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Lawrence Livermore National Laboratory performs continuous air sampling to evaluate its compliance with local, state, and federal laws and regulations and to ensure that human health and the environment are protected. Federal environmental air quality laws and U.S. DOE regulations include 40 CFR 61, Subpart H—the NESHAPs section of the Clean Air Act; applicable portions of DOE Order 5400.5; and ANSI standards. The *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (U.S. DOE 1991) provides the guidance for implementing DOE Order 5400.5.

The EPA Region IX has enforcement authority for LLNL compliance with radiological air emission regulations. Enforcement authority for the Clean Air Act regulations pertaining to nonradiological air emissions belongs to two local air districts: the BAAQMD and the SJVAPCD.

# 4.1 Air Effluent Monitoring

Air effluent monitoring of atmospheric discharge points is in place for compliance with 40 CFR 61, Subpart H and is used to determine the actual radionuclide releases from individual facilities during routine and nonroutine operations and to confirm the operation of facility emission control systems. Subpart H requires continuous monitoring of facility radiological air effluents if the potential off-site (fence-line) dose equivalent is greater than 1  $\mu$ Sv/y (0.1 mrem/y), as calculated using the EPA-mandated air dispersion dose model, CAP88-PC, without credit for emission control devices. The results of monitoring air discharge points provide the actual emission source information for modeling, which is used to ensure that the NESHAPs standard of 100  $\mu$ Sv/y (10 mrem/y) total site effective dose equivalent from the airborne pathway is not exceeded. See **Chapter 7** for further information on radiological dose assessment.

Currently, the air effluent sampling program measures only radiological emissions. For LLNL operations with nonradiological discharges, LLNL obtains permits from local air districts (i.e., BAAQMD and SJVAPCD) for stationary emission sources, and from the CARB for portable emission sources such as diesel air compressors and generators. Current permits do not require monitoring of air effluent but do require monitoring of equipment usage, material usage, and record keeping during operations. Based on air toxics emissions inventory and risk assessment required by the California Air Toxics "Hot Spots" Information and Assessment Act of 1987, BAAQMD and SJVAPCD have ranked LLNL as a low-risk facility for nonradiological air emissions.

#### 4.1.1 Air Effluent Radiological Monitoring Results and Impact on the Environment

In 2008, LLNL measured releases of radioactivity from air exhausts at five facilities at the Livermore site and at one facility at Site 300. Air effluent monitoring locations at the Livermore site and Site 300 are shown in **Figures 4-1** and **4-2**, respectively.

In 2008, a total of 137 GBq (3.7 Ci) of measured tritium was released from the Tritium Facility. Of this, approximately 93 GBq (2.5 Ci) of tritium was released as vapor (HTO). The remaining tritium released, 44 GBq (1.2 Ci), was gaseous tritium (HT).

During a DOE/NNSA Livermore Site Office walkthrough in May 2008, it was observed that the glycol bubbler from Stack 2 was not correctly assembled. It is believed that a construction project in February 2008 was the cause. There were no activities with tritium during the time frame the bubbler was offline and any tritium emissions to the environment from Stack 2 would be at outgassing levels. A conservative estimate of 3.7 GBq (0.1 Ci) of tritium in the vapor state is being applied. The estimate is based on an average of previous known outgassing results from Stack 2. The total tritium stack emissions from the facility for 2008 (measured plus estimate) were 141 GBq (3.8 Ci).

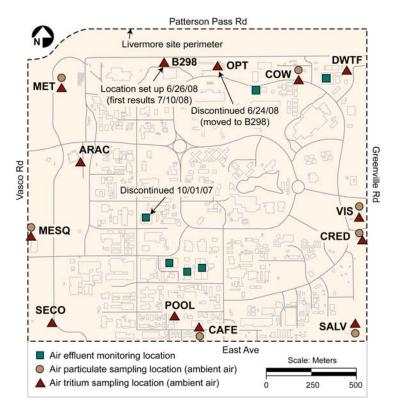
In 2008, a total of 3.2 GBq ( $8.6 \times 10^{-2}$  Ci) of measured tritium was released from the DWTF as HTO. Tritium in the gaseous state (HT) was not detected from the stack effluent in 2008. The emissions were a result of planned activities with tritium

The Contained Firing Facility (CFF) at Site 300 had measured depleted uranium emissions in 2008. A total of  $6.7 \times 10^{-8}$  GBq ( $1.8 \times 10^{-9}$  Ci) of uranium-234,  $5.9 \times 10^{-9}$  GBq ( $1.6 \times 10^{-10}$  Ci) of uranium-235, and  $4.8 \times 10^{-7}$  GBq ( $1.3 \times 10^{-8}$  Ci) of uranium-238 was released in particulate form. The emissions were a result of planned activities with depleted uranium.

None of the other facilities monitored for radionuclides had reportable emissions in 2008. The data tables in **Appendix A**, **Section A.1** provide summary results of all air effluent monitored facilities and include upwind locations (control stations) for gross alpha and gross beta background comparison to stack effluent gross alpha and gross beta results.

The dose to the hypothetical, site-wide maximally exposed individual (SW-MEI) member of the public caused by the measured air emissions from the Tritium Facility (modeling HT emissions as HTO as required by EPA) was  $3.3 \times 10^{-3}$  µSv/y ( $3.3 \times 10^{-4}$  mrem/y); the dose from the DWTF (modeling HT emissions as HTO) was  $3.3 \times 10^{-5}$  µSv/y ( $3.3 \times 10^{-6}$  mrem/y); and the dose from the CFF was  $4.4 \times 10^{-7}$  µSv/y ( $4.4 \times 10^{-8}$  mrem/y).

All of the reported SW-MEI doses at the Livermore site and Site 300 are less than one-tenth of one percent of the annual NESHAPs standard, which is  $100 \,\mu\text{Sv/y}$  ( $10 \,\text{mrem/y}$ ) total site effective dose equivalent. As shown in **Chapter 7**, the estimated radiological dose caused by measured air emissions from LLNL operations was minimal. See also the *LLNL NESHAPs 2008 Annual Report* (Bertoldo et al. 2009) for a complete description of air effluent monitoring.



**Figure 4-1.** Air effluent and ambient air monitoring locations at the Livermore site, 2008.

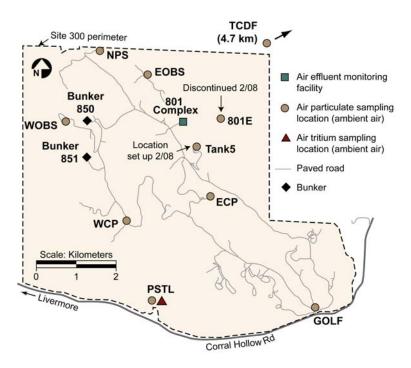


Figure 4-2. Air effluent and ambient air monitoring locations at Site 300, 2008.

#### 4.1.2 Nonradiological Air Releases and Impact on the Environment

In 2008, the Livermore site emitted approximately 133 kg/d of regulated air pollutants as defined by the Clean Air Act, including nitrous oxides (NOx), sulphur oxides (SOx), particulate matter (PM-10), carbon monoxide (CO), and reactive organic gases/precursor organic compounds (ROGs/POCs) (see **Table 4-1**). The stationary emission sources that released the greatest amount of regulated pollutants at the Livermore site were natural gas fired boilers, internal combustion engines (such as diesel generators), solvent cleaning, and surface coating operations (such as painting). Pollutant emission information was primarily derived from monthly material and equipment usage records.

<b>Table 4-1.</b> Nonradioactive air emissions, Livermore site	: and Site 300, 200	8.
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	Estimated releases (kg/d)			
Pollutant	Livermore site	Site 300		
ROGs/POCs	13.3	0.42		
Nitrogen oxides	63.0	2.32		
Carbon monoxide	49.0	0.51		
Particulates (PM-10)	5.8	0.49		
Sulfur oxides	1.7	0.21		
Total	132.8	3.95		

Livermore site air pollutant emissions were very low in 2008 compared to the daily releases of air pollutants from all sources in the entire Bay Area. For example, the average daily emission of NOx in the Bay Area was approximately  $4.32 \times 10^5$  kg/d, compared to the estimated daily release from the Livermore site of 63.0 kg/d, which is 0.015% of total Bay Area source emissions for NOx. The 2008 BAAQMD estimate for ROGs/POCs daily emissions throughout the Bay Area was  $3.21 \times 10^5$  kg/d, while the daily emission estimate for 2008 from the Livermore site was 13.3 kg/d, or 0.004% of the total Bay Area source emissions for ROGs/POCs.

Certain operations at Site 300 require permits from the SJVAPCD. The estimated daily air pollutant emissions during 2008 from operations (permitted and exempt stationary sources) at Site 300 are listed in **Table 4-1**. The stationary emission sources that release the greatest amounts of regulated air pollutants at Site 300 include internal combustion engines (such as diesel generators), a gasoline-dispensing facility, and general machine shop operations. Combustion pollutant emissions, such as NOx, CO, SOx, and PM-10, increased at Site 300 in 2008 primarily from the required, periodic preventative maintenance of emergency stand-by diesel generators and generator operation during emergency power outages.

# 4.2 Ambient Air Monitoring

LLNL conducts ambient air monitoring at on- and off-site locations to determine whether airborne radionuclides or beryllium are being released to the environs in measurable quantities by LLNL operations. Ambient air monitoring also serves to verify the air concentrations predicted by air dispersion modeling and to determine compliance with NESHAPs regulations.

The derived concentration guides (DCGs) in DOE Order 5400.5 specify the concentrations of a radionuclide that can be inhaled continuously 365 days a year without exceeding the DOE primary radiation protection standard for the public, which is 1 mSv/y (100 mrem/y) effective dose equivalent.

Beryllium is the only nonradiological emission from LLNL that is monitored in ambient air. LLNL requested and was granted a waiver by the BAAQMD for source-specific monitoring and record keeping for beryllium operations, provided that LLNL can demonstrate that monthly average beryllium concentrations in air are well below regulatory limits of 10,000 pg/m<sup>3</sup>. LLNL meets this requirement by sampling for beryllium at perimeter locations.

Based on air dispersion modeling using site-specific meteorological data, the ambient air samplers, particularly those on the site perimeters, have been placed to monitor locations where elevated air concentrations due to LLNL operations may occur. Sampling locations for each monitoring network are shown in **Figures 4-1**, **4-2**, and **4-3**.

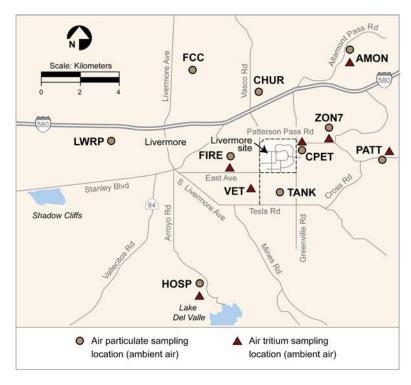


Figure 4-3. Air particulate and tritium monitoring locations in the Livermore Valley, 2008.

#### 4.2.1 Ambient Air Radioactive Particulates

By analyzing air samples for gamma-emitting radionuclides, LLNL verifies that there is no evidence of release of the small inventories of mixed-fission products and radiochemical tracers used by LLNL. Composite samples for the Livermore site and Site 300 were analyzed for an environmental suite of gamma-emitting radionuclide concentrations in air that include fission products, activation products, activities, and naturally occurring products. The isotopes detected at both sites in 2008 were beryllium-7 (cosmogenic), lead-210, radium-226, and potassium-40, all of which are naturally occurring in the environment.

Plutonium-239+240 was detected in 21 out of 216 samples taken in 2008. The highest values and percentage of the DCG were as follows:

- Livermore site perimeter: 17 nBg/m<sup>3</sup> (0.46 aCi/m<sup>3</sup>); 0.0023% of the DCG
- Livermore off-site locations: 56 nBq/m<sup>3</sup> (1.5 aCi/m<sup>3</sup>); 0.0076% of the DCG
- Site 300 composite: 6.3 nBq/m<sup>3</sup> (0.17 aCi/m<sup>3</sup>); 0.00085% of the DCG

The plutonium-239+240 detection at Site 300 is calculated to be from resuspended fallout from historic aboveground nuclear testing. Site 300 does not use or store plutonium on-site.

Uranium-235 and uranium-238 were detected at all sample locations. Uranium ratios are used to determine the type of uranium present in the environment. Natural uranium has a mathematical uranium-235/uranium-238 ratio of 0.00725, and depleted uranium has a uranium-235/uranium-238 ratio of 0.002. Uranium isotopes are naturally occurring. The annual median uranium-235/uranium-238 isotopic ratios for 2008 were as follows:

• Livermore site perimeter composite: 0.0073

• Site 300 sample locations: 0.0073

• Site 300 off-site location: 0.0073

The annual uranium-235/uranium-238 isotopic ratio medians are consistent with naturally occurring uranium. All of the individual uranium-235 and uranium-238 results were less than one-tenth of one percent of the DCG as shown in **Appendix A**, **Section A.2**.

Gross alpha and gross beta were sampled for at all locations. The primary sources of alpha and beta activities are naturally occurring radioisotopes. Routine isotopic gamma results indicate the activities are the result of naturally occurring isotopes (uranium, thorium, potassium, and lead), which are also routinely found in local soils. See **Appendix A, Section A.2**.

# 4.2.2 Ambient Air Tritium Concentrations

The biweekly air tritium data that are provided in **Appendix A**, <u>Section A.2</u> are summarized in **Table 4.2**. Area (diffuse) sources include stored containers of tritium waste or tritium-contaminated equipment from which HTO diffuses into the atmosphere.

Due to the very low HTO air concentrations observed at the Livermore site sample locations, where concentrations are expected to be greater than those at remote locations at greater distances, the remote sample locations are readily predicted to be below the minimum detectable concentration (MDC). However, some samples from these remote locations yielded results greater than the MDC. These results are attributed to the inability to discriminate between a true signal and a background signal in the observed data.

**Table 4-2.** Air tritium sampling summary for 2008.

Detection Sampling locations frequency	Detection	Concentration n (mBq/m³)				Median	Dose
	Mean	Median	IQR	Maximum	% DCG <sup>(a)</sup>	(nSv)	
Livermore site perimeter	176 of 307	31.9	18.1	25.4	1050	0.00049%	6.72
Livermore Valley	46 of 181	7.2	7.25	17.2	48.5	0.0002%	1.52
Site 300	7 of 24	3.31	1.32	17.2	31.9	0.000036%	0.697

<sup>(</sup>a) DCG = derived concentration guide of  $3.7 \times 10^6$  mBq/m<sup>3</sup> for tritium in air.

For a location at which the mean concentration is at or below the MDC, inhalation dose from tritium is assumed to be less than 5 nSv/y (0.5  $\mu$ rem/y) (i.e., the annual dose from inhaling air with a concentration at the MDC of about 25 mBq/m<sup>3</sup> [0.675 pCi/m<sup>3</sup>]).

#### 4.2.3 Ambient Air Beryllium Concentrations

LLNL measures the monthly concentrations of airborne beryllium at the Livermore site, Site 300, and at the off-site sampler northeast of Site 300. The highest value recorded at the Livermore site perimeter in 2008 for airborne beryllium was 69 pg/m³. This value is only 0.69% of the BAAQMD ambient concentration limit for beryllium (10,000 pg/m³). There is no regulatory requirement to monitor beryllium in San Joaquin County; however, LLNL analyzes samples from three Site 300 perimeter locations as a best management practice. The highest value recorded at the Site 300 perimeter in 2008 was 81 pg/m³ and the highest value at the off-site location was 120 pg/m³. These data are similar to data collected from previous years.

#### 4.2.4 Impact of Ambient Air Releases on the Environment

LLNL operations involving radioactive materials had minimal impact on ambient air during 2008. The measured radionuclide particulate and tritium concentrations in air at the Livermore site and Site 300 were all less than one-tenth of one percent of the DOE primary radiation protection standard for the public (DCG).

Beryllium is naturally occurring and has a soil concentration of approximately 1 part per million. The sampled results are believed to be from naturally occurring beryllium that was resuspended from the soil and collected by the sampler. Even if the concentrations of beryllium detected were from LLNL activities, the amount is still less than one percent of the BAAQMD ambient air concentration limit.