# 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 U.S. Department of Energy (DOE) regulations. Applicable sections of 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 Emissions 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. DOE 1991), 40 CFR 60, and NESHAPs-cited American National Standards Institute (ANSI) standards. The U.S. 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 has 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 1–9 for Site 300.

## 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 non-monitored operations with the potential to release radionuclides are also evaluated according to NESHAPs regulations.

To determine radiological NESHAPs compliance, corresponding doses are added to those obtained by modeling monitored emissions.

## 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 nonroutine operations, to confirm the operation of facility emission control systems, and to corroborate and aid in the resolution of ambient air measurement results for the site. (The relationship can 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.

In 2002, LLNL operated 74 sampling systems for radioactivity from air exhausts at 7 facilities at the Livermore site (see **Figure 4-1**) and 1 sampling system at Site 300 (see **Figure 4-2**). These systems are listed in **Table 4-1** along with the analytes of

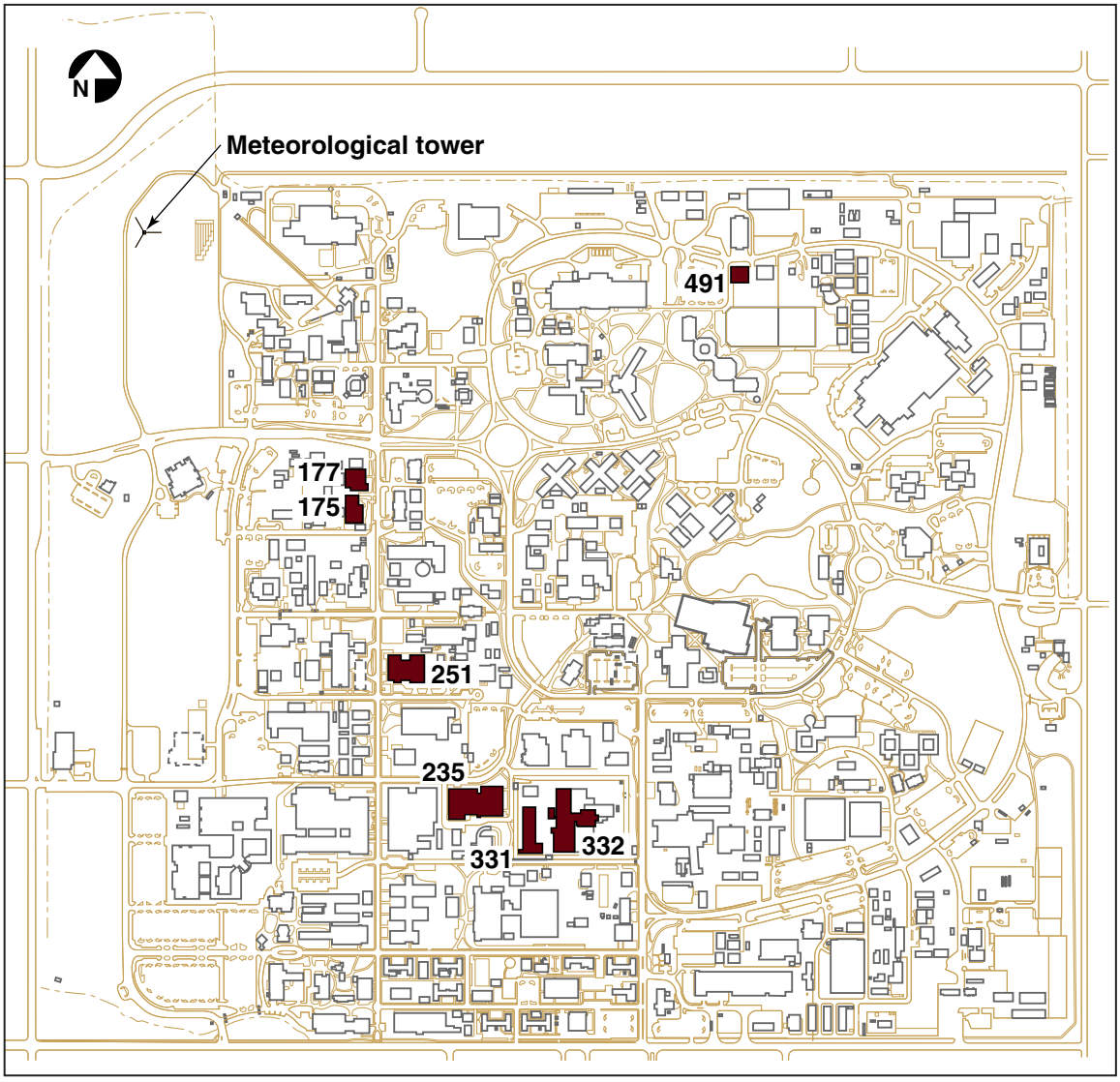


**Figure 4-1. Facilities at the Livermore site with air monitoring systems for effluent gas streams during all or part of 2002**

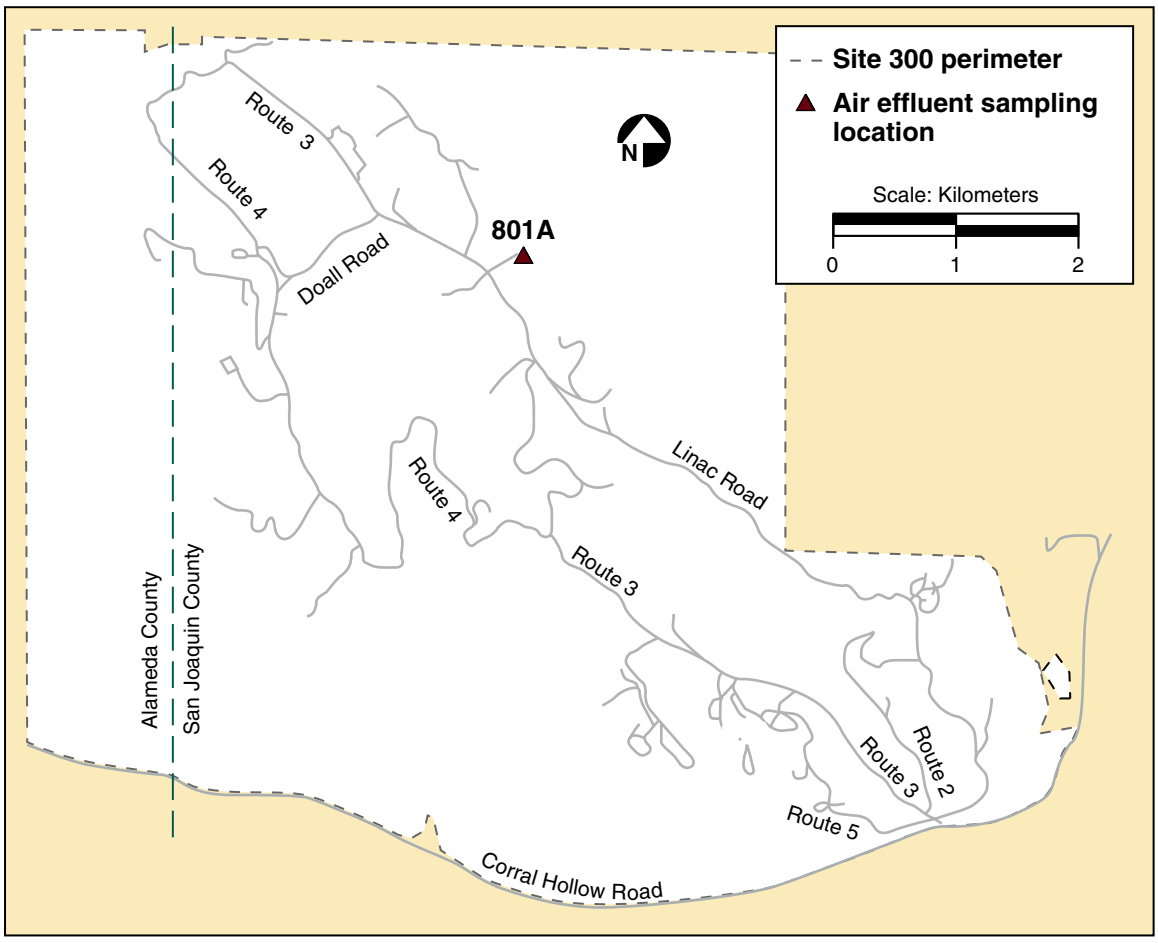


**Figure 4-2. Building 801A at Site 300 with an air monitoring system for effluent gas streams**

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 2002, one additional discharge point, a new operation in Building 801A at Site 300, was found to require continuous sampling.

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. In February 2002, decontamination activities at Building 177 were completed and the sampling system was deactivated. Air effluent sampling systems at Buildings 175 and 491 continue to operate as part of the maintenance and surveillance shutdown plan for AVLIS facilities.

Sampling for particles containing radioactivity was conducted in seven of the facilities and 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

**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 |
| 235 | Chemistry and Materials Science | Gross α, β on particles | Filter | 1 |
| 251 | Heavy elements | Gross α, β on particles | Filter | 28 |
| 331 | Tritium | Tritium | Stack ionization chamber[a] | 4 |
| | | Gaseous tritium and tritiated water vapor | Molecular sieves | 4 |
| 332 | Plutonium | Gross α, β on particles | Stack CAM[a,b] | 12 |
| | | Gross α, β on particles | Filter | 16 |
| 491 | Laser isotope separation | Gross α, β on particles | Filter | 1 |
| 801A | Contained Firing Facility | Gross α, β on particles | Filter | 1 |

a Alarmed systems

b CAM = Eberline continuous air monitors

samples for particulate emissions were extracted downstream of high-efficiency particulate air (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 was 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 detectable 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).

## Measured Radioactive Air Emissions

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

### Livermore Site

In 2002, a total of 1.3 TBq (36.3 Ci) of tritium was released from the Tritium Facility (Building 331). Of this, approximately 1.2 TBq (32.8 Ci) were released as tritiated water vapor (HTO). The remaining tritium released, 0.13 TBq (3.5 Ci), was elemental tritium gas (HT). Weekly HTO emissions from the facility ranged from 0 Bq/m$^3$ (0 Ci/m$^3$) to $2.4 \times 10^4$ Bq/m$^3$ ($6.6 \times 10^{-7}$ Ci/m$^3$), while HT emissions ranged from 0 Bq/m$^3$ (0 Ci/m$^3$) to $4.4 \times 10^3$ Bq/m$^3$

($1.2 \times 10^{-7}$ Ci/m$^3$). The highest single weekly stack emission from the facility was 141.3 GBq (3.82 Ci), of which 140.6 GBq (3.80 Ci) was HTO.

Emissions from Building 331 for 2002 continued to remain considerably lower than those during the 1980s. **Figure 4-3** illustrates the HTO and HT emissions from the facility since 1981.

Most sample results from the continuously sampled discharge points that have the potential for releasing particulate radionuclides were below the MDC of the analysis. Sometimes as few as 1 to 4 samples (out of 25 to 50 samples per year) exhibited concentrations greater than the MDC. Generally, these few samples 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 as well as HEPA-filtered air from facility operations, which means that background atmospheric radioactivity was also collected. When gross alpha is detected, a check is performed to determine if the blowers were operational at the time of the detection. If the blowers were operational, the sample result is considered a valid detection, otherwise the result is considered to be background atmospheric radioactivity.

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

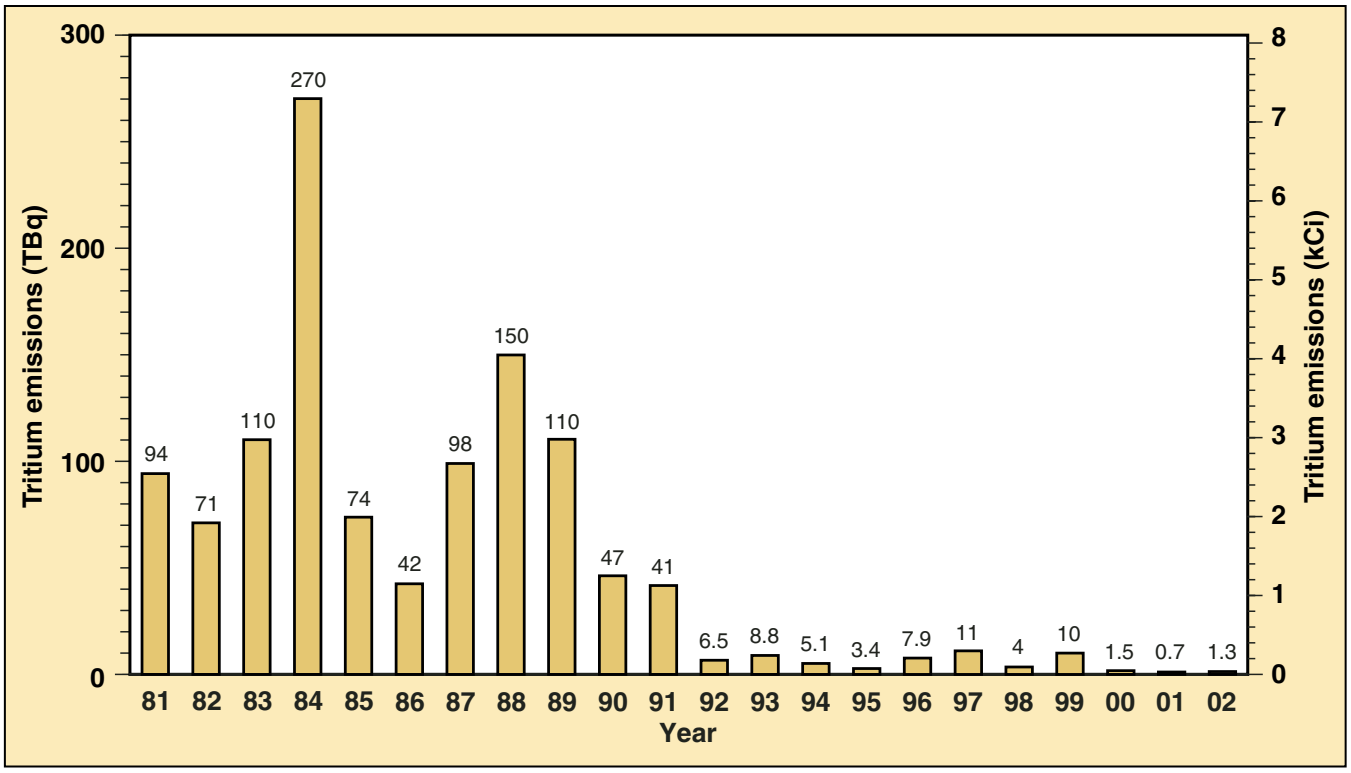


**Figure 4-3. Tritium Facility combined HTO and HT emissions from 1981 through 2002**

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. None of the facilities monitored for gross alpha and beta had emissions in 2002.

Radioactive effluent concentrations from individual discharge points at all monitored facilities are reported in Chapter 4 of the Data Supplement.

### Site 300

An effluent sampling system was installed in Building 801A at Site 300 in early 2002. Although all facility operations are HEPA filtered, this building has a large high bay room that exhausts to the stack without HEPA filtration. Consequently, some of the air sampled by the effluent sampling system is essentially outside, ambient air. In order to determine if any releases actually occurred from this facility, the sampling results must be compared to ambient air. In 2002, five samples out of 38 had concentrations greater than the MDC. The median concentration of the Building 801A detections, $1.3 \times 10^{-4}$ Bq/m$^3$ ($3.6 \times 10^{-15}$ Ci/m$^3$), is lower than the median concentrations of the detections from two offsite sampling locations that are used to establish background levels of gross alpha and beta activity for direct comparison to results from the air effluent samplers (See Chapter 5 for a description of the offsite sampling systems and data results). The median of all 38 of the Building 801A samples, $3.0 \times 10^{-5}$ Bq/m$^3$ ($8.0 \times 10^{-16}$ Ci/m$^3$), is approximately three times lower than the median of all of the offsite sampling location samples. Therefore, it is reasonable to conclude that Building 801A operations did not have radioactive emissions.

## All Potential Sources of Radioactive Air Emissions

This section discusses the evaluation of all potential 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. LLNL conducts this evaluation annually to assess both the potential dose to the public from all LLNL operations and the need for continuous sampling of individual discharge points.

For 2002, LLNL evaluated potential emissions of radionuclides from facilities and/or diffuse sources 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 2002 Annual Report* (Harrach et al. 2003) and are summarized in Chapter 13 of this report.

The radionuclide isotope responsible for the majority of the 2002 EDE was tritium. Emissions from the Tritium Facility, in the form of HTO, accounted for 36% of the potential EDE to the maximally exposed member of the public from the

Livermore site. A brief discussion of the relative dose impacts from HTO and HT is given in *LLNL NESHAPs 2002 Annual Report*.

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).

## Nonradioactive Air Emissions

The Livermore site currently emits approximately 109 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 4-2** lists the estimated Livermore site 2002 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 2002 were approximately $8.3 \times 10^4$ kg/day, compared with an estimate for LLNL releases of 67 kg/day for the Livermore site (0.08% of total Bay Area emissions from stationary sources). The BAAQMD estimate for reactive organic emissions was $9.8 \times 10^4$ kg/day for 2002, versus the Livermore site's estimated releases of 16 kg/day (0.02% of total Bay Area emissions from stationary sources) in 2002.

**Table 4-2. Nonradioactive air emissions, Livermore site and Site 300, 2002**

| Pollutant | Estimated releases (kg/day) | |
|---|---|---|
| | Livermore site | Site 300 |
| Organics/volatile organics | 16 | 0.23 |
| Nitrogen oxides | 67 | 1.1 |
| Carbon monoxide | 17 | 1.0 |
| Particulates (PM-10) | 6.1 | 0.09 |
| Sulfur oxides | 2.8 | 0.07 |

Certain operations at Site 300 require permits from SJVUAPCD. The total estimated air emissions during 2002 from operations (permitted and exempt air sources) at Site 300 are given in **Table 4-2**. 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 2002 are well below levels that would 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 and modeling HT emissions as HTO as required by EPA) is 0.081 µSv/y (0.0081 mrem/y). Evaluating the emissions with NEWTRIT, a model that expressly treats the HT emissions and incorporates the dose from organically bound tritium (see Chapter 13), the dose to the hypothetical maximally exposed member of the public is 0.056 µSv/y (0.0056 mrem/y).

In either case, the dose is far below the NESHAPs standard of 100 µSv/y (10 mrem/y), and the doses are below those from naturally occurring radiation. Thus, the estimated radiological dose caused by measured air emissions from LLNL operations is minimal. See **Table 13-2** for a summary of doses.

Estimated nonradioactive air emissions, which are also very small compared with emissions in surrounding areas, are well below standards and pose no threat to the environment or public health.

## Contributing Authors Acknowledgment

The author acknowledges Barbara Nisbet as a contributor to the "Nonradioactive Air Emissions" section of this chapter.