5. Air Effluent Monitoring

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Introduction

Air effluent emissions from facility operations are assessed to evaluate compliance with local, state, and federal regulations and to ensure that human health and the environment are protected from hazardous and radioactive air emissions. LLNL complies with local, state, and federal environmental air quality laws and DOE regulations previously discussed in Chapter 4. In general, LLNL analyzes for most constituents at levels that are far below regulatory standards in order to determine any environmental impact. Air surveillance measurements (see Chapter 4) are also made to assess environmental impact.

Assessment of air effluent emissions is performed by monitoring emissions and/or evaluating potential emissions. Currently, the air effluent sampling program measures only radiological emissions. LLNL has operations with the potential for nonradiological discharges; however, permits for these operations are obtained through local agencies having enforcement authority for the Clean Air Act, and stack monitoring is not required. The agencies governing LLNL compliance are EPA Region IX, the Bay Area Air Quality Management District (BAAQMD) for the Livermore site and the San Joaquin Valley Unified Air Pollution Control District (SJVUAPCD) for Site 300.

Historically, monitoring of radionuclide air effluents at LLNL has been implemented according to the DOE ALARA (as low as reasonably achievable) policy. This policy is meant to ensure that DOE facilities have the capabilities consistent with the types of operations to monitor routine and nonroutine radiological releases, so that the dose to members of the public can be assessed and that doses are ALARA. The more recent National Emission Standards for Hazardous Air Pollutants (NESHAPs) 40 CFR 61, Subpart H regulations require that monitoring of facility radionuclide air effluents must be performed if the potential off-site dose equivalent is greater than 1 μSv/y (0.1 mrem/y), as calculated using the EPA-mandated air dispersion dose model and assuming no emission control devices. Air effluent monitoring provides the actual source term for modeling to ensure that the NESHAPs standard, 100 μSv/y (10 mrem/y) total site effective dose equivalent, is not exceeded. Discharges that have a potential to release radionuclides from operations but that are not monitored are also evaluated according to the NESHAPs regulations.

A wide variety of radioisotopes are used for research purposes at LLNL, including transuranics, biomedical tracers, tritium, mixed fission products, and others. The major radionuclide released to the atmosphere from the Livermore

site is tritium. In addition to effluent sampling for tritium, a number of facilities at the Livermore site have air effluent samplers to detect the release of transuranic aerosols. The air effluent sampling systems described in this chapter apply to stationary and point source discharges. Sampling methods to evaluate LLNL diffuse sources are described in Chapter 4, Volume 2.

LLNL conducts air effluent monitoring at atmospheric discharge points of some facilities to determine the actual emissions from individual facilities and to confirm the operation of emission control systems. Air monitoring involves measurement of particles collected on filters or of vapor chemically trapped on a collection medium. Concentrations of various airborne radionuclides (including particles and tritiated water vapor) are measured at the Livermore site. Point sources as well as diffuse, or nonpoint sources, are monitored to fulfill NESHAPs requirements.

Methods

For air effluent monitoring, LLNL maintains 103 radionuclide samplers on air exhausts at 9 facilities at the Livermore site (see **Figure 5-1**). These systems are listed in **Table 5-1** along with the analytes of interest, the type of sampler, and the number of samplers and discharge points monitored. Sampling for particles containing radionuclides is conducted in eight of the facilities; sampling for tritium is conducted in one facility. All sampling systems operate continuously. Samples are collected weekly or biweekly depending on the facility. Air samples for particulate emissions are extracted downstream of high efficiency particulate air (HEPA) filters and prior to the discharge point to the atmosphere. Particles in the extracted air are collected on sample filters and analyzed for gross alpha and beta activity. Tritium is collected using molecular sieves. In addition to sample collection for environmental reporting, some facilities have real-time monitors at discharge points to provide faster notification in the event of a release of radioactivity. Further details of LLNL air effluent sampling systems are included in Chapter 4 of the *Environmental Monitoring Plan* (Tate et al. 1995).

The need for continuous air effluent monitoring at other air discharge points that can potentially release radionuclides to the atmosphere is evaluated according to the NESHAPs regulations. The evaluation is based on estimated releases using radionuclide inventories specific to individual discharge points and does not take into account reduction by emission control systems (according to the regulations). The most recent NESHAPs evaluation for LLNL operations is reported in the LLNL NESHAPs 1995 Annual Report (Gallegos et al. 1996). Many of the existing sampling systems now in place (**Table 5-1**) are not required by law; however, LLNL has continued to operate these systems as a best management practice.

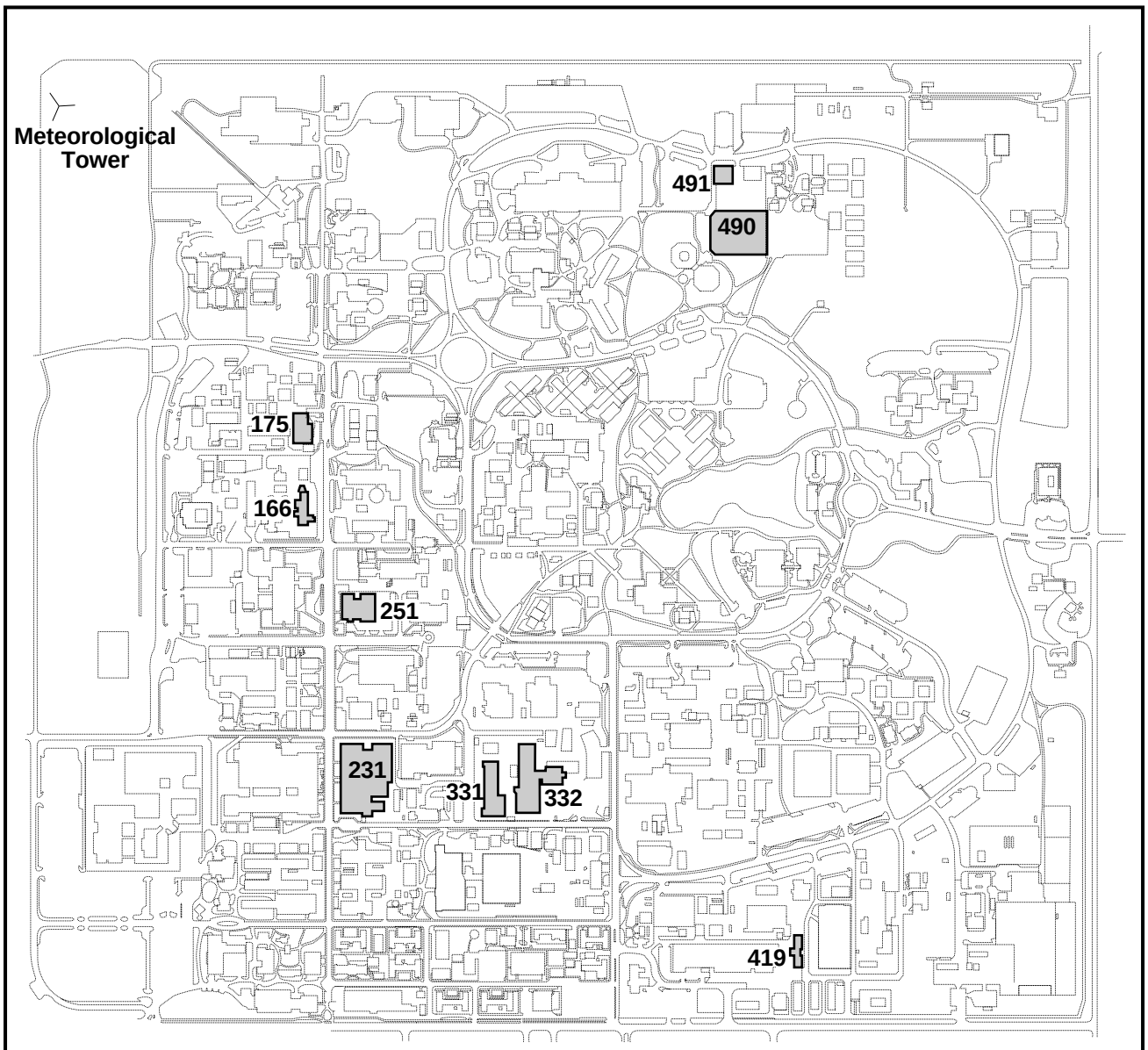

Figure 5-1. Buildings at the LLNL site having air monitoring systems for effluent gas streams.

The California Air Toxics "Hot Spots" legislation requires facilities to prepare an air toxics emissions inventory and risk assessment, which LLNL has completed. Based on these data, the BAAQMD and the SJVUAPCD have ranked LLNL as a low-risk facility. Each year LLNL completes a review of the air toxics inventory and updates the annual permit. Currently, nonradiological emissions (with the exception of beryllium) are permitted through the local air districts and air toxics monitoring is not required.

5. Air Effluent Monitoring

In 1995, a new radiological sampling system was installed in Building 166 to monitor emissions from glove-box operations. The system was installed based on a NESHAPs assessment of operations involving uranium that were begun in early 1995. In addition, 8 new filter type samplers were installed, 4 in the Building 251 hardened area and 4 in Building 332 as part of improvements made to LLNL sampling systems. Release points where these samplers were installed had existing sampling systems; the added samplers provide increased sampling capabilities.

All analytical results are reported as a measured concentration per volume of air, or at the minimum detection concentration (MDC) when no activity is detected. In all cases, the MDC is more than adequate for demonstrating compliance with the pertinent regulatory requirements for radionuclides that are present or may be present in the air sample.

Table 5-1. Air effluent sampling locations and systems.

Building	Facility	Analytes	Sample type	Number of samplers	Number of discharge points
166	Pyrochemistry demonstration facility	Gross α, β on particles	Filters	1	1
175	MARS	Gross α, β on particles	Filters	6	6
231	Vault	Gross α, β on particles	Filter	1	1
251	Heavy elements				
	Unhardened area	Gross α, β on particles	Filters	44	55[a]
	Hardened area	Gross α, β on particles	CAM[b]	4	4
		Gross α, β on particles	Filters	4	4
331	Tritium	Tritium	Ionization chamber[b]	4	4
		Gaseous tritium and tritiated water vapor	Molecular sieves	4	2
332	Plutonium	Gross α, β on particles	CAM[b]	12	11
		Gross α, β on particles	Filters	16	11
419	Decontamination	Gross α, β on particles	Filters	2	2
490	Laser isotope separation	Gross α, β on particles	Filters	4	4
491	Laser isotope separation	Gross α, β on particles	Filters	1	1

Note: "CAM" denotes Eberline continuous air monitors.

a Alternate blower system measured by the same sampler.

b Alarmed systems.

Results: Measured Emissions

This section discusses the air effluent monitoring results at the Livermore site.

Livermore Site

Radioactive Air Emissions

Actual measurements of air radioactivity and effluent flow are the basis for reported emissions from continuously monitored sources. LLNL facilities that have continuously monitored discharge points are Buildings 166, 175, 231-vault, 251, 331, 332, 419, 490, and 491.

Tritium emissions from operations at the Tritium Facility (Building 331) account for nearly all the radioactive discharges to the atmosphere from monitored facilities. In 1995, operations there released a total of 3.4×10^{12} Bq (92 Ci) of tritium, or approximately 97% of the tritium released from the Livermore site. Of this, approximately 2.3×10^{12} Bq (63 Ci) were released as tritiated water (HTO). The remaining tritium released, 1.1×10^{12} Bq (29 Ci), was elemental tritium gas. The highest single weekly stack emission from the facility was 1.4×10^{11} Bq (3.8 Ci), of which 5.6×10^{10} Bq (1.5 Ci) was tritiated water. The potential dose from tritium gas is approximately 25,000 times lower than the dose from a comparable release of tritiated water. Therefore, the tritiated hydrogen gas did not contribute significantly in calculations of the overall tritium dose. Building 331 tritium emissions over the period 1981 to 1995 are shown in **Figure 5-2**. Reduced operations in the facility have lead to continuing declining emissions in the latter years.

For most of the continuously sampled discharge points having the potential for particulate radionuclide releases, sample results are below the MDC of the analysis. Sometimes as few as 1 or 2 samples (out of 25 to 50 per year) have concentrations greater than the MDC. Generally, these few samples having results above the MDC are only marginally above the MDC. Use of zero values for this type of data can be justified based on facility knowledge, the use of multiple-stage HEPA filters in all significant release pathways, and alpha-spectroscopy-based isotopic analyses of selected air-sampling filters. These isotopic analyses have demonstrated the presence of naturally occurring radionuclides, such as radon daughters, e.g., polonium, on air-sampling filters. In addition, because of exhaust configurations at some facilities, the monitoring systems sometimes sample air from the ambient atmosphere in addition to the HEPA-filtered air from facility operations, which gives rise to background atmospheric radioactivity being collected. Because of these considerations, the emissions from such facility operations are reported as zero. Furthermore, even

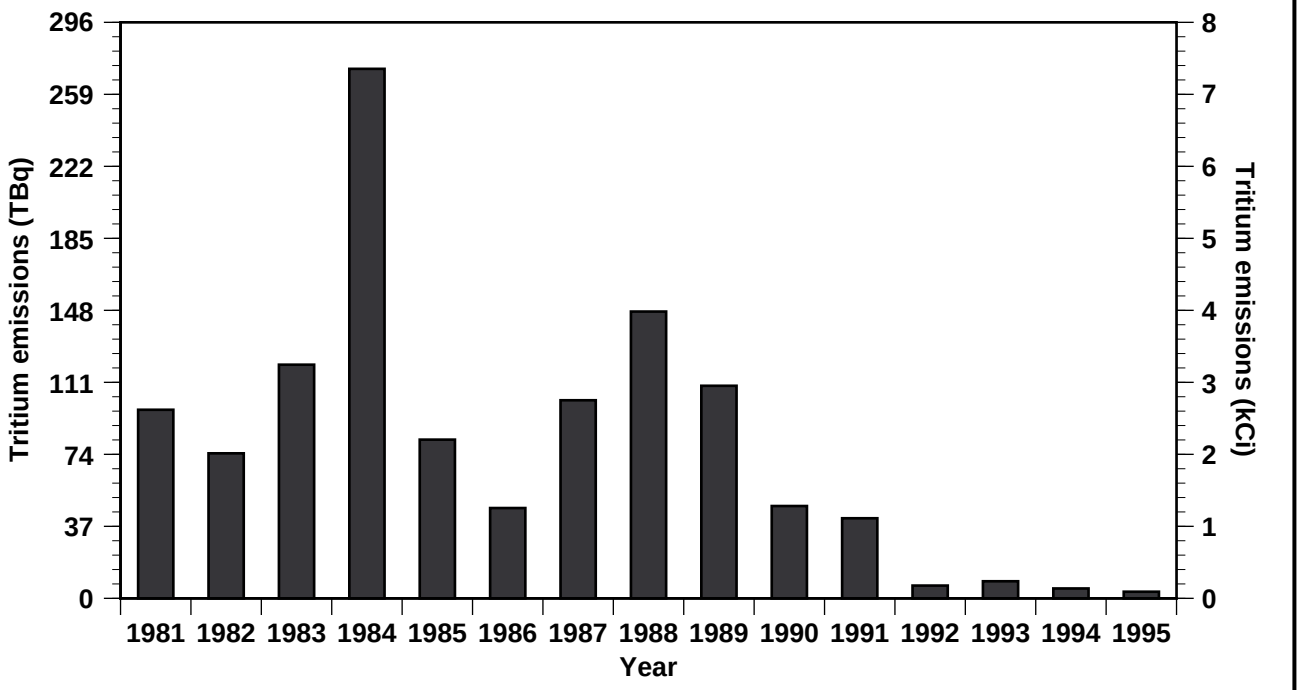

Figure 5-2. Tritium Facility emissions between 1981 and 1995.

if the MDC values were to be used in calculations of the emission estimates for these facilities, an extremely conservative approach, the total dose to a member of the public attributable to LLNL activities should not be significantly affected.

In 1995, samples from three emission points at two facilities, Buildings 251 (Unhardened Area) and 419, yielded gross alpha results greater than the MDC on a majority of the samples collected throughout the year. We use gross alpha as the primary indicator of potential emissions for operations, such as those at Buildings 251 and 419, that involve the use of transuranic materials. The gross alpha monitoring concentrations for these buildings ranged from 7.0×10^{-6} Bq / m^3 (1.9×10^{-16} Ci / m^3) to 2.3×10^{-4} Bq / m^3(6.2×10^{-15} Ci / m^3). Because of the number of samples with values above the MDC, we have taken a conservative approach and reported gross alpha and gross beta measurements as actual emissions. The gross alpha and gross beta emissions for Building 251 were determined to be 6.9×10^{2} Bq / y (1.9×10^{-8} Ci / y) and 7.4×10^{3} Bq / y (2.0×10^{-7} Ci / y), and the gross alpha and gross beta emissions derived from the measured concentrations for Building 419 were 8.7×10^{3} Bq / y (2.3×10^{-7} Ci / y) and 8.5×10^{4} Bq / y (2.3×10^{-6} Ci / y). Emissions for Building 251 here are less than those reported in the *LLNL NESHAPs 1995 Annual Report* (Gallegos et al. 1996) because subsequent investigation of the greater than MDC emissions for the sampler warranted correction to the data. **Table 5-2** lists a summary of

radioactive emissions for 1995. We have not confirmed these to be actual facility emissions by isotopic analysis, so it is possible that these, too, are due to naturally occurring, or background, radioactivity, as discussed above. In any case, the radiological dose from the emissions at these facilities is far less than the dose due to other Livermore site emissions.

Table 5-2. Measured radioactive air effluent emissions for 1995 for the Livermore site.

Tritium			
Building	**Facility**	**Elemental, HT (Bq)**	**Tritiated water, HTO (Bq)**
B331	Tritium	1.1×10^{12}	2.3×10^{12}

Gross alpha and gross beta			
Building	**Facility**	**Gross alpha (Bq)**	**Gross beta (Bq)**
B251	Heavy Element	6.9×10^{2}	7.4×10^{3}
B419	Decontamination	8.7×10^{3}	8.5×10^{4}
Total	**Livermore site**	**9.4×10^{3}**	**9.2×10^{4}**

Radioactive effluent concentrations from individual discharge points at these facilities are reported in Chapter 5 of Volume 2. Activity concentrations are comparable to the concentrations of gross alpha and gross beta activities as measured by LLNL air surveillance samplers and reported in Chapter 4.

Site 300

Radioactive Air Emissions

Currently, there is no air effluent monitoring of facilities at Site 300. Air surveillance monitoring is performed for Site 300, and results are reported in Chapter 4.

Results: All Potential Sources

This section discusses the evaluation of all sources of radionuclide emissions to air at the Livermore site and Site 300. All discharge points having a potential to release radionuclides to the air are evaluated according to 40 CFR 61, Subpart H of the NESHAPs regulations. This evaluation, performed on an annual basis, uses radionuclide inventories and / or monitoring data along with EPA-accepted release factors for operations and EPA-suggested reduction factors for emission

control devices to estimate the potential release for each individual discharge point. Results for 1995 have been published in *LLNL NESHAPs 1995 Annual Report* (Gallegos et al. 1996).

Estimates of emissions are also made for nonradioactive effluents.

Livermore Site Radioactive Emissions

All Potential Sources of Radioactive Air Emissions

An abbreviated isotope summary of measured and calculated emissions for 1995 is presented in **Table 5-3.** There were 45 buildings involved in the evaluation of emissions; these buildings, their operations, and effective dose equivalents to a member of the public are listed in Chapter 13 (Radiological Dose Assessment). The total estimated release from both point and diffuse sources for all isotopes used was 3.9×10^{12} Bq (105 Ci). Tritium emissions from both point and diffuse sources account for 94% of the total estimated emissions. Primary diffuse sources include tritium storage areas at Building 331, Hazardous Waste Management operations at Buildings 514 and 612, contaminated soil near Building 292, and contaminated soil in the southeast quadrant of the site. The diffuse tritium sources at Buildings 292, 331, 514, and 624 have a localized effect; no elevated tritium concentrations were detected at the site perimeter or off site (See Chapter 4). Operations involving tritium at facilities other than the Tritium Facility had estimated releases totaling 1.05×10^{11} Bq (2.8 Ci) during 1995. These releases were assumed conservatively to be HTO.

A complete isotope listing of calculated emissions appears in Volume 2, Table 5-1. The radioactive atmospheric emissions from these Livermore site operations during 1995 are generally lower than previous years.

Site 300 Radioactive Emissions

All Potential Sources of Radioactive Air Emissions

The estimated radioactive air emissions from Site 300 for 1995 are presented in **Table 5-4.** The total estimated release from both point and diffuse sources was 4.1×10^{10} Bq (1.1 Ci). Point sources, which included explosives testing operations at Buildings 801 and 851, accounted for 87% of the total estimated emissions at Site 300. The remaining 13% of the emissions were from diffuse sources and included subsurface tritium contamination and resuspension of uranium in

contaminated soil. Both types of contamination are from previous explosives testing. Details of the calculations and assumptions involved in obtaining the estimates are contained in the LLNL NESHAPs 1995 Annual Report (Gallegos, 1996).

Table 5-3. Calculated radioactive air emissions from the Livermore site for 1995.

Radionuclide[a]	Calculated emissions[b] (Bq)	Radionuclide	Calculated emissions[b] (Bq)
^{3}H (HTO)[c]	2.6×10^{12}	^{228}Th	1.6×10^{3}
^{238}U	5.4×10^{5}	^{237}Np	4.8×10^{2}
^{234}U	2.6×10^{5}	^{242}Pu	4.1×10^{2}
Gross alpha[c,d]	2.1×10^{4}	^{238}Pu	3.7×10^{2}
^{13}N	1.6×10^{11}	^{32}P	1.9×10^{7}
^{63}Ni	1.1×10^{9}	^{243}Am	8.2×10^{1}
^{235}U	1.4×10^{4}	^{239}Pu	7.1×10^{1}
^{241}Am	2.7×10^{3}	Gross beta[c,d]	1.0×10^{5}
^{15}O	8.5×10^{10}	^{3}H (HT)[c]	1.1×10^{12}
Total			**3.9×10^{12}**

a Radionuclides have been ordered by weighting the emissions according to the inhalation dose rate conversion factor for the isotope.

b Calculated emissions are estimates made according to NESHAPs 40 CFR 61, Subpart H except those noted as measured. Values are considered to be conservative.

c Includes measured emissions from continuously monitored facilities.

d Gross alpha and gross beta activities are reported in inventories where specific isotopic content is not determined.

Table 5-4. Calculated radioactive air emissions from Site 300 for 1995.

Radionuclide[a]	Calculated emissions[b] (Bq)
^{238}U	2.0×10^{9}
^{234}U	1.9×10^{8}
^{235}U	2.6×10^{7}
^{3}H (HTO)	3.8×10^{10}
Total	**4.1×10^{10}**

a Radionuclides have been ordered by weighting the emissions according to the inhalation dose rate conversion factor for the isotope.

b Calculated emissions are estimates made according to NESHAPs 40 CFR 61, Subpart H. Values are considered to be conservative.

Nonradioactive Effluents

The Livermore site currently emits approximately 100 kg / day of criteria air pollutants (nitrogen oxides, sulfur oxides, particulate matter (PM10), carbon monoxide, and lead). The largest sources of criteria pollutants from the Livermore site are surface coating operations, internal combustion engines, solvent operations, and, when grouped together, boilers (oil and natural gas fired).

The estimated releases from exempt and permitted sources of air pollutants at the Livermore site can be compared to the most recent estimated 1994 daily release of air pollutants for the entire Bay Area. For example, the total emissions of oxides of nitrogen released in the Bay Area is approximately 4.4×10^5 kg / day compared to an estimate for LLNL releases of 56 kg / day for the Livermore site (0.00013 of total Bay Area emissions). The BAAQMD estimate for reactive organic emissions is 7.5×10^5 kg / day, versus Livermore site's estimated releases of 25 kg / day (0.00003 of total Bay Area emissions) in 1995. **Table 5-5** lists the estimated Livermore site 1995 total airborne releases for criteria pollutants.

Certain operations at Site 300 require permits from San Joaquin Valley Unified Air Pollution Control District. The total estimated air emissions during 1995 from operations (permitted and exempt air sources) at Site 300 are given in **Table 5-5**. Criteria sources at Site 300 include a gasoline dispensing operation, open burning, paint spray booths, and soil vapor extraction.

Table 5-5. Nonradioactive air emissions, Livermore site and Site 300, 1995.

Pollutant	Estimated releases (kg/day)	
	Livermore site	Site 300
Organics/volatile organics	25	1.3
Oxides of nitrogen	56	0.60
Carbon monoxide	9.6	0.40
Particulates (PM10)	8.4	2.3
Oxides of sulfur	7.2×10^{-1}	2.5×10^{-2}

Environmental Impact

Radioactive air effluents from the Livermore site and Site 300 operations for 1995 are well below levels which should cause concern to the environment or public health according to existing regulatory standards. The doses to the hypothetical maximally exposed members of the public due to measured and potential air emissions, as reported in Chapter 13 (Radiological Dose Assessment), are 0.41 μSv (0.041 mrem) for the Livermore site and 0.23 μSv (0.023 mrem) for Site 300. When compared to the NESHAPs standard of 100 μSv / y (10 mrem / y) and dose from naturally occurring radiation, the estimated doses due to the LLNL radionuclide air emissions reported here are minimal. Nonradioactive air effluents, which are also very small compared to emissions in surrounding areas, are well below standards and do not indicate threats to the environment or public health.