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. LLNL assesses air effluent emissions from facility operations 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.

Air Quality Laws

LLNL complies with local, state, and federal environmental air quality laws and Department of Energy (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 American National Standards Institute (ANSI) standards. The Environmental Protection Agency (EPA) Region IX has oversight responsibility for LLNL compliance with regulations regarding radiological air emissions.

Enforcement authority of the Clean Air Act regulations for nonradiological air emissions have been delegated to the local air districts: 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. Applicable regulations and permitting requirements are contained in the BAAQMD Regulations 1-12 for the Livermore site and the SJVUAPCD Regulations Rules 1010-9120 for Site 300.

4 Air Effluent Monitoring

Monitored Emissions

LLNL uses a variety of radioisotopes—including uranium, transuranics, biomedical tracers, tritium, and mixed-fission products—for research purposes. 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. LLNL also monitors diffuse, or nonpoint, sources to fulfill NESHAPs requirements. Sampling methods to evaluate LLNL diffuse sources are described in Chapter 5 of the Data Supplement. Summary data from these diffuse sources can be found in Chapter 5 of this volume.

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, BAAQMD, and SJVUAPCD, and monitoring of the effluent 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 the assessment, BAAQMD and SJVUAPCD have ranked LLNL as a low-risk facility for nonradiological air emissions.

Historically, monitoring of radionuclide air effluents at LLNL has been implemented according to the DOE as low as reasonably achievable (ALARA) policy. This policy is meant to ensure that DOE facilities are capable of monitoring 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 facility radiological air effluents must be continuously monitored 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 that there are no emission control devices. The results from monitoring the air discharge points provide the actual emission source information for modeling, which is used 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.

Operation of Monitoring Systems

Air effluent monitoring of atmospheric discharge points is used to determine the actual radionuclide releases from individual facilities and processes during routine and non-routine operations, to confirm the operation of facility emission control systems, and to 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.

Methods

Air effluent monitoring involves the extraction of a measured volume of air from the exhaust of a facility or process and subsequent collection of particles 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 1999, LLNL operated 101 sampling systems for radioactivity from air exhausts at eight 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. LLNL reassesses the need for continuous monitoring on an annual basis and more often if warranted by new operations or changes in operations. From NESHAPs assessments of operations during 1999, no additional discharge points were found to require continuous sampling.

In 1999, sampling at several air effluent locations was terminated. In the past, sampling operations performed in Buildings 175, 177, 490, and 491 have supported research and development for the separation of uranium isotopes under the Advanced Vapor Laser Isotope Separation (AVLIS) Program. In 1999, the AVLIS Program was shut down, and samplers on a Building 490 exhaust system were deactivated because the operation of the ventilation system was stopped. Air effluent sampling systems at Buildings 175, 177, and 491 continue to operate as part of the maintenance and surveillance shutdown plan for AVLIS facilities. At the Heavy Element Facility (Building 251), 20 samplers were deactivated. This facility has been in a standby mode of operation for some time, and activities involving the use radiological materials are not expected to resume in the areas previously monitored by the deactivated samplers. Finally, a sampling system located at the Expedited Technology of Molten Salt Oxidation project in Building 292 was removed because the project was completed. At the end of 1999, LLNL was operating 76 air effluent sampling systems at six facilities.

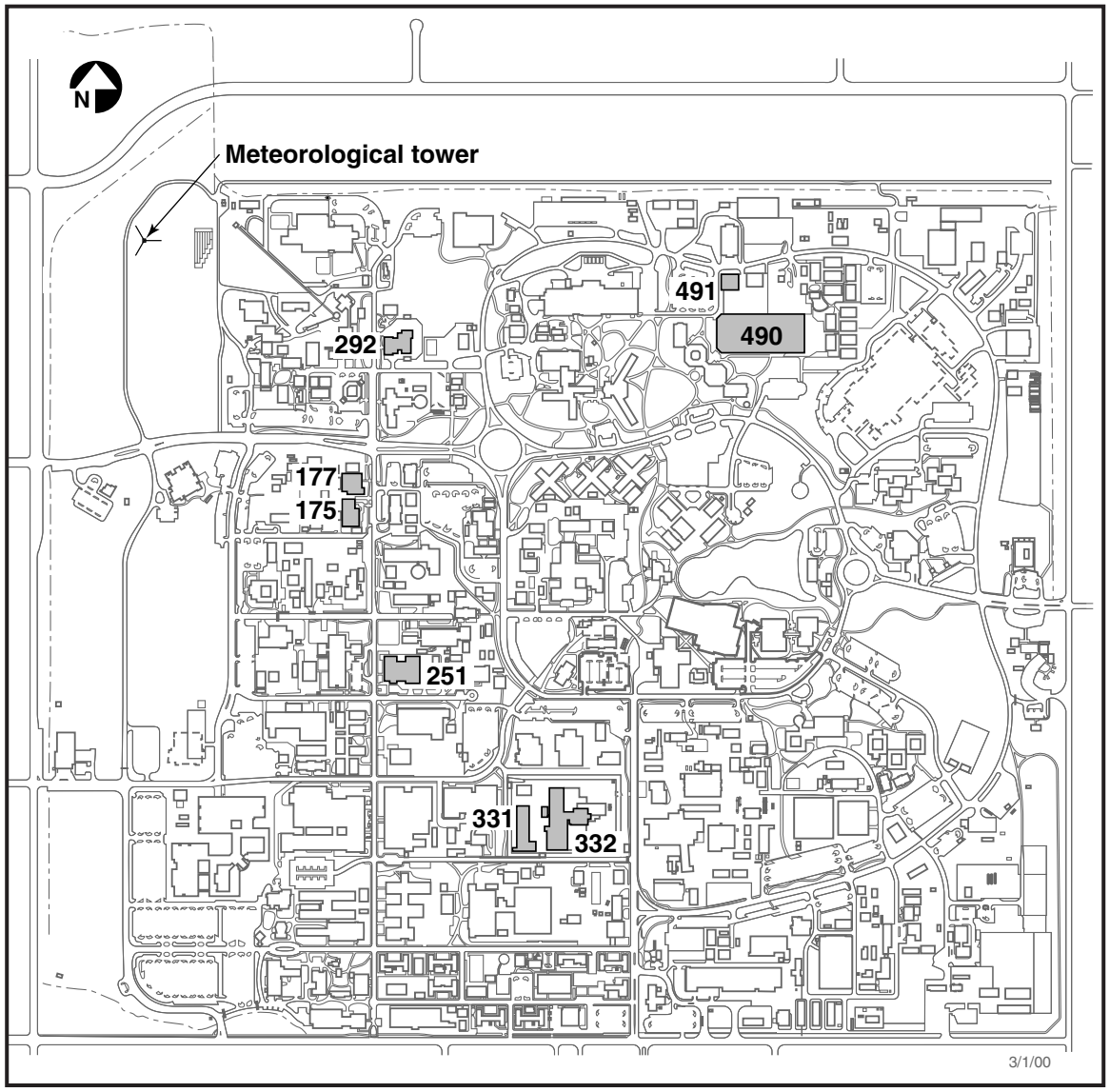


Figure 4-1. Facilities at the Livermore site with air monitoring systems for effluent gas streams during all or part of 1999.

Sampling for particles containing radioactivity was conducted in seven of the facilities; sampling for tritium was conducted in the Tritium Facility (Building 331). All sampling systems operated continuously. Samples were collected weekly or biweekly depending on the facility. Most air samples for particulate emissions were extracted downstream of HEPA filters and before the emissions were discharged to the atmosphere. Particles in the extracted air were 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 used real-time alarm monitors (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 as less than 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. 1999).

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

Building	Facility	Analytes	Sampler type	Number of samplers
175	MARS	Gross α, β on particles	Filter	6
177	Extractor Test Facility	Gross α, β on particles	Filter	1
251	Heavy elements			
	Unhardened area	Gross α, β on particles	Filter	44[a]
	Hardened area	Gross α, β on particles	Filter	4
	Hardened area	Gross α, β on particles	CAM[b,c]	4[d]
292	Molten salt oxidation	Gross α, β on particles	Filter	1[d]
331	Tritium	Tritium	Ionization chamber[c]	4
		Gaseous tritium and tritiated water vapor	Molecular sieves	4
332	Plutonium	Gross α, β on particles	CAM[b,c]	12
		Gross α, β on particles	Filter	16
490	Laser isotope separation	Gross α, β on particles	Filter	4[d]
491	Laser isotope separation	Gross α, β on particles	Filter	1

a Sixteen of these sampling systems were deactivated in 1999.

b CAM = Eberline continuous air monitors.

c Alarmed systems.

d Sampling at these locations was terminated in 1999.

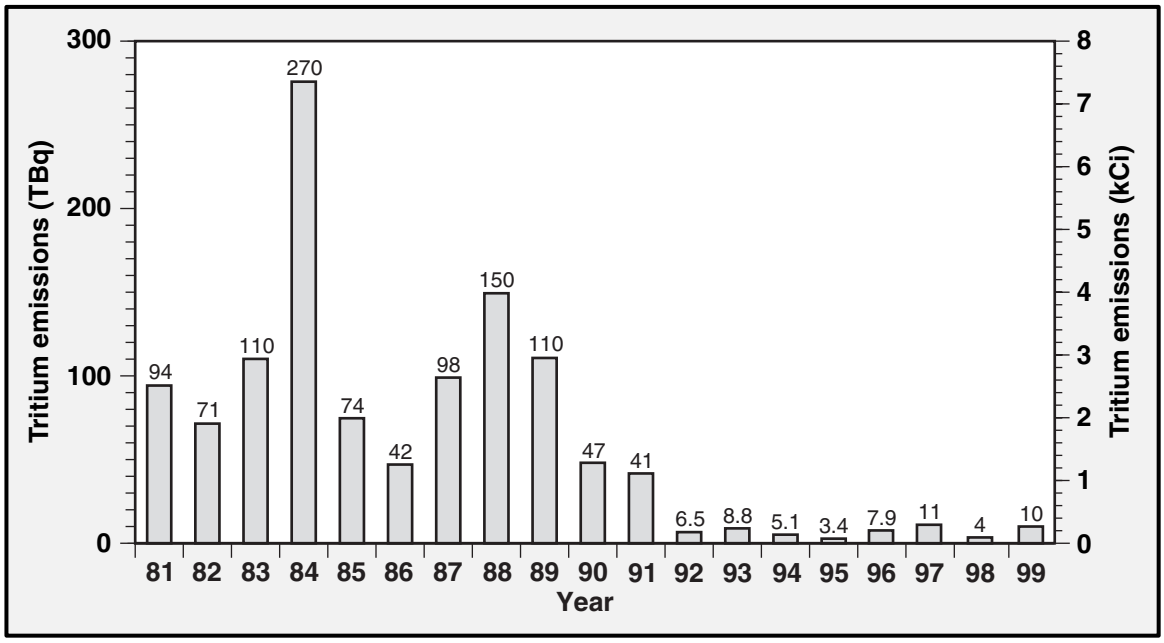
Measured Radioactive Air Emissions

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

Livermore Site

In 1999, a total of 10.4 TBq (281 Ci) of tritium was released from the Tritium Facility (Building 331). Of this, approximately 7.9 TBq (214 Ci) were released as tritiated water vapor (HTO). The remaining tritium released, 2.5 TBq (67 Ci), was elemental tritium gas (HT). HTO emissions from the facility ranged from 26 Bq/m^3 (7.0×10^{-10} Ci/m^3) to 1.6×10^5 Bq/m^3 (4.2×10^{-6} Ci/m^3), while HT emissions ranged from 6.8 Bq/m^3 (1.8×10^{-10} Ci/m^3) to 1.3×10^5 Bq/m^3 (3.5×10^{-6} Ci/m^3). The highest single weekly stack emission from the facility was 1.5 TBq (41 Ci), of which 0.54 TBq (15 Ci) was HTO. The emissions from Building 331 accounted for 96% of the estimated potential tritium emissions from the Livermore site.

Emissions from Building 331 for 1999 continued to remain considerably lower than those during the 1980s. **Figure 4-2** illustrates the HTO and HT emissions from the facility since 1981. The tritium emissions for 1999 were greater than 1998 emissions because of programmatic activities and an equipment malfunction that occurred in the facility. The tritium air surveillance sample results (see Data Supplement, Chapter 5) accurately confirmed the facility emissions, including a slightly elevated release period from January to March.



Note: The plot of Tritium Facility emissions differs in this report as compared with similar plots in previous reports, in which the Tritium Facility emissions were overstated for the years 1981–1987. The previous reports contained the total LLNL emissions of HTO and HT, not just Tritium Facility emissions.

Figure 4-2. Tritium Facility HTO and HT emissions between 1981 and 1999.

In 1999, most sample results from the continuously sampled discharge points that have the potential for releasing particulate radionuclides were below the MDC of the analysis. This was the case for all of the sampled discharge points at Buildings 175, 177, 332, 490, and 491 and for sampled discharge points at Building 251 except for the one discussed below. Sometimes as few as one to four samples (out of 25 to 50 samples per year) exhibited concentrations greater than the MDC. Generally, these few samples with results above the MDC were only marginally above the MDC. In addition, because of the way some exhaust systems were configured, the monitoring systems sometimes sampled air from the ambient atmosphere and HEPA-filtered air from facility operations, which means that background atmospheric radioactivity was also collected. LLNL uses zero values for these results based on knowledge of the facility, the use of HEPA filters in all significant release pathways, and alpha-spectroscopy-based isotopic analyses of selected air sampling filters. These analyses demonstrate the presence of naturally occurring radionuclides, such as radon daughters, like polonium. Even if LLNL used the MDC values to calculate 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.

At Building 251 (in the unhardened area), one discharge point had a significant number of samples collected throughout the year with gross alpha results greater than the MDC. 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. We use gross beta results to further corroborate those gross alpha results with concentrations above the MDC. The gross alpha and gross beta activity emissions for Building 251 were 1.4×10^{2} Bq/y (3.7×10^{-9} Ci/y) and 2.5×10^{3} Bq/y (6.8×10^{-8} 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 -7.8×10^{-5} Bq/m^3 (-2.1×10^{-15} Ci/m^3) to 5.7×10^{-4} Bq/m^3 (1.5×10^{-14} Ci/m^3). The Building 251 facility was in a standby, limited mode of operation throughout 1999, so emissions were not anticipated. Also, the gross alpha and gross beta average activity concentrations do not significantly differ from those of the low-volume air particulate surveillance samplers (see Chapter 5). Therefore, it is likely that Building 251 measurements were caused by naturally occurring or background radioactivity and by the facility exhaust configuration, as previously mentioned. In any case, the gross alpha and gross beta emissions from operations did not a significantly contribute to the radiological dose to the public.

Table 4-2 lists total radiological emissions as determined from the continuous sampling of facility exhausts for 1999. 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, 1999.

Tritium			
Building	**Facility**	**Elemental, HT (Bq)**	**Tritiated water, HTO (Bq)**
331	Tritium	2.5×10^{12}	7.9×10^{12}
Gross alpha and gross beta			
Building	**Facility**	**Gross alpha (Bq)**	**Gross beta (Bq)**
251	Heavy element	1.4×10^{2}	2.5×10^{3}

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, Air Monitoring.

All Potential Sources of Radioactive Air Emissions

This section discusses the evaluation of all sources of radionuclide emissions to air at the Livermore site and Site 300. LLNL evaluates all discharge points with the potential to release radionuclides to the air according to 40 CFR 61, Subpart H, of the NESHAPs regulations. LLNL uses radionuclide usage 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 calculated using radionuclide usage inventories as distinguished from emissions-based air effluent sampling. We conduct this evaluation annually to assess the potential dose to the public from all LLNL operations and the need for continuous sampling of individual discharge points.

For 1999, LLNL evaluated potential emissions of radionuclides from approximately 50 facilities to determine their contribution of dose to a member of the public. Potential emissions were estimated based on radionuclide usage inventories specific to individual discharge points, physical state of the materials involved in the processes, and reductions caused by emission control systems. The effective dose equivalent (EDE) to a

member of the public from specific operations at the Livermore site and Site 300 were published in *LLNL NESHAPs 1999 Annual Report* (Gallegos et al. 2000) and are summarized in Chapter 13 of this report.

The radionuclide isotope responsible for the majority of the 1999 EDE was tritium. Emissions from the Tritium Facility in the form of HTO accounted for 56% of the potential EDE to the maximally exposed member of the public from the Livermore site. Emissions from the facility in the form of HT, when modeled as HTO emissions as mandated by EPA, accounted for 18% of the potential EDE. A discussion of the relative dose impacts from HTO and HT is given in Chapter 13 in the section entitled "Assessment Assumptions Regarding Tritium." The other measured emissions shown in **Table 4-2** (Building 251) contributed negligibly to the EDE for the maximally exposed member of the public.

When determining if continuous sampling is needed at a discharge point, LLNL evaluates operations to determine if the potential dose to the maximally exposed member of the public will exceed 0.1 mrem for the calendar year. This evaluation is similar to the evaluation of EDE previously described except no credit is allowed for emission control systems (according to the regulations). For 1999, LLNL reported more than 200 potential discharge points and/or discharges at the Livermore site and Site 300. As a result of the evaluation, no additional discharge points other than those already being continuously sampled were found to require continuous sampling.

Nonradioactive Air Emissions

The Livermore site currently emits approximately 140 kg/day of criteria air pollutants (e.g., nitrogen oxides, sulfur oxides, particulate matter [PM-10], carbon monoxide, and lead, as defined by the Clean Air Act). 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). **Table 4-3** lists the estimated Livermore site 1999 total airborne releases for criteria pollutants.

When comparing the estimated releases from exempt and permitted sources of air pollutants at the Livermore site with daily releases of air pollutants for the entire Bay Area, LLNL emissions are very low. For example, the total emissions of nitrogen oxides released in the Bay Area for 1997 were approximately 1.4×10^5 kg/day compared with an estimate for LLNL releases of 81 kg/day for the Livermore site (0.06% of total Bay

Table 4-3. Nonradioactive air emissions, Livermore site and Site 300, 1999.

Pollutant	Estimated releases (kg/day)	
	Livermore site	Site 300
Organics/volatile organics	24	1.2
Nitrogen oxides	81	3.2
Carbon monoxide[a]	24	0.71
Particulates (PM-10)	8.6	0.33
Oxides of sulfur	0.98	0.28

a In 1999 the emission factor used to calculate carbon monoxide was 0.035lb/1000 ft^3 for large boilers and 0.021 lb/ft^3 for small boilers. In previous years the emission factor used was 0.017 lb/ft^3 for both large and small boilers. This resulted in a significant change in monoxide emissions reported for 1999.

Area emissions). The BAAQMD estimate for reactive organic emissions was 1.3×10^5 kg/day for 1997, versus the Livermore site's estimated releases of 24 kg/day (0.02% of total Bay Area emissions) in 1999.

Certain operations at Site 300 require permits from SJVUAPCD. The total estimated air emissions during 1999 from operations (permitted and exempt air sources) at Site 300 are given in **Table 4-3**. The largest sources of criteria pollutants at Site 300 include internal combustion engines, boilers, a gasoline-dispensing operation, open burning, paint spray booths, drying ovens, and soil vapor extraction operations.

Environmental Impact

Measured radiological air emissions from the Livermore site operations for 1999 are well below levels that should cause concern for public health according to existing regulatory standards for radioactive dose. The dose to the hypothetical maximally exposed member of the public caused by the measured air emissions reported here (that is, caused by emissions from monitored stacks) is 0.67 µSv/y (0.067 mrem/y). Including the measured HT emissions (with HT emissions modeled as HTO emissions as mandated by EPA), the dose to the hypothetical maximally exposed member of the public is 0.88 µSv/y (0.088 mrem/y). In either case, the dose is far below the NESHAPs standard of 100 µSv/y (10 mrem/y) and doses from naturally occurring radiation. Thus, the estimated radiological dose caused by measured air emissions from LLNL operations is minimal. See **Table 13-2** in Chapter 13 for a summary of all doses, monitored or otherwise. Nonradioactive air effluents, which are also very small compared with emissions in surrounding areas, are well below standards and are not a threat to the environment or public health.