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NESHAPs CY19 Report

K. Wilson, H. Byrnes, T. Wegrecki

June 8, 2020

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.



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U.S. Department of Energy
Radionuclide Air Emission Report for 2019
(in compliance with 40 CFR 61, Subpart H)

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Executive Summary

Lawrence Livermore National Security, LLC operates facilities at Lawrence Livermore National Laboratory (LLNL) in which radionuclides are handled and stored. These facilities are subject to the U.S. Environmental Protection Agency (EPA) National Emission Standards for Hazardous Air Pollutants (NESHAPs) in Code of Federal Regulations (CFR) Title 40, Part 61, Subpart H, which regulates radionuclide emissions to air from Department of Energy (DOE) facilities. Specifically, NESHAPs limits the emission of radionuclides to the ambient air to levels resulting in an annual effective dose equivalent of 10 mrem (100 μ Sv) to any member of the public. Using measured and calculated emissions, and building-specific and common parameters, LLNL personnel applied the EPA-approved computer code, CAP88-PC, Version 4.0.1.17, to calculate the dose to the maximally exposed individual member of the public for the Livermore Site and Site 300.

In 2019, LLNL maintained its compliance with 40 CFR 61, Subpart H. All radioactive air emissions resulted in calculated doses far below the annual 10 mrem (100 μ Sv) site-wide standard. The annual doses to the site-wide maximally exposed individual member of the public at the Livermore Site and Site 300 from planned and unplanned operations in 2019 are:

- Livermore Site: 4.3×10^{-3} mrem (4.3×10^{-2} μ Sv)
- Site 300: 9.5×10^{-8} mrem (9.5×10^{-7} μ Sv)

Background Information

LLNL is a premier research laboratory that is part of the National Nuclear Security Administration (NNSA) within DOE. As a national security laboratory, LLNL is responsible for ensuring that the nation's nuclear weapons remain safe, secure, and reliable. The Laboratory also meets other national security needs, including countering the proliferation of weapons of mass destruction and strengthening homeland security, and conducts major research in atmospheric, earth, and energy sciences; bioscience and biotechnology; and engineering, basic science, and advanced technology. The Laboratory serves as a scientific resource to the U.S. government and a partner to industry and academia.

Because LLNL is a DOE facility, it is subject to the requirements of 40 CFR 61, Subpart H, National Emission Standards for Emissions of Radionuclides Other than Radon from Department of Energy Facilities. This regulation limits emissions of radionuclides to ambient air to levels resulting in an annual effective dose equivalent of 10 mrem (100 μ Sv) to any member of the public. The regulation also requires annual reporting of the emissions and resulting dose.

1.1 SITE DESCRIPTION

LLNL consists of two sites—an urban site in Livermore, California, referred to as the “Livermore Site;” and a rural experimental test site, referred to as “Site 300,” near Tracy, California (**Figure 1**).



Figure 1. Locations of LLNL's Livermore Site and Site 300.

The Livermore Site is just within the eastern city limits of Livermore, a city of about 85,000 in Alameda County. The site occupies 1.3 square miles, including the land that serves as a buffer zone around much of the site. Within a 50-mile radius of the Livermore Site are communities such as Tracy and Pleasanton and the more distant (and more densely populated) cities of Oakland, San Jose, and San Francisco. Of the 7.8 million people within 50 miles of the Laboratory, only about 10% are within 20 miles.

Site 300, LLNL's Experimental Test Site, is in the Altamont Hills of the Diablo Range and straddles the San Joaquin and Alameda county line. The site is 12 miles east of the Livermore Site and occupies 10.9 square miles. The city of Tracy, with a population of about 86,000, is approximately 6 miles to the northeast (measured from the northeastern border of Site 300 to Sutter Tracy Community Hospital). Of the 7.1 million people who live within 50 miles of Site 300, 95% are more than 20 miles away in metropolitan areas such as Oakland, San Jose, and Stockton.

The weather conditions at the Livermore Site and Site 300 are very similar. The climate at both sites is best described as Mediterranean, characterized by mild, rainy winters and warm-to-hot, dry summers. However, the complex topography of Site 300 does influence local wind and temperature patterns. The stronger winds that occur at the higher elevations of Site 300 (**Figure 2**), result in warmer nights and slightly cooler days than the Livermore Site.

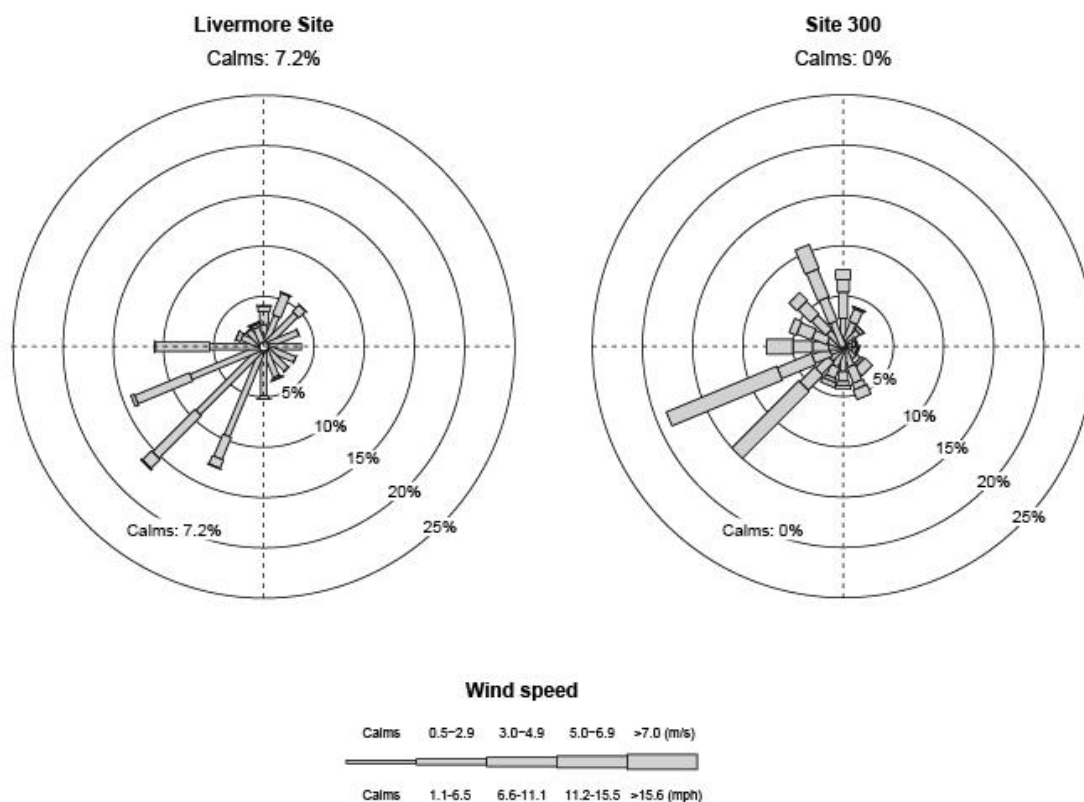


Figure 2. Wind roses for the Livermore Site and Site 300 for 2019.

The 2019 annual wind data for both sites are displayed as wind roses 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 spoke widths within each spoke represent wind speed classes. These data show that at the Livermore Site, winds blew from the south-southwest through the west-southwest about 43.7% of the time; and at Site 300, from the southwest to the west about 41.4% of the time. The average wind speed in 2019 at the Livermore Site was 2.2 m/s (4.9 mph), and the average wind speed at Site 300 was 5.7 m/s (12.7 mph). In 2019, the Livermore Site received 39.6 cm (15.6 in) of rain and Site 300 received 28.7 cm (11.3 in).

1.2 SOURCE DESCRIPTION

Many different radioisotopes were present at LLNL in 2019 including biomedical tracers, tritium, mixed fission products, transuranic isotopes, and others—see **Table 1**. Radioisotope handling procedures and work enclosures are determined for each project or activity, depending on the isotopes, the quantities being used, and the types of operations being performed. Work enclosures include glove boxes, exhaust hoods, and laboratory bench tops. Exhaust paths to the atmosphere include High Efficiency Particulate Air (HEPA) filtered ventilation systems, roof vents and stacks without abatement devices, resuspension of deposited depleted uranium in the soil from previous open-air explosives testing at Site 300, and releases to ambient air from a variety of diffuse sources. **Table 2** identifies the buildings, by managing organization, at LLNL where there is a potential for release of radioactive materials to the air in 2019.

Table 1. Radionuclides used at LLNL during 2019.

Ac-227	Ac-228	Ag-108m	Ag-110m	Am-241	Am-242m
Am-243	Au-194	Au-195	Au-196	Au-196m	Au-198
Ba-133	Ba-140	Be-10	Be-7	Bi-207	Bi-212
Bi-214	C-14	Ca-41	Ca-45	Cd-109	Ce-139
Ce-141	Ce-143	Ce-144	Cf-249	Cf-252	Cl-36
Cm-243	Cm-244	Cm-246	Cm-248	Co-55	Co-57
Co-58	Co-60	Cr-51	Cs-134	Cs-135	Cs-137
Eu-152	Eu-154	Eu-155	Fe-52	Fe-55	Fe-59
Fe-60	Gd-148	Gd-153	H-3	Hg-203	Ho-166
Ho-166m	I-125	I-129	I-131	I-132	I-133
Ir-190	Ir-192	Ir-194m	K-40	Kr-85	La-140
Lu-177m	Mn-54	Mo-93	Mo-99	Na-22	Nb-92
Nb-94	Nb-95	Nb-97	Nd-147	Ni-56	Ni-57
Ni-59	Ni-63	Np-236s	Np-237	Np-239	P-32
P-33	Pa-231	Pa-233	Pb-210	Pb-212	Pb-214
Pm-147	Pm-149	Po-208	Po-210	Pr-143	Pt-193m
Pt-195m	Pu-236	Pu-238	Pu-239	Pu-240	Pu-241
Pu-242	Pu-244	Ra-226	Ra-228	Rh-105	Rh-106
Ru-103	Ru-105	Ru-106	S-35	Sb-124	Sb-125
Se-75	Sm-145	Sm-151	Sn-113	Sn-121m	Sn-126
Sr-85	Sr-90	Sr-91	Tc-99	Te-129m	Te-132
Th-228	Th-229	Th-230	Th-232	Th-234	Tl-204
Tl-208	Tm-168	U-232	U-233	U-234	U-235

Table 1. (cont.) Radionuclides used at LLNL during 2019.

U-236	U-237	U-238	Xe-133	Xe-135	Y-88
Y-91	Y-93	Zn-65	Zr-88	Zr-89	Zr-93
Zr-95	Zr-97				

Table 2. Buildings at LLNL, by managing organization, where there is a potential for the release of radioactive materials to the air.

Director's Office	Physical & Life Sciences	Engineering	Weapons & Complex Integration	National Ignition Facility & Photon Science	Operations & Business
B253	B041 ^a	B231	B131HB	B165	B212
B254	B132 ^a	B321	B169 WAA	B298	B292
	B151	B322	B191	B381	B597
	B151 WAA	B327	B239	B391	
	B152	B341	B331 ^b	B490	
	B154		B332 ^b	B491	
	B190		B334	B581 ^b	
	B194		B612	B582	
	B235 ^b		B625		
	B262 ^a		B693		
	B282		B695/696 ^b		
	B361		B697		
	B364		B801 ^b		
	B378		B804		
	B379		B810A		
	B432		B810B		
			B836C		
			B836D		
			B851		

^a Managed by Global Security.

^b Continuous monitoring occurs at one or more exhaust points at the building.

Emissions Data

LLNL groups radionuclide emission sources into two categories; major sources or minor sources. Major sources are defined as those that have the potential to emit radionuclides that could result in an annual potential effective dose of 0.1 mrem (1 μ Sv) or more to a member of the public at an off-site location; the radionuclide NESHAPs regulation requires continuous monitoring of the stack effluent when the annual potential effective dose exceeds 0.1 mrem (1 μ Sv) to an off-site member of the public. Minor sources are defined as sources that do not have the potential to cause an annual effective dose of 0.1 mrem (1 μ Sv) to an off-site member of the public. At LLNL, all major sources of emissions are point sources, i.e., stack emission points; however, minor sources include both point sources and diffuse sources.

2.1 MAJOR SOURCES: MEASURED EMISSIONS

In 2019, there were five facilities at the Livermore Site and one facility at Site 300 that had radionuclide air effluent continuous monitoring systems. These facilities are listed in **Table 3**, along with the number of samplers, the types of samplers, and the analytes of interest. Some of these facilities have the potential to emit radionuclides that would cause an annual effective dose greater than the 0.1 mrem (1 μ Sv) NESHAPs standard; these sources are major sources following the definition given above. Other facilities have had the potential from radioactive air emissions in the past requiring monitoring, and the monitoring continues to be maintained to assure that the potential impact to the public and the environment is well understood. Additionally, monitoring may be in place for site-wide environmental impact statement commitments made to the public regarding the potential for radioactive air emissions.

Many of the monitored stacks at LLNL have effluent controls, such as HEPA filters, to collect materials before they are emitted to the atmosphere. Air samples for particulate emissions are extracted downstream of HEPA filters and prior to the discharge point to the atmosphere. Particles are collected on high efficiency cellulose membrane filters. The sample filters are removed and analyzed for radioactive particulate activity on a weekly or bi-weekly frequency depending on the facility. In all cases, continuous passive filter aerosol collection systems are used. At some facilities, continuous air monitors (CAMs) also sample the stack air exhaust for radionuclide activity. CAMs have an alarm capability in the event of an unplanned release of radionuclide activity. CAMs are used for facility personnel safety; they are not used for NESHAPs compliance demonstration.

Table 3. Air effluent sampling systems and locations.

Building	Facility	Analytes	Sample type	Number of samplers
235	Building in Physical and Life Sciences Directorate	Gross α , β on particles	Filter	1
331	Tritium Facility	Gaseous tritium/ tritiated water vapor	Ionization Chamber ^a	4
		Gaseous tritium/ tritiated water vapor	Glycol Bubblers	2
332	Plutonium Facility	Gross α , β on particles	Filters	15
		Gross α , β on particles	CAM ^a	14
581	National Ignition Facility	Gross α , β , Gamma suite on particles	Filter	1
		Radioiodine (volatile)	TEDA cartridge	1
		Gaseous tritium/ tritiated water vapor	Glycol Bubbler	1
		Gaseous tritium/ tritiated water vapor	Ionization Chamber ^a	1
695/696	Decontamination and Waste Treatment Facility	Gross α , β on particles	Filter	1
801A	Contained Firing Facility (Site 300)	Gross α , β on particles	Filter	1

Note: "CAM" denotes continuous air monitors. "TEDA" denotes triethylenediamine.

^a Alarmed systems used for notification for any unplanned release.

Gas flow proportional counters and gamma spectroscopy are used to detect radioactive particulate activity collected on the filters. For verification of the operation of the counting system, calibration sources, and background samples are interspersed among the sample filters for analysis. The Radiological Measurements Laboratory (RML) in LLNL's Radiation Protection Functional Area and the Environmental Monitoring Radio-analytical Laboratory (EMRL) in the Physical and Life Sciences Directorate perform the analyses.

When the result for gross alpha or gross beta on a particulate filter is greater than the minimum detectable concentration (MDC), the filter is recounted. If the second result is also above the MDC, the filter is submitted to the EMRL for isotopic analysis to determine whether the activity on the filter is the result of naturally occurring radionuclides or is reportable as a radionuclide emission from the facility.

Glycol bubblers are used to monitor for tritium releases from the two Tritium Facility (Building 331) stacks, and the National Ignition Facility (NIF) stack. In addition to this NESHAPs compliance monitoring, the two Tritium Facility stacks, and the NIF stack are monitored using ion chambers. The ion chamber monitors are set to alarm at designated tritium concentrations to identify accidental or off-normal releases. Ion

chambers are in place for notification only so that any unplanned release may be detected and responded to real-time; all the stack samplers monitor continuously.

Because tritium can be released in the form of either tritiated water vapor (HTO) or tritiated hydrogen gas (HT), glycol bubblers employ a two-stage glycol impinging process to capture both physical forms. Stack air to be sampled enters the instrument and flows through the first stage impingers, capturing the HTO present. Next, the sampled air is directed through a heated palladium catalyst where oxidation of any HT in the sample takes place, converting gaseous tritium to HTO, which is then collected in the second stage impingers. The impingers are analyzed by the RML using liquid scintillation analysis. This type of sampling quantifies the amount of tritium for both species, HT and HTO.

Tritium in particulate form is monitored when required using high efficiency (better than HEPA grade) cellulose membrane filters. Measurements indicate that tritium exchange (adsorption of tritium in HTO and/or HT captured in the filter medium via a binding reaction) occurs; this was verified by placing two particulate filters in series and comparing the results (applying 2-sigma error) on each filter. If tritiated particulate were present, then the pre-stream filter (the filter that comes into contact with stack air first) would have a higher amount of tritium than the post-stream filter (the filter that comes into contact with stack air immediately after the pre-stream filter). This is because the pre-stream filter would have significantly filtered out particles prior to the post-stream; this would result in less tritium on the post-stream filter. The pre-stream filter and post-stream filter showed the same amount of tritium (within analytical error) indicating tritium exchange.

Triethylenediamine (TEDA) cartridges are used to sample for radioactive iodines in gaseous or vapor state. The TEDA is impregnated into carbon (activating the carbon) by the manufacturer and is housed in a plastic cartridge of standard industry size 2 ¼" diameter by 1" thick (30 × 50 Mesh). Stack air is directed through the TEDA cartridge, which is the second stage of the two-stage filter housing. Both the particulate filter (first stage) and the TEDA cartridge (second stage) are counted by gamma spectroscopy by the EMRL (see **Table 3**).

In 2019, stack measurements indicate that a total of 126.4 Ci (4677 GBq) of tritium was released from the Tritium Facility (B331). Of this, approximately 80% of tritium was released as HTO. The remaining 20% was HT.

The NIF (B581) 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 HT.

The Contained Firing Facility (B801A) at Site 300 had measured depleted uranium stack emissions in 2019 consisting of 1.2×10^{-7} Ci (4.4×10^{-6} GBq) of uranium-234, 1.7×10^{-8} Ci (6.4×10^{-7} GBq) of uranium-235, and 9.2×10^{-7} Ci (3.4×10^{-5} GBq) of uranium-238 in particulate form.

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

2.2 MINOR SOURCES: AMBIENT MEASUREMENT COMPARISON

With EPA's Region IX approval, LLNL uses ambient air monitoring data to demonstrate compliance for minor emissions sources (both non-monitored stack and area sources). The method entails comparing measured ambient air concentrations at the location of the site-wide maximally exposed individual (SW-MEI) to concentration limits set by the EPA in its Table 2 of Appendix E to 40 CFR 61. Comparisons are made for tritium and plutonium 239+240 for the Livermore SW-MEI and uranium-238 for the Site 300 SW-MEI (see **Table 6** in Section 3.3.2).

2.3 MINOR SOURCES: SOURCE TERM ESTIMATE

LLNL estimates the source terms for Livermore Site minor sources, both diffuse and non-monitored point, using a mathematical optimization routine that minimizes the root-mean-square (*rms*) differences between modeled and measured annual average ambient tritium concentrations (MacQueen et al. 2013; see **Figure 5** in Section 3.3.2 of this report for the ambient air sampling locations). This process has two parts.

First, CAP88-PC is used to model the contribution each source (monitored point, non-monitored point and diffuse) makes to the annual average tritium concentration in HTO at each of the ambient air monitoring locations. Each monitored point source is modeled using source-specific model parameters (activity, stack height, etc.). Each non-monitored point source is modeled using a unit source activity of 1 Ci (37 GBq) and stack-specific model parameters (stack height, etc.). Each diffuse source is modeled using a unit source activity, a 1-meter height, a 10-meter diameter, and a fixed plume rise across stability classes A through F. All models use the same LLNL 2019 wind file (see **Attachment 2**). The individual contributions (monitored point, non-monitored point, and diffuse) at each monitoring location are added to produce an all-sources-combined model estimate of the annual average ambient concentration at each location.

Second, the 1 Ci (37 GBq) source term for each non-monitored point and diffuse source is adjusted independently using the mathematical search routine. The source terms of the monitored point sources are held fixed with the measured source terms. The adjusted source terms that produce the best fit of the all-sources-combined model concentrations to the measured concentrations are then used to calculate the dose contribution from each non-monitored point and diffuse source.

In 2019, the best-fit source term estimates were 0.27 Ci (10 Gbq) for the Building 169 Waste Accumulation Area (WAA), 0.26 Ci (9.6 GBq) for the Building 298 area activities, 1.4 Ci (51.8 GBq) for the Building 331 WAA. The measured and best-fit model annual average ambient air concentrations are shown in **Figure 3**.

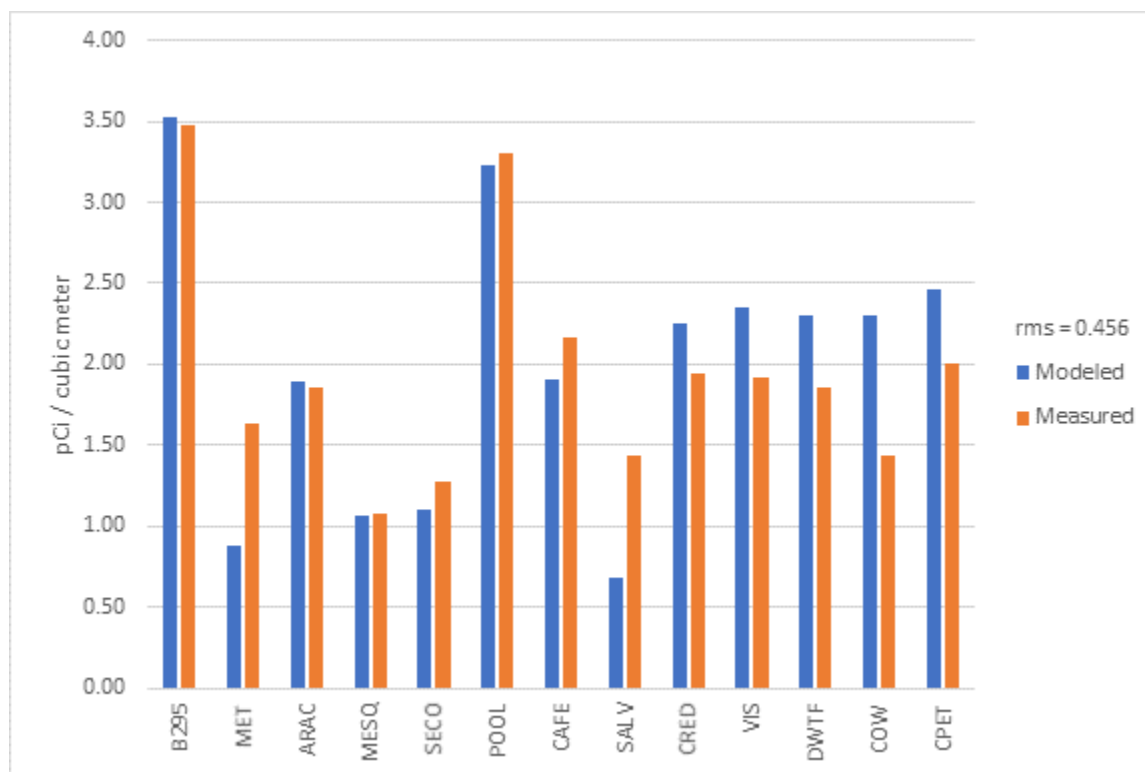


Figure 3. Comparison of measured and modeled annual average concentrations of tritiated water vapor (HTO) in units of pCi/m³ in air at Livermore Site ambient air locations, 2019.

2.4 MINOR SOURCES: OPEN-AIR TESTS

Another potential source of radioactive air emissions from LLNL operations at Site 300 is the emission of materials from open-air explosives tests. In 2019, there were no open-air explosives tests that contained radioactive materials.

Dose Assessment

3.1 GENERAL

To comply with NESHAPs regulations and DOE guidance, the EPA-approved atmospheric dispersion and radiation dose calculation computer code, CAP88-PC, Version 4.0.1.17, was used to calculate the dose at specific distances from release points. For diffuse sources having a significant contribution to total dose, in addition to comparing the emissions to the concentration limits set by the EPA in its Table 2 of Appendix E to 40 CFR 61, doses were calculated using either CAP88-PC or DOE standard (DOE-STD-1196-2011) breathing rates and dose conversion factors.

For LLNL to comply with the NESHAPs regulations, the SW-MEI cannot receive an effective dose equivalent greater than 10 mrem/y (100 μ Sv/y) per site. A SW-MEI is defined as a *hypothetical* member of the public at a single residence, school, business, church, or other such facility who receives the greatest LLNL induced dose from the combination of all evaluated radionuclide source emissions, as determined by modeling. At the Livermore Site, the 2019 SW-MEI is located at the Integrative Veterinary Care (CPET) facility, which is approximately 115 feet (35 m) outside the eastern fence line of the site. At Site 300, the 2019 SW-MEI is located at the Site 300 boundary with the Carnegie State Vehicle Recreation Area, managed by the California Department of Parks and Recreation, approximately 1.9 miles (3.2 km) south-southeast of the firing table at Building 851. The locations of the SW-MEI's for both LLNL sites are shown in **Figure 4**.

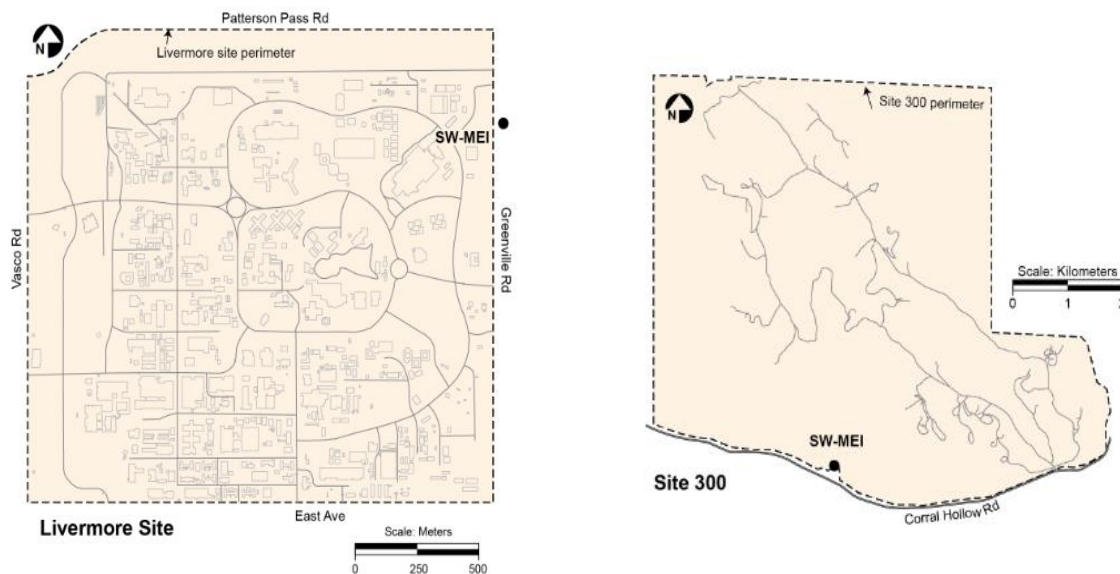


Figure 4. Location of Site-Wide Maximally Exposed Individual (SW-MEI) at the Livermore Site and Site 300, 2019.

3.2 CAP88-PC INPUT PARAMETERS

Input parameters to CAP88-PC include the emissions discussed in Section 2, and building-specific and common parameters, discussed below. To estimate dose, CAP88-PC, Version 4.0.1.17, provides a library of the radionuclides at LLNL. In addition, when calculating dose from particulate alpha- and beta-emitting radionuclides, LLNL assigns gross alpha and gross beta measurements to the radionuclides handled in the facility when they can be specifically identified, or to plutonium-239+240 and strontium-90, respectively. The use of plutonium-239+240 and strontium-90 to represent alpha and beta emissions provides a health-conservative estimate of the dose.

3.2.1 Building-Specific Parameters

For dose assessment, LLNL uses building-specific information about radionuclide releases, as well as building-specific parameters for stack height, stack exhaust rate, stack diameter, and distances to the fence line. The building specific parameters are presented in **Attachment 1**.

3.2.2 Common Parameters

The input parameters that are common among LLNL sources are the agricultural parameters. Meteorological data from the LLNL Livermore Site meteorological tower are used to model Livermore Site sources, and meteorological data from the LLNL Site 300 meteorological tower are used to model Site 300 sources (see **Section 1.1**). The CAP88-PC default for absolute humidity of 8 grams per cubic meter was used as a reasonable representation of conditions at LLNL. The value for lid (mixing) height of 2,461 feet (750 m) was chosen for the Livermore Site, whereas the lid height value for Site 300 was 3,281 feet (1,000 m). The 2019 wind data and wind file are provided in **Attachment 2**.

For agricultural parameters in CAP88-PC, LLNL used area-weighted mean values based on data from the U.S. Department of Agriculture (USDA 2012) and the U.S. Census Bureau (USCB 2014) for periodic update. These were last updated in 2014.

County-specific agricultural data were obtained from the USDA sources. The borders of California counties were downloaded from the USCB website. For each county that intersects an 80 km radius circle around LLNL, the area of the overlapping portion of the county was calculated using geographic information system (GIS) software. Each of these areas was divided by the sum of the areas to determine the proportion of the 80 km radius circle representing each county. These proportions were then used to calculate the area weighted averages of the county-specific agricultural data. The results are shown in **Table 4**.

Table 4. Agricultural parameter values representing LLNL used in CAP88-PC.

Parameter	Value
Beef cattle density (number of cows/km ²)	4.02
Milk cattle density (number of cows/km ²)	13.16
Land fraction cultivated for vegetable crops	0.0021

For individual and collective doses from ingestion, it was assumed that 100% of milk is imported (i.e., free from LLNL-generated radioactivity), and that vegetables and meat are 25% home-grown and 75% imported.

3.2.3 Distances and Directions

In 2014, LLNL reviewed and updated its database of distances from sources to the fence-line and potential receptor locations using GIS and Google Earth software. Adjustments of up to about 15 meters were made to a few source locations. No further adjustments have been made.

3.3 COMPLIANCE ASSESSMENT

3.3.1 Major Sources

Doses from LLNL's major sources, which are point sources, were evaluated using CAP88-PC and the input parameters discussed above. The modeled doses to the SW-MEI for the facilities where there were measured emissions are shown in **Table 5**. The specific results for all monitored sources are provided in **Attachment 1**.

Table 5. Monitored point source doses for 2019.

Site	Facility	Dose (mrem)
Livermore Site	Tritium Facility (B331)	3.9×10^{-3}
Livermore Site	NIF (B581)	1.6×10^{-4}
Site 300	CFF (B801A)	9.5×10^{-8}

3.3.2 Minor Sources

LLNL has many minor sources; that includes point sources and diffuse sources. As stated previously, with EPA's Region IX approval, LLNL demonstrates compliance for minor emission sources (both diffuse and non-monitored stack effluent sources) through the comparison of ambient air monitoring data with concentration limits set by the EPA in Table 2 of Appendix E to 40 CFR 61. This is done for tritium and plutonium-239+240 for the Livermore SW-MEI and uranium-238 for the Site 300 SW-MEI.

The 2019 average monitoring results for tritium and plutonium from the sampling location at the Livermore Site SW-MEI (CPET) were used for the purposes of this minor source comparison. (See **Figure 5** for a map of all Livermore Site sampling locations). The results of these comparisons are shown in **Table 6**. In 2019, there were no plutonium-

239+240 results above the detection limit at the CPET location. All results for plutonium-239+240 air samples in 2019 were below the detection limit at the SW-MEI location.

At Site 300, wind-driven re-suspension of soil contaminated with depleted uranium is in the minor source category. In 2019, but in contrast to previous years, there were no ambient air measurements for uranium that showed a contribution from depleted uranium at the SW-MEI (see **Figure 4** for the location of the Site 300 SW-MEI). The uranium-238 value in **Table 6** represents a natural background value (see footnote c).

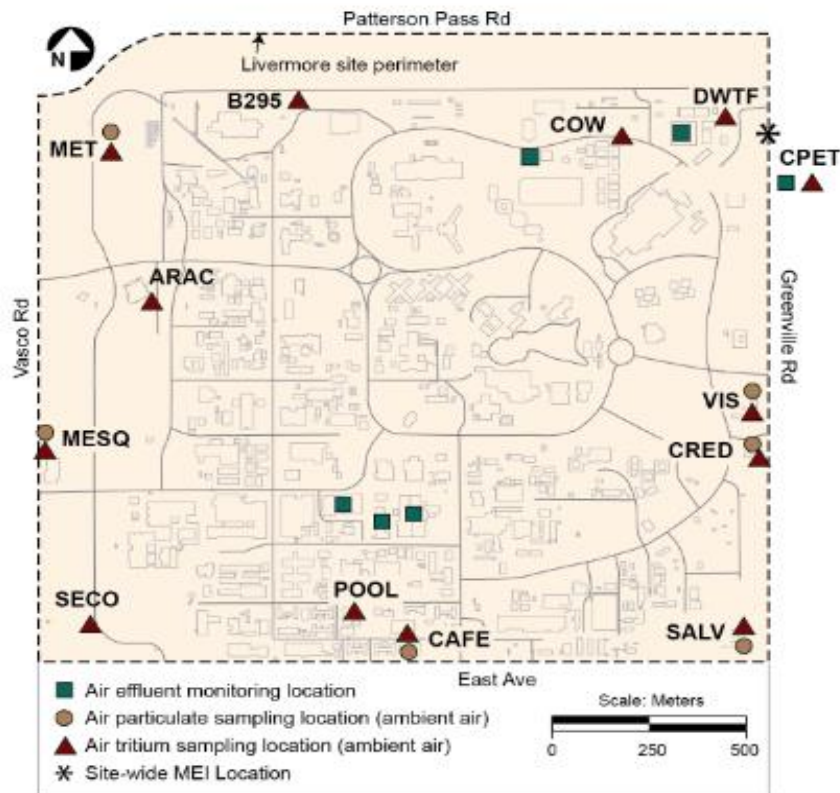


Figure 5. Radiological air monitoring locations at the Livermore Site.

The measured concentrations at the SW-MEI are presented in **Table 6**. Also shown in **Table 6** are the EPA's standards from Table 2 of Appendix E to 40 CFR 61. As demonstrated by the calculation of the fraction of the standard, LLNL's measured concentrations in air for tritium, plutonium-239+240, and uranium-238 are all less than one percent of the standard for these radionuclides.

Table 6. Mean concentrations of radionuclides of at the location of the SW-MEI in 2019 compared to EPA's concentration standard.

Location	Nuclide	EPA's Table 2 concentration standard	Mean measured concentration	Measured concentration as a fraction of the standard	Detection limit
Livermore Site SW-MEI	Tritium	1.5×10^{-9} Ci/m ³	2.7×10^{-12} Ci/m ^{3a}	1.8×10^{-3}	1×10^{-12} Ci/m ³
Livermore Site SW-MEI	Plutonium-239	2.0×10^{-15} Ci/m ³	Non-detect ^b	Non-detect ^b	5×10^{-19} Ci/m ³
Site 300 SW-MEI	Uranium-238	8.3×10^{-15} Ci/m ³	1.2×10^{-17} Ci/m ^{3c}	1.4×10^{-3}	3×10^{-20} Ci/m ³

^a The measured tritium value includes contributions from all major and minor sources including stack and diffuse releases measured at the location of the SW-MEI.

^b All results for Pu-239 at the SW-MEI location were below the analytical detection limit in 2019.

^c The average ratio of uranium-235 and uranium-238 concentrations for 2019 is 0.0072 at the SW-MEI location, which is consistent with naturally occurring uranium. This value for uranium-238 is from naturally occurring uranium resuspended to ambient air from the soil.

The source terms for the diffuse sources of tritium were developed as described in **Section 2.3**, and the doses were calculated using CAP88-PC. The total diffuse dose plus non-monitored stack source dose for 2019 at the SW-MEI location was 1.9×10^{-4} mrem (1.9×10^{-3} μSv) for the Livermore Site. Because there was no diffuse source term for Site 300, no minor diffuse dose was calculated. **Attachment 1** lists the doses from diffuse sources.

3.3.3 SW-MEI Dose

Doses from LLNL's airborne emissions are well below the 10 mrem (100 μSv) NESHAPs annual dose standard. The annual doses (point source plus diffuse source) to the SW-MEI at the Livermore Site and at Site 300 are:

- Livermore Site: 4.3×10^{-3} mrem (4.3×10^{-2} μSv)
- Site 300: 9.5×10^{-8} mrem (9.5×10^{-7} μSv)

The EPA-approved software calculates the dose assuming a person resides there all year for 24 hours a day and eats a specified fraction of meat and vegetables grown at the SW-MEI location (**Section 3.2.2**). Thus, the calculated dose to the SW-MEI is a hypothetical conservative dose in comparison to an actual resident.

Table 7 presents 2019 doses with those of previous years. Diffuse source doses were not reported for the Livermore Site for 1990 and 1991 and were not reported for Site 300 for 1990 through 1992.

Table 7. Doses (in mrem) calculated for the Site-Wide Maximally Exposed Individual (SW-MEI) for the Livermore site and Site 300, 1990 to 2019.

Year	Total Dose	Point Source Dose	Diffuse & Non-monitored Stack Source Dose
Livermore Site			
2019	4.3×10^{-3}	4.1×10^{-3}	1.9×10^{-4}
2018	6.7×10^{-3}	6.46×10^{-3}	2.49×10^{-4}
2017	1.9×10^{-3}	1.5×10^{-3}	3.6×10^{-4}
2016	2.8×10^{-3}	2.6×10^{-3}	2.3×10^{-4}
2015	1.7×10^{-3}	1.4×10^{-3}	3.3×10^{-4}
2014	2.7×10^{-3}	2.4×10^{-3}	2.8×10^{-4}
2013	1.8×10^{-3}	1.61×10^{-3}	1.44×10^{-4}
2012	0.0054 ^a	0.005 ^a	0.00041
2011	0.017 ^a	0.015 ^a	0.0019
2010	0.011 ^a	0.0033 ^a	0.0074
2009	0.0042 ^a	0.0015 ^a	0.0027
2008	0.0013 ^a	0.00033 ^a	0.00095
2007	0.0031 ^a	0.0013 ^a	0.0018
2006	0.0045 ^a	0.0016 ^a	0.0029
2005	0.0065 ^a	0.0027 ^a	0.0038
2004	0.0079 ^a	0.0021 ^a	0.0058
2003	0.044 ^a	0.024 ^a	0.02
2002	0.023 ^a	0.010 ^a	0.013
2001	0.017 ^a	0.0057 ^a	0.011
2000	0.038 ^a	0.017 ^a	0.021
1999	0.12 ^a	0.094 ^a	0.028
1998	0.055 ^a	0.031 ^a	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	0.04	0.026
1992	0.079	0.069	0.01
1991	0.234	— ^b	— ^b
1990	0.24	— ^b	— ^b

Table 7. (cont.) Doses (in mrem) calculated for the Site-Wide Maximally Exposed Individual (SW-MEI) for the Livermore Site and Site 300, 1990 to 2019.

Year	Total Dose	Point Source Dose	Diffuse Source Dose
Site 300			
2019	9.5×10^{-8}	9.5×10^{-8}	— ^c
2018	9.6×10^{-5}	9.9×10^{-8}	9.6×10^{-5}
2017	4.8×10^{-5}	2.8×10^{-7}	4.8×10^{-5}
2016	2.2×10^{-4}	9.3×10^{-8}	2.2×10^{-4}
2015	4.8×10^{-4}	1.3×10^{-7}	4.8×10^{-4}
2014	7.1×10^{-8}	7.1×10^{-8}	— ^c
2013	4.0×10^{-8}	4.0×10^{-8}	— ^c
2012	1.3×10^{-6}	1.3×10^{-6}	— ^c
2011	9.0×10^{-8}	9.0×10^{-8}	— ^c
2010	5.7×10^{-7}	5.7×10^{-7}	— ^c
2009	2.7×10^{-7}	2.7×10^{-7}	— ^c
2008	4.4×10^{-8}	4.4×10^{-8}	— ^c
2007	0.0035	0.0031	0.00035
2006	0.016	0.014	0.002
2005	0.018	0.0088	0.0094
2004	0.026	0.025	0.00086
2003	0.017	0.017	0.00034
2002	0.021	0.018	0.0033
2001	0.054	0.05	0.0037
2000	0.019	0.015	0.0037
1999	0.035	0.034	0.0012
1998	0.024	0.019	0.005
1997	0.02	0.011	0.0088
1996	0.033	0.033	0.00045
1995	0.023	0.02	0.003
1994	0.081	0.049	0.032
1993	0.037	0.011	0.026
1992	0.021	0.021	— ^d
1991	0.044	0.044	— ^d
1990	0.057	0.057	— ^d

- ^a The dose includes HT emissions modeled as HTO. Modeling HT emissions as such results in an overestimation of the dose. This methodology is used for purposes of compliance, as directed by EPA Region IX. Beginning in CY13, the EPA NESHAPs compliance modeling code CAP88-PC Version 4.0.1.17 now allows tritium to be modeled in both species, HT and HTO.
- ^b Point and diffuse source doses were not reported separately from the total dose for the Livermore Site for 1990 and 1991.
- ^c Diffuse emissions dose was not calculated because ambient monitoring did not yield SW-MEI results indicating the presence of depleted uranium. This is primarily due to no atmospheric shots with DU since 2008, and no significant amounts of contaminated soil disturbance at the location of the SW-MEI.
- ^d No diffuse emissions were evaluated at Site 300 for years before 1993.

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: Lisa M. Woodrow
Associate Director
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Lawrence Livermore National Laboratory
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Livermore, CA 94551

Signature: _____ **Date:** _____
Lisa M. Woodrow

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: Tom Grim
Assistant Manager for Environment, Safety & Health
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Signature: _____ **Date:** _____
Tom Grim

Additional Information

5.1 UNPLANNED RELEASES

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 (7.4×10^{-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 accidental valve misalignment from becoming a reoccurring event.

The unplanned release of 0.00002 Ci (7.4×10^{-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 (7.4×10^{-4} GBq) source term. The dose to the SW-MEI (CPET) was 5.3×10^{-5} mrem (5.3×10^{-4} μ 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 (1.5×10^{-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 (1.5×10^{-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 Ci (1.5×10^{-4} GBq) source term. The dose to the SW-MEI (CPET) was 1.1×10^{-6} mrem (1.1×10^{-5} μ 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.

Supplemental Information

6.1 COLLECTIVE DOSE ASSESSMENT

Collective population dose is calculated using CAP88-PC as the average radiation dose to a person in a specified area, multiplied by the number of people in that area. In accordance with DOE and EPA guidance documents, all radionuclides potentially emitted in 2019 were assumed to be released from a central location. The total population within 50 miles (80 km) of the Livermore Site is approximately 7.8 million, and the total population within 50 miles (80 km) of Site 300 is approximately 7.1 million. The populations were derived using Oak Ridge National Laboratory LandScan 2010 data and ESRI ARCMAP software. The population file is provided in Attachment 3. The estimated collective dose attributable to LLNL airborne emissions in 2019 to persons living within 50 miles (80 km) of the Livermore Site is 0.33 person-rem (0.0033 person-Sv) and to persons living within 50 miles (80 km) of Site 300 is 2.9×10^{-5} person-rem (2.9×10^{-7} person-Sv).

6.2 40 CFR 61 SUBPARTS Q AND T

LLNL does not have storage and disposal facilities for radium containing materials that would be a significant source of radon. LLNL does not have or store any uranium mill tailings.

6.3 PERIODIC CONFIRMATORY MEASUREMENT

Results of NESHAPs periodic confirmatory measurements (PCM) are intended to support or confirm two objectives: 1) that those operations not continuously monitored do not, in fact, need to be continuously monitored and 2) that radionuclide usage-inventory-based estimates of emissions and their corresponding doses are conservative.

For sources evaluated to have a potential to result in a dose less than the regulatory value of 0.1 mrem/y that requires continuous monitoring under Subpart H, LLNL achieves the PCM objectives by fulfilling the requirements stated in 40 CFR 61.93, paragraph (e) with its ambient air monitoring program. The ambient air monitoring effort includes more than 30 sampling locations with more than 40 samplers placed in strategic areas (see the Air Monitoring Programs section in the LLNL Site Annual Environmental Report [<https://saer.llnl.gov/>] for a description of LLNL's ambient air radiological monitoring).

6.4 FACILITY COMPLIANCE

In 2019, LLNL maintained its compliance with 40 CFR 61 Subpart H. All emissions resulted in calculated doses well below the 10 mrem (100 μ Sv) standard.

Attachment 1 provides the dose estimates for each individual source.

References

EPA 1989: U.S. Environmental Protection Agency, National Emission Standard for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities, 40 CFR Part 61, Subpart H (1989, as amended).

MacQueen, D., N. Bertoldo, and A. Wegrecki (2013). "A Best Fit Approach to Estimating Multiple Diffuse Source Terms Using Ambient Air Monitoring Data and an Air Dispersion Model." Operational Radiation Safety, Vol. 105, suppl. 2, August 2013.

LandScan™ Global Population Database, 2010,
http://www.ornl.gov/sci/landscan/landscan_documentation.shtml

National Council on Radiation Protection and Measurements (NCRP), Principles and Application of Collective Dose in Radiation Protection, NCRP Report No. 121 (1995).

USDA 2012. United States Department of Agriculture. The Census of Agriculture. 2012.
http://www.agcensus.usda.gov/Publications/2012/Full_Report/Volume_1,_Chapter_1_US/usv1.pdf (Issued May 2014).

Errata

There were no errata found in the calendar year 2018 report.

Attachments

ATTACHMENT 1: 2019 LLNL NESHAPs Annual Report Spreadsheet (11 x 17 table) – Placeholder page

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A.2.2 SITE 300 TOWER

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ATTACHMENT 3: POPULATION DATA

The source of the geographic population distribution data used for this report is Oak Ridge National Laboratory (ORNL) LandScan™ 2010 data and ESRI ARCMAP software. The data are placed into an annular grid that is created from sixteen 22.5-degree sectors centered on the cardinal wind directions and five distances spaced at 16 km to a total 80-km radius. In deriving the population for each site, the ORNL data set is input into ESRI ARCMAP with the 80-km grid for the Livermore Site centered at 37.686 N latitude, -121.7045 W longitude (near the center of the site) and Site 300 centered at the 52-m meteorological tower located at 37.675 N latitude, -121.541 W longitude. The first line of the input file is informational. Distances are shown in the second row. Population data begin in the third row starting with direction, N. There are 20 spaces reserved for each direction no matter how many are used, i.e., the next direction, NNW, starts approximately half-way through the fifth row, 21 values after the first value.

[illegible]

[illegible]

ATTACHMENT 4: UNITS OF MEASURE AND EQUIVALENTS

Symbols and Units of Measure

Bq	becquerel	mph	mile per hour
GBq	gigabecquerel (10^9 Bq)	mrem	millirem (10^{-3} rem)
°C	degree centigrade	m/s	meter per second
Ci	curie	mrem/y	millirem per year (10^{-3} rem/y)
cm	centimeter	pCi	picocurie (10^{-12} Ci)
°F	degree Fahrenheit	pCi/m ³	picocurie (10^{-12} Ci) per meter cubed
in.	inch	person-Sv	person-sievert
km	kilometer	Sv	sievert
m	meter	μSv	microsievert
mi	mile	μSv/y	microsievert per year

Metric and U.S. Customary Unit Equivalents

Category	From metric unit to U.S. customary equivalent unit		From U.S. customary unit to metric equivalent unit	
	Metric	U.S.	U.S.	Metric
Length	1 centimeter (cm)	0.39 inches (in.)	1 inch (in.)	2.54 centimeters (cm)
	1 millimeter (mm)	0.039 inches (in.)		25.4 millimeters (mm)
	1 meter (m)	3.28 feet (ft)	1 foot (ft)	0.3048 meters (m)
		1.09 yards (yd)	1 yard (yd)	0.9144 meters (m)
	1 kilometer (km)	0.62 miles (mi)	1 mile (mi)	1.6093 kilometers (km)
Volume	1 liter (L)	0.26 gallons (gal)	1 gallon (gal)	3.7853 liters (L)
		8.11×10^{-7} acre-feet	1 acre-foot	1.23×10^6 liters (L)
	1 cubic meter (m ³)	35.32 cubic feet (ft ³)	1 cubic foot (ft ³)	0.028 cubic meters (m ³)
		1.35 cubic yards (yd ³)	1 cubic yard (yd ³)	0.765 cubic meters (m ³)
Weight	1 gram (g)	0.035 ounces (oz)	1 ounce (oz)	28.6 gram (g)
	1 kilogram (kg)	2.21 pounds (lb)	1 pound (lb)	0.454 kilograms (kg)
	1 metric ton (MT)	1.10 short ton (2000 pounds)	1 short ton (2000 pounds)	0.90718 metric ton (MT)
Area	1 hectare (ha)	2.47 acres	1 acre	0.40 hectares (ha)
Radioactivity	1 becquerel (Bq)	2.7×10^{-11} curie (Ci)	1 curie (Ci)	3.7×10^{10} becquerel (Bq)
Radiation dose	1 gray (Gy)	100 rad	1 rad	0.01 gray (Gy)
Radiation dose equivalent	1 sievert (Sv)	100 rem	1 rem	0.01 sievert (Sv)
Temperature	°Fahrenheit = (°Centigrade $\times$ 1.8) + 32		°Centigrade = (°Fahrenheit – 32) / 1.8	