4

Air Effluent Monitoring

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Introduction

Lawrence Livermore National Laboratory performs continuous air effluent sampling of atmospheric discharge points at several facilities. 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. DOE Orders 5400.1, General Environmental Protection Program; and 5400.5, Radiation Protection of the Public and the Environment define standards for controlling exposures to the public from operations at DOE facilities. Subpart H of the National Emission Standards for Hazardous Air Pollutants (NESHAPs), 40 Code of Federal Regulations (CFR) 61, requires the continuous monitoring of certain discharge points and the estimation of dose to the public resulting from operations at DOE facilities. Guidance on air effluent sampling is provided in the Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance (U.S. Department of Energy 1991), 40 CFR 60, and NESHAPs-cited ANSI standards. In general, LLNL analyzes for most constituents at levels that are far below regulatory standards in order to determine any environmental impact.

Assessment of air effluent emissions and resulting dose to the public is performed by monitoring emissions and/or evaluating potential emissions. Currently, the air effluent sampling program measures only radiological emissions. LLNL has operations with nonradiological discharges; however, permits for these operations are obtained through local agencies having enforcement authority for the Clean Air Act, and monitoring of the effluent is not required. The agencies with oversight for LLNL compliance with air regulations 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 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 so that doses are ALARA. In addition, the NESHAPs 40 CFR 61, Subpart H regulations require that monitoring of facility

radiological 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. For air discharge points that are monitored, the results of the monitoring provide 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 from operations that have the potential to release radionuclides but that are not monitored are also evaluated according to the NESHAPs regulations, and the corresponding doses are added to those obtained by modeling monitored emissions to determine radiological NESHAPs compliance.

Air effluent monitoring of atmospheric discharge points determines the actual radionuclide releases from individual facilities and processes during routine and nonroutine operations, confirms the operation of facility emission control systems, and can corroborate and aid in the resolution of air surveillance measurement results for the site. (The relationship can also work the other way as well—air surveillance measurements can corroborate effluent monitoring.) Measurements made by the air surveillance samplers located on and off site are reported in Chapter 5.

A variety of radioisotopes are used for research purposes at LLNL; these include uranium, transuranics, biomedical tracers, tritium, and mixed fission products. 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 uranium and transuranic aerosols. The air effluent sampling systems described in this chapter apply to stationary and point source discharges. Diffuse, or nonpoint sources, are also monitored to fulfill NESHAPs requirements. Sampling methods to evaluate LLNL diffuse sources are described in Chapter 5, Data Supplement. Summary data from these diffuse sources can be found in Chapter 5 of this volume.

Methods

Air effluent monitoring involves the extraction of a measured volume of air from the exhaust of the facility or process and subsequent collection of particles in the extracted volume by filters or of vapors by a collection medium. After collection, the various radionuclides in the sample are measured by appropriate analytical methods.

At the beginning of 1997, LLNL operated 103 radionuclide samplers on air exhausts at 9 facilities at the Livermore site (see **Figure 4-1**). These systems are listed in **Table 4-1** along with the analytes of interest, the type of sampler, and the number of samplers.

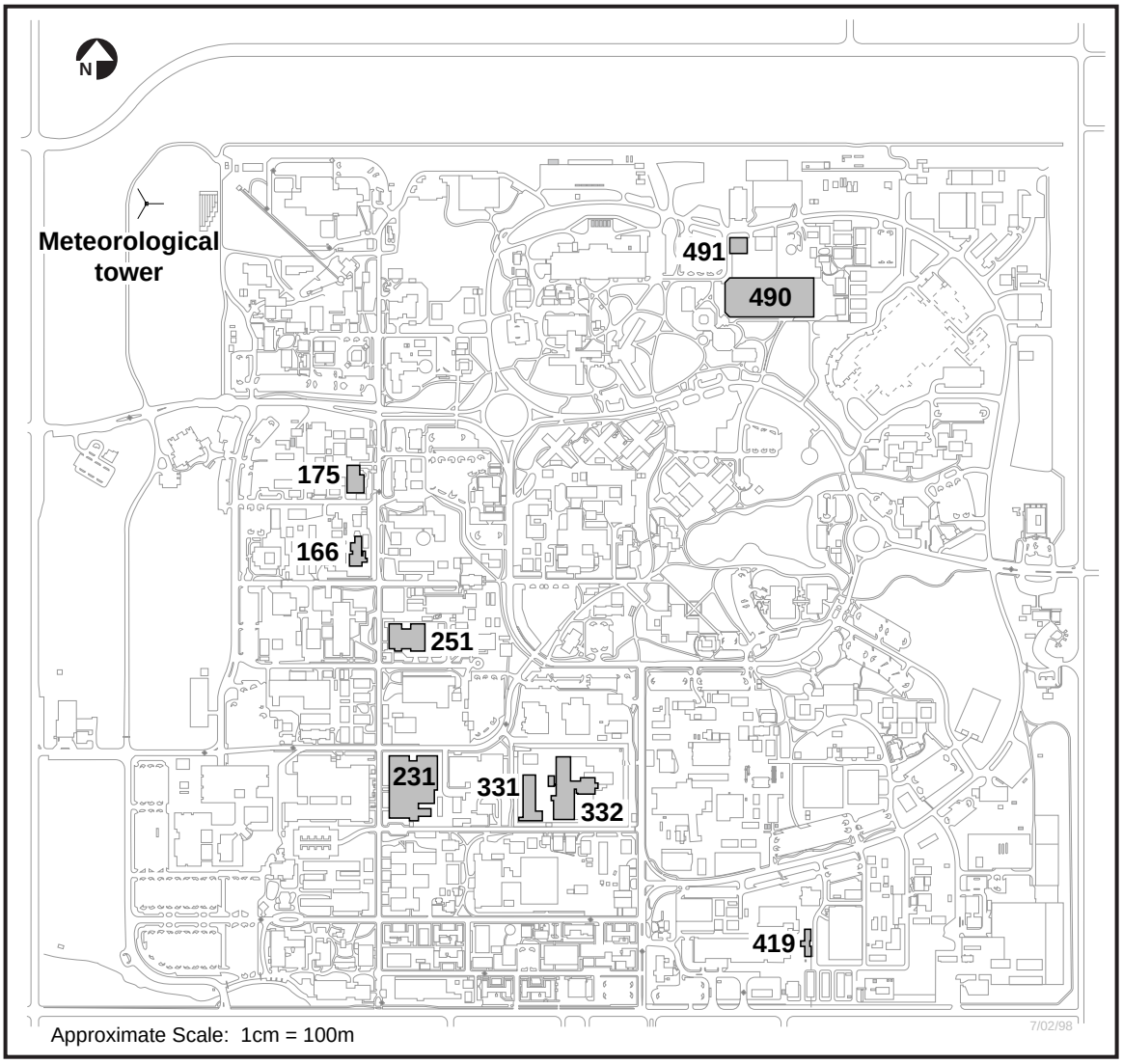

Figure 4-1. Buildings at the Livermore site having air monitoring systems for effluent gas streams during all or part of 1997 (see text).

LLNL reassesses the need for continuous monitoring on an annual basis and more often if warranted by new operations or changes in operations. During 1997, sampling at the locations in Buildings 166, 231, and 419 was discontinued. For Buildings 166 and 419 samplers were removed because the operations originally requiring sampling ceased. Continuous sampling at the Building 231 location was terminated because operations include the receipt, repackaging and shipping of only sealed or encapsulated sources. Therefore, normal operations are not a potential source of emissions and continuous sampling according to NESHAPs regulations is not required. Many of other sampling systems still in place (**Table 4-1**) are not required by regulation; however, LLNL continues to operate these systems as a best management practice.

Table 4-1. Air effluent sampling locations and systems.

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

Note: "CAM" denotes Eberline continuous air monitors.

[a] Sampling discontinued in 1997 due to programmatic changes and re-evaluation.

[b] Alarmed systems.

Sampling for particles containing radionuclides was 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. Most 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 (also listed in **Table 4-1**) at discharge points to provide faster notification in the event of a release of radioactivity. Analytical results from the continuous samplers 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 sampled air. Further details of LLNL air effluent sampling systems are included in Chapter 4 of the *Environmental Monitoring Plan* (Tate et al. 1995).

Currently, nonradiological emissions (with the exception of beryllium) are permitted through the local air districts, and monitoring of them is not required. 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.

Measured Radioactive Air Emissions

This section discusses the radiological air emissions from facilities that have continuously monitored discharge points.

Livermore Site

In 1997, operations in the Tritium Facility (Building 331) released a total of 1.1×10^{13} Bq (300 Ci) of tritium. Of this, approximately 9.9×10^{12} Bq (270 Ci) were released as tritiated water (HTO). The remaining tritium released, 1.2×10^{12} Bq (30 Ci), was elemental tritium gas (HT). The highest single weekly stack emission from the facility was 7.6×10^{11} Bq (21 Ci), of which 5.0×10^{11} Bq (13.6 Ci) was tritiated water. Building 331 emissions continue to remain considerably lower than during the 1980's. **Figure 4-2** illustrates the emissions from the facility, both HTO and HT, since 1981. For 1997, emissions from Building 331 account for 97% of the estimated potential tritium emissions from the Livermore site.

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 to 4 samples (out of 25 to 50 samples 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 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 if the

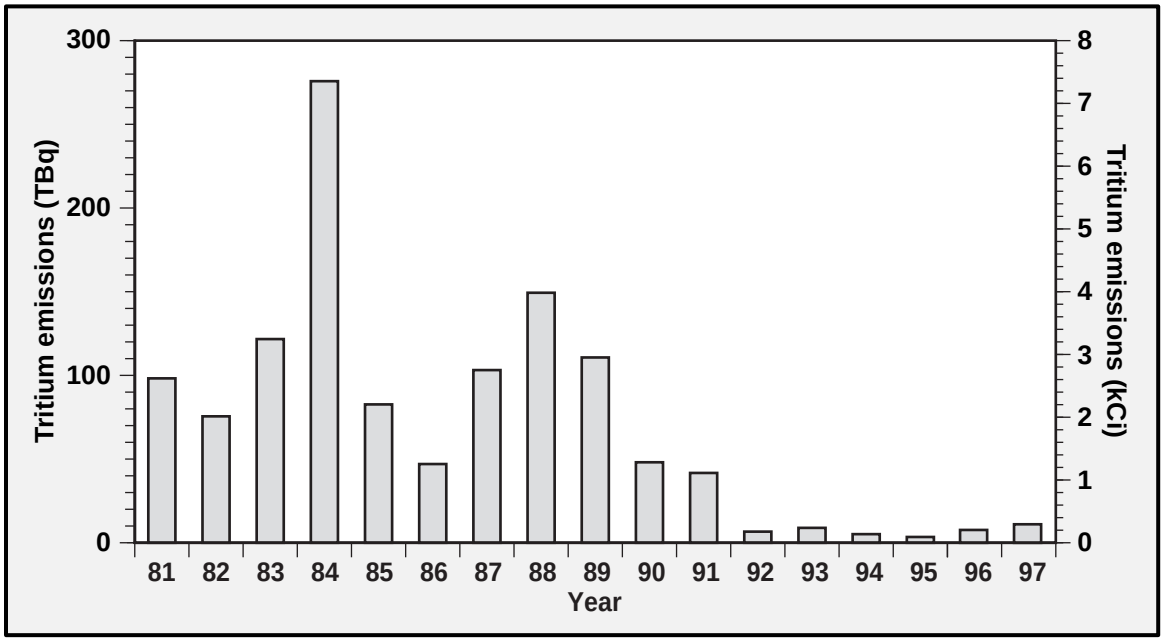

Figure 4-2. Tritium Facility emissions (HTO and HT) between 1981 and 1997.

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

In 1997, samples from four emission points at Building 251 (the unhardened area) yielded gross alpha results greater than the MDC on a significant number of the samples collected throughout the year. We use gross alpha as the primary indicator of potential emissions for operations such as those at Building 251 that involve the use of uranium and transuranic materials. Gross beta results are used as a further corroboration of those gross alpha results having concentrations above the MDC. The gross alpha and gross beta emissions for Building 251 were determined to be 6.0×10^{3} Bq / y (1.6×10^{-7} Ci / y) and 3.9×10^{4} Bq / y (1.1×10^{-6} Ci / y). Because of the number of samples with values above the MDC, we have taken a conservative approach and are reporting gross alpha and gross beta measurements as actual emissions.

The gross alpha monitoring concentrations for Building 251 ranged from -3.1×10^{-4} Bq / m^3 (-8.4×10^{-15} Ci / m^3) to 3.7×10^{-5} Bq / m^3 (1.0×10^{-15} Ci / m^3). These activity concentrations do not differ significantly from the results of low-volume air surveillance samplers reported in Chapter 5. The Building 251 facility is in a standby, limited mode, of operation and emissions are not anticipated. So it is likely that emissions reported here for Building 251 are due to naturally occurring, or background,

radioactivity, and to the facility exhaust configuration as previously mentioned. In any case, assessment of the gross alpha and gross beta emissions being reported here indicates the radiological dose is not a significant contributor to the dose to the public from all Livermore site operations.

Table 4-2 lists total radiological emissions as determined from the continuous sampling of facility exhausts for 1997. Radioactive effluent concentrations from individual discharge points at all monitored facilities are reported in Chapter 4, Data Supplement.

Table 4-2. Measured radiological air effluent emissions for the Livermore site, 1997.

Tritium			
Building	**Facility**	**Elemental, HT (Bq)**	**Tritiated water, HTO (Bq)**
331	Tritium	1.2×10^{12}	9.9×10^{12}
Gross alpha and gross beta			
Building	**Facility**	**Gross alpha (Bq)**	**Gross beta (Bq)**
251	Heavy Element	6.0×10^{3}	3.9×10^{4}

Site 300

Currently, there is no requirement for air effluent monitoring of facilities at Site 300. Air surveillance monitoring is performed for Site 300, and results are reported in Chapter 5.

All Potential Sources of Emissions

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. Potential emissions are those based upon the radionuclide inventories as distinguished from emissions based air effluent sampling. The evaluation is conducted to assess the dose to the public from all LLNL operations and the need for continuous sampling of individual discharge points.

For 1997, measured and potential emissions of radionuclides from 45 facilities were evaluated for their contribution of dose to a member of the public. The evaluation is based on estimated releases using radionuclide inventories specific to individual discharge points, physical state of the materials involved in the processes, and reductions due to emission control systems. The effective dose equivalent to a member of the public from specific operations at the Livermore site and Site 300 have been published in *LLNL NESHAPs 1997 Annual Report* (Gallegos et al. 1998a) and are summarized in Chapter 12 (Radiological Dose Assessment) of this report.

The radionuclide isotope responsible for the majority of the dose is tritium. Emissions from the Tritium Facility in the form of HTO account for 78% of the potential effective dose equivalent (EDE) to the maximally exposed member of the public from Livermore site (see Chapter 12). The 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 emissions did not contribute significantly to the overall tritium dose. The other measured emissions shown in **Table 4-2** (Building 251) contribute negligibly to the EDE for the maximally exposed member of the public.

Many other isotopes are also used at the Livermore site and Site 300. However, simple comparison of the potential radioactivity emissions does not take into account atmospheric dispersion, dose, and the biological response to the isotope. The importance of other isotopes is assessed in Chapter 12 on risk assessment.

To determine the need for continuous sampling of a discharge point, all operations with the potential to contribute emissions to a discharge point were evaluated to determine if the dose to the maximally exposed member of the public exceeded 0.1 mrem for the calendar year. This evaluation is similar to that already discussed except no credit is allowed for emission control systems (according to the regulations). The evaluation for 1997 involved approximately 150 discharge points and / or discharges at the Livermore site and Site 300. No discharge points not presently having continuous sampling were found to require continuous sampling.

Nonradioactive Effluents

The Livermore site currently emits approximately 100 kg / day of criteria air pollutants (nitrogen oxides, sulfur oxides, particulate matter [PM-10], 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 daily releases of air pollutants for the entire Bay Area. For example, the total emissions of oxides of nitrogen released in the Bay Area for 1995 was approximately 4.8×10^5 kg / day compared to an estimate for LLNL releases of 59 kg / day for the Livermore site (0.012% of total Bay Area emissions). The BAAQMD estimate for reactive organic emissions was 5×10^5 kg / day, versus Livermore site's estimated releases of 37 kg / day (0.007% of total Bay Area emissions) in 1997. **Table 4-3** lists the estimated Livermore site 1997 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 1997 from operations (permitted and exempt air sources) at Site 300 are given in **Table 4-3**. Criteria sources at Site 300 include internal combustion engines, boilers, a gasoline dispensing operation, open burning, paint spray booths, drying ovens, and soil vapor extraction.

Table 4-3. Nonradioactive air emissions, Livermore site and Site 300, 1997.

Pollutant	Estimated releases (kg/day)	
	Livermore site	Site 300
Organics/volatile organics	37	1.1
Oxides of nitrogen	59	1.8
Carbon monoxide	10	0.41
Particulates (PM-10)	5.7	0.52
Oxides of sulfur	0.92	0.15

Environmental Impact

Measured radiological air emissions from the Livermore site operations for 1997 are well below levels that should cause concern for the environment or public health according to existing regulatory standards. The dose to the hypothetical maximally exposed member of the public due to the measured air emissions reported here (that is, due to emissions from monitored stacks) is 0.75 μSv / y (0.075 mrem / y), far below the NESHAPs standard of 100 μSv / y (10 mrem / y) and doses from naturally occurring radiation. Thus, the estimated radiological dose due to measured air emissions from LLNL operations is minimal. See **Table 12-2** in Chapter 12 for a summary of all doses, monitored or otherwise. 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.