and Institutional (Deputy Director for Operations).

### SECTION III. Dose Assessment Methods & Concepts

#### Description of the Air Dispersion and Dose Model

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

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 (e.g., a business, church, school, or residence), for comparison to the 10 mrem/y (100  $\mu$ Sv/y) standard; (2) the maximum dose to any member of the public, in any direction (usually a point at the LLNL fence line), 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 the CAP88-PC model runs. These include Bldg. number; stack ID; isotope(s); emission rate in curies per year (1 Ci =  $3.7 \times 10^{10}$  Bq); and stack parameters, including height, diameter, and emission velocity.

**Meteorological Data:** All model runs used actual 2003 Livermore-site and Site 300 meteorological data, collected from the meteorological towers for each site. At these towers, wind speed and direction and temperature are sampled every one or two seconds, and are averaged into quarter-hour increments, time tagged, and computer recorded. The data are converted into a CAP88-PC input wind file using EPA guidelines.

**Surrogate Radionuclides:** CAP88-PC contains a library of 265 radionuclides; however, it does not contain all the radionuclides in use at LLNL. As a consequence, it was necessary in a few cases to use surrogate radionuclides to estimate EDEs. Attachment 2 shows the surrogate radionuclides used in CAP88-PC. 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, experimenters did not provide isotopic analyses of mixtures of radionuclides, and they identified the radionuclides used as "gross alpha," "gross beta," "gross gamma," or "mixed fission products" (MFP). In these cases,  $^{239}\text{Pu}$  was used as the surrogate for gross alpha,  $^{137}\text{Cs}$  was used as the surrogate for gross gamma, and  $^{90}\text{Sr}$  was used as the surrogate for gross beta and mixed fission products to provide conservative dose estimates.

**Population Inputs:** For the 2003 modeling effort, we updated the population distributions centered on the two LLNL sites. These population distributions are based on the LandScan Global Population 2001 Database (Dobson, J. E., E. A. Bright, P. R. Coleman, R.C. Durfee, B. A. Worley, LandScan: A Global Population Database for Estimating Populations at Risk, *Photogrammetric Engineering & Remote Sensing* Vol. 66, No. 7, July 2000, pp. 849-857; see also the Website

<http://www.ornl.gov/sci/gist/landscan/index.html>). 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 relatively nearby communities of Livermore and Tracy, and the more distant metropolitan areas of Oakland, San Francisco, and San Jose, as well as the San Joaquin Valley communities of Modesto and Stockton. Within the 80 km outer distance specified by 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:** Options for model inputs regarding agricultural characteristics and land use are established by the EPA, and the particular designation selected can strongly influence the ingestion dose received by the population being evaluated. The “user entered” option was again selected for the CAP88-PC modeling effort for 2003. The values entered corresponded to the “local agriculture” option (everything is home produced), with one exception—all milk consumed was assumed to be imported when assessing dose to individuals (as opposed to populations). An assumption that all milk comes from local cows would not be supported by the agricultural activities conducted in the area. For population dose assessments, all food is considered to be locally grown, i.e., grown within an 80 km radius about the site; default densities of agricultural products in California are used.

**Emission Source Terms:** The source term for each emission point in the calculations was determined by one of two methods: For continuously monitored sources, the sampling data (curies released per unit time) for each radionuclide were used directly. For unmonitored facilities, the radionuclide usage inventories, together with time factors and EPA-specified physical state factors, are used to estimate potential emissions to air from a source. Time factors are used to 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. Time factors are chosen to allow a more reasonable estimate of the amount of radioactive material released into the atmosphere. The EPA-specified factors for potential release to air of materials in different physical states (solid, liquid, powder, or gas) are those stated in 40 CFR Part 61, Appendix D. If the material was an unconfined gas, or any material heated above 100°C (with exceptions noted in Table 6), 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.

The U.S. EPA has granted approval for LLNL to use alternative physical state factors for elemental uranium, various uranium compounds/alloys, and elemental plutonium. In 2003, LLNL requested general permission to use physical state factors based on actual physical form. The U.S. EPA did not grant this request, stating they are open to further discussion on this issue, but that such a change may require modification of the regulations. **Table 6** provides the approved temperatures for application of the physical state factor for each material.

In addition to physical state factors, emission control abatement factors (40 CFR 61, Appendix D) were used, when applicable. Each HEPA filter stage was given a 0.01 abatement factor. (However, abatement factors were not used to evaluate compliance with the 0.1 mrem [1  $\mu$ Sv] standard that determines the need for continuous monitoring at a facility.) The use of actual stack effluent sampling data is much more direct, and presumably more accurate, than using assumptions based on usage inventory, time factors, release fractions, and emission control factors.

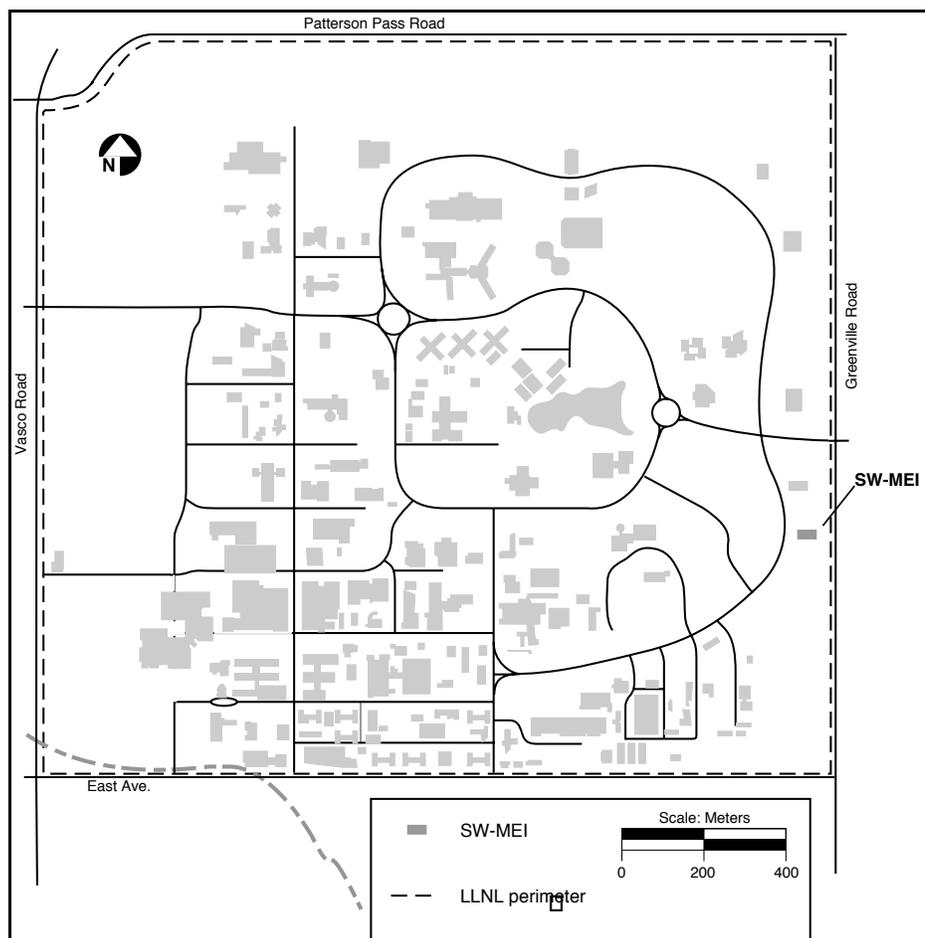
**Table 6.** List of materials exempted from the “treat as a gas above 100°C rule,” and temperatures at which the various physical state factors apply.

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

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

At the Livermore site, the SW-MEI for 2003 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 2003 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 Bldg. 851, as shown in **Figure 4**.

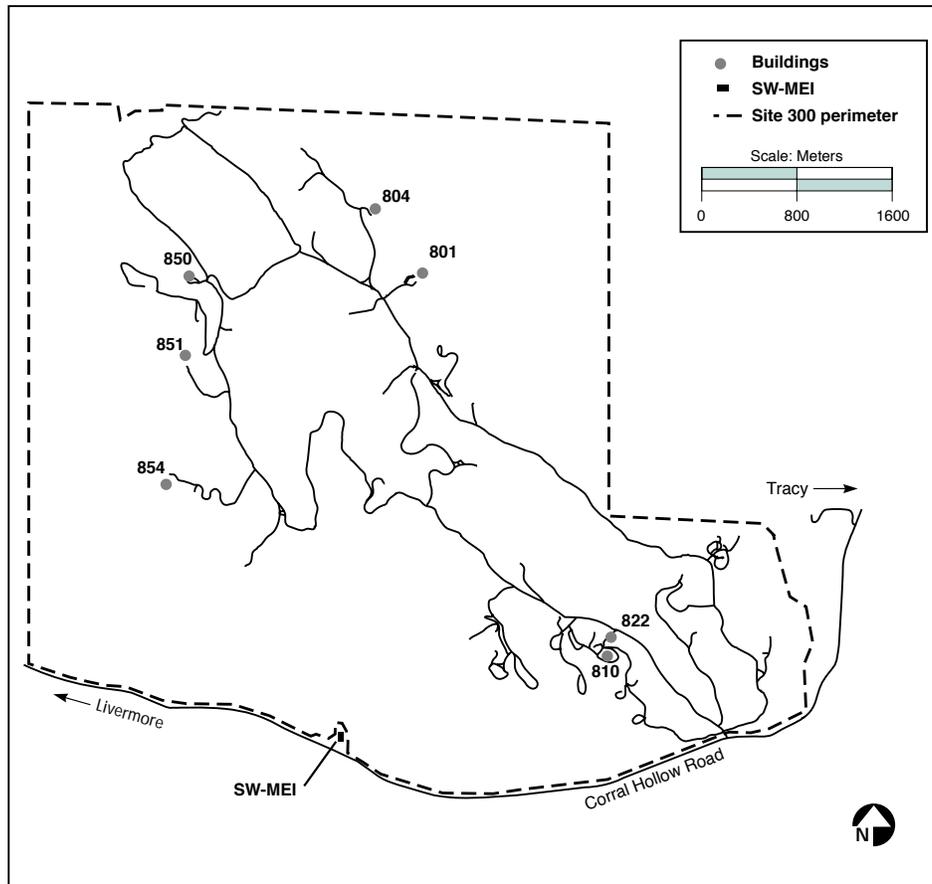


**Figure 3.** Location of Site-Wide Maximally Exposed Individual (SW-MEI) at the Livermore site, 2003.

In the Attachment 1 spreadsheet, the distance and direction to the respective SW-MEI are shown for each facility at each site. Doses to the SW-MEIs were evaluated for each source and then totaled for site-specific evaluations against the 10 mrem/y (100  $\mu$ Sv) dose standard (see “Total Dose to Site-Wide Maximally Exposed Individuals” in Section IV).

**Maximally Exposed Public Individual:** To assess compliance with the EPA requirement for continuous monitoring of a release point (potential dose greater than 0.1 mrem/y [1.0  $\mu$ Sv/y]), emissions must be individually evaluated from each point source; the location of the maximally exposed public individual (MEI) is

generally different for each emission point. The maximum dose at a location of unrestricted public access typically occurs at a point on the site perimeter. Therefore,



**Figure 4.** Location of Site-Wide Maximally Exposed Individual (SW-MEI) at Site 300, 2003.

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 a stack is close to the perimeter; generally, for all emission points at the Livermore site (and also at Site 300 with the exception of dispersals from some of the open-air explosives experiments), calculations show that ground level concentrations of radionuclides decline continuously beyond LLNL boundaries. As stipulated by the regulations in 40 CFR Section 61.93 (b)(4)(ii), modeling for assessment of continuous monitoring requirements assumed unabated emissions (i.e., no credit was taken for emission abatement devices, such as filters). Model runs typically include evaluation of the dose to the MEI and the distance and direction to the LLNL fence line where the MEI is located.

## SECTION IV. Results of 2003 Radiological Dose Assessment

This section summarizes the doses to the most exposed public individuals from LLNL operations in 2003, shows the comparison to previous years, presents the potential doses to the populations residing within 80 km of either the Livermore site or Site 300, and summarizes LLNL's compliance with 40 CFR 61, Subpart H (61.93).

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

The total dose to the Livermore site SW-MEI from operations in 2003 was 0.044 mrem (0.44  $\mu$ Sv). Of this, 0.024 mrem (0.24  $\mu$ Sv) or 55% was contributed by point sources, while diffuse emissions accounted for 0.020 mrem (0.20  $\mu$ Sv) or 45% of the total. The point source dose includes Tritium Facility HT emissions modeled as HTO, as directed by EPA Region IX. The SW-MEI dose calculated using CAP88-PC-T with its NEWTRIT model (see "Modeling Dose from Tritium" in Section VII), rather than the default CAP88-PC code, reduced the tritium component of the Livermore site dose from 0.041 mrem (0.41  $\mu$ Sv) to 0.030 mrem (0.30  $\mu$ Sv).

The total dose to the Site 300 SW-MEI from operations in 2003 was 0.017 mrem (0.17  $\mu$ Sv). Point source emissions from firing table explosives experiments accounted for 98%, of this total, while 0.00034 mrem (0.0034  $\mu$ Sv), or about 2%, was contributed by diffuse sources.

**Table 7** shows the facilities or sources that collectively accounted for more than 90% of the doses to the SW-MEI for the Livermore site and Site 300 in 2003. 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** compares 2003 doses with those of previous years. No diffuse emissions were reported at Site 300 for years before 1993, so comparison of total Site 300 dose can only be made for 1993 and later. In addition, diffuse source doses were not reported separately from the total dose for the Livermore site for 1990 and 1991.

### Doses from Unplanned Releases

There were no unplanned atmospheric releases of radionuclides at the Livermore site or Site 300 in 2003.

## Population Doses

Population doses, or collective EDEs, for both LLNL sites were calculated out to a distance of 80 km in all directions from the site centers using CAP88-PC. As noted earlier, in Section III under "Population Inputs," revised population data files were used for the 2003 assessment. CAP88-PC evaluates the four principal exposure pathways: ingestion through food and water consumption, inhalation, air immersion, and irradiation by contaminated ground surface.

The CAP88-PC result for potential population dose attributed to 2003 Livermore-site operations was 1.6 person-rem (0.016 person-Sv); the corresponding collective EDE from Site 300 operations was 3.2 person-rem (0.032 person-Sv). These values are both quite small and within the normal range of variation seen from year to year. By way of comparison, the population dose in the United States from exposure to the average level of natural background radioactivity is  $1.9 \times 10^6$  person-rem ( $1.9 \times 10^4$  person-Sv).

**Table 7.** List of facilities or sources whose emissions collectively accounted for more than 90% of the SW-MEI doses for the Livermore site and Site 300 in 2003.

Facility (Source Category)	CAP88-PC Dose in mrem/y	CAP88-PC Percentage Contribution to Total Dose
<b>Livermore site</b>		
Bldg. 331 stacks (point source)	0.022*	50%
Bldg. 612 Yard (diffuse source)	0.013*	30%
Bldg. 331 Outside (diffuse source)	0.0059*	13%
Bldg. 612, Room 102 (point source)	0.0014	3.2%
<b>Site 300</b>		
Bldg. 851 Firing Table (point source)	0.017	98%
Soil resuspension (diffuse source)	0.00034	2%

\* When LLNL's NEWTRIT model (see Section VII, subsection on "Modeling dose from tritium") is used in CAP88-PC in place of CAP88-PC's default tritium model, the doses for the diffuse Building 612 yard and Building 331 Outside sources are reduced to 0.75 of the values shown, and that for the Building 331 stacks is reduced to 0.73 of the value shown. Doses for other sources in the table are practically unchanged, since they have minor or no contribution from tritium.

**Table 8.** Doses (in mrem) calculated for the Site-Wide Maximally Exposed Individual (SW-MEI) for the Livermore site and Site 300, 1990 to 2003.

Year	Total Dose	Point Source Dose	Diffuse Source Dose
<b>Livermore site</b>			
2003	0.044 <sup>a</sup>	0.024 <sup>a</sup>	0.020
2002	0.023 <sup>a</sup>	0.010 <sup>a</sup>	0.013
2001	0.017 <sup>a</sup>	0.0057 <sup>a</sup>	0.011
2000	0.038 <sup>a</sup>	0.017 <sup>a</sup>	0.021
1999	0.12 <sup>a</sup>	0.094 <sup>a</sup>	0.028
1998	0.055 <sup>a</sup>	0.031 <sup>a</sup>	0.024
1997	0.097	0.078	0.019
1996	0.093	0.048	0.045
1995	0.041	0.019	0.022
1994	0.065	0.042	0.023
1993	0.066	0.040	0.026
1992	0.079	0.069	0.010
1991	0.234	— <sup>b</sup>	— <sup>b</sup>
1990	0.240	— <sup>b</sup>	— <sup>b</sup>
<b>Site 300</b>			
2003	0.017	0.017	0.00034
2002	0.021	0.018	0.0033
2001	0.054	0.050	0.0037
2000	0.019	0.015	0.0037
1999	0.035	0.034	0.0012
1998	0.024	0.019	0.005
1997	0.020	0.011	0.0088
1996	0.033	0.033	0.00045
1995	0.023	0.020	0.003
1994	0.081	0.049	0.032
1993	0.037	0.011	0.026
1992	0.021	0.021	— <sup>c</sup>
1991	0.044	0.044	— <sup>c</sup>
1990	0.057	0.057	— <sup>c</sup>

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

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

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

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

Calculations of effective dose equivalents for Livermore-site and Site 300 facilities having the potential to release radioactive material to the atmosphere were found to be well below the 10 mrem (100  $\mu$ Sv) NESHAPs dose standard for dose to the most-exposed individual members of the public. Tritium accounted for 93% of the Livermore-site calculated dose, while at Site 300 practically the entire calculated dose was due to the isotopes  $^{238}\text{U}$ ,  $^{235}\text{U}$ , and  $^{234}\text{U}$ , in depleted uranium.

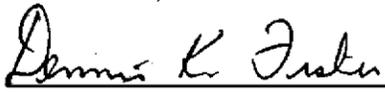
In 2003, there were seven buildings (Buildings 175, 235, 251, 331, 332, 491, and 695) at the Livermore site and one (Bldg. 801A, the Contained Firing Facility) at Site 300 that had radionuclide air effluent monitoring systems. These buildings are listed in **Table 2**, along with the number of samplers, the types of samplers, and the analytes of interest.

LLNL remains committed to monitoring stack effluent air from its Tritium Facility (Building 331), Plutonium Facility (Building 332), Decontamination and Waste Treatment Facility (Building 695), 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

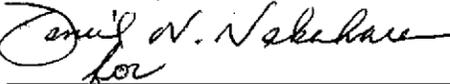
I certify under penalty of law that this document and all attachments were prepared under my direction or supervision in accordance with a system designed to assure that qualified personnel properly gather and evaluate the information submitted. Based on my inquiry of the person or persons who manage 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: Dennis K. Fisher  
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Safety and Environmental Protection  
Lawrence Livermore National Laboratory  
7000 East Avenue, L-668  
Livermore, CA 94550

Signature:  Date: 6/22/04  
Dennis K. Fisher

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:   
for  
Phillip Hill Date: 6/28/04

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

### **Use of Surveillance Air Monitoring in Demonstrating NESHAPs Compliance for LLNL's Numerous Minor Sources**

As noted earlier in Section II under the heading "Characterizing Minor Radiological Sources by Ambient Air Monitoring," the assessment of 2003 operations marked the first use of a new approach approved by EPA for evaluating NESHAPs compliance of LLNL's many minor sources of radiological releases to air. Greater reliance on surveillance air monitoring data and less on radiological usage inventories resulted in considerable simplification and savings in time and expense.

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

The LLNL NESHAPs quality assurance program is a multi-organizational effort that is described in the *Lawrence Livermore National Laboratory Quality Assurance Project Plan for National Emission Standards for Hazardous Air Pollutants (NESHAPs), 40 CFR 61, Subpart H (QAPP—Hall, L.C. and A.H. Biermann, UCRL-ID-13914, 2000)*. The QAPP is structured in the manner prescribed for quality assurance programs that is outlined in Appendix B, Method 114 of 40 CFR 61. The QAPP describes the organization structure and functional responsibilities, objectives of the quality assurance program, administrative controls in place for handling sample collection systems, sample collection and effluent flow rate measurement systems, corrective actions, and reporting.

The major components of this multi-organizational effort are the LLNL facilities/programs that have continuous monitoring systems, the Radiological Measurements Laboratory (RML) and the Analytical Laboratory (AL), both in the Hazards control Department (HCD), and the Environmental Protection Department (EPD). In addition to the QAPP, NESHAPs Agreement of Roles and Responsibilities (NARRs) documents are in place between EPD and the facilities and/or programs and HCD; these NARRs formalize responsibilities and obligations of the organizations regarding many tasks for the air effluent sample 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 archival of records.

EPD is responsible for an annual assessment and demonstration of LLNL's compliance with NESHAPs. The Department operates under a Quality Assurance Management Plan and associated procedures and guidance documentation. The Terrestrial and Atmospheric Monitoring and Modeling Group (TAMM) of EPD is responsible for environmental monitoring; air dispersion and dose assessment

modeling; assessment (in cooperation with Laboratory Program personnel) of usage and potential release of radioactive materials to air in operations throughout the Laboratory; and reporting to EPA and DOE to demonstrate the Laboratory's compliance with NESHAPs. Detailed records are kept of all measurements, computer model runs and other calculations, and selected model runs are validated. The TMM group is informed of proposed new operations, and modified operations where significant changes in radiological usage inventories occur, by several mechanisms. These include reviews of National Environmental Policy Act (NEPA) documentation, Integration Worksheets, Occupational Safety Plans (describing facility-specific safety procedures and plans), and knowledge derived from participation on EPD's Environmental Support Teams. All NESHAPs evaluations and calculations, along with supporting information, are archived for at least the period of time specified in 40 CFR 61 Subpart H.

### **Quality Control (QC) for 2003 Air Dispersion and Dose Assessment Model Runs and Radiological Usage Inventories**

Under the new protocol mentioned in the leading paragraph of this section, the only radiological facilities or projects providing an accounting by means of radionuclide inventory forms were ones commencing operation in 2003, or ones that contributed significantly to last year's dose to the public. The former underwent NESHAPs evaluation in which NEPA or related documents such as Integration Work Sheets and Occupational Safety Plans were examined both prior to start-up of operations and in a follow-up at year's end; none of these projects produced a significant radiological release to air. The latter were the five leading sources operated by Radioactive and Hazardous Waste Management (RHWM) Division. All inventory information specifying release potential for the RHWM sources were checked independently, and one of the 5 model runs was validated.

Model runs were performed for some two dozen sources in the 2003 assessment, including the activities mentioned above and one stack-monitored facility whose data showed a non-zero release to air (the Contained Firing Facility (CFF) at Site 300). Approximately 15% of the model runs were selected for validation, which entailed confirmation of both the source emission data and dose modeling calculations. Two sources, one from each of the two LLNL sites, were selected because they represented the most significant contributions to 2003 potential dose to the public; one was selected from the RHWM Division set; one from the set of continuously monitored sources; and one from the category of diffuse sources. Specifically, the sources chosen for quality control review were the following: the Tritium Facility's two 30-m stacks; one explosives experiment conducted at Site 300's Firing Table 851; the CFF at Site 300; one source reported by RHWM; and the Bldg. 612 Yard waste tritium storage area. Copies of individual model runs, including input parameters and resultant calculated doses, are archived in the records kept by the Terrestrial & Atmospheric

Monitoring & Modeling (TAMM) Group of the Environmental Protection Department.

Based on these QC efforts, we believe that the data, results, and conclusions presented in this report meet EPD's quality assurance objectives.

## **EPA Inspection**

EPA conducted a multi-media inspection on November 4 – 7, 2003, that included radiological NESHAPs stack and surveillance monitoring activities. EPA inspectors concluded that the overall evaluation of LLNL's sampling and analysis program with regard to compliance issues was very favorable, and that "the facility's level of compliance with the radionuclide NESHAPs was excellent." EPA's final report was issued May 21, 2004 (*Compliance Evaluation Inspection Report for Lawrence Livermore National Laboratory*, EPA RCRA No. CA2890012584).

## **SECTION VII: Supplementary Information on Radiological Dose Assessment for 2003**

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

The dose evaluations for diffuse sources at the Livermore site in 2003 required several different modeling approaches. Building 331 Outside Yard and Building 612 Yard emissions estimates were based on facility personnel knowledge and "back calculations" (in which the source terms in model runs were adjusted to reproduce the concentrations determined from environmental surveillance air monitoring data). Building 514 Tank Farm emissions estimates were derived from radiological usage inventory data. The dose in each of these cases was calculated using CAP88-PC. Air surveillance monitoring data for plutonium from a monitor located at the location of the SW-MEI was used directly (sans model run) to evaluate the dose from plutonium contamination in the Southeast Quadrant.

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

As the Tritium Facility (Bldg. 331) conducts operations, tritium-contaminated equipment and material slated for disposal is removed from the building, packaged in a waste accumulation and storage area, removed from the building to an outside storage container, and finally sent to Radioactive and Hazardous Waste Management Division (RHWM) facilities. During 2003, outgassing from such waste released an estimated 8.7 Ci ( $3.2 \times 10^{11}$  Bq) of tritium to the atmosphere outside Building 331. This amount was derived from a combination of environmental surveillance monitoring data and air dispersion back-calculation, and concurred with estimates based on process and facility knowledge. Its release was modeled in CAP88-PC as a 1 m<sup>2</sup> area source, leading to a calculated 2003 dose to the SW-MEI of

$5.9 \times 10^{-3}$  mrem ( $5.9 \times 10^{-2}$   $\mu$ Sv); a dose 0.75 times this amount was calculated when the NEWTRIT model was implemented in CAP88-PC.

### ***Building 514 Tank Farm***

Another potential source of diffuse emissions of a variety of radionuclides was RHWL waste storage and treatment operations. Bldg. 514 houses the RHWL "Tank Farm," consisting of six 7,170-liter tanks with ancillary equipment such as pumps, mixers, probes, and a bulking station. The tanks are used to store and treat liquid and solid radioactive and/or mixed wastes. Treatment is performed on a batch basis. Chemicals and waste are added to the tanks to achieve the desired treatment objectives. A 2003 radionuclide usage inventory was conducted for the facility to determine the diffuse source term (see Attachment 1 spreadsheet). CAP88-PC modeling gave a 2003 SW-MEI dose from Tank Farm releases to air to be  $5.9 \times 10^{-4}$  mrem ( $5.9 \times 10^{-3}$   $\mu$ Sv).

### ***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 median annual concentration of tritium in air for 2003 in this area was 2.4 pCi/m<sup>3</sup> ( $8.9 \times 10^{-2}$  Bq/m<sup>3</sup>). These data were used to calculate the total tritium emissions from the area, using a conservative approach that assumed the source to be 60 m south-southwest of the air sampler. With this assumption, a diffuse source emission of 3.4 Ci/y ( $1.3 \times 10^{11}$  Bq/y) was required to produce the concentrations measured at the air sampler. This source term produced a CAP88-PC-calculated 2003 dose to the SW-MEI from the Building 612 Yard of  $1.3 \times 10^{-2}$  mrem ( $1.3 \times 10^{-1}$   $\mu$ Sv); a dose 0.75 times this amount was calculated when the NEWTRIT model was implemented in CAP88-PC.

### ***Southeast Quadrant***

The Southeast Quadrant of the Livermore site has elevated levels of plutonium in the surface soil (from historic waste management operations) and air (from resuspension). A high volume air particulate sampler is located adjacent to the UNCLE Credit Union (the location of the SW-MEI) to monitor the plutonium levels in this area. Monitoring data from this air sampler were used as a direct measurement of potential dose via the air pathway. The median annual concentration of <sup>239+240</sup>Pu (the analytical technique used, namely alpha spectroscopy, does not distinguish between <sup>239</sup>Pu and <sup>240</sup>Pu) in air was  $1.3 \times 10^{-19}$  Ci/m<sup>3</sup> ( $4.9 \times 10^{-9}$  Bq/m<sup>3</sup>). Using the dose conversion factor of  $3.08 \times 10^5$  mrem/ $\mu$ Ci ( $8.32 \times 10^{-5}$  Sv/Bq) from Federal Guidance Report No. 11, EPA-520/1-88-020, U.S. Environmental Protection Agency (1988) for <sup>239</sup>Pu and <sup>240</sup>Pu, and the

standard man breathing rates of 8400 m<sup>3</sup>/y, the dose was determined to be 3.4 x 10<sup>-4</sup> mrem (3.4 x 10<sup>-3</sup> μSv) for 2003.

### **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<sup>3</sup>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<sup>3</sup>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 2003, all measurements in ambient air at the Site 300 perimeter location were consistent with natural background measurements.

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

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

A model was developed to distinguish between the contribution to measured uranium activities arising from naturally occurring uranium (NU) and that from depleted uranium (DU) contributed by LLNL operations. (A derivation of the model was presented in *LLNL NESHAPs 1995 Annual Report*, Gallegos et al., 1996.) We base our dose estimate for resuspended depleted uranium (DU) on the measured environmental surveillance monitoring total concentration in air of uranium-238, subtracting out the part contributed by NU, from the following equation:

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

where  $\mu$  is the fraction (by weight) of uranium contributed by operations, CU is composite uranium (both DU and NU),  $M(\text{CU-235})$  the mass of U-235 in the composite (measured) uranium, and  $M(\text{CU-238})$  the mass of U-238 in the composite (measured) uranium.

For 2003, 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  $3.4 \times 10^{-4}$  mrem ( $3.4 \times 10^{-3} \mu\text{Sv}$ ) for the SW-MEI dose resulting from resuspension of DU in soil for 2003.

## Estimating Temporal Effects on Dose from Explosive Experiments

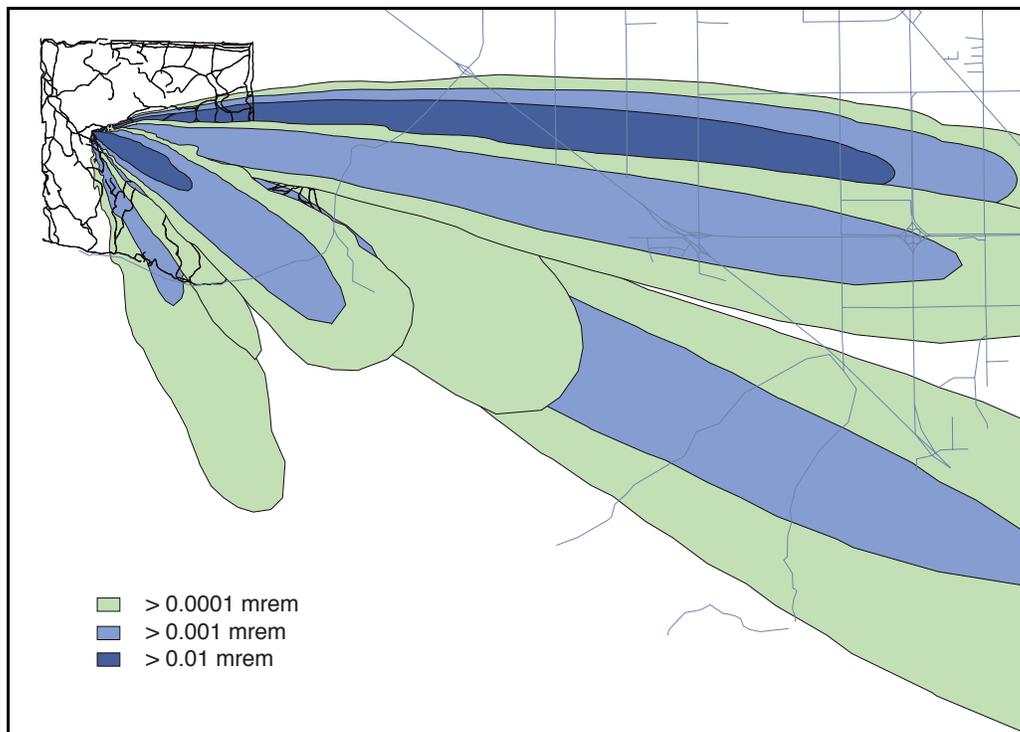
CAP88-PC, a continuous emission model with annual wind fields, though admittedly not well suited to model explosives testing events at Site 300, has been used for this purpose by LLNL since the beginning of its NESHAPs compliance efforts. In 1992, LLNL proposed to use the model INPUFF or a similar transient puff model that considered meteorological conditions prevailing at the time of the practically instantaneous release. The EPA rejected this approach, preferring the consistency that comes from having all releases modeled in the same manner for compliance purposes.

Recently, the National Atmospheric Release Advisory Center (NARAC), a programmatic research center at LLNL specializing in air dispersion modeling, made available some of its sophisticated modeling capabilities to an increased user base in the form of client server tools. Specifically, a NARAC modeling tool called iClient provides access to the three-dimensional hazardous material atmospheric transport and diffusion modeling capabilities of NARAC. Using iClient, the modeler creates input files on his local computer, then sends those files via the Internet to NARAC, where site-specific, computationally intensive meteorological calculations are performed, with the results returned to the user; see <http://narak.llnl.gov/iclient.html>.

In 2003, LLNL staff began to investigate the application of iClient to the explosives tests at Site 300. The dose output plumes for the model runs corresponding to the seven tests are qualitatively displayed in **Figure 5**. Because CAP88-PC and iClient are such different models, it is difficult to compare the results from them. One useful comparison is to look at the population dose estimates of each model, shown in **Table 9**. The total population dose estimated by CAP88-PC from the 2003 explosives tests at Site 300 was 3.2 person•rem/y. In contrast, the total population dose estimated by iClient for the same tests was  $3.4 \times 10^{-3}$  person•rem/y, three orders of magnitude smaller. A small part of this difference can be explained by the fact that

iClient does not include a calculation of ingestion dose. If the ingestion dose component were added to the iClient estimate, the dose would increase slightly, to  $3.9 \times 10^{-3}$  person•rem/y. The primary reason the dose estimates are so different is that CAP88-PC calculates dose to everyone within an 80-km (50-mile) radius of Firing Table 851 at Site 300, weighted by the frequency and strength of winds in particular directions. This 360-degree dispersal in CAP88-PC encompasses large population centers, in particular the San Francisco Bay Area, whereas iClient only calculates doses to people in the sparsely populated areas to the east through south directions from Site 300, where the transient plumes are transported.

The estimate from CAP88-PC is almost surely the less accurate of the two. Nonetheless the use of CAP88-PC accomplishes what is intended for regulatory compliance, i.e., the calculation errs on the “health protective” side of over-predicting dose.



**Figure 5.** Results from NARAC iClient model for tests at Site 300’s Firing Table 851 in 2003, qualitatively showing plumes from each of the seven experiments.

**Table 9.** Comparison of population doses from explosives experiments<sup>a</sup> at Site 300 in 2003, as evaluated using the CAP88-PC code<sup>b</sup> and iClient code.<sup>c</sup>

Shot Number (Date)	CAP88-PC Code Population Dose <sup>d</sup> (person-rem/y)	iClient Code Population Dose <sup>e</sup> (person-rem/y)
1 (27Mar03)	0.141	$4.2 \times 10^{-6}$
2 (9April03)	0.137	$7.9 \times 10^{-5}$
3 (24June03)	0.572	$3.9 \times 10^{-5}$
4 (10July03)	0.521	$4.1 \times 10^{-4}$
5 (7Aug03)	0.144	$5.5 \times 10^{-4}$
6 (13Aug03)	0.134	$5.8 \times 10^{-5}$
7 (15Oct03)	1.52	$2.3 \times 10^{-3}$
Totals	3.17	$3.4 \times 10^{-3}$

<sup>a</sup> In 2003, all open-air explosives experiments were conducted on Firing Table 851.

<sup>b</sup> CAP88-PC models the release as continuous, using meteorological data covering one year.

<sup>c</sup> The iClient code models the release as a short duration puff, using meteorological data appropriate to the period of release.

<sup>d</sup> Population dose includes all persons within 80 km (50 mi) of F.T. 851.

<sup>e</sup> Population dose includes persons within plume out to 80 km (50 mi) from F.T. 851.

## 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 tritiated gas (HT) or organically bound tritium (OBT) are not calculated. CAP88-PC's tritium model is based on the specific activity model, which assumes that the tritium-to-hydrogen ratio in body water is the same as in air moisture. Because the specific activity model is linked in CAP88-PC with relatively high dose coefficients for HTO, the model's dose predictions generally err on the high side.

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

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

A simple tritium model, NEWTRIT, has been developed that calculates ingestion dose from both HTO and OBT and accounts for conversion of HT to HTO in the environment following releases of HT (Peterson, S-R. and P.A. Davis, *Health Physics* 82(2): 213-225, 2002). For calculating doses in this report, LLNL has used the NEWTRIT model in CAP88-PC, in addition to the default CAP88-PC code, to estimate doses from significant sources of tritium emissions; see, e.g., Table 4. A brief discussion of the NEWTRIT model was presented in Attachment 2 of the 2000 NESHAPs annual report (*LLNL NESHAPs 2000 Annual Report*, Gallegos et al. June 2001).

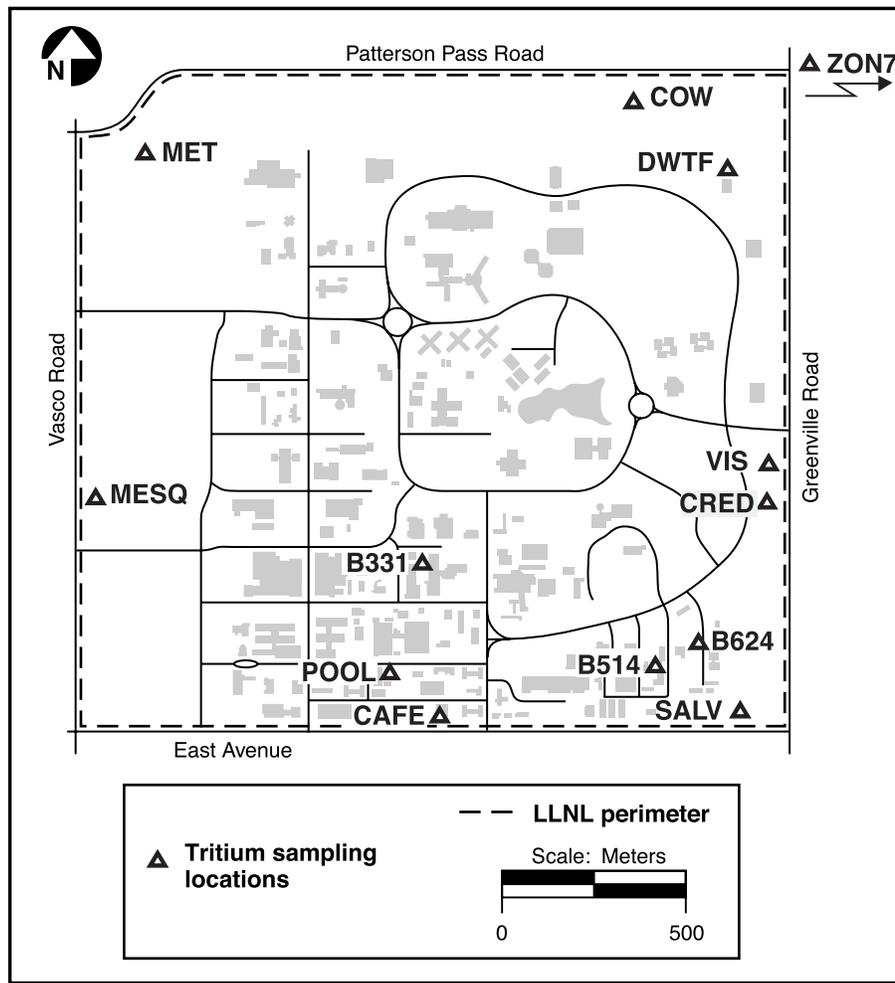
In October 2001, LLNL sent a letter to EPA Region IX requesting consideration of NEWTRIT as an alternative methodology for calculating doses from atmospheric releases of tritiated water vapor (HTO) and tritiated gas (HT), for use in demonstrating compliance with radionuclide NESHAPs (40 CFR 61 Subpart H). In late 2002, the EPA had NEWTRIT coded into GENII-NESHAPs, a version of GENII (Napier et al. 1988) that the EPA plans to approve as a regulatory model for evaluating radionuclide NESHAPs compliance. At this writing, GENII-NESHAPs is undergoing peer review and should be approved in late 2004.

## Comparison of 2003 Modeling Results with Tritium Air Surveillance Monitoring Data

A comparison was made between CAP88-PC-predicted concentrations of tritium in air and ambient air monitoring data for eleven tritiated water vapor samplers on the Livermore site (designated, CAFE, DWTF, MESQ, MET, COW, POOL, SALV, VIS, B331, B514, and B624) and one off-site sampler (ZON7). Sampling at B292 was discontinued in 2003, so no model predictions for B292 were calculated. Monitor locations are shown in **Figure 6**. Modeled predictions have been compared with monitoring observations since 1997.

Only concentrations from the three most significant sources of tritium releases to air at the Livermore site were included in the model-data comparison. The largest point source is the Tritium Facility (Bldg. 331), where tritium is emitted from two 30-m-high, continuously monitored stacks. Based on stack monitoring and emission reconstruction, a total of 110 Ci ( $4.07 \times 10^{12}$  Bq) of HTO was emitted from Bldg. 331 stacks in 2003. (The estimated 6.33 Ci [ $2.34 \times 10^{11}$  Bq] of HT emitted from the Tritium Facility stacks is not included in the comparison because the tritium air surveillance monitors register only HTO.) Generally one would expect the Tritium Facility stacks to make the largest contribution to concentrations of tritium at distant monitors (e.g., ZON7), because the emissions are cast high into the air and carried with the wind. Diffuse-source emissions are lower to the ground, primarily affecting those monitors in close proximity. The other two principal sources in our modeling/measurement comparison are of this type: open-air diffuse emission areas associated with the Bldg. 612 Yard and the Tritium Facility (Bldg. 331) outside yard waste accumulation and storage areas. Emissions from the Bldg. 612 Yard source were estimated to be 3.4 Ci ( $1.3 \times 10^{11}$  Bq), based on calibrating CAP88PC-predictions of tritium concentrations at the tritium monitor B624 closest to it. (Thus the Bldg. 624 data do not provide a test of the modeling.) Emissions from the B331 outside yard source were estimated to be 8.7 Ci ( $3.2 \times 10^{11}$  Bq) in 2003, based on facility knowledge and environmental monitoring data (primarily the Bldg. 331 monitor near this yard). While these two diffuse sources contribute significantly to tritium concentrations in all of the monitors, all other potential sources of tritiated water vapor release were too minor to influence the overall model-data comparison.

Annual average concentrations of HTO in air ( $\text{pCi}/\text{m}^3$ ) at the locations of the twelve monitors were modeled for the three sources individually, and the sum of the three contributions was compared to the measured annual mean concentrations. The results, displayed in **Table 10**, show that by taking into account the leading sources releasing tritiated water vapor to air, fairly good agreement is obtained between model runs and data for all of the air tritium monitors.



**Figure 6.** Tritiated water vapor surveillance sampling locations, Livermore site.

With the exception of the air tritium monitoring locations DWTF and ZON7, all predictions are equal to or greater than the measured concentrations of tritium in air. The under-estimation at DWTF, which is located near the newly opened Decontamination and Waste Treatment Facility, is due to activities at the facility. The small under-estimation at ZON7 is not meaningful due to the large uncertainty on the observed value, given that only 56% of the observations were above detection limits. Thus, in 2003, as in the past, CAP88-PC over-predicts HTO in air from LLNL releases of HTO. This consistent over-prediction since 1997, especially at those locations to the west and south, is probably caused by the relative importance of the diffuse sources for these years (S-R. Peterson, "Testing CAP88-PC's Predicted Air Concentrations Against Historical Air Tritium Monitoring Data, 1986–2001, at

Lawrence Livermore National Laboratory," LLNL Report UCRL-ID-155505, 2003). A comparison of AIRDOS-EPA predictions of air concentrations for various radionuclides ( $^{234}\text{U}$ ,  $^{238}\text{U}$ ,  $^{85}\text{Kr}$ , and  $^3\text{H}$ ) with measurements at six different sites concluded that the 90% confidence interval for the accuracy of the CAP88-PC dispersion model ranged from a factor of 0.3 to 4.4, based on 51 samples (Jack Faucett Associates, Report JACKFAU-341/12-87; 1987). Similarly, the Peterson study cited above compared CAP88-PC predictions with air tritium concentrations at 13 perimeter and off-site locations for 1986–2001; it found that 96% of all predictions fell within a factor of three of the observations, and slightly more than half of the predicted air concentrations were greater than the observed air concentrations.

**Table 10.** Comparison of measured and modeled annual mean concentrations of tritiated water vapor (HTO) in air at selected Livermore site locations, 2003.

Air monitor (name)	Mean measured concentration ( pCi/m <sup>3</sup> )	Modeled* average concentration (pCi/m <sup>3</sup> )	Ratio of modeled- to-measured concentrations	Modeled concentration of tritium in air contributed by the indicated source (pCi/m <sup>3</sup> )		
				B331 Stacks	B612 Yard	B331 Outside
B331	90.8	93	1.0	0.19	2.2	91
B624	84.4	89	1.1	4.1	84	1.1
POOL	7.85	18	2.3	4.5	1.8	12
DWTF	7.02	4.9	0.70	3.8	0.32	0.73
B514	6.09	17	2.8	1.6	15	0.86
VIS	4.89	6.3	1.3	3.5	1.9	0.92
COW	3.54	5.4	1.5	4.2	0.31	0.88
CAFÉ	3.01	8.4	2.8	2.1	1.9	4.40
SALV	2.46	3.0	1.2	1.20	1.3	0.48
ZON7**	2.05	1.7	0.83	1.30	0.17	0.21
MET**	0.967	1.8	1.9	0.33	0.25	1.2
MESQ**	0.956	5.0	5.2	0.50	0.55	3.9
(CRED)***		7.3		3.8	2.4	1.1

\* This result takes into account the three most significant tritium sources; it is the annual-average concentration comprising the sum of the three contributions shown in the far right columns.

\*\* At these locations, more than 25% of the samples were below detection limits. The annual mean includes negative concentrations at CAFE, MESQ, MET, and ZON7. MESQ has the lowest percentage of detections (41%).

\*\*\* A tritium surveillance air monitor at the CRED location, which marks the location of the SW-MEI, began operating in July 2003.

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

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

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

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

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

LLNL does not have or store any uranium mill tailings.

#### ***Information on Radon-220 and Radon-222 Emissions***

Radon emissions occur naturally by emanation from the earth. Radon-222 emissions that were reported in past NESHAPs annual reports from research experiments at the Livermore site did not occur in 2003.

## **ATTACHMENT 1. LLNL NESHAPs 2003 Annual Report Spreadsheet**

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

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

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

### ***Radionuclides***

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

### ***Radionuclide Usage Inventories with Potential for Release***

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

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

The physical state factors listed are EPA potential release fractions from 40 CFR 61, Appendix D, whereby emissions are estimated from radionuclide usage inventories

depending on their physical states for use in dispersion/dose assessment modeling. A physical state factor of  $1.0 \times 10^{-6}$  is used for solids,  $1.0 \times 10^{-3}$  is used for liquids and powders, and 1.0 is used for unconfined gases and substances heated above 100°C. Regarding the latter, U.S. EPA has granted LLNL approved alternative emissions factors for elemental uranium, uranium/niobium alloy, and elemental plutonium. (See **Table 6** 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***

Engineering surveys conducted from 1990 through 1992 laid the basis for the stack physical parameters shown in the spreadsheet, which were checked and validated by facility experimenters and managers for 1994 and 1995, and in later years as changes were made. Stack physical parameters for sources evaluated in 2003 were updated, as necessary, by experimenters and managers for those facilities.

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

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

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

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

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

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

Actual emission measurements are the basis for reported emissions from continuously monitored facilities. LLNL facilities that had continuous monitoring systems in 2003 were Buildings 175, 235, 251, 331, 332, 491, and 695 at the Livermore site, and Building 801A at

Site 300, as noted earlier. See the discussion below under “0.1 mrem/y Monitoring Requirement” regarding the use of emissions measurements for monitored sources.

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

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

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

### **0.1 mrem/y Monitoring Requirement**

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

The unabated EDE cannot be calculated for HEPA-filtered facilities monitored for radioactive particles. Because the monitoring equipment is placed after HEPA filtration, there is no way to obtain an estimate for what the emissions might have been had there been no filtration. It is not reasonable to apply factors for the effects of the HEPA filters on the emission rate because most of what is measured on the HEPA filters is the result of the radioactive decay of 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 2003; (2) Unmonitored or non-continuously monitored Livermore site facilities with a previous radionuclide usage inventory update (this category is not used in years with complete usage inventory updates, such as 2000); (3) Continuously monitored Livermore site and Site 300 facilities; (4) Site 300 explosives experiments; (5) Diffuse sources where emissions and subsequent doses were estimated using inventory processes; (6) Diffuse sources where emission and dose estimates were supported by environmental surveillance measurements; and (7) Sources whose emissions estimates and subsequent doses were estimated by confirmatory air sampling rather than continuous sampling.

Attachment 1 - 2003 LLNL NESHAPs Annual Report Spreadsheet

Building	Room/Area	Stack ID	Operation	Radionuclides	Annual Inventory with Potential for Release (Ci)	Physical State Factor	Stack Height (m)	Stack Diameter (m)	Stack Velocity (m/s)	Control Device(s)	Control Device Abatement Factor	Estimated Annual Emissions (Ci)	10 mrem/y Site-Wide Dose Requirement			0.1 mrem/y Monitoring Requirement			Source Category
													Distance to SWMEI (m)	Direction to SWMEI	EDE (mrem)	Distance to MEI (m)	Direction to MEI	Unabated EDE (mrem)	
<b>LIVERMORE SITE POINT SOURCES</b>																			
Building 175 was part of the Uranium Atomic Vapor Laser Isotope Separation (U-AVLIS) program, affiliated with The United States Enrichment Corporation (USEC). In June 1999, USEC suspended further development of the U-AVLIS technology. In May 2003, sampling at Building 175 was discontinued because the facility no longer possessed a radionuclide inventory and there are no plans to conduct activities with radionuclides in the facility in the foreseeable future.																			
*Gross alpha and Gross beta emissions are continuously monitored at the stack.																			
**Because monitoring takes place after HEPA filtration, an unabated EDE cannot be determined (see discussion in Section II, subsection "Stack Monitoring for Gross Alpha and Gross Beta Radiation.")																			
175	103	FFE-02	Operations discontinued	Gross alpha	*	NA	9.4	0.61	4.5	HEPA	1.0E-02	0.0E+00	**	**	0.0E+00	**	**	**	3
	103	FFE-01		Gross beta	*	NA	9.4	0.61	4.6			0.0E+00							
	112	FHE-02					6.8	0.36	6.4										
	112	FHE-01					6.7	0.33	6.4										
	128	FHE-2000					8.9	0.59	4.6										
	128	FHE-1000					8.9	0.59	5.2										
Building 235 is part of the Chemistry and Materials Sciences Directorate. Operations in the facility include examination of material structure, surface, and subsurface; precision cutting, ion implanting, and metallurgical studies.																			
*Gross alpha and Gross beta emissions are continuously monitored at the stack.																			
**Because monitoring takes place after HEPA filtration, an unabated EDE cannot be determined (see discussion in Section II, subsection "Stack Monitoring for Gross Alpha and Gross Beta Radiation.")																			
235	1130	FHE-1A/1B, FHE2A/2B, FGBE-1A/1B	Preparation of plutonium samples for diamond anvil studies	Gross alpha	*	NA	10.7	2.75	4.0	Double HEPA	0.0001	0.0E+00	**	**	0.0E+00	**	**	**	3
				Gross beta	*	NA						0.0E+00							
Building 251, the Heavy Element Facility, is managed by the Safety, Security and Environmental Protection Directorate for the Institutions as a non-operational facility in which transuranic isotopes are stored until they can be disposed. One area of the facility has been "hardened" to resist damage from earthquakes. Room exhausts from this hardened area are double HEPA filtered; glove box exhausts are triple HEPA filtered. Exhausts from the unhardened area, also HEPA filtered, are continuously sampled by simple filter systems.																			
*Stack emissions have been combined as permitted by the EPA/DOE Memorandum of Understanding.																			
**Because monitoring takes place after HEPA filtration, an unabated EDE cannot be determined (see discussion in Section II, subsection "Stack Monitoring for Gross Alpha and Gross Beta Radiation.")																			
	Unhardened Area*																		
251	1003	FHE-5	General chemistry	Gross alpha	*	NA	4.3	0.26	8.6	HEPA	0.01	0.0E+00	1188	E	0.0E+00	**	**	**	3
	1003	FHE-4		Gross beta			4.3	0.27	4.2			0.0E+00							
	1142	FHE-8					4.3	0.32	4.1										
	1142	FHE-9					4.3	0.26	5.1										
	1142	FHE-10					4.3	0.28	13.7										
	1150	FGBE-33,34					8.0	0.15	12.8										
	1150	FFE-15					4.3	0.31	7.6										
	1165	FGBE-31,32					5.5	0.87	0.1										
	1211	FHE-6					6.4	0.25	8.0										
	1211	FHE-7					6.4	0.25	4.3										
	1212	FGBE-15,16					5.5	0.10	8.0										
	1232	FGBE-38,39					7.2	0.15	5.1										
	1234	FFE-9					4.3	0.19	14.7										
	1235	FFE-12					4.3	0.25	7.6										
	1235	FGBE-29,30					5.5	0.13	7.1										
	1363	FHE-12					4.3	0.32	9.1										
	1363	FHE-13					6.4	0.28	6.8										
	1364	FFE-23					4.3	0.34	9.1										
	1364	FGBE-35,36					4.3	0.13	11.2										
	1314, 1354	FGBE-44,45					10.2	0.15	10.2										
	Hot cells	FGBE-40,41					5.5	0.23	5.6										
	Hot cells	FGBE-42,43					5.5	0.36	12.7										
	1150	FFE-13					5.5	0.28	4.1										
	Hardened Area																		
251	Glove Boxes*	FGBE-1000	Previous transuranic research	Gross alpha	*	NA	7.8	0.30	4.8	Triple HEPA	0.000001	0.0E+00	1188	E	0.0E+00	**	**	**	3
		FGBE-2000		Gross beta			7.8	0.30	4.8			0.0E+00							
	Room Exhaust*	FFE-1000		Gross alpha	*	NA	7.8	0.50	11.7	Double HEPA	0.0001	0.0E+00	1188	E	0.0E+00	**	**	**	3
		FFE-2000		Gross beta			7.8	0.50	11.7			0.0E+00							
Building 331 is operated by the Defense and Nuclear Technologies Directorate. The building houses the tritium research facility and associated laboratories.																			
*Tritium HT and HTO emissions from the two 30-m stacks are continuously monitored in compliance with NESHAPs regulations. Monitoring data, rather than the inventory approach, are used to determine emissions.																			
**Stack emissions have been combined as permitted by the EPA/DOE Memorandum of Understanding.																			
***Calculated dose of 2.2E-02 mrem includes modeling the HT emissions as HTO, as directed by U.S. EPA, Region IX. The dose from HT and HTO emissions calculated appropriately using the NEWTRIT model is 1.6E-02. See discussion in Section VIII, subsection "																			
331	All**	Stack 1	Tritium research and development	H-3	*	1.0E+00	30.0	1.22	7.6	None	1	5.8E-01	957	ENE	2.2E-02	957	ENE	2.2E-02	3
		Stack 2	Decontamination of parts	H-3	*	1.0E+00	30.0	1.22	10.5	None	1	1.1E+02			***1.6E-02			***1.6E-02	



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Building	Room/Area	Stack ID	Operation	Radionuclides	Annual Inventory with Potential for Release (Ci)	Physical State Factor	Stack Height (m)	Stack Diameter (m)	Stack Velocity (m/s)	Control Device(s)	Control Device Abatement Factor	Estimated Annual Emissions (Ci)	10 mrem/y Site-Wide Dose Requirement			0.1 mrem/y Monitoring Requirement			Source Category			
													Distance to SWMEI (m)	Direction to SWMEI	EDE (mrem)	Distance to MEI (m)	Direction to MEI	Unabated EDE (mrem)				
514	108	(continued)		Tl-204	7.5E-10	1.0E-03						7.5E-13										
				U-232	8.7E-11	1.0E-03								8.7E-14								
				U-233	8.8E-07	1.0E-03								8.8E-10								
				U-234	8.7E-05	1.0E-03								8.7E-08								
				U-235	1.2E-05	1.0E-03								1.2E-08								
				U-236	9.3E-13	1.0E-03								9.3E-16								
				U-237	5.2E-10	1.0E-03								5.2E-13								
				U-238	1.0E-03	1.0E-03								1.0E-06								
				Zn-65	8.8E-15	1.0E-03					8.8E-18											
514	Evaporator	Room Air	Waste consolidation	Am-241	1.8E-06	1.0E-03	NA	NA	NA	None	1	1.8E-09	528	NE	2.1E-04	217	SW	7.0E-04	1			
				Am-243	1.7E-09	1.0E-03								1.7E-12								
				Ba-133	8.5E-11	1.0E-03								8.5E-14								
				C-14	1.5E-03	1.0E-03								1.5E-06								
				Ce-144	1.6E-15	1.0E-03								1.6E-18								
				Cl-36	3.7E-09	1.0E-03								3.7E-12								
				Co-57	9.3E-15	1.0E-03								9.3E-18								
				Co-60	4.4E-16	1.0E-03								4.4E-19								
				Cr-51	3.3E-13	1.0E-03								3.3E-16								
				Cs-134	1.4E-09	1.0E-03								1.4E-12								
				Cs-137	3.0E-07	1.0E-03								3.0E-10								
				Eu-152	2.6E-16	1.0E-03								2.6E-19								
				Fe-55	3.4E-12	1.0E-03								3.4E-15								
				H-3	3.3E-02	1.0E-03								3.3E-05								
				I-125	9.2E-04	1.0E-03								9.2E-07								
				I-131	9.2E-04	1.0E-03								9.2E-07								
				K-40	2.5E-10	1.0E-03								2.5E-13								
				Mn-54	8.5E-08	1.0E-03								8.5E-11								
				Na-22	1.4E-06	1.0E-03								1.4E-09								
				Ni-63	1.1E-05	1.0E-03								1.1E-08								
				Np-237	3.1E-07	1.0E-03								3.1E-10								
				P-32	9.4E-04	1.0E-03								9.4E-07								
				P-33	9.2E-04	1.0E-03								9.2E-07								
				Pb-210	1.7E-09	1.0E-03								1.7E-12								
				Pm-147	3.4E-12	1.0E-03								3.4E-15								
				Pu-236	1.1E-11	1.0E-03								1.1E-14								
				Pu-238	3.1E-07	1.0E-03								3.1E-10								
				Pu-239	1.2E-06	1.0E-03								1.2E-09								
				Pu-240	3.1E-07	1.0E-03								3.1E-10								
				Pu-241	3.1E-07	1.0E-03								3.1E-10								
				Pu-242	3.2E-07	1.0E-03								3.2E-10								
				Ra-226	2.7E-12	1.0E-03								2.7E-15								
				Ra-228	6.8E-13	1.0E-03								6.8E-16								
				Re-187	6.4E-09	1.0E-03								6.4E-12								
				S-35	9.2E-04	1.0E-03								9.2E-07								
				Se-75	1.1E-05	1.0E-03								1.1E-08								
				Sr-89	3.0E-16	1.0E-03								3.0E-19								
				Sr-90	1.2E-05	1.0E-03								1.2E-08								
				Tc-99	2.8E-07	1.0E-03								2.8E-10								
				Th-228	6.8E-13	1.0E-03								6.8E-16								
				Th-229	2.3E-11	1.0E-03								2.3E-14								
Th-232	1.1E-08	1.0E-03								1.1E-11												
Tl-204	2.7E-10	1.0E-03								2.7E-13												
U-232	3.1E-11	1.0E-03								3.1E-14												
U-233	3.1E-07	1.0E-03								3.1E-10												
U-234	3.1E-05	1.0E-03								3.1E-08												
U-235	4.2E-06	1.0E-03								4.2E-09												
U-236	3.3E-13	1.0E-03								3.3E-16												
U-237	1.8E-10	1.0E-03								1.8E-13												
U-238	3.6E-04	1.0E-03								3.6E-07												
				Zn-65	3.1E-15	1.0E-03					3.1E-18											
612	101	FHE-4	Laboratory analysis of waste treatment and treatability samples	Am-241	1.3E-05	1.0E-03	10.5	0.31	5.6	HEPA	0.01	1.3E-10	444	NE	1.4E-05	384	NE	1.6E-03	1			
				Am-243	1.3E-08	1.0E-03								1.3E-13								
				Ba-133	6.2E-10	1.0E-03								6.2E-15								
				C-14	1.1E-02	1.0E-03								1.1E-07								
				Ce-144	1.2E-14	1.0E-03								1.2E-19								
				Cl-36	2.7E-08	1.0E-03								2.7E-13								
				Co-57	6.8E-14	1.0E-03								6.8E-19								
				Co-60	3.2E-15	1.0E-03								3.2E-20								
				Cr-51	2.4E-12	1.0E-03								2.4E-17								
				Cs-134	1.0E-08	1.0E-03								1.0E-13								
				Cs-137	2.2E-06	1.0E-03								2.2E-11								
				Eu-152	1.9E-15	1.0E-03								1.9E-20								
				Fe-55	2.5E-11	1.0E-03								2.5E-16								
				H-3	2.4E-01	1.0E-03								2.4E-06								
I-125	6.7E-03	1.0E-03								6.7E-08												
				I-131	6.7E-03	1.0E-03					6.7E-08											

NOTE: To convert curies to becquerels use 1 Ci=3.7E+10 Bq and to convert millirem to sieverts use 1 Sv=1.0E+05 mrem.

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Building	Room/Area	Stack ID	Operation	Radionuclides	Annual Inventory with Potential for Release (Ci)	Physical State Factor	Stack Height (m)	Stack Diameter (m)	Stack Velocity (m/s)	Control Device(s)	Control Device Abatement Factor	Estimated Annual Emissions (Ci)	10 mrem/y Site-Wide Dose Requirement			0.1 mrem/y Monitoring Requirement			Source Category						
													Distance to SWMEI (m)	Direction to SWMEI	EDE (mrem)	Distance to MEI (m)	Direction to MEI	Unabated EDE (mrem)							
612	101	(continued)		K-40	1.9E-09	1.0E-03						1.9E-14													
				Mn-54	6.2E-07	1.0E-03								6.2E-12											
				Na-22	1.0E-05	1.0E-03								1.0E-10											
				Ni-63	8.2E-05	1.0E-03								8.2E-10											
				Np-237	2.3E-06	1.0E-03								2.3E-11											
				P-32	6.9E-03	1.0E-03								6.9E-08											
				P-33	6.7E-03	1.0E-03								6.7E-08											
				Pb-210	1.2E-08	1.0E-03								1.2E-13											
				Pm-147	2.5E-11	1.0E-03								2.5E-16											
				Pu-236	7.9E-11	1.0E-03								7.9E-16											
				Pu-238	2.3E-06	1.0E-03								2.3E-11											
				Pu-239	9.1E-06	1.0E-03								9.1E-11											
				Pu-240	2.3E-06	1.0E-03								2.3E-11											
				Pu-241	2.3E-06	1.0E-03								2.3E-11											
				Pu-242	2.3E-06	1.0E-03								2.3E-11											
				Ra-226	2.0E-11	1.0E-03								2.0E-16											
				Ra-228	5.0E-12	1.0E-03								5.0E-17											
				Re-187	4.7E-08	1.0E-03								4.7E-13											
				S-35	6.7E-03	1.0E-03								6.7E-08											
				Se-75	8.2E-05	1.0E-03								8.2E-10											
				Sr-89	2.2E-15	1.0E-03								2.2E-20											
				Sr-90	8.9E-05	1.0E-03								8.9E-10											
				Tc-99	2.0E-06	1.0E-03								2.0E-11											
				Th-228	5.0E-12	1.0E-03								5.0E-17											
				Th-229	1.7E-10	1.0E-03								1.7E-15											
				Th-232	7.9E-08	1.0E-03								7.9E-13											
				Tl-204	1.9E-09	1.0E-03								1.9E-14											
				U-232	2.3E-10	1.0E-03								2.3E-15											
				U-233	2.3E-06	1.0E-03								2.3E-11											
				U-234	2.2E-04	1.0E-03								2.2E-09											
				U-235	3.1E-05	1.0E-03								3.1E-10											
				U-236	2.4E-12	1.0E-03								2.4E-17											
				U-237	1.3E-09	1.0E-03								1.3E-14											
				U-238	2.7E-03	1.0E-03								2.7E-08											
				Zn-65	2.3E-14	1.0E-03								2.3E-19											
				612	102	Room Air	Laboratory analysis of waste treatment and treatability samples	Am-241	1.3E-05	1.0E-03	NA	NA	NA	None	1	1.3E-08	444	NE	1.4E-03	384	NE	1.7E-03	1		
								Am-243	1.3E-08	1.0E-03								1.3E-11							
								Ba-133	6.2E-10	1.0E-03								6.2E-13							
								C-14	1.1E-02	1.0E-03								1.1E-05							
								Ce-144	1.2E-14	1.0E-03								1.2E-17							
								Cl-36	2.7E-08	1.0E-03								2.7E-11							
Co-57	6.8E-14	1.0E-03												6.8E-17											
Co-60	3.2E-15	1.0E-03												3.2E-18											
Cr-51	2.4E-12	1.0E-03												2.4E-15											
Cs-134	1.0E-08	1.0E-03												1.0E-11											
Cs-137	2.2E-06	1.0E-03												2.2E-09											
Eu-152	1.9E-15	1.0E-03												1.9E-18											
Fe-55	2.5E-11	1.0E-03												2.5E-14											
H-3	2.4E-01	1.0E-03												2.4E-04											
I-125	6.7E-03	1.0E-03												6.7E-06											
I-131	6.7E-03	1.0E-03												6.7E-06											
K-40	1.9E-09	1.0E-03												1.9E-12											
Mn-54	6.2E-07	1.0E-03												6.2E-10											
Na-22	1.0E-05	1.0E-03												1.0E-08											
Ni-63	8.2E-05	1.0E-03												8.2E-08											
Np-237	2.3E-06	1.0E-03												2.3E-09											
P-32	6.9E-03	1.0E-03												6.9E-06											
P-33	6.7E-03	1.0E-03												6.7E-06											
Pb-210	1.2E-08	1.0E-03												1.2E-11											
Pm-147	2.5E-11	1.0E-03												2.5E-14											
Pu-236	7.9E-11	1.0E-03												7.9E-14											
Pu-238	2.3E-06	1.0E-03												2.3E-09											
Pu-239	9.1E-06	1.0E-03												9.1E-09											
Pu-240	2.3E-06	1.0E-03												2.3E-09											
Pu-241	2.3E-06	1.0E-03												2.3E-09											
Pu-242	2.3E-06	1.0E-03												2.3E-09											
Ra-226	2.0E-11	1.0E-03												2.0E-14											
Ra-228	5.0E-12	1.0E-03												5.0E-15											
Re-187	4.7E-08	1.0E-03								4.7E-11															
S-35	6.7E-03	1.0E-03								6.7E-06															
Se-75	8.2E-05	1.0E-03								8.2E-08															
Sr-89	2.2E-15	1.0E-03								2.2E-18															
Sr-90	8.9E-05	1.0E-03								8.9E-08															
Tc-99	2.0E-06	1.0E-03								2.0E-09															
Th-228	5.0E-12	1.0E-03								5.0E-15															
Th-229	1.7E-10	1.0E-03								1.7E-13															
Th-232	7.9E-08	1.0E-03								7.9E-11															

NOTE: To convert curies to becquerels use 1 Ci=3.7E+10 Bq and to convert millirem to sieverts use 1 Sv=1.0E+05 mrem.

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Building	Room/Area	Stack ID	Operation	Radionuclides	Annual Inventory with Potential for Release (Ci)	Physical State Factor	Stack Height (m)	Stack Diameter (m)	Stack Velocity (m/s)	Control Device(s)	Control Device Abatement Factor	Estimated Annual Emissions (Ci)	10 mrem/y Site-Wide Dose Requirement			0.1 mrem/y Monitoring Requirement			Source Category
													Distance to SWMEI (m)	Direction to SWMEI	EDE (mrem)	Distance to MEI (m)	Direction to MEI	Unabated EDE (mrem)	
612	102	(continued)		Tl-204	1.9E-09	1.0E-03						1.9E-12							
				U-232	2.3E-10	1.0E-03						2.3E-13							
				U-233	2.3E-06	1.0E-03						2.3E-09							
				U-234	2.2E-04	1.0E-03						2.2E-07							
				U-235	3.1E-05	1.0E-03						3.1E-08							
				U-236	2.4E-12	1.0E-03						2.4E-15							
				U-237	1.3E-09	1.0E-03						1.3E-12							
				U-238	2.7E-03	1.0E-03						2.7E-06							
				Zn-65	2.3E-14	1.0E-03						2.3E-17							
Building 695/696 is the Decontamination Waste Treatment Facility operated by Radiological and Hazardous Waste Management Division. It began operations in 2003.																			
*Gross alpha and Gross beta emissions are continuously monitored at the stack.																			
**Because monitoring takes place after HEPA filtration, an unabated EDE cannot be determined (see discussion in Section II, subsection "Stack Monitoring for Gross Alpha and Gross Beta Radiation.")																			
All operations are HEPA filtered and have pre-filters in place; some operations have additional HEPA filtration.																			
695/696	DWTF	FHE 1000/2000/3000	Waste treatment	Gross alpha	*	NA	20.0	1.98	10.9	HEPA	0.01	0.0E+00	**	**	0.0E+00	**	**	**	3
				Gross beta	*	NA				Pre-filter	0.1	0.0E+00							
<b>SITE 300 POINT SOURCES</b>																			
Building 801 is the Contained Firing Facility, where explosives tests are conducted.																			
*Gross alpha and gross beta emissions are continuously monitored at the stack.																			
**Except for high-bay exhaust that is not HEPA-filtered, monitoring takes place after HEPA filtration, and an unabated EDE cannot be determined (see discussion in Section II, subsection "Stack Monitoring for Gross Alpha and Gross Beta Radiation.")																			
801	Contained Firing Facility	FEFH-1, FE-2	Explosive tests	U-238	*	NA	16.8	1.60	9.4	HEPA	0.01	4.6E-07	3770	S	1.3E-06	**	**	**	3
				U-235	*	NA				Pre-filter	0.1	5.9E-09							
				U-234								4.3E-08							
Explosives tests in which radionuclides may be present are conducted on open-air firing tables located at Bunker 851. These tests have depleted uranium material as part of the material inventory. There are multiple tests per year.																			
851	Firing Table	None	Explosive tests	U-238	2.0E-02	1.0E+00	NA	NA	NA	None	1	2.0E-02	3170	SSE	1.7E-02	1285	W	2.7E-02	4
				U-235	2.6E-04	1.0E+00						2.6E-04			3836	ENE			
				U-234	1.9E-03	1.0E+00						1.9E-03							
<b>LIVERMORE SITE DIFFUSE SOURCES</b>																			
Building 331 - As part of D&D operations, contaminated equipment outside the facility is awaiting transport and storage by Radioactive and Hazardous Waste Management.																			
***The dose from HTO emissions calculated using the NEWTRIT model; see discussion in Section VIII, subsection "Modeling Dose from Tritium."																			
331	Outside	None	Storage of contaminated parts	H-3	NA	1	NA	NA	NA	None	1	8.7E+00	957	ENE	5.9E-03	441	SSW	2.9E-02	6
															***4.4E-03			***2.2E-02	
Building 514 is operated by the Radioactive and Hazardous Waste Management Division. The wastewater treatment tank farm and storage tank area processes the liquid waste from facilities on site. The treatment process may involve batch chemical treatment consisting of neutralization, flocculation, oxidation, reduction, precipitation, separation, and filtration. Areas used for storage are not considered to release radionuclides because the wastes are fully contained.																			
514	Tank Farm	Area Source	Process liquid hazardous mixed and radioactive wastes in open topped tanks.	Am-241	5.0E-06	1.0E-03	NA	NA	NA	None	1	5.0E-09	528	NE	5.9E-04	217	SW	2.3E-03	5
				Am-243	4.9E-09	1.0E-03						4.9E-12							
				Ba-133	2.4E-10	1.0E-03						2.4E-13							
				C-14	4.3E-03	1.0E-03						4.3E-06							
				Ce-144	4.7E-15	1.0E-03						4.7E-18							
				Cl-36	1.1E-08	1.0E-03						1.1E-11							
				Co-57	2.7E-14	1.0E-03						2.7E-17							
				Co-60	1.3E-15	1.0E-03						1.3E-18							
				Cr-51	9.3E-13	1.0E-03						9.3E-16							
				Cs-134	3.9E-09	1.0E-03						3.9E-12							
				Cs-137	8.6E-07	1.0E-03						8.6E-10							
				Eu-152	7.4E-16	1.0E-03						7.4E-19							
				Fe-55	9.7E-12	1.0E-03						9.7E-15							
				H-3	9.3E-02	1.0E-03						9.3E-05							
				I-125	2.6E-03	1.0E-03						2.6E-06							
				I-131	2.6E-03	1.0E-03						2.6E-06							
				K-40	7.2E-10	1.0E-03						7.2E-13							
				Mn-54	2.4E-07	1.0E-03						2.4E-10							
				Na-22	3.9E-06	1.0E-03						3.9E-09							
				Ni-63	3.2E-05	1.0E-03						3.2E-08							
				Np-237	8.8E-07	1.0E-03						8.8E-10							
				P-32	2.7E-03	1.0E-03						2.7E-06							
				P-33	2.6E-03	1.0E-03						2.6E-06							
				Pb-210	4.8E-09	1.0E-03						4.8E-12							
				Pm-147	9.7E-12	1.0E-03						9.7E-15							
				Pu-236	3.1E-11	1.0E-03						3.1E-14							
				Pu-238	8.8E-07	1.0E-03						8.8E-10							
				Pu-239	3.5E-06	1.0E-03						3.5E-09							
				Pu-240	8.8E-07	1.0E-03						8.8E-10							
				Pu-241	8.9E-07	1.0E-03						8.9E-10							
				Pu-242	9.0E-07	1.0E-03						9.0E-10							
				Ra-226	7.8E-12	1.0E-03						7.8E-15							

NOTE: To convert curies to becquerels use 1 Ci=3.7E+10 Bq and to convert millirem to sieverts use 1 Sv=1.0E+05 mrem.

Attachment 1 - 2003 LLNL NESHAPs Annual Report Spreadsheet

Building	Room/Area	Stack ID	Operation	Radionuclides	Annual Inventory with Potential for Release (Ci)	Physical State Factor	Stack Height (m)	Stack Diameter (m)	Stack Velocity (m/s)	Control Device(s)	Control Device Abatement Factor	Estimated Annual Emissions (Ci)	10 mrem/y Site-Wide Dose Requirement			0.1 mrem/y Monitoring Requirement			Source Category	
													Distance to SWMEI (m)	Direction to SWMEI	EDE (mrem)	Distance to MEI (m)	Direction to MEI	Unabated EDE (mrem)		
514	Tank Farm	(continued)		Ra-228	1.9E-12	1.0E-03						1.9E-15								
			Re-187	1.8E-08	1.0E-03								1.8E-11							
			S-35	2.6E-03	1.0E-03								2.6E-06							
			Se-75	3.2E-05	1.0E-03								3.2E-08							
			Sr-89	8.4E-16	1.0E-03								8.4E-19							
			Sr-90	3.4E-05	1.0E-03								3.4E-08							
			Tc-99	7.9E-07	1.0E-03								7.9E-10							
			Th-228	1.9E-12	1.0E-03								1.9E-15							
			Th-229	6.4E-11	1.0E-03								6.4E-14							
			Th-232	3.1E-08	1.0E-03								3.1E-11							
			Tl-204	7.5E-10	1.0E-03								7.5E-13							
			U-232	8.7E-11	1.0E-03								8.7E-14							
			U-233	8.8E-07	1.0E-03								8.8E-10							
			U-234	8.7E-05	1.0E-03								8.7E-08							
			U-235	1.2E-05	1.0E-03								1.2E-08							
			U-236	9.3E-13	1.0E-03								9.3E-16							
			U-237	5.2E-10	1.0E-03								5.2E-13							
			U-238	1.0E-03	1.0E-03								1.0E-06							
Zn-65	8.8E-15	1.0E-03								8.8E-18										
The Building 612 Yard is operated by the Radioactive and Hazardous Waste Management Division. The Yard consists of several areas where containers having radioactive wastes are stacked outdoors. The containers, which are not air tight, can outgas tritium.																				
*The drum sampling operation takes place at all site Waste Accumulation Areas. Inventories were combined and modeled as if the operation occurred at the center of the site.																				
***The dose from HTO emissions calculated using the NEWTRIT model; see discussion in Section VIII, subsection "Modeling Dose from Tritium."																				
612	Yard	Area Source	Storage of low level waste	H-3	NA	NA	NA	NA	NA	None	1	3.4E+00	444	NE	1.3E-02	212	SSW	4.4E-02	6	
															***9.9E-03			***3.3E-02		
The Southeast Quadrant of the Livermore Site has slightly elevated levels of Pu-239 in the surface soil and air (presumably from resuspension). The source of the Pu-239 was past waste management operations.																				
Southeast Quadrant		Area Source	Resuspension	Pu-239	NA	NA	NA	NA	NA	None	1	NA	0	NA	4.7E-04	NA	NA	NA	6	
<b>SITE 300 DIFFUSE SOURCES</b>																				
Diffuse sources consist of resuspension of depleted uranium and waste handling.																				
Site 300	All	Area Source	Soil resuspension	U-238	NA	NA	NA	NA	NA	None	1	NA	NA	NA	3.3E-03	NA	NA	NA	6	
				U-235	NA	NA	NA	NA	NA			NA								
				U-234	NA	NA	NA	NA	NA			NA								
<b>EMISSION SOURCES THAT ACCOUNT FOR MORE THAN 90% OF THE POTENTIAL EFFECTIVE DOSE EQUIVALENT AT EACH SITE.</b>																				
<b>LIVERMORE SITE SOURCES</b>																				
612	Yard	Area Source	Storage of low level waste	H-3	NA	NA	NA	NA	NA	None	1	3.4E+00	444	NE	1.3E-02	212	SSW	4.4E-02	6	
331	All	Stack 1	Tritium research and development	H-3	*	1	30	1.22	7.59	None	1	5.8E-01	957	ENE	2.2E-02	957	ENE	2.2E-02	3	
		Stack 2	Decontamination of parts	H-3	*	1	30	1.22	10.5	None	1	1.1E+02			***1.6E-02			***1.6E-02		
612	102	Room Air	Laboratory analysis	Various nuclides	1.3E-05	1.0E-03	NA	NA	NA	None	1	1.3E-08	444	NE	1.4E-03	384	NE	1.7E-03	1	
331	Outside	None	Storage of contaminated parts	H-3	NA	1	NA	NA	NA	None	1	8.7E+00	957	ENE	5.9E-03	441	SSW	0.029	6	
															***4.4E-03			***2.2E-02		
<b>SITE 300 SOURCES</b>																				
851	Firing Table	None	Explosive tests	U-238	2.0E-02	1	NA	NA	NA	None	1	2.0E-02	3170	SSE	1.7E-02	1285	W	2.7E-02	4	
				U-235	2.6E-04	1							3.1E-04							
				U-234	1.9E-03	1							2.3E-03							
Site 300	All	Area Source	Soil resuspension	U-238	NA	NA	NA	NA	NA	None	1	NA	NA	NA	3.3E-03	NA	NA	NA	6	
				U-235	NA	NA	NA	NA	NA			NA								
				U-234	NA	NA	NA	NA	NA			NA								

NOTE: To convert curies to becquerels use 1 Ci=3.7E+10 Bq and to convert millirem to sieverts use 1 Sv=1.0E+05 mrem.

## ATTACHMENT 2. Surrogate Radionuclides List

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

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

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

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

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

**Table 2-1.** List of surrogate radionuclides.

Isotope	Half-Life	Lung Class <sup>a</sup>	ALI (inh) $\mu\text{Ci}$	DAC (inh) $\mu\text{Ci}/\text{m}^3$	Surrogate	Half-Life	Lung Class <sup>a</sup>	ALI (inh) $\mu\text{Ci}$	DAC (inh) $\mu\text{Ci}/\text{m}^3$
<b>Ca-108m</b>	127 y	Y	$2.0 \times 10^1$	$1.0 \times 10^{-8}$	<b>Co-60</b>	5.271 y	Y	$3.0 \times 10^1$	$1.0 \times 10^{-8}$
<b>Bi-207</b>	38 y	W	$4.0 \times 10^2$	$1.0 \times 10^{-7}$	<b>Bi-214</b>	19.9 min	W	$9.0 \times 10^2$	$4.0 \times 10^{-7}$
<b>Ca-45</b>	163 d	W	$8.0 \times 10^2$	$4.0 \times 10^{-7}$	<b>Sr-90</b>	29.12 y	D	$2.0 \times 10^1$	$8.0 \times 10^{-9}$
<b>Cd-109</b>	464 d	Y	$1.0 \times 10^2$	$5.0 \times 10^{-8}$	<b>Co-60</b>	5.271 y	Y	$3.0 \times 10^1$	$1.0 \times 10^{-8}$
<b>Cf-249</b>	350.6 y	Y	$1.0 \times 10^{-2}$	$4.0 \times 10^{-12}$	<b>Cm-245</b>	8500 y	W	$6.0 \times 10^{-3}$	$3.0 \times 10^{-12}$
<b>Cf-250</b>	13.1 y	W	$9.0 \times 10^{-3}$	$4.0 \times 10^{-12}$	<b>Am-241</b>	432.2 y	W	$6.0 \times 10^{-3}$	$3.0 \times 10^{-12}$
<b>Cl-36</b>	$3.01 \times 10^5$ y	W	$2.0 \times 10^2$	$1.0 \times 10^{-7}$	<b>Cs-137</b>	30 y	D	$2.0 \times 10^2$	$6.0 \times 10^{-8}$
<b>Es-254</b>	275.7 d	W	$7.0 \times 10^{-2}$	$3.0 \times 10^{-11}$	<b>Pu-239</b>	24065 y	Y	$2.0 \times 10^{-2}$	$7.0 \times 10^{-12}$
<b>Eu-149</b>	93.1 d	W	$3.0 \times 10^3$	$1.0 \times 10^{-6}$	<b>Pm-151</b>	28.4 hr	Y	$3.0 \times 10^3$	$1.0 \times 10^{-6}$
<b>Gd-148</b>	93 y	D	$8.0 \times 10^{-3}$	$3.0 \times 10^{-12}$	<b>La-140</b>	40.272 h	W	$1.0 \times 10^3$	$5.0 \times 10^{-7}$
<b>Os-185</b>	94 d	D	$5.0 \times 10^2$	$2.0 \times 10^{-7}$	<b>Mo-99</b>	66 h	Y	$1.0 \times 10^3$	$6.0 \times 10^{-7}$
<b>P-33</b>	25.4 d	W	$3.0 \times 10^3$	$1.0 \times 10^{-6}$	<b>P-32</b>	14.29 d	D	$9.0 \times 10^2$	$4.0 \times 10^{-7}$
<b>Re-184</b>	38 d	W	$1.0 \times 10^3$	$6.0 \times 10^{-7}$	<b>Mo-99</b>	66 h	Y	$1.0 \times 10^3$	$6.0 \times 10^{-7}$
<b>Se-75</b>	119.8 d	W	$6.0 \times 10^2$	$3.0 \times 10^{-7}$	<b>As-76</b>	26.32 h	W	$1.0 \times 10^3$	$6.0 \times 10^{-7}$
<b>Sr-85</b>	64.8 d	D	$3.0 \times 10^3$	$1.0 \times 10^{-6}$	<b>Sr-90</b>	29.12 y	D	$2.0 \times 10^1$	$8.0 \times 10^{-9}$
<b>Ta-182</b>	115 d	Y	$1.0 \times 10^2$	$6.0 \times 10^{-8}$	<b>Hf-181</b>	42.4 d	W	$4.0 \times 10^2$	$2.0 \times 10^{-7}$
<b>Tb-157</b>	110 y	W	$3.0 \times 10^2$	$1.0 \times 10^{-7}$	<b>La-140</b>	40.272 h	W	$1.0 \times 10^3$	$5.0 \times 10^{-7}$
<b>Tb-158</b>	180 y	W	$2.0 \times 10^1$	$8.0 \times 10^{-9}$	<b>La-140</b>	40.272 h	W	$1.0 \times 10^3$	$5.0 \times 10^{-7}$
<b>Tl-204</b>	3.78 y	D	$2.0 \times 10^3$	$9.0 \times 10^{-7}$	<b>Pb-214</b>	26.8 min	D	$8.0 \times 10^2$	$3.0 \times 10^{-7}$
<b>Tm-168</b>	93.1 d	W	$2.0 \times 10^3$	$8.0 \times 10^{-7}$	<b>La-140</b>	40.272 h	W	$1.0 \times 10^3$	$5.0 \times 10^{-7}$
<b>Tm-171</b>	1.92 y	Y	$3.0 \times 10^2$	$1.0 \times 10^{-7}$	<b>La-140</b>	40.272 h	W	$1.0 \times 10^3$	$5.0 \times 10^{-7}$
<b>Y-88</b>	106.64 d	Y	$2.0 \times 10^2$	$1.0 \times 10^{-7}$	<b>Y-90</b>	64 h	Y	$6.0 \times 10^2$	$3.0 \times 10^{-7}$
<b>Am-244</b>	10.1 h	W	$2.0 \times 10^2$	$8.0 \times 10^{-8}$	<b>Cm-244</b>	18.11 y	W	$1.0 \times 10^{-2}$	$5.0 \times 10^{-12}$
<b>Au-195</b>	183 d	Y	$4.0 \times 10^2$	$2.0 \times 10^{-7}$	<b>Ba-133</b>	10.74 y	D	$7.0 \times 10^2$	$3.0 \times 10^{-7}$
<b>Co-56</b>	78.76 d	Y	$2.0 \times 10^2$	$8.0 \times 10^{-8}$	<b>Co-60</b>	5.271 y	Y	$3.0 \times 10^1$	$1.0 \times 10^{-8}$
<b>Gd-146</b>	48.3 d	W	$3.0 \times 10^2$	$1.0 \times 10^{-7}$	<b>Sm-147</b>	$1.06 \times 10^{11}$ y	W	$4.0 \times 10^{-2}$	$2.0 \times 10^{-11}$
<b>Kr-85</b>	10.72 y	Gas	See Note	$1.0 \times 10^{-4}$					
<b>Rh-102</b>	2.9 y	Y	$6.0 \times 10^1$	$2.0 \times 10^{-8}$	<b>Rh-106m</b>	29.9 s	Y	$4.0 \times 10^4$	$1.0 \times 10^{-5}$
<b>U-239</b>	23.54 min	Y	$2.0 \times 10^5$	$6.0 \times 10^{-5}$	<b>U-240</b>	14.1 h	Y	$2.0 \times 10^3$	$1.0 \times 10^{-6}$
<b>Zr-90</b>	809 ms	W	N/A	N/A	<b>Y-90</b>	64 h	Y	$6.0 \times 10^2$	$3.0 \times 10^{-7}$
<b>Po-209<sup>b</sup></b>	102 y	N/A	N/A	N/A	<b>Pu-239</b>	24065 y	Y	$2.0 \times 10^{-2}$	$7.0 \times 10^{-12}$

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

<sup>a</sup> D = days, W = weeks, Y = years.

<sup>b</sup> No ALI or DAC information available. Pu-239 used to provide a conservative alpha-emitter dose.

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