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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. 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 enforcement authority 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 Air Pollution Control District (SJVAPCD).

Air effluent monitoring of atmospheric discharge points is conducted to measure the quantities of radionuclides released from individual facilities during routine and nonroutine operations; ambient air monitoring at LLNL-site and off-site locations determines if airborne radionuclides or beryllium are being released in measurable quantities to its environs by these and other LLNL operations. 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 2004 Annual Report* [Harrach et al. 2005].)

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 during routine and non-routine 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 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 SJVAPCD, and monitoring of the effluent is not required. Based on air toxics emissions inventory and risk assessment required by the California Air Toxics "Hot Spots" legislation, BAAQMD and SJVAPCD 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. 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. 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 2004, LLNL operated 67 sampling systems for radioactivity from air exhausts at 6 facilities at the Livermore site (see **Figure 3-1**) and 1 sampling system at Site 300 (see **Figure 3-2**). From NESHAPs assessments of operations during 2004, one additional discharge point, a new operation in the Building 695 yard, the TRU Mover, was found to require continuous sampling. 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.

Sampling for particles containing radioactivity was conducted in all six 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. Analytical results from the continuous samplers are reported as a measured concentration per volume of air or as less than the

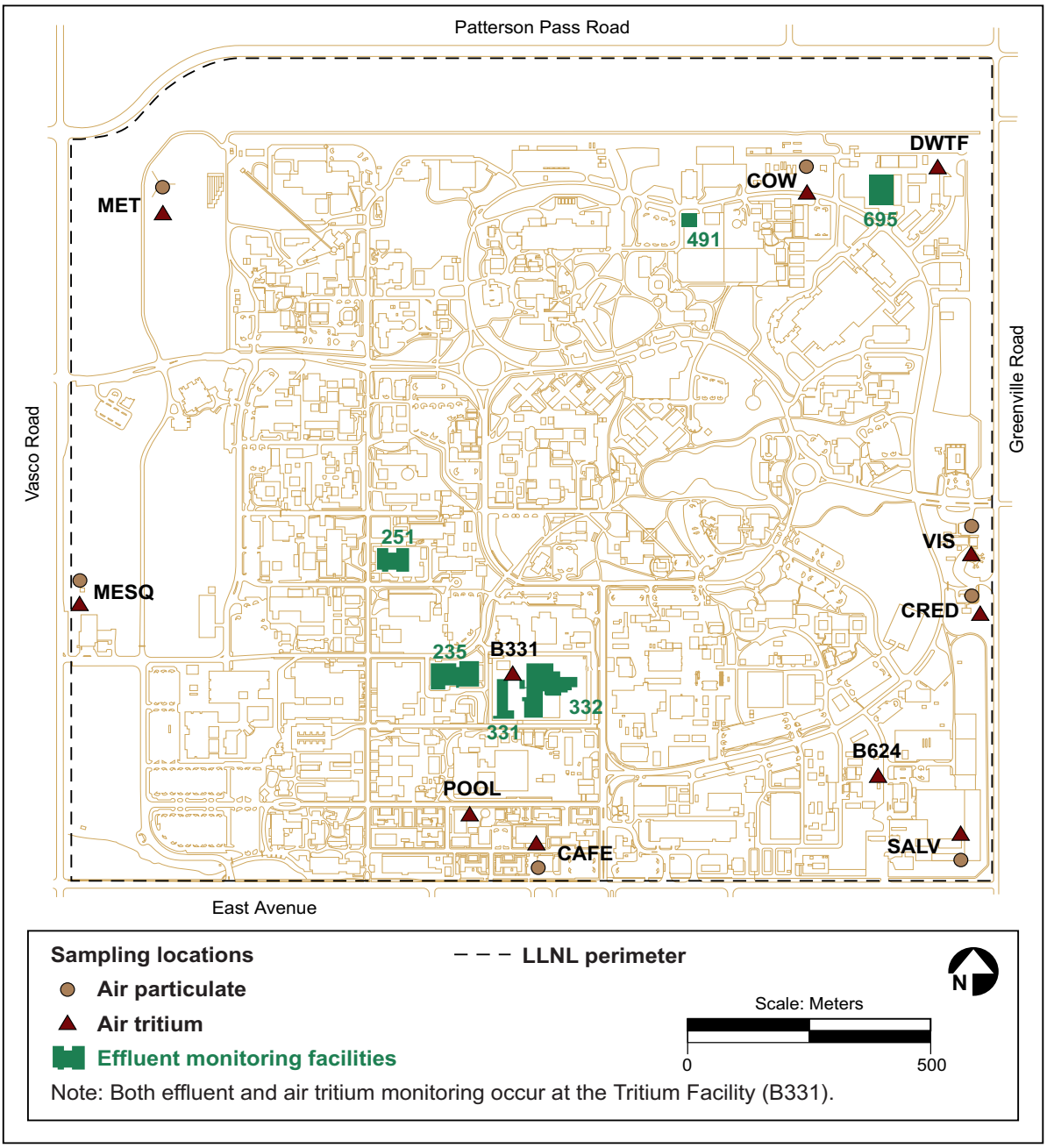

Figure 3-1. Livermore site air monitoring locations, 2004

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 2005).

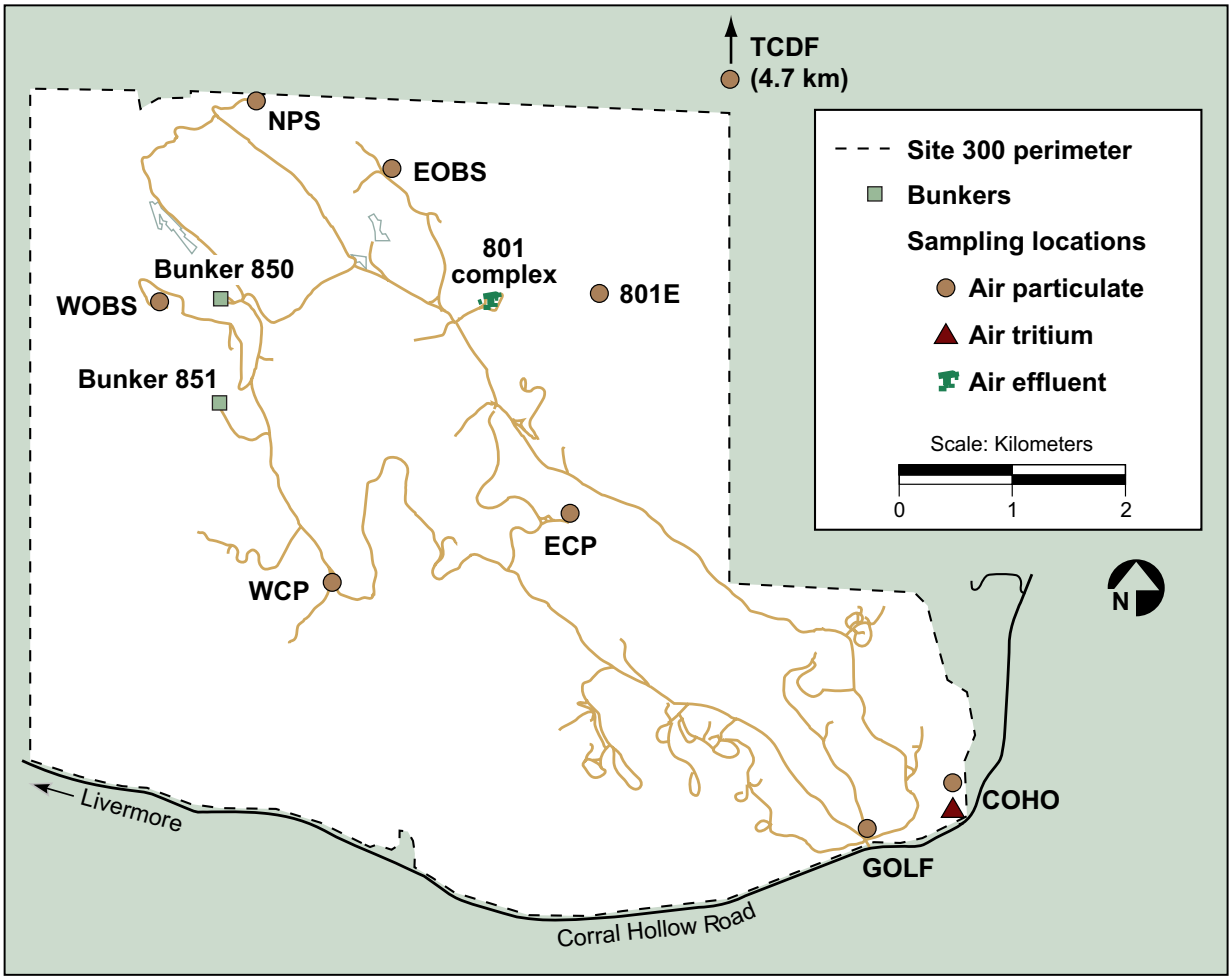

Figure 3-2. Site 300 air monitoring locations, 2004

To establish the background levels of gross alpha and beta activity that are used to determine if a release has occurred from monitored stacks, LLNL operates three low-volume radiological air particulate samplers at locations HOSP and FCC in the Livermore Valley and NPS at Site 300. These samplers collect particulate on membrane filters at a continuous rate of 0.03 m^3/min. The low-volume samplers are not part of the ambient air network.

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.

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

Building	Facility	Analytes	Sampler type	Number of samplers
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[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	15
491	Laser isotope separation[c]	Gross α, β on particles	Filter	1
695	Decontamination and Waste Treatment Facility	Gross α, β on particles	Filter	1
695 Yard	TRU Mover	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

c Operations discontinued; however, the air effluent sampling system at this building continues to operate as part of the maintenance and surveillance shutdown plan for the Advanced Vapor Laser Isotope Separation (AVLIS) program.

Air Effluent Radiological Monitoring Results

In 2004, a total of 0.61 TBq (16 Ci) of tritium was released from the Tritium Facility (Building 331). Of this, approximately 0.45 TBq (12 Ci) were released as tritiated water vapor (HTO). The remaining tritium released, 0.16 TBq (4.0 Ci), was elemental tritium gas (HT). The median emissions from the facility were 1.9×10^{3} Bq/m^3 (5.1×10^{-8} Ci/m^3) for HTO, and 1.3×10^{2} Bq/m^3 (3.5×10^{-9} Ci/m^3) for HT. The highest single weekly stack emission from the facility was 9.6×10^{-2} TBq (2.6 Ci), of which 8.5×10^{-2} TBq (2.3 Ci) was HT. Emissions from Building 331 for 2004 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. Some sampling systems may exhibit as few as one to four values (out of 26 to 52 samples per year) greater than the MDC. Generally, these samples are only marginally above the MDC. In addition, due to the way some of the exhaust systems are configured, the monitoring systems sometimes sample air from the atmosphere in addition to HEPA-

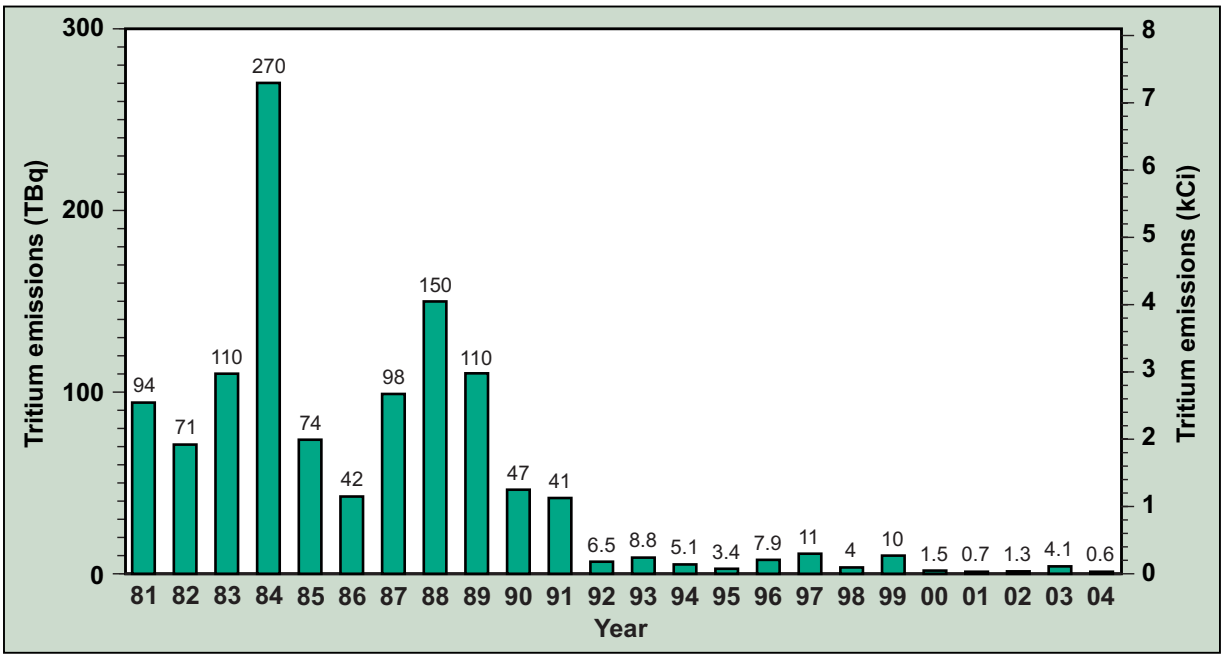

Figure 3-3. Tritium Facility combined HTO and HT emissions from 1981 through 2004

filtered air from facility operations, thereby collecting 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 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.

In 2004, a significant number of samples collected throughout the year from two release emission points at Building 251 (the unhardened area) yielded gross alpha results greater than the MDC. Gross alpha is used as the primary indicator of potential emissions for operations, such as those at Building 251 that involve the use of uranium and transuranic materials. The gross alpha and gross beta activity emissions for Building 251 were 1.9×10^{2} Bq/y (5.0×10^{-9} Ci/y) and 1.6×10^{3} Bq/y (4.3×10^{-8} Ci/y). Because of the number of samples with values above the MDC, gross alpha and gross beta measurements are being reported as actual emissions.

Table 3-2 summarizes total radiological emissions as determined from the continuous sampling of facility exhausts for 2004.

Table 3-2. Measured radiological air effluent emissions above the detection limit for Livermore site, 2004

Building (Facility)	HT (Bq)	HTO (Bq)	Gross alpha (Bq)	Gross beta (Bq)
331 (Tritium Facility)	1.6×10^{11}	4.5×10^{11}	—	—
251 (Heavy Element Facility)	—	—	1.9×10^{2}	1.6×10^{3}

Nonradiological Results

The Livermore site currently emits approximately 153 kg/day of regulated air pollutants as defined by the Clean Air Act, including nitrogen oxides, sulfur oxides, particulate matter [PM-10], and carbon monoxide (see **Table 3-3**). Carbon monoxide emissions appear to have increased in 2004 because a higher emission factor, required by the Synthetic Minor Operating Permit, was used in estimations for small boilers on site. The emission sources that release the greatest amount of regulated pollutants at the Livermore site are surface-coating, internal combustion engines, solvent wiping, and, natural gas fired boilers. **Table 3-3** lists estimated airborne releases for regulated pollutants from the Livermore site.

Table 3-3. Nonradioactive air emissions, Livermore site and Site 300, 2004

Pollutant	Estimated releases (kg/day)	
	Livermore site	Site 300
Organics/volatile organics	16.0	0.47
Nitrogen oxides	75.1	1.84
Carbon monoxide	54.7	0.40
Particulates (PM-10)	5.7	0.41
Sulfur oxides	1.5	0.53

LLNL air pollutant emissions are very low compared with daily releases of air pollutants for the entire Bay Area. For example, the total emissions of nitrogen oxides released in the Bay Area for 2004 were approximately 6.9×10^4 kg/day, compared with the estimated release from the Livermore site of 75.1 kg/day, which is 0.11% of total Bay Area emissions from stationary sources. The 2004 BAAQMD estimate for reactive organic emissions was 9.1×10^4 kg/day, while the estimated releases for 2004 from the Livemore site were 16.0 kg/day, or 0.02% of the total Bay Area emissions from stationary sources.

Certain operations at Site 300 require permits from SJVAPCD. The total estimated air pollutant emissions during 2004 from operations (permitted and exempt sources) at Site 300 are given in **Table 3-3**. The emission sources that release the greatest amounts

of criteria pollutants at Site 300 include internal combustion engines, boilers, a gasoline-dispensing facility, prescribed burns, paint spray booths, drying ovens, and soil vapor extraction equipment.

Impact of Air Effluent on the Environment

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 1.4×10^{-2} µSv/y (1.4×10^{-3} mrem/y) and the dose from Building 251 is 6.8×10^{-6} µSv/y (6.8×10^{-7} 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 beryllium 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 area sources, are monitored to fill 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. 2004). 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. 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 Pollutants.

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 at a background location near 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 monitor concentrations at the perimeters of the sites or at distant background locations. Throughout the year at selected locations, additional samplers are placed next to permanent samplers. Duplicate samples thus obtained provide quality control of the data. Trip blanks are also taken on the air particulate sampling routes to help identify any contaminate introduced during the sampling process.

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 2005).

Sample Collection

The air particulate networks use high-volume air sampling units, which collect airborne particulate weekly 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, draw air through sampling flasks containing silica gel that traps the air moisture. These flasks are changed every two weeks.

Sampling Locations

Based on historical meteorological data, all ambient air samplers have been positioned to detect any significant concentration of radioactive or beryllium effluents from LLNL operations with reasonable probability. Before startup of a new operation, the need for a new sampling location is assessed using air dispersion modeling.

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 near the city of Tracy. There are 7 air particulate samplers on the Livermore site, 9 in the Livermore Valley, and 8 at Site 300. There are 11 air tritium samplers at the Livermore site, 6 in the Livermore Valley, and 1 at Site 300. In December 2003, the air particulate location TFIR was removed and replaced (in March 2004) by a more suitable background location for Site 300. This station is called TCDF and is approximately 4.7 kilometers north of Site 300.

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]	Onsite	X	X	X	X	X
DWTF, POOL	Onsite					X
B331, B624	Diffuse/onsite					X
CRED[b]	SW-MEI[c]	X	X			X
ZON7, PATT, AMON	Downwind	X	X			X
CHUR, FCC[d], TANK	Upwind	X	X			
FIRE, HOSP[d]	Upwind	X	X			X
VET	Upwind					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[d], 801E	Onsite[b]	X	X	X		
COHO	Onsite[b]	X		X		X
TCDF[e]	Offsite[b]	X		X	X	

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

b On the Livermore site, samplers VIS and CRED represent the location of the site-wide maximally exposed individual (SW-MEI), and concentrations obtained from them are averaged for compliance with minor sources; at Site 300, the average of all locations is applied.

c SW-MEI for NESHAPs compliance based on air dispersion modeling.

d Low-volume sampler also operated at this location; particles are collected on millipore filters. These samplers are operated to provide background values for the air effluent monitoring program.

e Location TFIR was removed at the end of 2003 and replaced by TCDF, which began in March 2004.

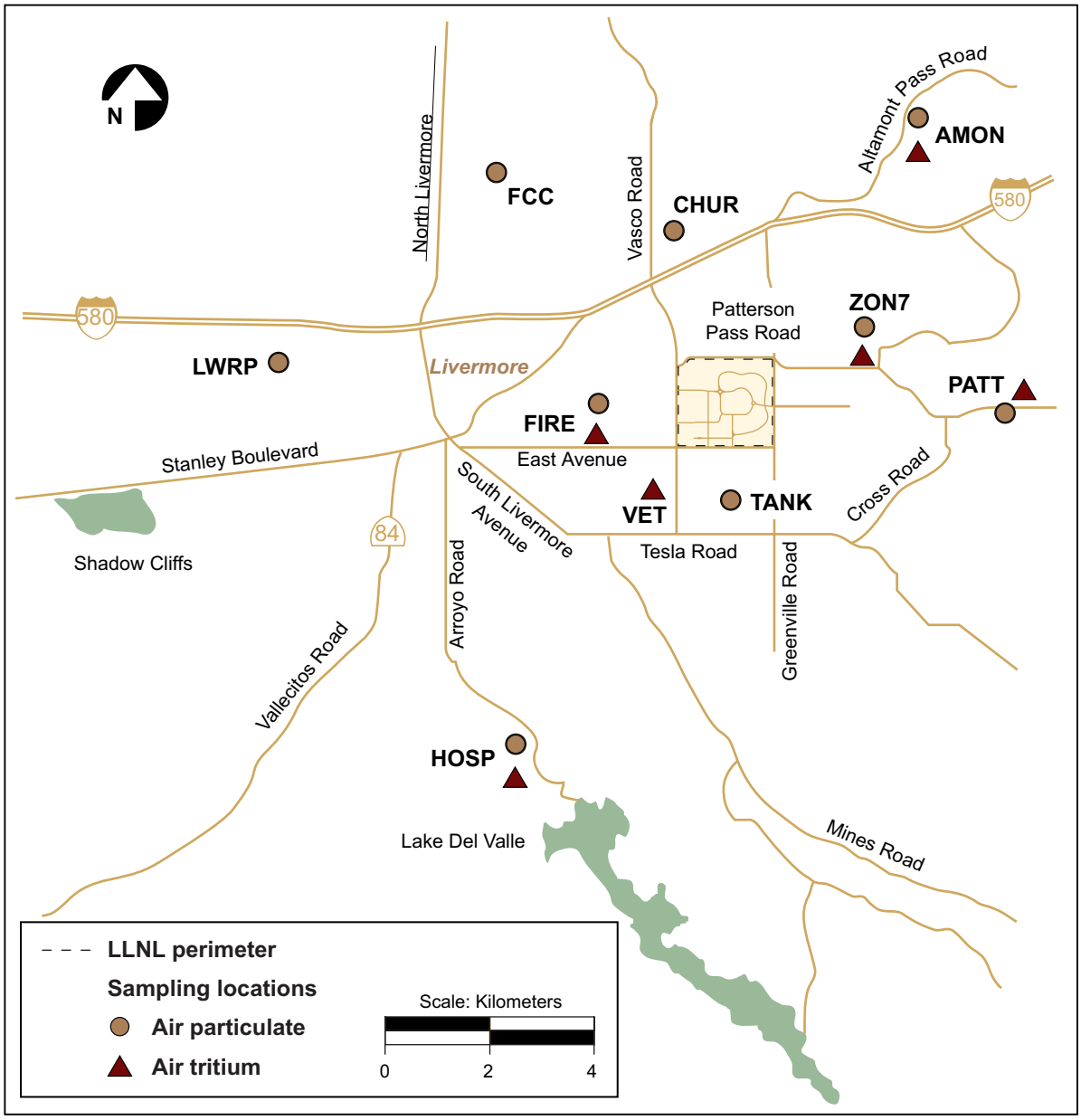

Figure 3-4. Air particulate and tritium sampling locations in the Livermore Valley, 2004

In general, air sampling locations are grouped in categories representing the following areas; perimeter, upwind, downwind, diffuse sources or areas of known contaminaton, and special interest locations. The mean 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 on-site locations serves as the SW-MEI for NESHAPs minor source compliance.

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 the new location (TCDF) north of Site 300.

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 counting. Procedures for analysis are summarized in the *Environmental Monitoring Plan* (Woods 2005). Beryllium metal concentration is determined by inductively coupled plasma-mass spectrometry. 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 of the samples.

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, which display monthly and biweekly data, are measured concentrations and their associated $\pm 2\sigma$ counting errors.

Particle size distribution of 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 total particles collected.

Gross Alpha and Gross Beta Concentrations

The primary sources of alpha and beta activities are naturally occurring radioisotopes. **Figure 3-5** shows the three-year history of median monthly gross alpha and gross beta activities for the Livermore site perimeter, Livermore Valley, and Site 300 sampling locations. These data are slightly lower than last year but follow a pattern similar to previous years with a seasonal increase in the fall and early winter months. As soils dry out during the summer months, the resuspended particulate can build up and increase until the winter rains begin. In many cases there is an inverse relationship between rainfall and particulate activity indicating that the increases in activity may be from particulate mass from resusupended soils rather than LLNL airborne sources. Routine isotopic gamma results of site composite samples indicate that higher activities are the result of naturally occurring isotopes (uranium, thorium, potassium, and lead) which are also routinely found in local soils.

In 2004, the typical gross alpha activity (annual median value) for the Livermore site perimeter was 21 µBq/m^3 (0.57 fCi/m^3); for the upwind and downwind Livermore Valley stations, the value was 20 µBq/m^3 (0.54 fCi/m^3); and for Site 300, the value was 26 µBq/m^3 (0.70 fCi/m^3). The annual gross beta median for all upwind and downwind locations was 260 µBq/m^3 (7.0 fCi/m^3); for the Livermore site perimeter it was 270 µBq/m^3 (7.3 fCi/m^3; and for Site 300 it was 310 µBq/m^3 (8.5 fCi/m^3). Location CHUR (an upwind location) recorded high gross alpha and beta activity during November; samples were recounted but remained higher than normal. A gamma scan is being performed to determine what isotope is causing this spike in activity. See the section "Gamma-Emitting Radionuclides" in this chapter for more information.

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 site air samples with a increase in the fall and early winter months then a decrease during the winter as rains reduce the resuspension effect. The highest weekly gross alpha sample measured at Site 300 was 240 µBq/m^3 (6.5 fCi/m^3) at WOBS. This sampler is near locations where open-air shots have occurred (Building 851 bunker and the Contained Firing Facility [Building 801]). In addition, there were two shots during December that most likely contributed to the elevated gross alpha values. The overall annual median gross alpha value at Site 300 was 26 µBq/m^3 (0.70 fCi/m^3).

The highest Site 300 onsite weekly gross beta value was 1432 µBq/m^3 (39 fCi/m^3) recorded at WOBS which also coincides with a shot at Site 300. The overall annual median beta value for Site 300 was 310 µBq/m^3 (8.4 fCi/$m^{3)}$.

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 also reveals emissions from global fallout sources such as aboveground tests and the Chernobyl accident (Holland et al. 1987). Composite

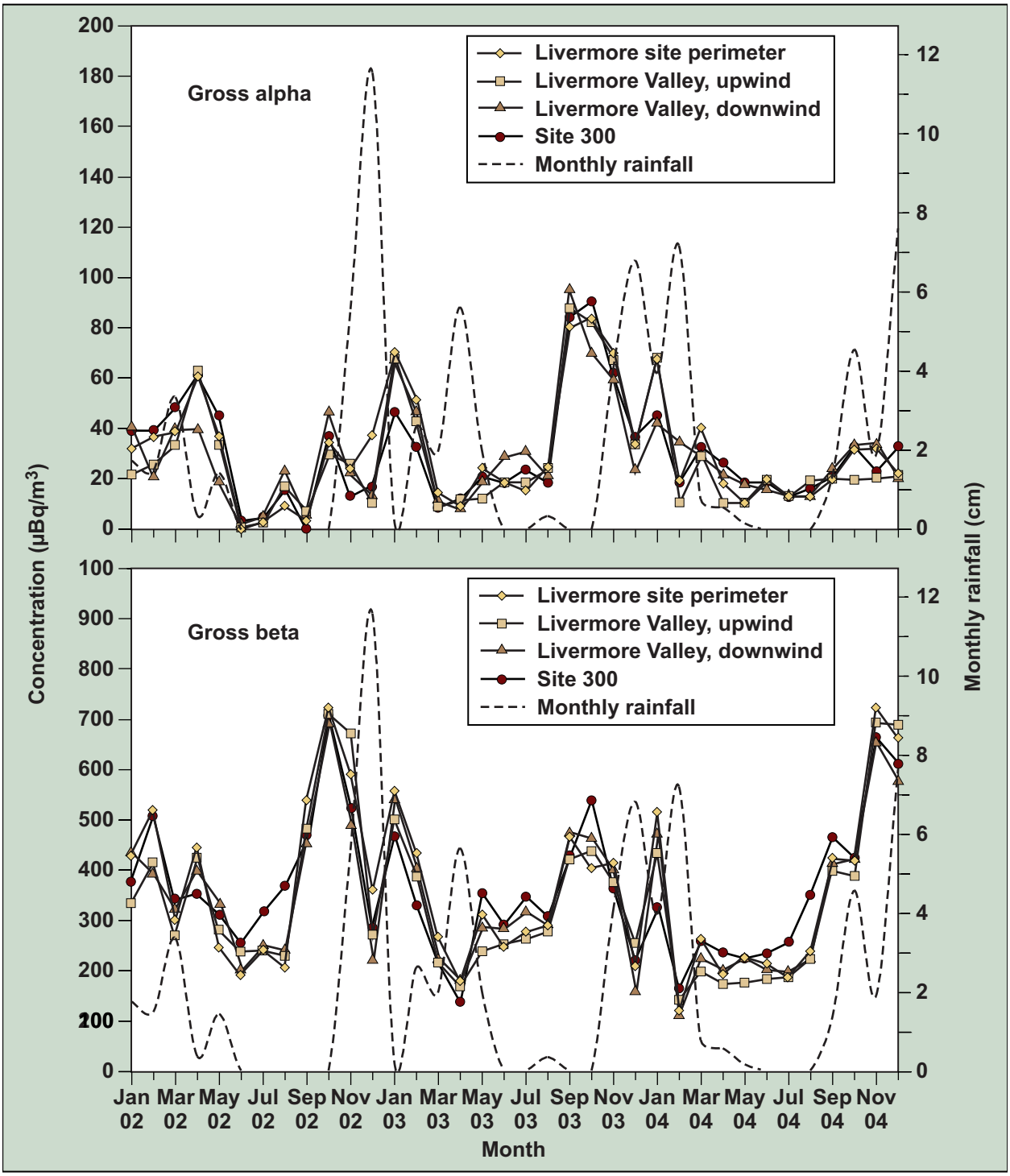

Figure 3-5. Three-year history of monthly median gross alpha and gross beta activities for all particulate samples grouped by area, along with corresponding monthly rainfall totals, 2002-2004

samples for the Livermore site and Site 300 are analyzed for an environmental suite of gamma-emitting radionuclide concentrations in air. Site composite samples are scanned for 47 isotopes with over 350 gamma rays. These include fission products, activation products, actinides, and naturally occurring products. The results for gamma composites for 2004 were within known background levels (see file "Ch3 Ambient Air" on report CD for analytical results). Occasionally weekly samples that are screened for gross alpha and beta are also gamma scanned to determine what isotope may be the cause of higher than usual activity. Such was the case for the sample mentioned above (CHUR). In this case the activity was determined to be caused by an increase in a naturally occurring isotope and not by LLNL operations.

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 concentrations at both of these sites have been decreasing as fallout diminishes and on-site surface areas of potential resuspension have been covered with pavement or buildings. LLNL analyzes all Livermore area samples individually, while a composite is created from all on-site Site 300 samples.

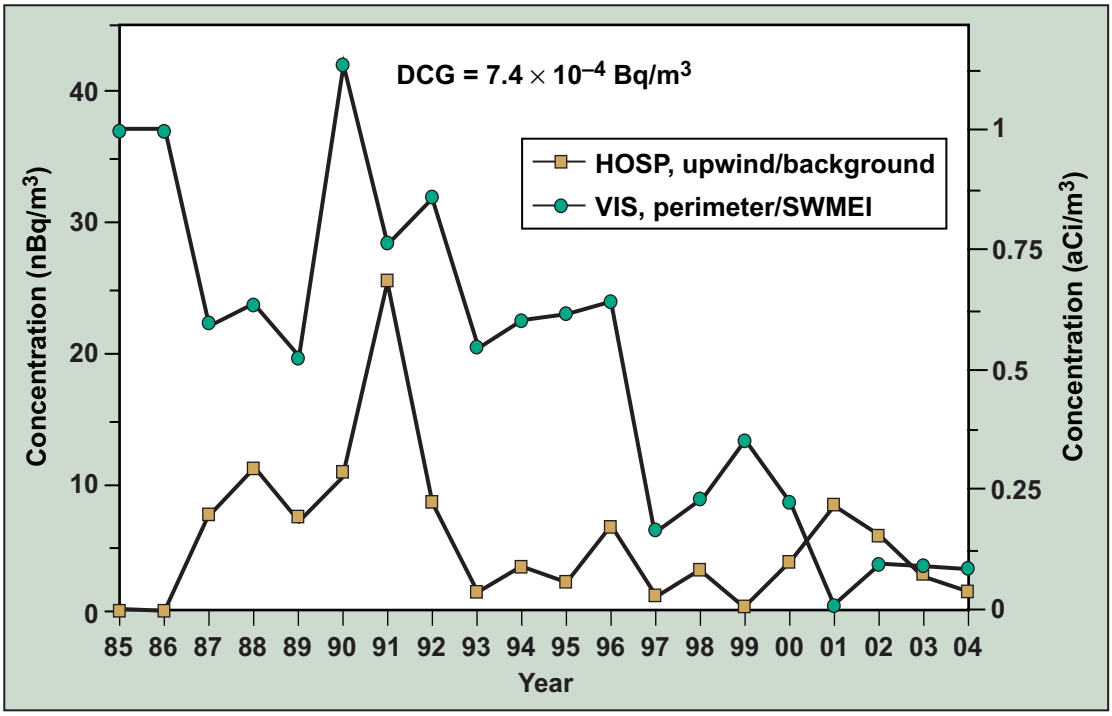

Figure 3-6. Calculated annual median concentrations of plutonium-239+240 for HOSP and VIS for the last 20 years

Plutonium-239+240 was detected in 13 of the 234 samples tested from Livermore area air samples. Six of those positive samples came from on-site samplers. These detections all came between June and October, when resuspension is potentially greatest. The highest recorded onsite plutonium-239+240 detection was at the SW-MEI (CRED) of 21 nBq/m^3 (0.57 aCi/m^3) (0.003% of the DCG), while the highest off-site plutonium value was recorded as 14 nBq/m^3 (0.38 aCi/m^3) at the TANK location in August. Plutonium was detected in only 1 of the 12 composite samples collected from Site 300 and this value was very close to the minimum detection limit. This value of 4.5 nBq/m^3 (0.12 aCi/m^3) (0.0006% of the DCG) was recorded in September and was lower than all but one (MET) of the maximum values for samples collected in Livermore. 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 monthly composites had a uranium-235 median concentration of 0.14 pg/m^3 and a uranium-238 median concentration of 22 pg/m^3. This has a median ratio of 0.007, which is considered natural uranium and typical of what has been recorded in the past. Only one sample, which was collected on the Livermore site during December, showed anything other that natural activity; in this case, a ratio of 0.002 was recorded which indicated the presence of depleted uranium. This activity is highly unlikely at the Livermore site and was suspicious because two very high uranium samples were collected from Site 300 in December. An investigation of the data was performed, and it was determined that handling and analytical sample processing most likely resulted in cross contamination between the Livermore site and Site 300 composites. The Livermore site composite was normal in January 2005. The standard operating procedures have been amended to eliminate the possibility for this type of cross contamination from occurring again.

The annual median uranium-235 concentration for all Site 300 locations was 0.17 pg/m^3 (or less than 0.00003% of the DCG) and the uranium-238 median concentration was 24 pg/m^3 (or less than 0.0008% of the DCG). As with the Livermore site, the Site 300 isotopic ratio for the annual median was 0.007, which is considered natural uranium. As with the December Livermore site composite, 7 of 9 samples collected from Site 300 during December recorded a uranium-235/uranium-238 ratio with a depleted uranium signature. These depleted uranium signatures are likely since there were several outdoor test shots with depleted uranium over a two month period. The highest uranium-238 value was 8660 pg/m^3 in December at WOBS (the second highest was 174 pg/m^3 at NPS, also in December). 8660 pg/m^3 is 3% of the DCG and is significantly higher than any other sample collected in recent years.

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 the B331 and B624 samplers on the Livermore site near stored containers of tritium waste or tritium-contaminated equipment (the Building 331 waste accumulation area and the Building 612 Yard) that outgas tritium as HTO. The annual median concentration for 2004 for the B331 diffuse-source sampler was more than a factor of five times lower than in 2003 reflecting the large decrease in tritium operations at the Tritium Facility in 2004. The median concentration at the B624 sampler in the Building 612 Yard was only slightly lower than in 2003. Sampling at the Building 514 Tank Farm, continuous between August 1991 and December 2003, was discontinued because the facility underwent RCRA closure.

Table 3-5. Tritium in air samples (mBq/m^3), 2004

Sampling locations	Detection frequency	Mean	Median	IQR	Maximum	Median Percent of DCG[a]
Diffuse on-site sources	50 of 50	1420	435	1990	7470	0.0117
Livermore site	174 of 231	42.2	34.6	39.5	718	0.000935
Livermore Valley	48 of 155	6.35	4.59	21.1	57.7	0.000124
Site 300	3 of 26	–2.27	–0.325	21.5	31.5	_[b]

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

b Median percent DCG not calculated because the median is negative.

Samplers near the perimeter of the Livermore site exhibit the next highest air tritium concentrations. Of these locations, POOL exhibited the highest median concentration at just 0.0021% of the DCG. Concentrations at POOL were on average much lower than in 2003. Median concentrations for 2004 for on-site locations were on average about half of those for 2003. Much less variability was seen in the concentrations for 2004 compared with 2003. Because releases from the Tritium Facility were markedly reduced in 2004 compared with 2003, the high peak air tritium concentrations seen in 2003 are not seen in 2004—the mean of all maximum concentrations for all on-site locations for 2003 was 718 mBq/m^3 (19.4 pCi/m^3); for 2004 it was 161 mBq/m^3 (4.35 pCi/m^3).

For 2004, two of the locations near the perimeter (MESQ and MET) had median concentrations below the detection limit (about 25 mBq/m^3), while all of the median concentrations in the Livermore Valley and at Site 300 (**Table 3-5**; see also file "Ch3 Ambient Air" on report CD for biweekly data) were below the detection limit. Given the low tritium concentrations observed at the Livermore site perimeter, all samples from locations distant from the Livermore site are expected to exhibit tritium background concentrations that are below the detection limit. Similarly, because no operations at

LLNL release tritium to the environment at Site 300, concentrations at COHO are expected to be below the detection limit. Detections occurring at these sampling locations are artifacts of scintillation counting with a high counter background.

Beryllium Metal Concentrations

LLNL measures the monthly concentrations of airborne beryllium for the Livermore site, Site 300, and the off-site sampler located north of Site 300. (See file "Ch3 Ambient Air" on report CD for data.) The highest value at the Livermore site was 21 pg/m^3 which was recorded at location SALV in September. This value is only 0.21% of the BAAQMD ambient concentration limit for beryllium (10,000 pg/m^3). These data are 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 2004. 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.

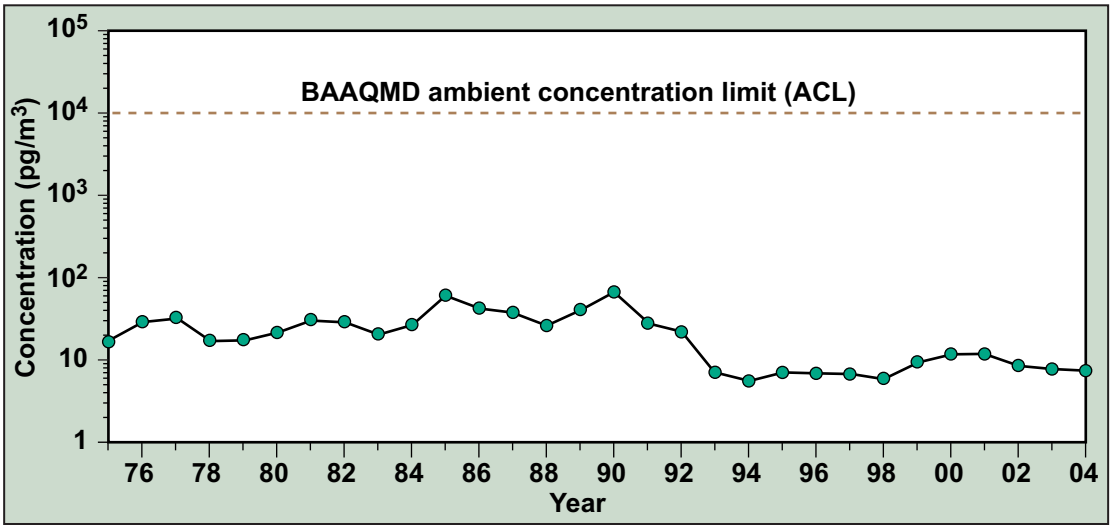

Figure 3-7. Median concentration of beryllium in air particulate samples taken at the Livermore site perimeter, 1975–2004

There is no regulatory requirement to monitor beryllium in 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 7.2 pg/m^3. The highest value for the Site 300 area samples occurred in the September sample at TCDF. This sample recorded a value of 23 pg/m^3, which is 0.12% of the ambient concentration limit.

Environmental Impact of Ambient Air

LLNL operations involving radioactive materials had little impact on radionuclide concentrations in ambient air during 2004. 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.

The diffuse tritium sources at Building 331 and the Building 612 Yard had a small, localized effect with minimal impact on the public. Any potential dose received by a member of the public from the diffuse sources is accounted for when doses are calculated based on tritium concentrations at the Livermore site perimeter. The mean tritium concentration for all Livermore site perimeter air tritium sampling locations in 2004 was about one-third lower than in 2003. Both mean and median concentrations of tritium in the Livermore Valley or at Site 300 were all well below detection limits. For a location at which the mean concentration is at or below the detection limit, inhalation dose from tritium is assumed to be less than 5 nSv/y (i.e., the dose from the detection limit of about 25 mBq/m^3).

There are two Livermore site locations (CRED and VIS) with public access, at least during working hours. If it were assumed that a member of the public inhaled air continuously for a year at the maximum biweekly concentration at CRED (120 mBq/m^3) or VIS (72.2 mBq/m^3), the resulting doses would still be tiny (25 nSv/y and 15 nSv/y, respectively). Put another way, the maximum concentration at CRED is just 0.2% of concentration limits for minor sources set by the U.S. EPA in Table 2, Appendix E to 40 CFR 61 (Harrach 2005).

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 median for the Livermore site and Site 300 (excluding the off-site location, TCDF) are both 7.3 pg/m^3. These data are lower than estimated for natural background, well below standards, and do not indicate the presence of a threat to the environment or public health.