

## 4. Air Monitoring and Dose Assessment

*Heather Byrnes • Anthony Wegrecki • Kent Wilson*

---

Lawrence Livermore National Laboratory (LLNL) 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. Department of Energy (DOE) regulations include Title 40, Code of Federal Regulations, Part 61 (40 CFR 61), Subpart H—the National Emission Standards for Hazardous Air Pollutants (NESHAPs) section of the Clean Air Act; applicable portions of DOE Order 458.1; and American National Standards Institute (ANSI) standards (N13.1-1969, 1999 [reaffirmed 2011]). The *Environmental Radiological Effluent Monitoring and Environmental Surveillance* (DOE 2015) handbook provides the guidance for implementing DOE Order 458.1.

The U.S. Environmental Protection Agency (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 Bay Area Air Quality Management District (BAAQMD) and the San Joaquin Valley Air Pollution Control District (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  $\mu\text{Sv}/\text{y}$  (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  $\mu\text{Sv}/\text{y}$  (10 mrem/y) total site effective-dose equivalent from the airborne pathway is not exceeded. See **Appendix D** for the *LLNL 2019 NESHAPs Annual Report* (Wilson et al., 2020).

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 the California Air Resources Board (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**

In 2019, 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.

Three facilities had measurable emissions in 2019. A total of 126.4 Ci (4,677 GBq) of measured tritium was released from the stack exhausts at the Tritium Facility. Of this, approximately 80% of tritium was released as vapor (HTO). The remaining 20% released was gaseous tritium (HT). The National Ignition Facility (NIF) released a total of 2.8 Ci (104 GBq) of tritium from the stack exhaust in 2019. Of this, approximately 90% of tritium was released as HTO. The remaining 10% was released as HT. The Contained Firing Facility (B801A) at Site 300 had measured depleted uranium stack emissions in 2019 consisting of  $1.2 \times 10^{-7}$  Ci ( $4.4 \times 10^{-6}$  GBq) of uranium-234,  $1.7 \times 10^{-8}$  Ci ( $6.4 \times 10^{-7}$  GBq) of uranium-235, and  $9.2 \times 10^{-7}$  Ci ( $3.4 \times 10^{-5}$  GBq) of uranium-238 in particulate form.

There were two unplanned radioactive air releases from the Livermore Site in 2019. On October 11, 2019, the Physical and Life Sciences Directorate (B235) had a uranium fire as a result of material transfer when one ball valve was accidentally left open on a two-valve system. When the container was moved out of alignment, approximately 0.00002 Ci ( $7.4 \times 10^{-4}$  GBq) of depleted uranium dropped into a photo tray under the ball valve of the machine. The material began to spark and was placed inside a fume hood. The fire was extinguished with a spray bottle and the sash was lowered. Work was suspended until a Group Management Concerns/Issues process was completed. This included the Environmental Functional Area Health Physicist and facility management that implemented proper corrective actions to reduce the possibility of an accidental valve misalignment from becoming a reoccurring event.

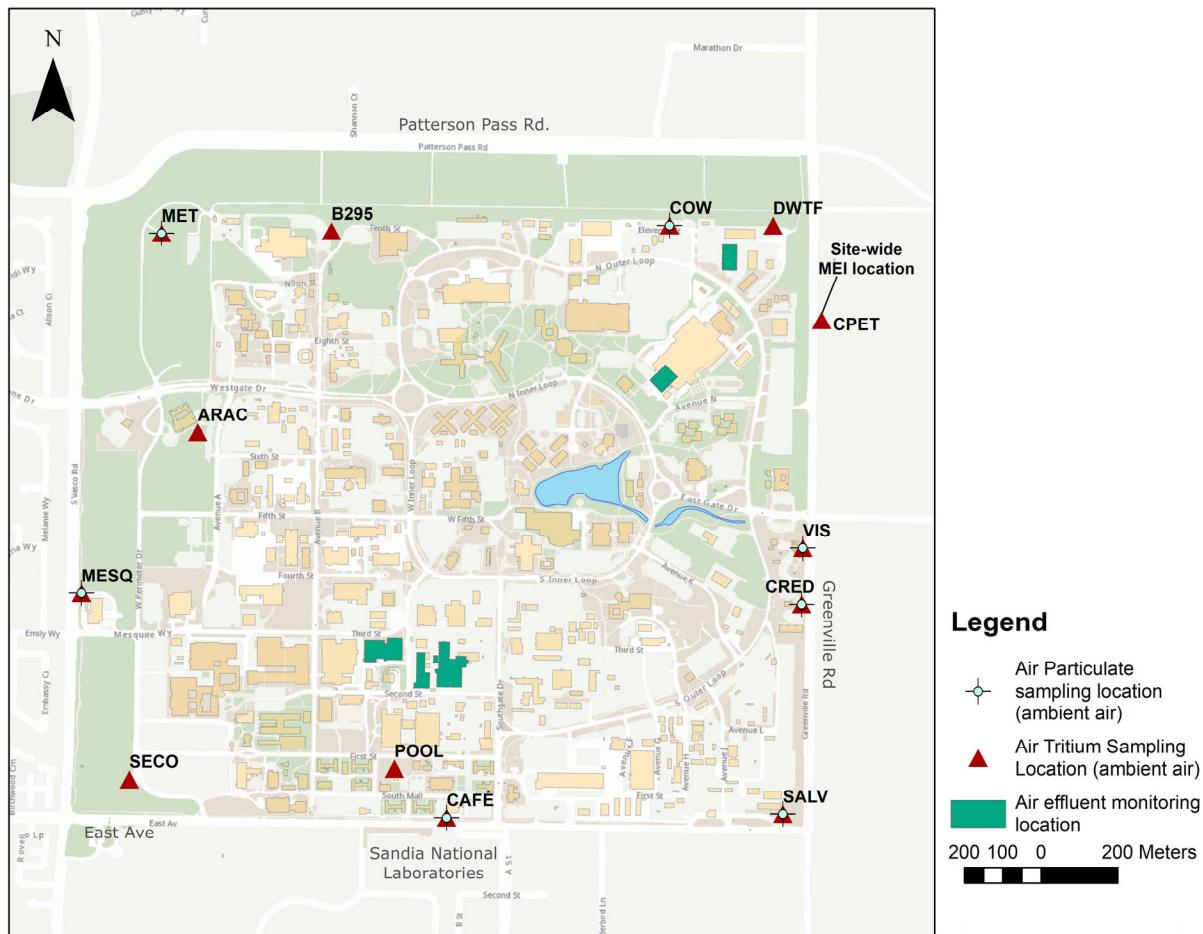
The unplanned release of 0.00002 Ci ( $7.4 \times 10^{-4}$  GBq) of depleted uranium is less than one-tenth of a percent of the U.S EPA Reportable Quantities (40 CFR 302). CAP88-PC Version 4.0.1.17 was used to model the 0.00002 Ci ( $7.4 \times 10^{-4}$  GBq) source term. The dose to the site-wide maximally exposed individual (SW-MEI) (CPET) was  $5.3 \times 10^{-5}$  mrem ( $5.3 \times 10^{-4}$   $\mu$ Sv). The modeled dose is well below the NESHAPS 10 mrem/y site-wide standard dose to members of the public; it is included in the total site-wide reported dose.

On October 26, 2019, the Physical and Life Sciences Directorate (B235) had a second uranium fire that occurred during the cleaning of a plasma-based particle spheroidizer. Approximately 0.000004 Ci ( $1.5 \times 10^{-4}$  GBq) of uranium dropped onto a contamination barrier and began to smolder. The beginning fire was extinguished with a Metal-X extinguisher. The work was suspended with no routine access allowed in the room until a Group Management Concerns/Issues process was completed. This included the Environmental Functional Area Health Physicist and facility management that implemented proper corrective actions to reduce the possibility of an accidental uranium release from maintenance activities becoming a reoccurring event.

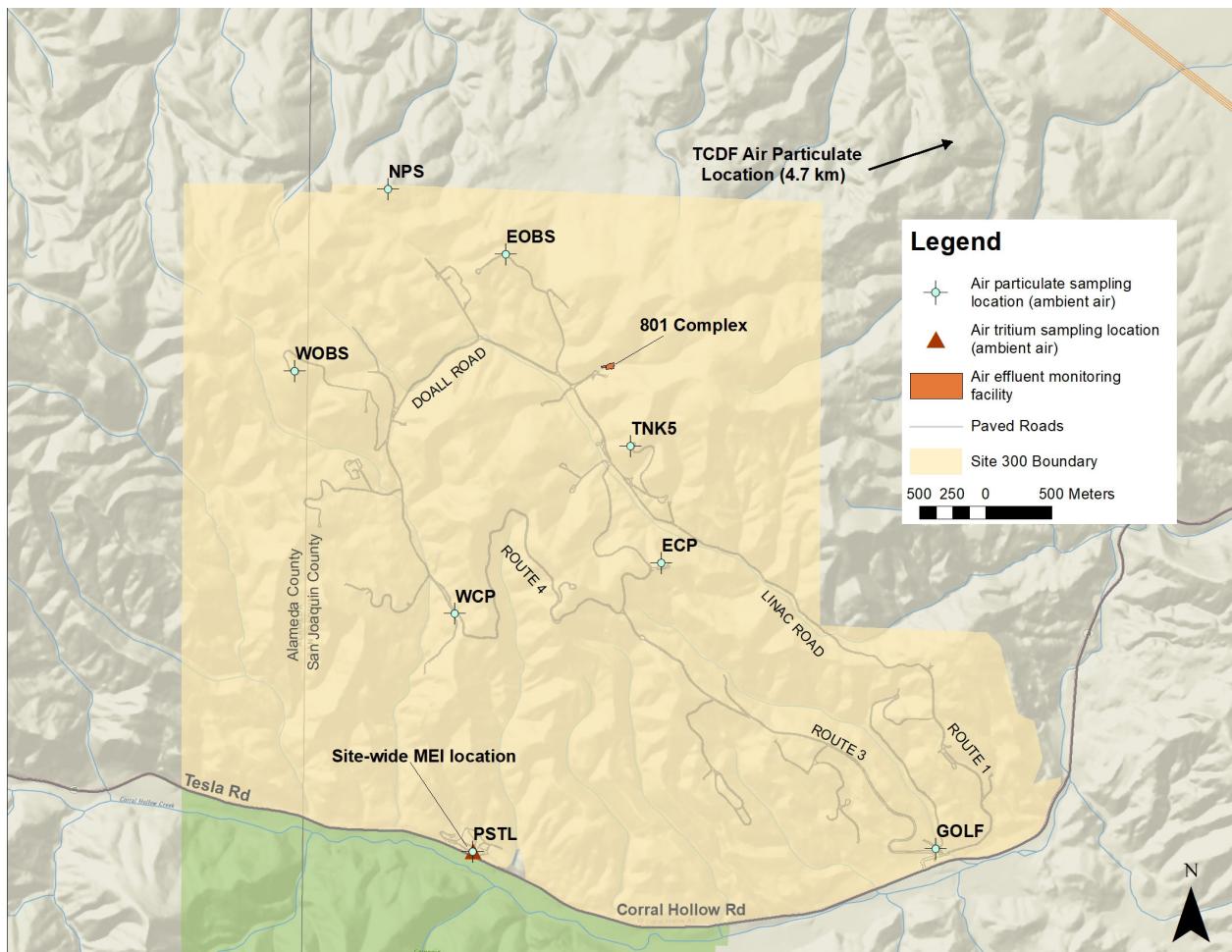
The unplanned release of 0.000004 Ci ( $1.5 \times 10^{-4}$  GBq) of depleted uranium is less than one-tenth of a percent of the U.S EPA Reportable Quantities (40 CFR 302). CAP88-PC Version 4.0.1.17

was used to model the  $0.000004 \text{ Ci}$  ( $1.5 \times 10^{-4} \text{ GBq}$ ) source term. The dose to the SW-MEI (CPET) was  $1.1 \times 10^{-6} \text{ mrem}$  ( $1.1 \times 10^{-5} \mu\text{Sv}$ ). The modeled dose is well below the NESHAPS 10 mrem/y site-wide standard dose to members of the public; it is included in the total site-wide reported dose.

None of the other facilities monitored for radionuclides had reportable emissions in 2019. 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.



**Figure 4-1.** Air effluent and ambient air monitoring locations at the Livermore Site, 2019.



**Figure 4-2.** Air effluent and ambient air monitoring locations at Site 300, 2019.

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

In 2019, the Livermore Site emitted approximately 99.0 kg/d of regulated air pollutants as defined by the Clean Air Act, including nitrous oxides (NOx), sulfur oxides (SOx), particulate matter (PM10), 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, 2019.

Pollutant	Estimated releases (kg/d)	
	Livermore Site	Site 300
ROGs/POCs	17.1	0.15
Nitrogen oxides	35.3	0.79
Carbon monoxide	40.5	0.25
Particulates (PM10)	4.4	0.29
Sulfur oxides	1.7	0.04
<b>Total</b>	<b>99.0</b>	<b>1.52</b>

Livermore Site air pollutant emissions were very low in 2019 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 is estimated to be  $2.3 \times 10^5$  kg/d, compared to the estimated daily release from the Livermore Site of 35.3 kg/d, which is 0.015% of total Bay Area source emissions for NOx. The 2019 BAAQMD estimate for ROGs/POCs daily emissions throughout the Bay Area was approximately  $2.2 \times 10^5$  kg/d, while the daily emission estimate for 2019 from the Livermore Site was 17.1 kg/d, or 0.0078% of the total Bay Area source emissions for ROGs/POCs.

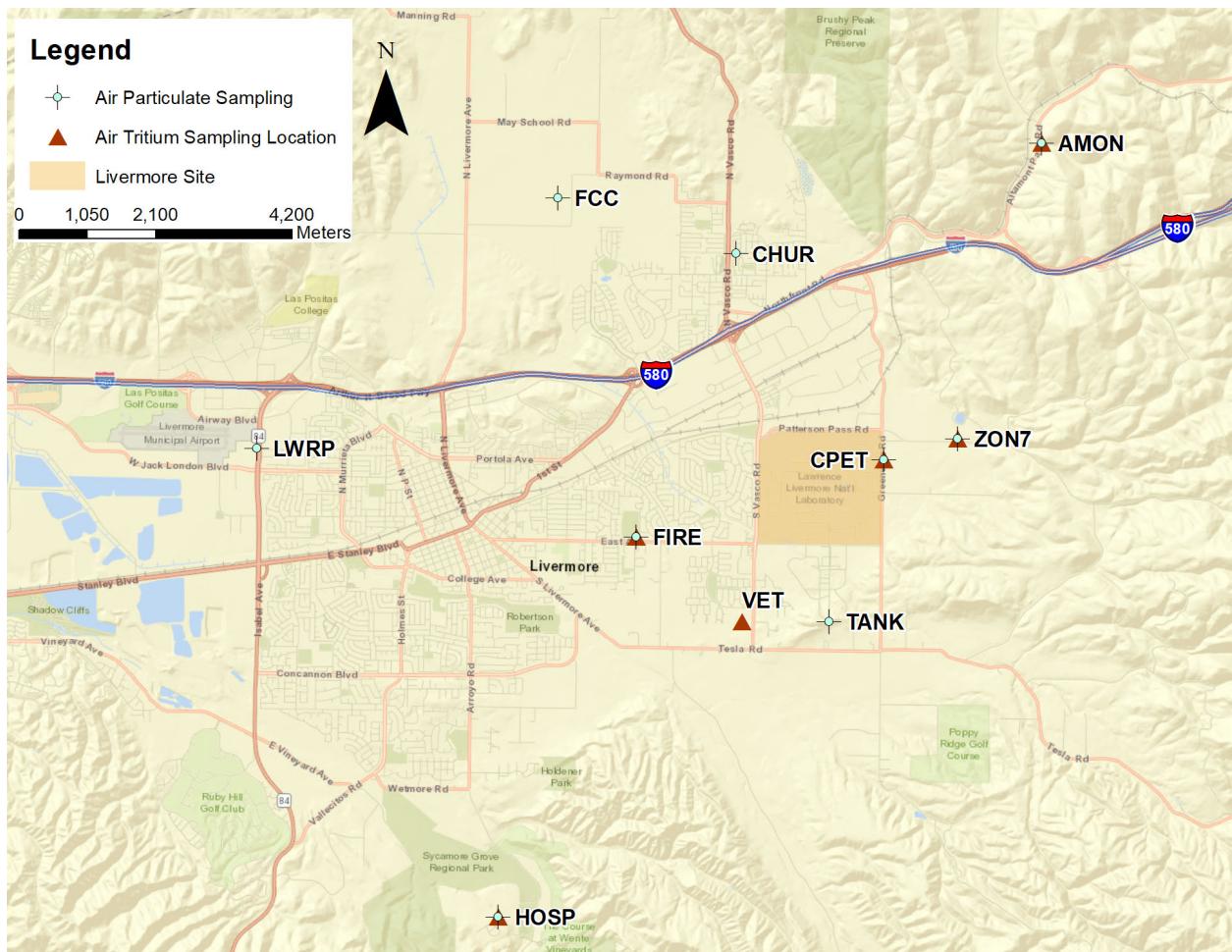
Certain operations at Site 300 require permits from the SJVAPCD. The estimated daily air pollutant emissions during 2019 from operations (permitted and exempt stationary sources) at Site 300 are listed in **Table 4-1**. The stationary emission sources that release the greatest amounts of regulated air pollutants at Site 300 include internal combustion engines (such as diesel-powered generators), a gasoline-dispensing facility, and general research operations. Combustion pollutant emissions, including NOx, CO, PM10, SOx, and ROGs/POCs decreased in 2019. The diesel-powered generators were the primary source of the pollutants.

## 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 NESHPAs regulations.

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, 2019.

#### 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 isotopes. The isotopes detected at both sites in 2019 were beryllium-7 (cosmogenic), lead-210, and potassium-40, all of which are naturally occurring in the environment. The composite samples for both sites were non-detections for potassium-40 in 2019.

Composite samples were analyzed by alpha spectroscopy for plutonium-239+240, which was detected in 4 out of 204 samples taken in 2019. Detections at the Livermore Site, Site 300, and Livermore off-site locations for plutonium-239+240 are attributed to a number of factors that include: resuspension of plutonium-contaminated soil (see Chapter 6), resuspended fallout from previous atmospheric testing, or resuspended fallout from the Fukushima nuclear accident.

The derived concentration 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.

The DCS was formerly published in DOE Order 5400.5 (Radiation Protection of the Public and the Environment) 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).

The highest values and percentage of the DCS for the plutonium-239+240 detections were as follows:

- Livermore Site perimeter: 26.8 nBq/m<sup>3</sup> (0.72 aCi/m<sup>3</sup>), 0.0003% of the DCS.
- Livermore off-site locations: All results were non-detections in 2019.
- Site 300 composite: All results were non-detections in 2019.

Uranium-235 and uranium-238 were detected at all sample locations. Uranium ratios, which can be calculated by mass or by atom, 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 typical uranium-235/uranium-238 ratio of 0.002. The annual median uranium-235/uranium-238 isotopic ratios for 2019 at the Livermore Site and off-site location were:

- Livermore Site perimeter composite: 0.00724.
- Off-site TCDF (located 4.7 km northeast from Site 300): 0.00726.

The annual uranium-235/uranium-238 isotopic ratio medians are consistent with naturally occurring uranium.

Site 300 has not had open-air depleted uranium shots since September 2007. However, there are still areas of depleted uranium contaminated soil. Wind-driven resuspension as well as soil disturbance from construction-type activities and fire road maintenance has occasionally shown a depleted uranium signature at the location of the SW-MEI member of the public (see **Figure 4-2**). The uranium-235 to uranium-238 isotopic ratio at the SW-MEI was consistent with naturally occurring uranium.

All of the individual uranium-235 and uranium-238 results, including on-site samples showing a depleted uranium signature, were less than one tenth of one percent of the DCS as shown in **Appendix A, Section A.2**.

All locations were sampled for gross alpha and gross beta. 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, radium, and lead), which are also routinely found in local soils. See **Appendix A, Section A.2**.

## 4.2.2 Ambient Air Tritium Concentrations

LLNL emits tritium to the air from multiple sources. These sources include monitored stack sources, such as the Tritium Facility and NIF, unmonitored stack sources having minor emissions of tritium, and area sources. Area (diffuse) sources include stored containers of tritium waste or tritium-contaminated equipment from which HTO diffuses into the atmosphere. LLNL does not directly measure diffuse emissions, but estimates the radiation dose to the public from these sources given measurements taken using the ambient air tritium sampling network. The ambient air tritium sampling network measures HTO concentrations in the air from all sources. This information, along with measured stack emissions, is used to provide an estimate of the dose to the public from diffuse area tritium emissions. The approach used to characterize the area emission sources is stated in the *LLNL NESHAPs 2019 Annual Report* (Wilson et al., 2020). See **Appendix D** for a copy of this report. The biweekly air tritium data that are provided in **Appendix A, Section A.2** are summarized in **Table 4-2**.

**Table 4-2.** Ambient air tritium sampling summary for 2019.

Sampling location	Detection frequency <sup>(a)</sup>	Concentration (mBq/m <sup>3</sup> )				Median as % of DCS <sup>(d)</sup>	Mean dose <sup>(e)</sup> (nSv)
		Mean	Median	IQR <sup>(b)</sup>	Maximum <sup>(c)</sup>		
Livermore Site perimeter	291 of 309	81	47	69	740	0.00060	19.0
Livermore Valley	121 of 150	36.2	22.6	22.2	729	0.00029	8.49
Site 300	13 of 25	10.5	6.1	11.6	65.5	0.000078	<5

(a) Detection frequency indicates the number of samples that measure greater than 100% of 2-Sigma uncertainty (see Chapter 8).

(b) IQR = Interquartile Range

(c) The maximum concentration in 2019 was 0.0095% of the DCS. (DCS for tritium is 7.8E+03 Bq/m<sup>3</sup>, DOE-STD-1196-2011).

(d) Median as a percent of DCS is not used when the median is a negative value (see Chapter 8).

(e) Based on an annual breathing rate of 8103 m<sup>3</sup> and inhalation dose conversion factor of  $1.93 \times 10^{-11}$  Sv/Bq (DOE-STD-1196-2011). Dose due to HTO absorption through skin is accounted for. It is estimated as equaling one-half of the dose due to inhalation (2001 Environmental Report, Appendix A).

For a location at which the mean concentration is at or below the minimal detectable concentration, dose from tritium is assumed to be less than 5 nSv/y (0.5  $\mu$ rem/y).

## 4.2.3 Ambient Air Beryllium Concentrations and Impact on the Environment

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 2019 for airborne beryllium was 62 pg/m<sup>3</sup>. This value is 0.62% of the BAAQMD ambient concentration limit for beryllium (10,000 pg/m<sup>3</sup>). 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 2019 was 74 pg/m<sup>3</sup> and the highest value at the off-site location was 98 pg/m<sup>3</sup>. These data are similar to data collected from previous years.

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 samplers. 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.

## 4.3 Radiological Air Dose Assessment

Dose is assessed for two types of receptors. First is the dose to the SW-MEI member of the public. Second is the collective or “population” dose received by people who reside within 80 km of either of the two LLNL sites.

In 2019, the SW-MEI at the Livermore Site was located at the Integrative Veterinary Care facility (CPET) about 35 meters outside the site’s eastern perimeter. The SW-MEI at Site 300 was located on the site’s south-central perimeter (PSTL), which borders the Carnegie State Vehicular Recreation Area. The two SW-MEI locations are shown in Figures 4-1 and 4-2. Table 4-3 shows average doses received in the United States from exposure to sources of radiation as well as the collective dose for people residing within 80 km of the Livermore Site.

**Table 4-3.** Radiation doses from ubiquitous background and man-made sources of radiation.

Source category <sup>(a)</sup>	Individual dose ( $\mu$ Sv) <sup>(b, c)</sup>	Collective dose <sup>(d)</sup> (person-Sv) <sup>(e)</sup>
Natural radioactivity <sup>(f)</sup>		
Cosmic radiation	330	2,570
Terrestrial radiation	210	1,640
Internal (food and water consumption)	290	2,260
Radon and Thoron	2,280	17,800
Medical radiation procedures	3,000	23,400
Consumer	130	1,010
Industrial plus occupational	8	62

(a) From National Council on Radiation Protection and Measurements, Report No. 160, Table 8.1 (NCRP 2009).

(b) 1  $\mu$ Sv = 0.1 mrem.

(c) This dose is an average over the U.S. population.

(d) The collective dose is the combined dose for all individuals residing within an 80-km radius of LLNL (approximately 7.8 million people for the Livermore Site and 7.1 million for Site 300), calculated with respect to distance and direction from each site. The Livermore Site population estimate of 7.8 million people was used to calculate the collective doses for the source categories.

(e) 1 person-Sv = 100 person-rem.

(f) These values vary with location.

The annual radiological doses from all air emissions at the Livermore Site and Site 300 in 2019 were found to be well below the applicable standards for radiation protection of the public, in particular the NESHAPs 100  $\mu$ Sv/y (10 mrem/y) site-wide standard. Using an EPA-mandated

computer model and LLNL site-specific meteorology appropriate to the two sites, the doses to the LLNL SW-MEI members of the public from LLNL operations in 2019 were:

- Livermore Site:  $4.3 \times 10^{-2}$   $\mu\text{Sv}$  ( $4.3 \times 10^{-3}$  mrem).
- Site 300:  $9.5 \times 10^{-4}$   $\mu\text{Sv}$  ( $9.5 \times 10^{-5}$  mrem).

The collective effective dose equivalent (EDE) attributable to LLNL airborne emissions in 2019 was calculated to be 0.0033 person-Sv (0.33 person-rem) for the Livermore Site and  $2.9 \times 10^{-7}$  person-Sv ( $2.9 \times 10^{-5}$  person-rem) for Site 300. These doses include potentially exposed populations of 7.8 million people for the Livermore Site and 7.1 million people for Site 300 living within 80 km of the site centers.

The doses to the SW-MEI, which represent the maximum doses that could be received by members of the public where there is a residence, school, business, or office, resulting from Livermore Site and Site 300 operations in 2019, were less than one percent of the NESHAPs 100  $\mu\text{Sv}/\text{y}$  (10 mrem/y) site-wide standard.

LLNL operations involving radioactive materials had minimal impact on ambient air during 2019. The measured radionuclide particulate and tritium concentrations in ambient 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). The SW-MEI doses from both sites for 2019 are much less than one-tenth of one percent of the total dose from sources of natural occurring radioactivity shown in **Table 4-3**.