4. Air Monitoring Programs

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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 458.1; 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 458.1.

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 µSv/y (0.1 mrem/y), as calculated using the U.S. 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 µSv/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 and registrations from local air districts (i.e., BAAQMD and SJVAPCD) for stationary emission sources and from CARB for portable emission sources such as diesel air compressors and generators and for off-road diesel vehicles. Current permits and registrations do not require monitoring of air effluent but do require monitoring of equipment inventory, equipment usage, material usage, and/or 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 2011, LLNL measured releases of radioactivity from air exhausts at six 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.

Four facilities had measureable emissions in 2011. A total of 6978 GBq (188.6 Ci) of measured tritium was released from the stack exhausts at the Tritium Facility. Of this, approximately 32% of tritium was released as vapor (HTO). The remaining 68% released was gaseous tritium (HT).

The DWTF released a total of 0.56 GBq (0.015 Ci) of measured tritium from the stack exhaust. The tritium released was approximately 75% vapor (HTO) and 25% gaseous tritium (HT).

The National Ignition Facility (NIF) released a total of 45.1 GBq (1.22 Ci) of measured tritium from the stack exhaust in 2011. A total of 30.9 GBq (0.836 Ci) was released as vapor (HTO), 14.2 GBq (0.385 Ci) as gaseous (HT), and 5.2×10^{-3} GBq (1.4×10^{-4} Ci) of tritiated particulate.

The Contained Firing Facility (CFF) at Site 300 had measured depleted uranium stack emissions in 2011. A total of 2.1×10^{-7} GBq (5.6×10^{-9} Ci) of uranium-234, 1.5×10^{-8} GBq (4.1×10^{-10} Ci) of uranium-235, and 1.2×10^{-6} GBq (3.2×10^{-8} Ci) of uranium-238 was released in particulate form.

The measured emissions from monitored facilities were a result of planned activities with radioactive material.

None of the other facilities monitored for radionuclides had reportable emissions in 2011. The data tables in **Appendix A, Section A.1** provide summary results of all air effluent monitored facilities and include upwind locations (control stations) which are used 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 1.5×10^{-1} µSv./y (1.5×10^{-2} mrem/y); the dose from the DWTF (modeling HT emissions as HTO) was 5.8 x 10^{-6} µSv/y (5.8×10^{-7} mrem/y); the dose from the NIF (modeling HT emissions as HTO) was 2.8×10^{-4} µSv/y (2.8×10^{-5} mrem/y), and the dose from the CFF was 9.0×10^{-7} µSv/y (9.0×10^{-8} mrem/y).

All of the reported SW-MEI doses at the Livermore site and Site 300 are less than one percent of the annual NESHAPs standard, which is 100 µSv/y (10 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 2011 Annual Report* (Wilson et al. 2012) for a complete description of air effluent monitoring, this report is located in **Appendix D**.

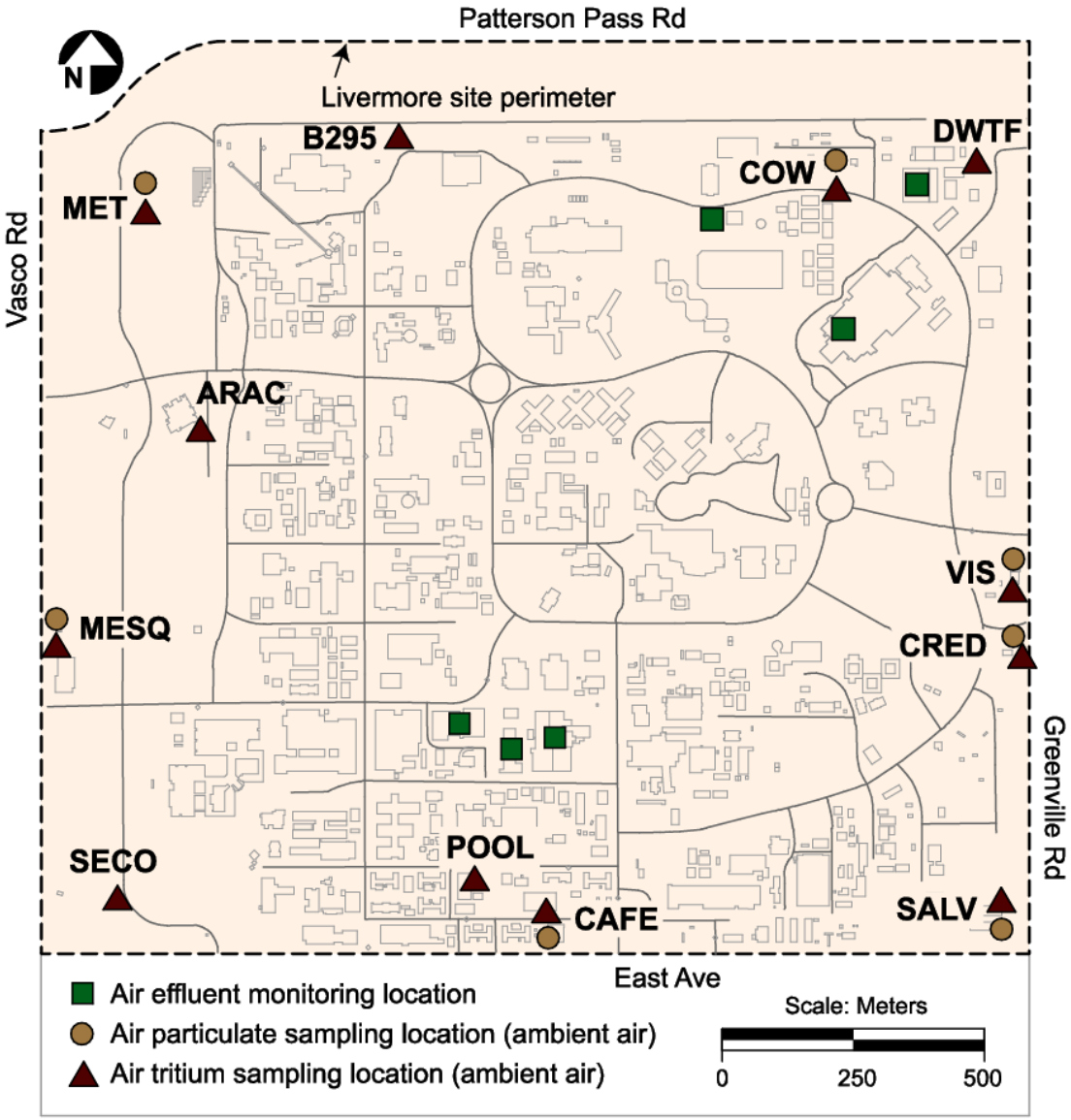

Figure 4-1. Air effluent and ambient air monitoring locations at the Livermore site, 2011.

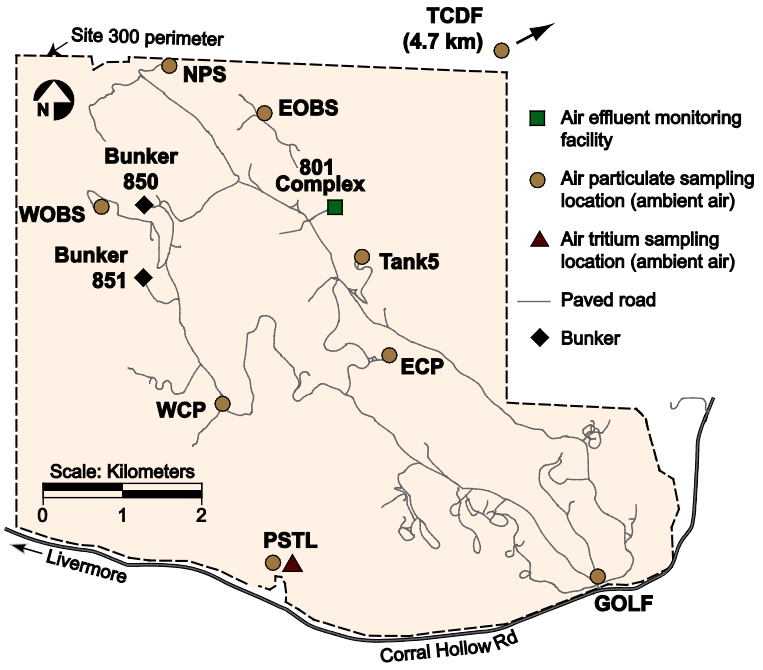

Figure 4-2. Air effluent and ambient air monitoring locations at Site 300, 2011.

4.1.2 Nonradiological Air Releases and Impact on the Environment

In 2011, the Livermore site emitted approximately 109 kg/d of regulated air pollutants as defined by the Clean Air Act, including nitrous oxides (NOx), sulfur 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.

Table 4-1. Nonradioactive air emissions, Livermore site and Site 300, 2011.

	Estimated releases (kg/d)	
Pollutant	**Livermore site**	**Site 300**
ROGs/POCs	10.8	0.33
Nitrogen oxides	48.1	2.70
Carbon monoxide	43.3	0.71
Particulates (PM-10)	4.8	0.56
Sulfur oxides	1.5	0.17
Total	108.5	4.47

Livermore site air pollutant emissions were very low in 2011 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 3.63×10^5 kg/d, compared to the estimated daily release from

the Livermore site of 48.1 kg/d, which is 0.013% of total Bay Area source emissions for NOx. The 2011 BAAQMD estimate for ROGs/POCs daily emissions throughout the Bay Area was 2.71×10^5 kg/d, while the daily emission estimate for 2011 from the Livermore site was 10.8 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 2011 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-powered generators), a gasoline-dispensing facility, and general research operations. Combustion pollutant emissions, such as NOx, CO, PM-10 and SOx, increased in 2011 primarily from the increased usage of two diesel-powered generators in response to two 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 technical standard (DCS), which complements DOE Order 458.1, specifies 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^3. 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**.

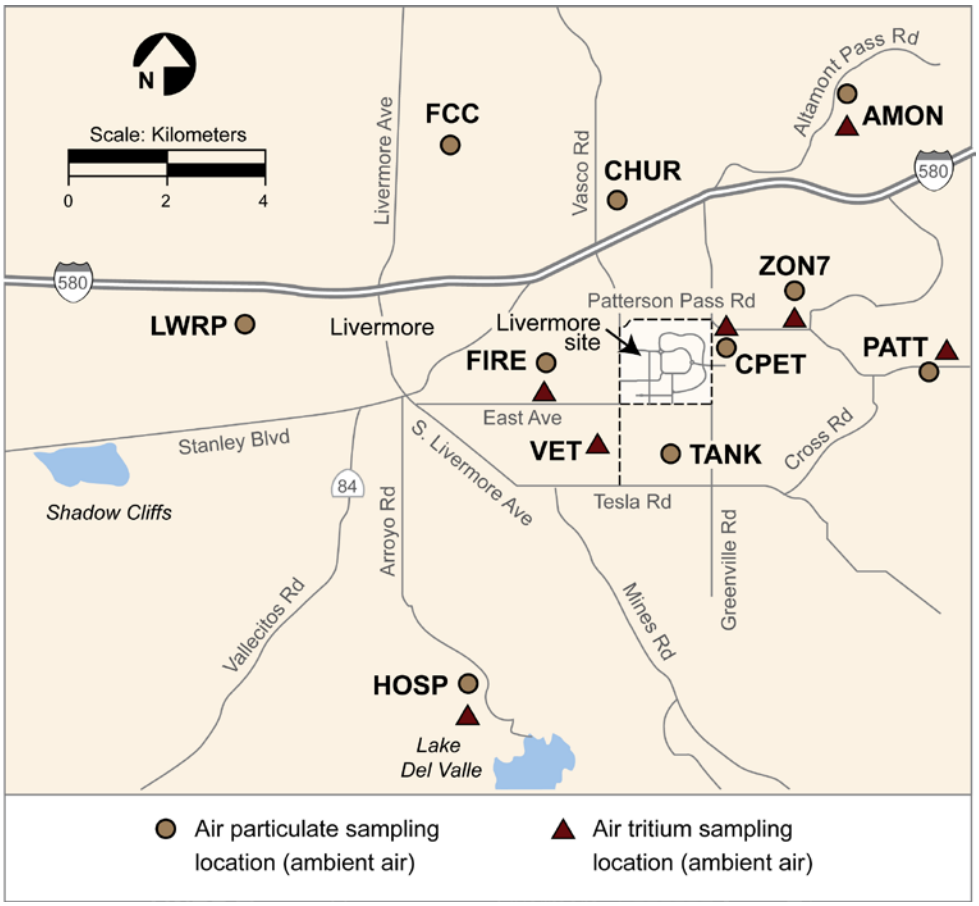

Figure 4-3. Air particulate and tritium monitoring locations in the Livermore Valley, 2011.

4.2.1 Ambient Air Radioactive Particulates

Composite samples for the Livermore site and Site 300 were analyzed by gamma spectroscopy for an environmental suite of gamma-emitting radionuclide concentrations in air that include fission products, activation products, actinides, and naturally occurring products. The isotopes detected at both sites throughout 2011 were beryllium-7 (cosmogenic), lead-210, radium-226, and potassium-40, all of which are naturally occurring in the environment.

On March 11 of 2011, the Japanese Fukushima reactor crisis began. Detections of iodine-131, cesium-134, and cesium-137 in ambient air were seen in March, April, and May of 2011. After that time, sampled air returned to non-detections for these isotopes. The detections are attributed to the Fukushima reactors (see **Appendix A**, **Section A.2** for data tables); the sampled results were consistent with the EPA's RadNet air concentration measurement data from fallout of the Fukushima reactors (U.S. EPA, 2011).

EPA stated that *it is important to note that all of the radiation levels detected by RadNet monitors and sampling have been very low, well below any level of public health concern* (U.S. EPA, 2011).

Composite samples were analyzed by alpha spectroscopy for plutonium-239+240, which was detected in 9 out of 216 samples taken in 2011. Detections at the Livermore site and Livermore off-site locations for plutonium-239+240 are attributed to resuspension of plutonium-contaminated soil (see Chapter 6) to ambient air from historical operations. The highest values and percentage of the DCS for the plutonium-239+240 detections were as follows:

- Livermore site perimeter: 16.5 nBq/m^3 (0.44 aCi/m^3); 0.00018% of the DCS
- Livermore off-site locations: 16.7 nBq/m^3 (0.45 aCi/m^3); 0.00019% of the DCS
- Site 300 composite: There were no detections in 2011.

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 2011 were as follows:

- Livermore site perimeter composite: 0.0072
- Site 300 sample locations: 0.0071
- Site 300 off-site location: 0.0072

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 percent of the DCS 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**, **Section A.2** 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. Because HTO air concentrations observed at the Livermore site sample locations are low, the concentrations at 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. The Derived Concentration Standards were formerly published in DOE Order 5400.5 in 1993. The current radiation protection standards approach, which has changed from the previously adopted 1993 guidance, uses age and gender specific attributes for the population subgroups of members of the public subject to exposure incorporating more sophisticated biokinetic and dosimetric information from the International Commission on Radiological Protection (ICRP).

Table 4-2. Air tritium sampling summary for 2011.

Sampling locations	Detection frequency	Concentration (mBq/m^3)				Median % DCS[(a)]	Mean Dose (nSv)
		Mean	Median	IQR	Maximum		
Livermore site perimeter	267 of 311	87.3	53.6	66.8	799	0.00069	21.0
Livermore Valley	114 of 180	29.5	19.6	27.3	336	0.00025	7.08
Site 300	6 of 25	3.39	6.77	13.1	28.3	0.000087	<5

(a) DCS = Derived Concentration Technical Standard = 7.8×10^6 mBq/m^3 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 µrem/y) (i.e., the annual dose from inhaling air with a concentration at the MDC of about 25 mBq/m^3 [0.675 pCi/m^3]).

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 2011 for airborne beryllium was 12 pg/m^3 This value is only 0.12% of the BAAQMD ambient concentration limit for beryllium (10,000 pg/m^3). 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 2011 was 16 pg/m^3 and the highest value at the off-site location was 23 pg/m^3. 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 2011. The measured radionuclide particulate and tritium concentrations in air at the Livermore site and Site 300 were all less than one percent of the DOE primary radiation protection standard for the public (DCS).

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.