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Air Monitoring Programs

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Lawrence Livermore National Laboratory performs continuous air sampling to evaluate its compliance with local, state, and federal laws and regulations and to ensure that human health and the environment are protected from hazardous and radioactive air emissions. Federal environmental air quality laws and U.S. Department of Energy (DOE) regulations include Title 40 of the Code of Federal Regulations (CFR) Part 61, the National Emissions Standards for Hazardous Air Pollutants (NESHAPs) section of the Clean Air Act, and applicable portions of DOE Order 5400.5, Radiation Protection of the Public and the Environment, and American National Standards Institute (ANSI) standards. The *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (U.S. DOE 1991) provides the guidance for implementing DOE Order 5400.5.

The U.S. Environmental Protection Agency (EPA) Region IX has oversight responsibility for LLNL compliance with radiological air emissions regulations. Enforcement authority for the Clean Air Act regulations pertaining to nonradiological air emissions belongs to two local air districts, the Bay Area Air Quality Management District (BAAQMD) and the San Joaquin Valley Unified Air Pollution Control District (SJVUAPCD).

Whereas LLNL conducts air effluent monitoring of atmospheric discharge points to determine the actual radionuclide releases from individual facilities and processes during routine and nonroutine operations, ambient air monitoring at LLNL-site and off-site locations determines if airborne radionuclides or hazardous materials are being released by LLNL operations to its environs. Ambient air monitoring also serves to verify the air concentrations predicted by air dispersion modeling and to determine compliance with the NESHAPs regulation. (See *LLNL NESHAPs 2003 Annual Report* [Harrach et al. 2004] and Chapter 6.)

Air Effluent Monitoring

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 section apply to stationary point source discharges.

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.) It 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. 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.

Methods

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. 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. 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. 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 2003, LLNL operated 72 sampling systems for radioactivity from air exhausts at 8 facilities at the Livermore site (see **Figure 3-1**) and 1 sampling system at Site 300 (see **Figure 3-2**). These systems are listed in **Table 3-1** along with the analytes of interest, the type of sampler, and the number of samplers. LLNL periodically reassesses the need for continuous monitoring and assesses new operations or changes in operations. From NESHAPs assessments of operations during 2003, one additional discharge point, a new operation in Building 695, was found to require continuous sampling.

LLNL also operates a low-volume radiological air particulate network that consists of two samplers located at HOSP and FCC. The results from these samplers are used to establish background levels of gross alpha and beta activity for direct comparison to emissions from the air effluent samplers. These low-volume samplers collect particulate at a continuous rate of 0.03 m^3/min using membrane filters.

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

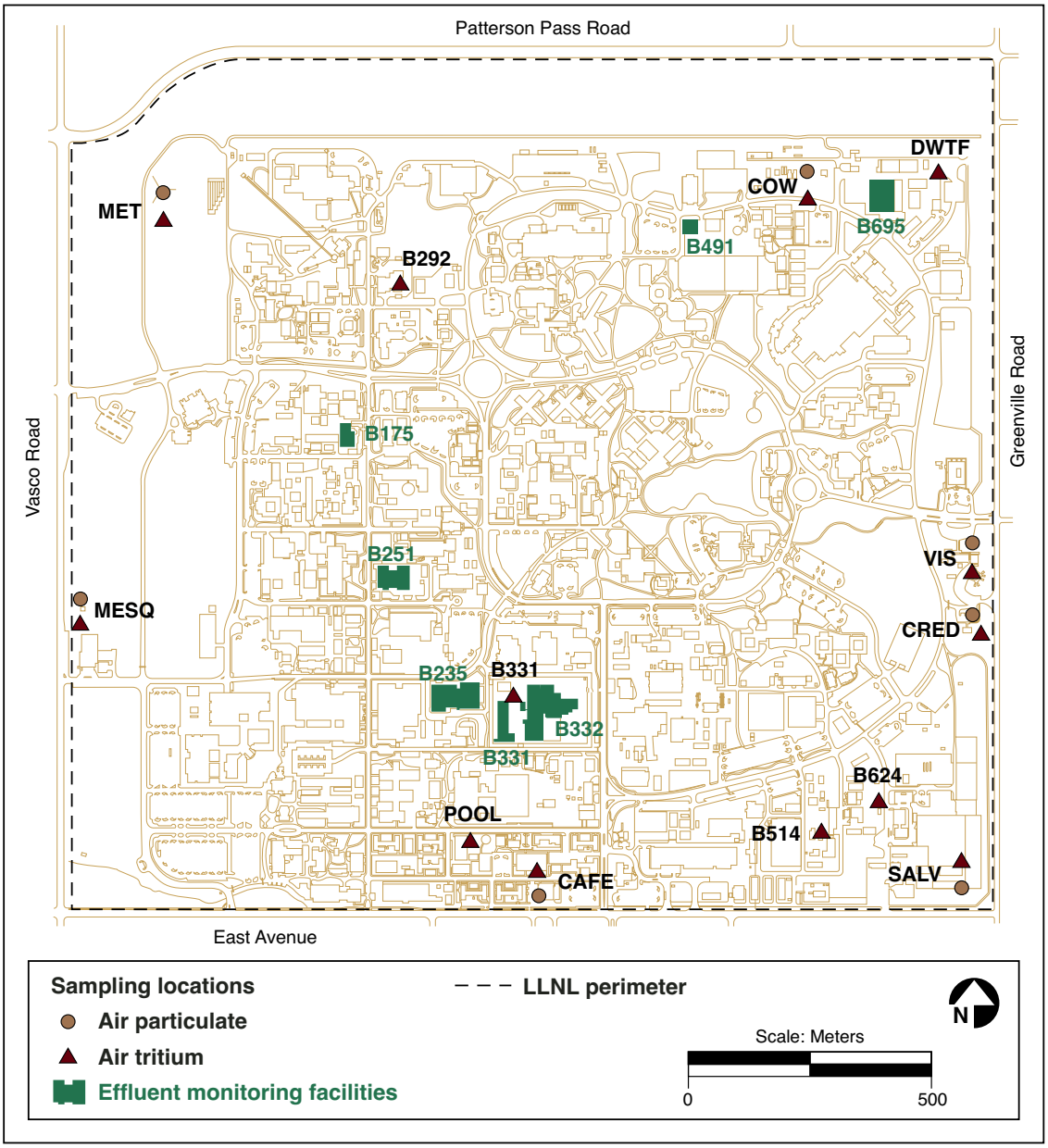

Figure 3-1. Livermore site air monitoring locations, 2003

Program was terminated, 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. In May 2003, sampling at Building 175 was discontinued because the facility no longer possessed a radionuclide inventory or plans to conduct activities with

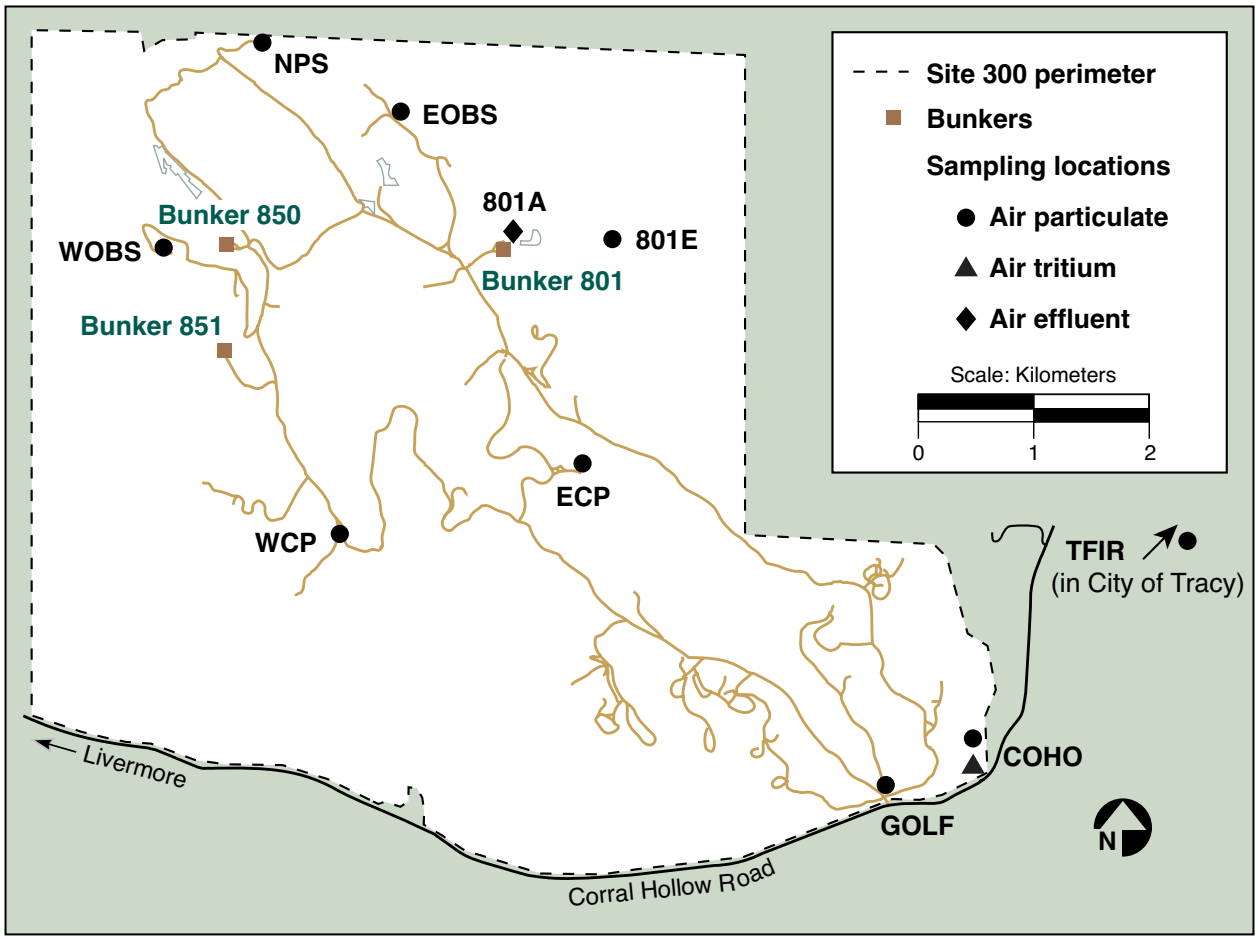

Figure 3-2. Site 300 air monitoring locations, 2003

radionuclides in the forseeable future. The air effluent sampling system at Building 491 continues 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 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 3-1**) at discharge points to provide faster notification in the event of a release of radioactivity.

Table 3-1. Air effluent sampling locations and sampling systems

Building	Facility	Analytes	Sampler type	Number of samplers
175	Mars[a]	Gross α, β on particles	Filter	6
235	Chemistry and Materials Science	Gross α, β on particles	Filter	1
251	Heavy Element	Gross α, β on particles	Filter	27
331	Tritium	Tritium	Stack ionization chamber[b]	4
		Gaseous tritium and tritiated water vapor	Molecular sieves	4
332	Plutonium	Gross α, β on particles	Stack CAM[b,c]	12
		Gross α, β on particles	Filter	15
491	Laser isotope separation	Gross α, β on particles	Filter	1
695	Decontamination and Waste Treatment Facility	Gross α, β on particles	Filter	1
801A	Contained Firing Facility	Gross α, β on particles	Filter	1

a Operations discontinued; however, air effluent sampling systems at this building continued to operate as part of the maintenance and surveillance shutdown plan for the facilities. The sampling system in Building 175 was removed from service in May 2003; the building no longer contained an inventory of radioactive materials.

b Alarmed systems

c CAM = Eberline continuous air monitors

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. Air effluent samples were obtained in accordance with written standardized procedures summarized in the *Environmental Monitoring Plan* (Woods 2002).

The following sections discuss the radiological air emissions from facilities that have continuously monitored discharge points. All effluent air analytical results are summarized in the file "Ch3 Air Effluent" included on the report CD.

Air Effluent Radiological Monitoring Results

In 2003, a total of 4.1 TBq (110 Ci) of tritium was released from the Tritium Facility (Building 331). Of this, approximately 3.8 TBq (104 Ci) were released as tritiated water vapor (HTO). The remaining tritium released, 0.22 TBq (6.0 Ci), was elemental tritium gas (HT). The median emissions from the facility were 6.0×10^3 Bq/m^3 (1.6×10^{-7} Ci/m^3) for HTO, and 5.1×10^2 Bq/m^3 (1.4×10^{-8} Ci/m^3) for HT. The

highest single weekly stack emission from the facility was 0.38 TBq (10.2 Ci), of which 0.37 TBq (10.05 Ci) was HTO. Emissions from Building 331 for 2003 continued to remain considerably lower than those during the 1980s. **Figure 3-3** illustrates the combined 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 a sample will exhibit concentrations greater than the MDC. Generally, these samples are only marginally above the MDC. Due to the way some of the exhaust systems are configured, the monitoring systems sometimes sample air from the atmosphere in addition to HEPA-filtered air from facility operations, thereby collecting background atmospheric radioactivity. If gross alpha is detected, a check is performed to determine if the ventilation blowers were operational at the time of the detection and the results are compared to the low-volume sampling results. If the blowers were operational, the sample result is considered a valid detection; otherwise the result is considered to be background atmospheric radioactivity. None of the facilities monitored for gross alpha and beta at the Livermore site had emissions in 2003.

Building 801A has a continuously monitored air effluent sampling system because depleted uranium is used during facility operations. Building 801 facility activities involving radionuclides are performed in the area of the building where the air is exhausted through HEPA filtration. Operations conducted in the building high bay do

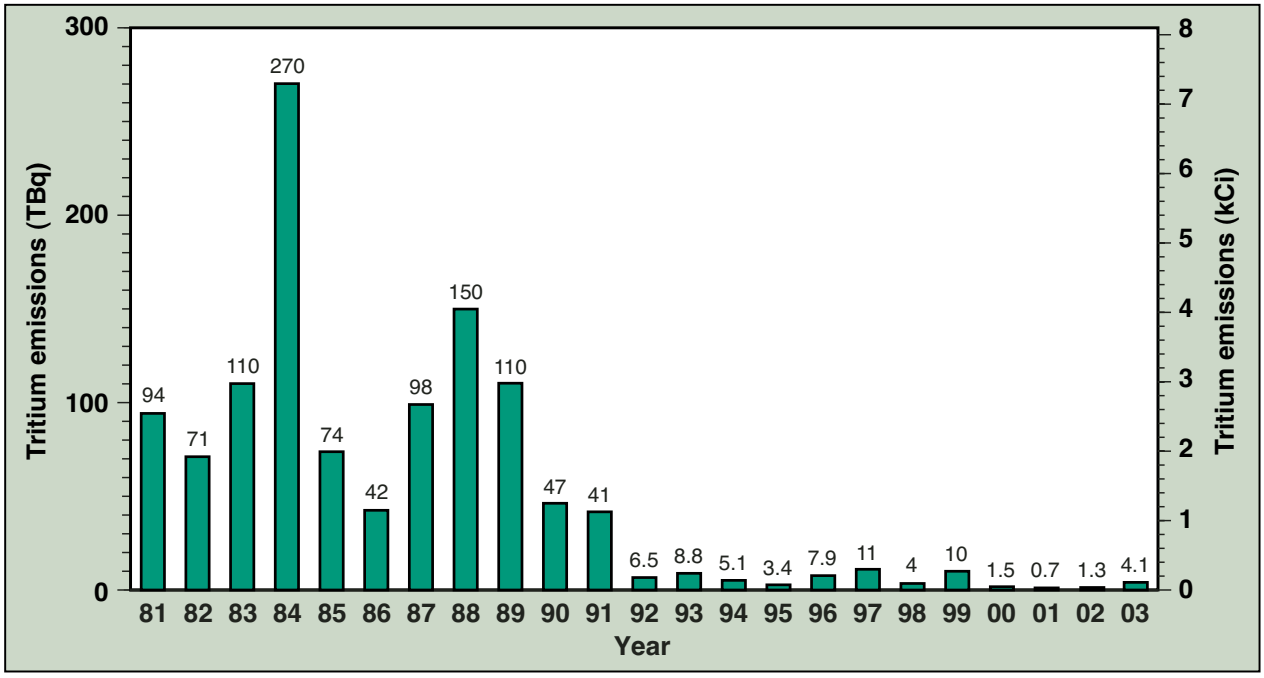

Figure 3-3. Tritium Facility combined HTO and HT emissions from 1981 through 2003

not use radionuclides and exhaust to the stack without HEPA filtration. Consequently, some of the air sampled by the effluent sampling system is unfiltered. In order to determine if any emissions occurred from this facility, the sampling results must be compared to results from the ambient air samplers. In 2003, ten samples out of 48 had concentrations greater than the MDC. The median concentration of the Building 801A alpha detections was 2.4×10^{-4} Bq/m^3 (6.6×10^{-15} Ci/m^3), which is approximately three times higher than the median concentration of the off-site sampling locations. The conservative approach is being taken by reporting this as actual emissions.

The gross alpha emissions for Building 801A was 1.9×10^4 Bq/y (5.1×10^{-7} Ci/y). **Table 3-2** summarizes total radiological emissions as determined from the continuous sampling of facility exhausts for 2003.

Nonradiological Results

The Livermore site currently emits approximately 101 kg/day of regulated air pollutants as defined by the Clean Air Act (e.g., 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). **Table 3-3** lists the estimated Livermore site 2003 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 2003 were approximately 7.1×10^4 kg/day, compared with an estimate for LLNL releases of 63 kg/day for the Livermore site (0.09% of total Bay Area emissions from stationary sources). The BAAQMD estimate for reactive organic emissions was 9.4×10^4 kg/day for 2003, versus the Livermore site's estimated releases of 14 kg/day (0.01% of total Bay Area emissions from stationary sources) in 2003.

Certain operations at Site 300 require permits from SJVUAPCD. The total estimated air emissions during 2003 from operations (permitted and exempt air sources) at Site 300 are given in **Table 3-3.** 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.

Table 3-2. Measured radiological air effluent emissions above the detection limit for Livermore site and Site 300, 2003

Building (Facility)	HT (Bq)	HTO (Bq)	Gross alpha (Bq)	Gross beta (Bq)
331 (Tritium Facility)	2.2×10^{11}	3.8×10^{12}	—	—
801A (Contained Firing Facility)	—	—	1.9×10^4	$9.7 \times 10^{4(a)}$

a Value is consistent with background

Table 3-3. Nonradioactive air emissions, Livermore site and Site 300, 2003

Pollutant	Estimated releases (kg/day)	
	Livermore site	Site 300
Organics/volatile organics	14	0.57
Nitrogen oxides	63	0.97
Carbon monoxide	16	0.22
Particulates (PM-10)	5.8	0.31
Sulfur oxides	1.7	0.98

Environmental Impact on Air Effluent

Measured radiological air emissions from the Livermore site operations for 2003 are well below levels that would cause concern for public health and are well below existing regulatory standards for radioactive dose. The site-wide dose to the hypothetical maximum exposed individual of the public from Building 801A is 1.3×10^{-5} µSv/y (1.3×10^{-6} mrem/y). The dose to the hypothetical maximally exposed member of the public caused by the measured air emissions from the Tritium Facility (modeling HT emissions as HTO as required by EPA) is 0.22 µSv/y (0.022 mrem/y). Thus, the estimated radiological dose caused by measured air emissions from LLNL operations is minimal. See Chapter 6 for a discussion 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.

Ambient Air Monitoring

LLNL monitors ambient air to determine if airborne radionuclides or hazardous materials are being released by Laboratory operations, what the concentrations are, and what the trends are in the LLNL environs. In the ambient air monitoring program, LLNL collects particles on filters and physically traps vapors on a collection medium. Concentrations of various airborne radionuclides (including particles and tritiated water vapor) and beryllium metals are measured at the Livermore site, Site 300, and at off-site locations throughout the Livermore Valley and in the City of Tracy. In addition, some point sources and diffuse, or nonpoint sources, are monitored to fulfill NESHAPs requirements. In 2003, the EPA approved use of the air surveillance monitoring data from the location of the site-wide maximally exposed individual (SW-MEI) to demonstrate compliance with NESHAPs for minor emission point sources (Harrach et al.

2003). In addition, the Derived Concentration Guides (DCGs) found in DOE Order 5400.5 specify the concentrations of radionuclides that can be inhaled continuously 365 days a year without exceeding the DOE primary radiation protection standard for the public, which is 1 mSv/y (100 mrem/y) effective dose equivalent. (Chapter 6 provides an explanation of this and other units of dose.) Data tables in this chapter present the DCG and the percent of the DCG for the given isotope. For beryllium metals, an ambient air concentration limit of 10,000 pgm/m^3 is established by the BAAQMD under Regulation 11 for the Hazardous Air Polltants.

Methods

Monitoring networks are established for surveillance of air particulates and tritium in the environs of the Livermore site and Site 300, as well as in the surrounding Livermore Valley and in the City of Tracy. All monitoring networks use continuously operating samplers.

The sampling locations for each monitoring network are listed in **Table 3-4** and shown on **Figures 3-1**, **3-2** and **3-4**. Several locations target specific areas of known contamination while other locations are considered perimeter or background locations. Duplicate quality control samplers operate in parallel with the permanent sampler at a given site, and these samples are analyzed to confirm results. Trip blanks are also taken on the air sampling routes to help identify any contaminate introduction during the sampling process.

In 2003, an LLNL state-certified analytical laboratory performed all sample analyses. Samples were analyzed for gross alpha and beta activity, gamma-emitting radionuclides, plutonium, uranium, tritium and beryllium metals. **Table 3-4** provides the requested analysis for each ambient air sampling station. Ambient air samples were obtained in accordance with written standardized procedures summarized in the *Environmental Monitoring Plan* (Woods 2002).

Sample Collection

The air particulate networks use high-volume air sampling units, which collect airborne particulate at a continuous rate of 0.42 m^3/min using Whatman 41 cellulose filters. The tritium samplers, operating at a flow rate of 500 cm^3/min, use a continuous vacuum pump to capture air moisture on silica gel contained in sampling flasks. These flasks are changed every two weeks.

Table 3-4. Sampling locations and type and frequency of analyses for ambient air

Livermore site						
	Target location	**Weekly gross alpha & beta (high volume)**	**Monthly $^{239+240}$Pu**	**Monthly Gamma & $^{235, 238}$U[a]**	**Monthly beryllium**	**Biweekly tritium**
Network	**Air particulate**					**Air vapor**
Collection Media	**Cellulose**					**Silica gel**
SALV, MET MESQ, COW CAFE, VIS[b]	Perimeter	X	X	X	X	X
DWTF	Perimeter					X
B331, B514, B624 POOL, B292[c]	Diffuse					X
ZON7	Downwind	X	X			X
PATT	Downwind	X	X			X
AMON	Downwind	X	X			X
CHUR	Upwind	X	X			X
FCC[d], VET, FIRE, CRED[b]	Upwind	X	X			X
HOSP[d], TANK	Upwind	X	X			X
LWRP	Special Interest	X	X			
Site 300						
		Weekly gross alpha & beta (high volume)	**Monthly Gamma & $^{239+240}$Pu[a]**	**Monthly $^{235, 238}$U**	**Monthly beryllium**	**Biweekly tritium**
Network		**Air particulate**				**Air vapor**
Collection Media		**Cellulose**				**Silica gel**
EOBS, GOLF, WOBS	Onsite[b]	X	X	X	X	
ECP, WCP, NPS, 801E	Onsite[b]	X	X	X		
COHO	Onsite[b]	X		X		X
TFIR	Offsite[b]	X		X	X	

a Perimeter composite samples include portions of weekly filters from the specified locations.

b SW-MEI

c Removed from Air Tritium Network in July 2003

d Low-volume sampler also operated at this location, the collection media is millipore filters. These samplers are operated to provide background values for the air effluent monitoring program.

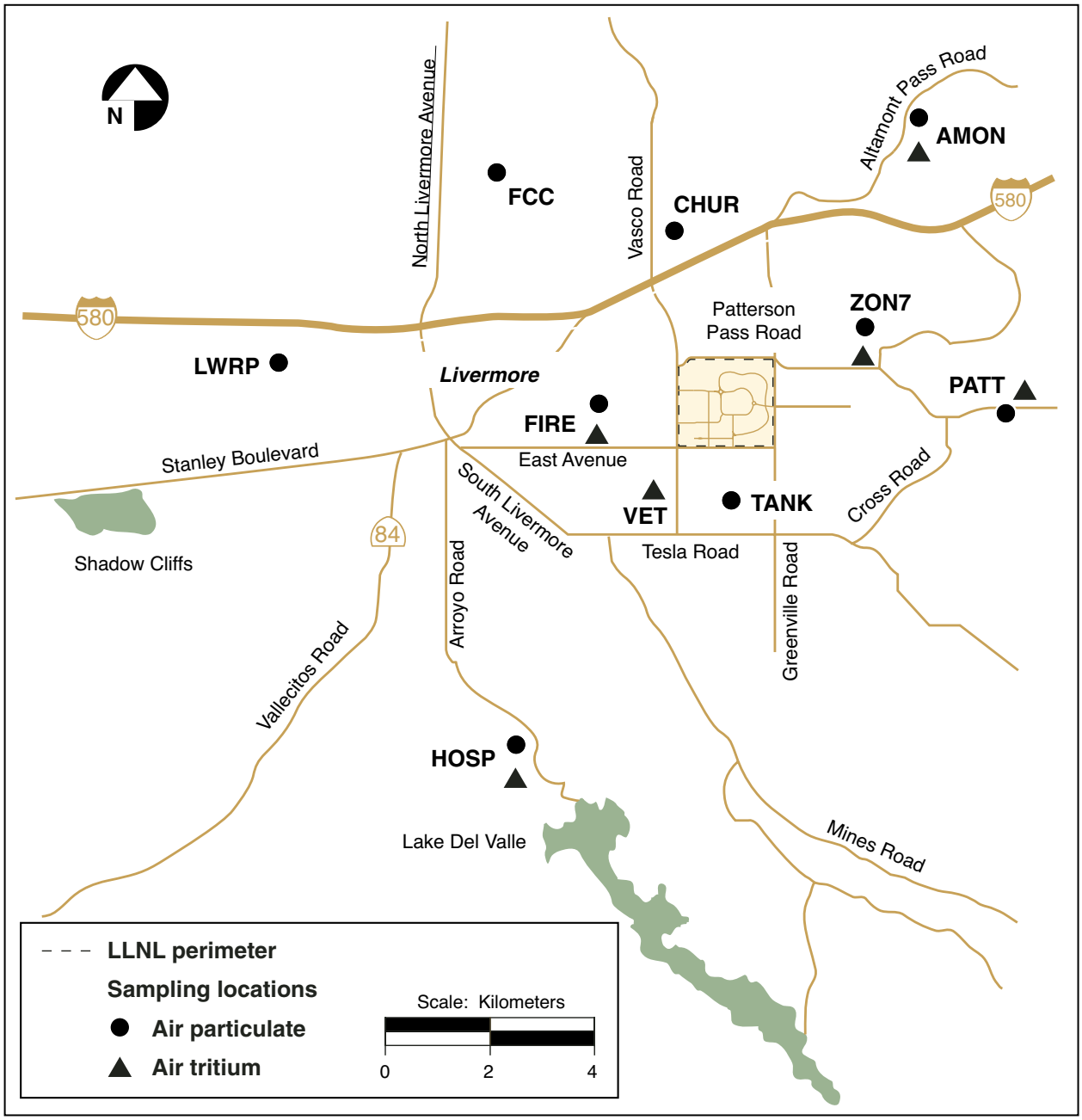

Figure 3-4. Air particulate and tritium sampling locations in the Livermore Valley, 2003

Sampling Locations

Using the historical meteorological data to determine prevailing wind direction, LLNL staff has positioned all ambient air samplers to provide reasonable probability that any significant concentration of radioactive or beryllium effluents from LLNL operations will be detected. LLNL activities are routinely evaluated though air dispersion modeling to ensure the sampling units are positioned properly.

Monitoring networks are established for surveillance of air particulates and tritium in the environs of the Livermore site and Site 300, as well as in the surrounding Livermore Valley and in the City of Tracy. There are 7 air particulate samplers on the Livermore site, 9 in the Livermore Valley, 1 in the City of Tracy, and 8 at Site 300. There are 12 air tritium samplers at the Livermore site, 6 in the Livermore Valley, and 1 at Site 300.

In general, air sampling locations are grouped in categories representing the following areas; perimeter, upwind, downwind, diffuse soures or areas of known contaminaiton, and special interest locations. The results from locations CRED and VIS serve as the SW-MEI for NESHAPs minor source compliance. Because resuspension of soil at Site 300 is the minor source of greatest interest, the average of all 8 locations serves as the SW-MEI for NESHAPs minor source compliance.

The air tritium sampling location B292 was removed in July and a new air tritium sampling location was added at CRED.

Beryllium is monitored at six Livermore site perimeter locations as required by the BAAQMD. Although there is no requirement to monitor beryllium at Site 300, as a best management practice, it is monitored at three locations on-site and at one location in the City of Tracy (see **Figure 3-2**).

Sample Analysis

Gross alpha and gross beta activities are determined by gas flow proportional counting; plutonium isotopes by alpha spectrometry; uranium isotopes by inductively coupled plasma-mass spectrometry; gamma emitters by gamma spectroscopy; and tritium by freeze-dried vacuum distillation followed by liquid scintillation. Procedures for analysis are summarized in the *Environmental Monitoring Plan* (Woods 2002). Beryllium metal concentration is determined by inductively coupled plasma-mass spectrometry as described in UCRL-POST-201469. See **Table 3-4** for the frequency of analysis at each location. In addition to using the analytical methods summarized in this section, the analytical laboratory also runs a series of quality control tests that include laboratory control spikes, blanks and duplicates. The analytical laboratory reports the actual instrumentation values, including negative results that arise when background measurements are higher than those for the sampled filters.

Because plutonium research occurs at the Livermore site, plutonium analyses are performed individually for all Livermore locations. However, plutonium is not used at Site 300; therefore, a composite from all locations is analyzed.

Uranium use at the Livermore site is very minimal so a composite from all the Livermore site perimeter locations is created and analyzed for uranium activity. However, at Site 300, where depleted uranium is used in explosives testing, specific locations are analyzed for uranium activity.

Results

As outlined in *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (U.S. DOE 1991), gross alpha, gross beta, and gamma emitters on air filters are used as trend indicators; specific radionuclide analysis is done for plutonium, uranium, and tritium. Radiological analytical results are reported as a measured activity per volume of air. Regardless of whether any activity is considered to have been detected, the result of the analysis is reported. The activities shown in the tables, located in the file "Ch3 Ambient Air" included on the report CD, displaying monthly and biweekly data are measured concentrations and their associated $\pm 2\sigma$ counting errors.

Particle size distribution on air samples is not determined because the estimated effective dose equivalent to the maximally exposed individual (from the total particulate) is well below the 0.01-mSv (1-mrem) environmental regulatory guide allowable limit (U.S. DOE 1991) using the total particles collected.

Gross Alpha and Gross Beta Concentrations

The primary sources of the alpha and beta activities are the naturally occurring radioisotopes. **Figure 3-5** shows the three-year history of monthly gross alpha and gross beta median activities for the Livermore site perimeter, Livermore Valley, and Site 300 sampling locations. In 2003 the typical gross alpha activity (annual median value) for the Livermore site perimeter was 37 $\mu Bq/m^3$ (0.99 fCi/m^3); for the upwind Livermore Valley stations, the value was 31 $\mu Bq/m^3$ (0.84 fCi/m^3); for the downwind Livermore Valley stations, the value was 36 $\mu Bq/m^3$ (0.97 fCi/m^3); and for Site 300, the value was 31 $\mu Bq/m^3$ (0.84 fCi/m^3). The annual gross beta median for all downwind locations was 320 $\mu Bq/m^3$ (8.6 fCi/m^3); for upwind locations it was 300 $\mu Bq/m^3$ (8.2 fCi/m^3); for the Livermore site perimeter it was 330 $\mu Bq/m^3$ (8.8 fCi/m^3; and for Site 300 it was 320 $\mu Bq/m^3$ (8.6 fCi/m^3).

These data follow a pattern similar to previous years with a seasonal increase in the fall and early winter months. As local soils dry out during the summer months, the resuspended particulate can build up and increase until the winter rains begin. From May to November 2003, LLNL measured less than 0.5 inches of rainfall, while November and December recorded 3.5 and 6.7 inches, respectively. This increase in rainfall most likely reduced the resuspension contribution on air filters, subsequently showing a sharp decline in the gross alpha and gross beta activity detected once the rains started. Isotopic

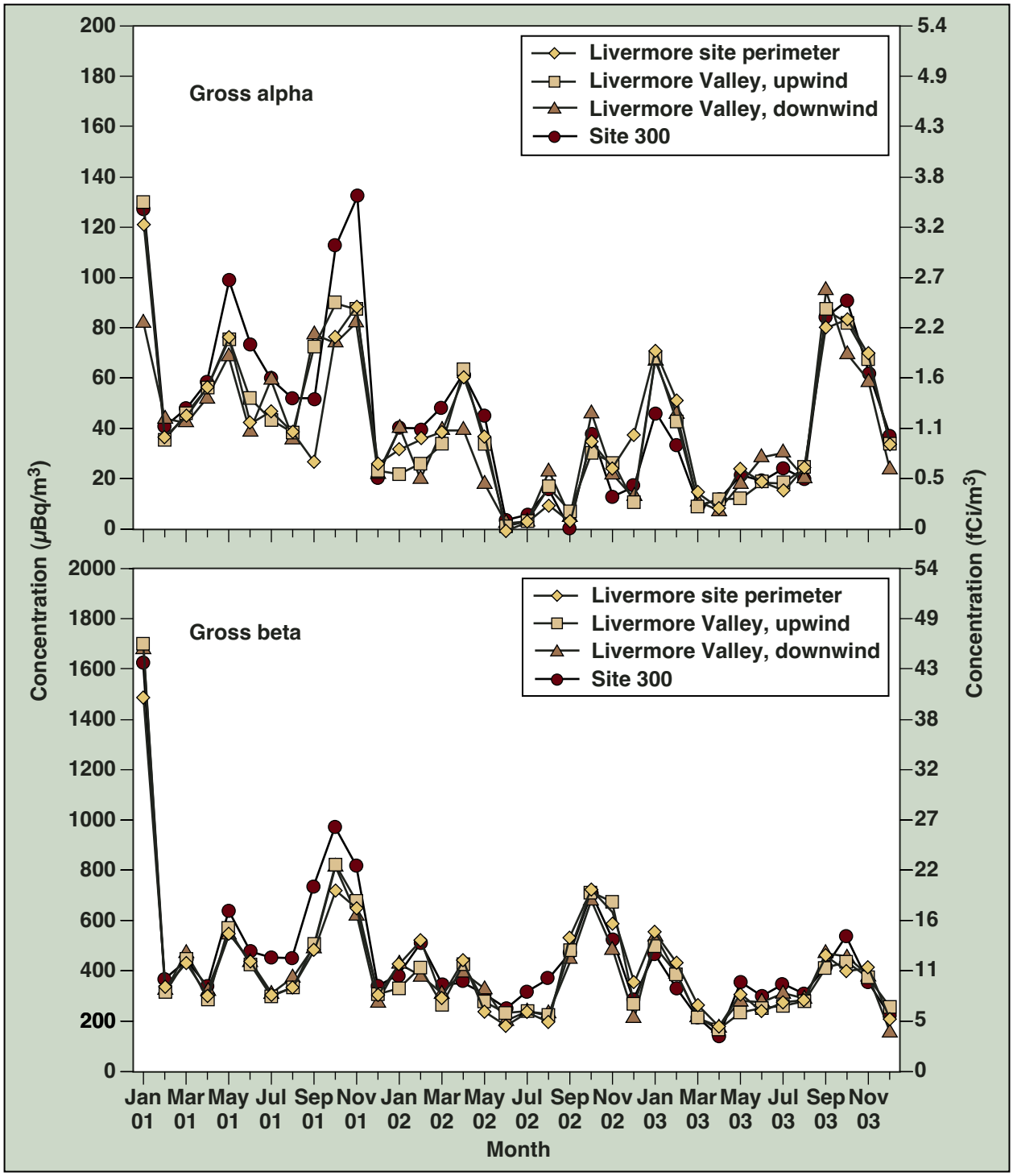

Figure 3-5. Three-year history of the median gross alpha and gross beta activities for all particulate samples grouped by area, 2001-2003

gamma results of these samples indicate that the higher activities are the result of naturally occurring isotopes (uranium, thorium, potassium, and lead) which are also routinely found in local soils.

Site 300 is less developed and has more barren soil compared to the Livermore site. As a result, Site 300 air samples tend to collect more particulate from resusupended soils. The pattern of activity as seen in **Figure 3-5** however is very similar to the Livermore air samples with a increase in the fall and early winter months then decrease during the winter time as rains reduce the resuspension effect. The highest weekly gross alpha sample measured at Site 300 was 218 µBq/m^3 (5.9 fCi/m^3) at WOBS. This sampler is near the active B851 bunker and the Contained Firing Facility (Building 801) where historical operations occurred in open-air shots.

The highest Site 300 onsite weekly gross beta value was 1060 µBq/m^3 (28.6 fCi/m^3) at 801E, also near the Contained Firing Facility. The offsite sampler for the Site 300 route, located in an industrial area, recorded the highest overall concentrations with a weekly maximum of 1360 µBq/m^3 (36.7 fCi/m^3), which is consistent with historical measurements from this sampling location. Because this sampler was in an industrial area, it does not represent a typical background location for Site 300. It was replaced in January 2004 with a more suitable location closer to Site 300 in a more rural area.

Gamma-Emitting Radionuclides

By analyzing air samples for gamma-emitting radionuclides, LLNL verifies that there is no evidence of release of the small inventories of mixed fission products and radiochemical tracers used by LLNL. This analysis will also reveal emissions from global fallout sources such as aboveground tests and the Chernobyl accident (Holland et al. 1987). Composite samples for the Livermore site and Site 300 are analyzed for an environmental suite of gamma-emitting radionuclide concentrations in air. Of those isotopes, beryllium-7 (cosmogenic in origin), lead-210 and potassium-40, all of which are naturally occurring in the environment, were consistently detected at both sites. The results are within known background levels (see file "Ch3 Ambient Air" on report CD for analytical results).

Plutonium Concentrations

Historical environmental plutonium 239+240 activity for the past 20 years is shown in **Figure 3-6**. Locations HOSP and VIS represent typical upwind and onsite sampling locations. Plutonium activity at both of these sites has been decreasing as fallout diminishes and surface areas of potential resuspension have been covered with pavement or buildings. These positive detections are likely due to resuspended soils. LLNL monitors all Livermore area samplers and a composite is created from all onsite Site 300 samplers.

Plutonium 239+240 was detected in 12 of the 187 samples tested from Livermore area air samples. Seven of those positive samples came from on-site samplers. The majority of these were from the SALV location that is in the southeast quadrant of LLNL. The southeast quadrant is an area of known plutonium contamination (see *Environmental*

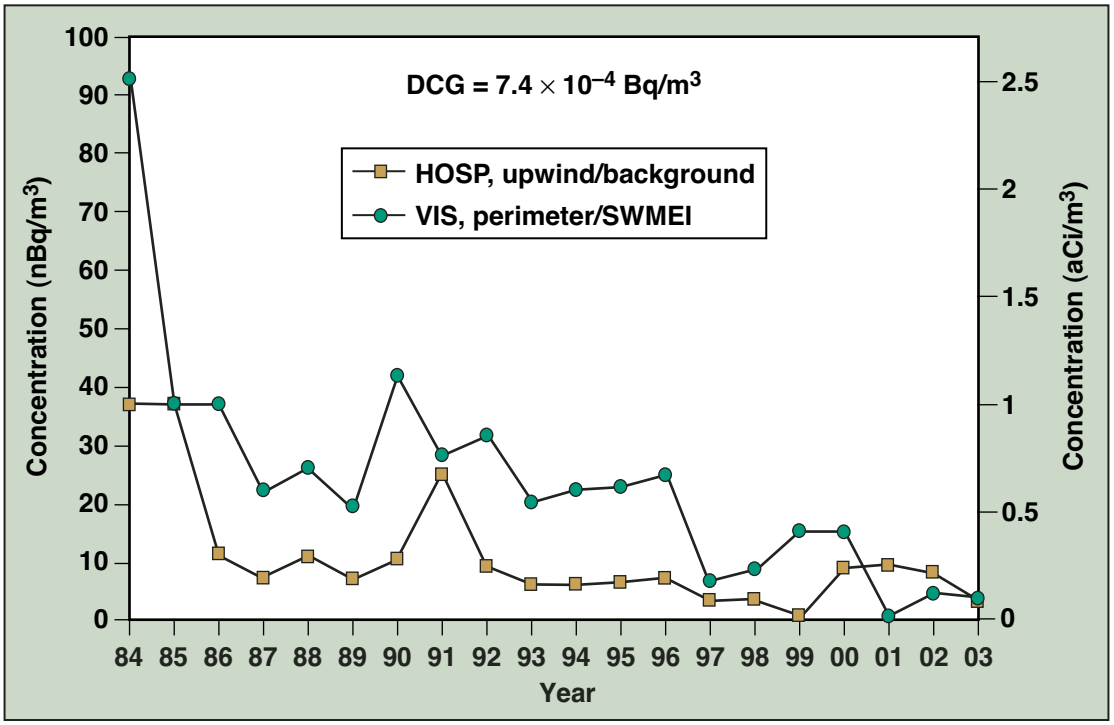

Figure 3-6. Calculated annual median concentrations of plutonium-239+240 for HOSP and VIS for the last 20 years

Report 1990 [Sims et al. 1991]). These plutonium detections can be attributed to the increased construction activities here during summer 2003 that created higher resuspension of localized soils. The highest recorded onsite plutonium 239+240 detection was in December at COW of 65.5 nBq/m^3 (1.77 aCi/m^3) (0.009% of the DCG), while the highest offsite detection occurred in June at the downwind location AMON. This result of 25.5 nBq/m^3 (0.69 aCi/m^3) is only 0.003% of the DCG. Plutonium was detected in 2 of the 12 composite samples collected from Site 300. The highest of these occurred in October with a value of 5.92 nBq/m^3 (0.16 aCi/m^3). This concentration is only 0.0008% of the DCG. All positive detections for plutonium from either site were far below the DCG of 0.74 mBq/m^3.

Uranium Concentrations

Uranium ratios are used to determine the type of uranium present in the environment. Natural uranium has a mathematical ratio of uranium-235/uranium-238 of 0.00725 and depleted uranium has a uranium-235/uranium-238 ratio of 0.002.

Uranium isotopes are naturally occurring and all but one of the uranium-235 analyses had positive detections. The Livermore site composite had a uranium-235 median concentration of 0.17 µg/m^3 and a uranium-238 median concentration of 21.5 µg/m^3. This composite had a median ratio of 0.007, which is considered natural uranium.

The annual median uranium-235 concentration for the all Site 300 locations was 0.15 pg/m^3 (or less than 0.00003% of the DCG) and the uranium-238 median concentration was 22 pg/m^3 (or less than 0.0008% of the DCG). As with the Livermore site isotopic ratio for the annual median was 0.007, which is considered natural uranium.

Tritium Concentrations

Tritium data presented in **Table 3-5** summarize the biweekly tritium data provided in data tables on the report compact disk (see file "Ch3 Ambient Air" on the report CD). Locations are grouped by expected concentrations of tritium. The highest concentrations of tritium are from samplers on the Livermore site near locations of diffuse tritium (B292, B331, B514, and B624). The sources of tritium in these locations are mostly stored containers of tritium waste or tritium-contaminated equipment; B292 is near a pine tree that acts as a diffuse source of tritium because its roots are growing in water contaminated with tritium from an underground retention tank that previously leaked. Median concentrations for 2003 at the B292 location and the B514 location were comparable to those from 2002. The B292 sampling location was taken off-line in July 2003 because the results from this location have not been used since 1997 to demonstrate compliance with NESHAPS for the diffuse source at this location (see Chapter 5). The annual median concentrations for 2003 for the remaining two diffuse-source samplers (B331 and B624) were slightly elevated from concentrations in 2002 with the more substantial change at the B331 location.The highest biweekly concentration, 21.1 Bq/m^3 (5.6×10^{-10} Ci/m^3), was observed in October when cleanup activities involved the temporary storage of contaminated obsolete equipment outside Building 331.

Samplers on the perimeter of the Livermore site exhibit the next highest air tritium concentrations. Of the perimeter locations, POOL and DWTF (at the Decontamination and Waste Treatment Facility) exhibited the highest concentrations and the same median concentration which is 0.0034% of the DCG. Concentrations at POOL were comparable to those in 2002, but DWTF results were elevated by comparison. The increased concentrations at DWTF correlate to the Radioactive and Hazardous Waste Management Division starting to use the new Decontamination and Waste Treatment

Table 3-5. Tritium in air samples (mBq/m^3), 2003

Sampling locations	Detection frequency	Mean	Median	IQR	Maximum	Median Percent of DCG[a]
Diffuse on-site sources	95 of 95	1920	407	2600	21100	0.011
Livermore perimeter	194 of 223	138	64.4	105	1760	0.0017
Livermore Valley	70 of 161	28.2	13.2	29.6	744	0.00036
Site 300	5 of 26	2.83	3.13	12.9	29.3	0.000085

a DCG = Derived Concentration Guide of 3.7×10^6 mBq/m^3 for tritium in air.

Facility for operations that may include such items as storing containers of tritium waste or tritium-contaminated equipment. Median concentrations for 2003 for the perimeter locations were generally comparable to those for 2002. The several locations with slightly higher concentrations reflect higher emissions from the Tritium Facility (see "Air Effluent Monitoring" section). One new perimeter tritium sampling location was added to the network in July 2003. The CRED location was added as a monitoring point of compliance in lieu of inventory-based modeling for minor sources. See *NESHAPS 2002 Annual Report* (Harrach et al. 2003).

The median perimeter concentrations for 2003 (even when data from POOL and DWTF are omitted) are statistically higher than concentrations of tritium in air from the Livermore Valley. Sampling locations in the Livermore Valley demonstrate that LLNL tritium has only a small impact past the perimeter fence. Fifty-seven percent of the Valley samples had concentrations indistinguishable from zero. The median concentrations for the Valley locations for 2003 are comparable to those for 2002 except for AMON. Concentrations at AMON reflect the higher 2003 emissions from the Tritium Facility.

Table 3-5 shows the median concentration of tritium in air that was observed at the sampling location at Site 300 (see file "Ch3 Ambient Air" on report CD for biweekly data). Site 300 concentrations are mostly below the detection limit and most likely represent background levels of tritium unaffected by local sources of tritium.

Beryllium Metal Concentrations

LLNL measures the monthly concentrations of airborne beryllium for the Livermore site, Site 300, and the downtown Tracy sampling location (TFIR). (See file "Ch3 Ambient Air" on report CD for data.) The highest value at the Livermore site was 37.4 pg/m^3 which was recorded at location COW in March. This value is only 0.37% of the BAAQMD ambient concentration limit for beryllium (10,000 pg/m^3). With the exception of the slightly higher value in March, all data were similar to data collected from previous years.

Figure 3-7 is a plot of the median beryllium concentration at the Livermore site perimeter from 1975 through 2003. The decrease in median concentration in 1993 and the slight increase in 1999 were likely the result of a change in the analytical laboratory used to perform this analysis. LLNL monitors beryllium metals in air samples on the Livermore site as part of an agreement with the local BAAQMD.

There is no regulatory requirement to monitor beryllium in the San Joaquin County; however, LLNL analyzes samples from several Site 300 locations as a best management practice. The monthly median beryllium concentration for all Site 300 locations was 6.64 pg/m^3. The highest value for the Site 300 area samples occurred again in the October sample at the off-site location in Tracy (TFIR). The concentration at this location is typically higher than at all other locations (including Livermore area samplers) because it is located in a congested part of town and accumulates a greater amount of industrial particulate pollutants. This maximum value was 34.3 pg/m^3 with an annual median of 10.7 pg/m^3.

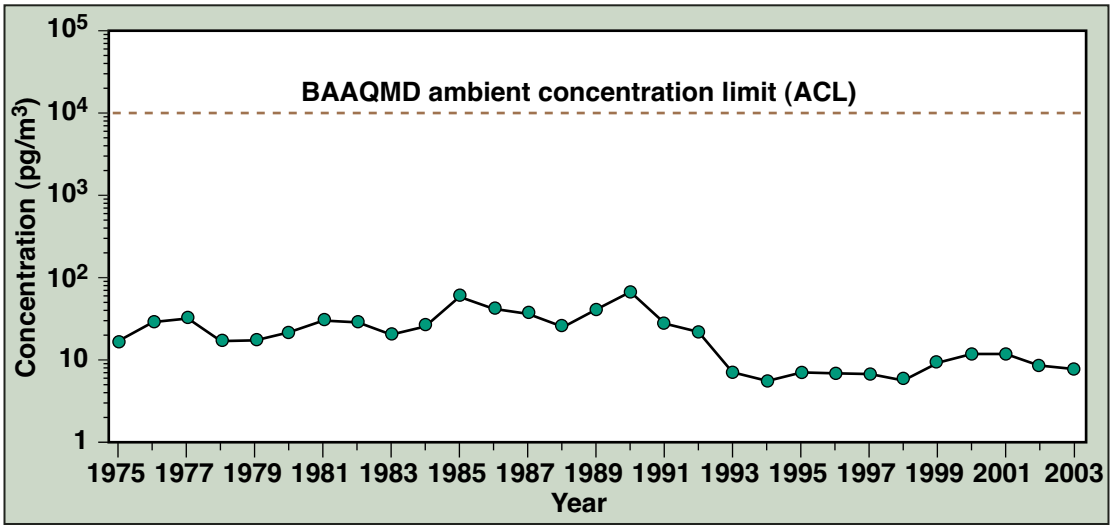

Figure 3-7. Median concentration of beryllium in air particulate samples taken at the Livermore site perimeter, 1975–2003

Environmental Impact on Ambient Air

LLNL operations involving radioactive materials had little impact on radionuclide concentrations in ambient air during 2003. Radionuclide particulate concentrations in air at the Livermore site and in the Livermore Valley were well below the levels that would cause concern for the environment or public health according to existing regulatory standards.

The diffuse tritium sources at DWTF, B292, B331, B514, and B624 had a small, localized effect with minimal impact, if any, on the public. Any potential dose received by a member of the public from the diffuse sources is included in doses calculated for tritium concentrations at the Livermore site perimeter. Tritium concentrations at the Livermore site perimeter which were slightly greater in 2003 than in 2002 correlate well with increased stack emissions in 2003. The increased tritium concentrations observed at the Livermore site perimeter had minimal impact on off-site concentrations.

A maximum tritium dose of 370 nSv/y to a member of the public at the Livermore site perimeter can be estimated based on the extraordinarily conservative assumption that the maximum biweekly concentration (1760 mBq/m^3) is maintained for an entire year and that a member of the public breathes that concentration for the entire year. This improbable inhalation dose to the public is just 0.37% of NESHAPs standard of 0.1 mSv/y arising as a result of releases of radionuclides to air from DOE facilities.

The concentrations of beryllium at both the Livermore site and Site 300 can be attributed to resuspension of surface soil containing naturally occurring beryllium. Local soils contain approximately 1 ppm of beryllium, and the air of the Livermore area and the Central Valley typically contains 10 to 100 μg/m^3 of particulates. Using a value of 50 μg/m^3 for an average dust load and 1 ppm for beryllium content of dust, a conservative airborne beryllium concentration of 50 pg/m^3 can be predicted. The overall average for the Livermore site and Site 300 (including TFIR location in Tracy) are 7.81 pg/m^3 and 6.76 pg/m^3, respectively. These data are lower than predicted, well below standards, and do not indicate the presence of a threat to the environment or public health.