Air Monitoring Programs

CHAPTER 4

Kent Wilson
Steven Cerruti
S. Ring Peterson

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), 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 the environs by LLNL operations. Ambient air monitoring also serves to verify the air concentrations predicted by air dispersion modeling and to determine

compliance with NESHAPs regulations. (See *LLNL NESHAPs 2005 Annual Report* [Larson et al. 2006].)

Air Effluent Monitoring

For research purposes, LLNL uses a variety of radioisotopes including uranium, transuranic radionuclides, biomedical tracers, tritium, and mixed-fission products. The principal 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 and to confirm the operation of facility emission control systems. Air effluent and ambient air monitoring measurements can be compared to confirm the expected relationship between them and to help resolve unexpected differences. Air effluent monitoring 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. When applicable, LLNL obtains permits for the operations from local air districts (i.e., BAAQMD or SJVAPCD). Current permits do not require monitoring of air effluent, but do require monitoring of equipment usage, material usage, and record keeping during operations. 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 2005, LLNL operated 71 sampling systems that measured releases of radioactivity from air exhausts at 6 facilities at the Livermore site and 1 sampling system at Site 300. These systems are listed in **Table 4-1** along with the analytes of interest, the type of sampler, and the number of samplers (see **Figures 4-1** and **4-2** for Livermore site and Site 300 air monitoring locations). LLNL periodically reassesses the need for continuous monitoring and assesses new operations or changes in operations.

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

Facility	Analytes	Sampler type	Number of samplers
Chemistry and Materials Science	Gross α, β on particles	Filter	1
Heavy Element	Gross α, β on particles Gross α, β on particles	Stack CAM[a,b] Filter	2 28
Tritium	Tritium Gaseous tritium and tritiated water vapor	Stack ionization chamber[a] Molecular sieves	4 4
Plutonium	Gross α, β on particles Gross α, β on particles	Stack CAM[a,b] Filter	12 15
Laser isotope separation[c]	Gross α, β on particles	Filter	1
Decontamination and Waste Treatment Facility TRU Mover	Gross α, β on particles Gaseous tritium and tritiated water vapor Gross α, β on particles	Filter Glycol bubbler Filter	1 1 1
Contained Firing Facility	Gross α, β on particles	Filter	1

a Alarmed systems (real-time)
b CAM = Eberline continuous air monitors (real-time)
c Isotopic separation operations were discontinued; area now used for storage of contaminated parts

Sampling for radioactive particles was conducted in all facilities except for the Tritium Facility, where only tritium is measured. Both radioactive particulates and tritium are sampled at the Decontamination and Waste Treatment Facility. 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 and glycol bubblers.

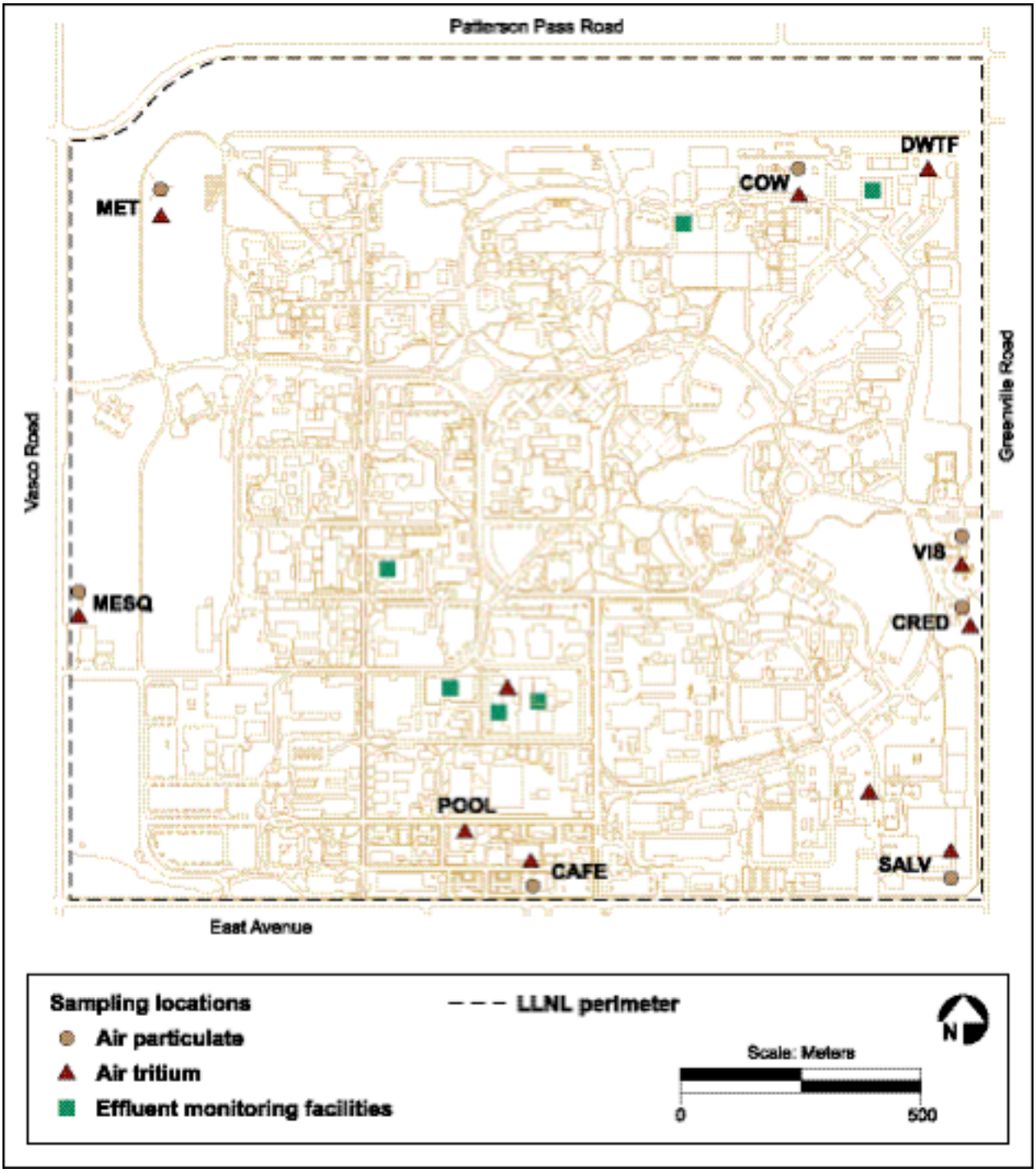


Figure 4-1. Livermore site air monitoring locations, 2005

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. Air effluent samples were obtained in accordance with written standardized procedures summarized in the *Environmental Monitoring Plan* (Woods 2005).

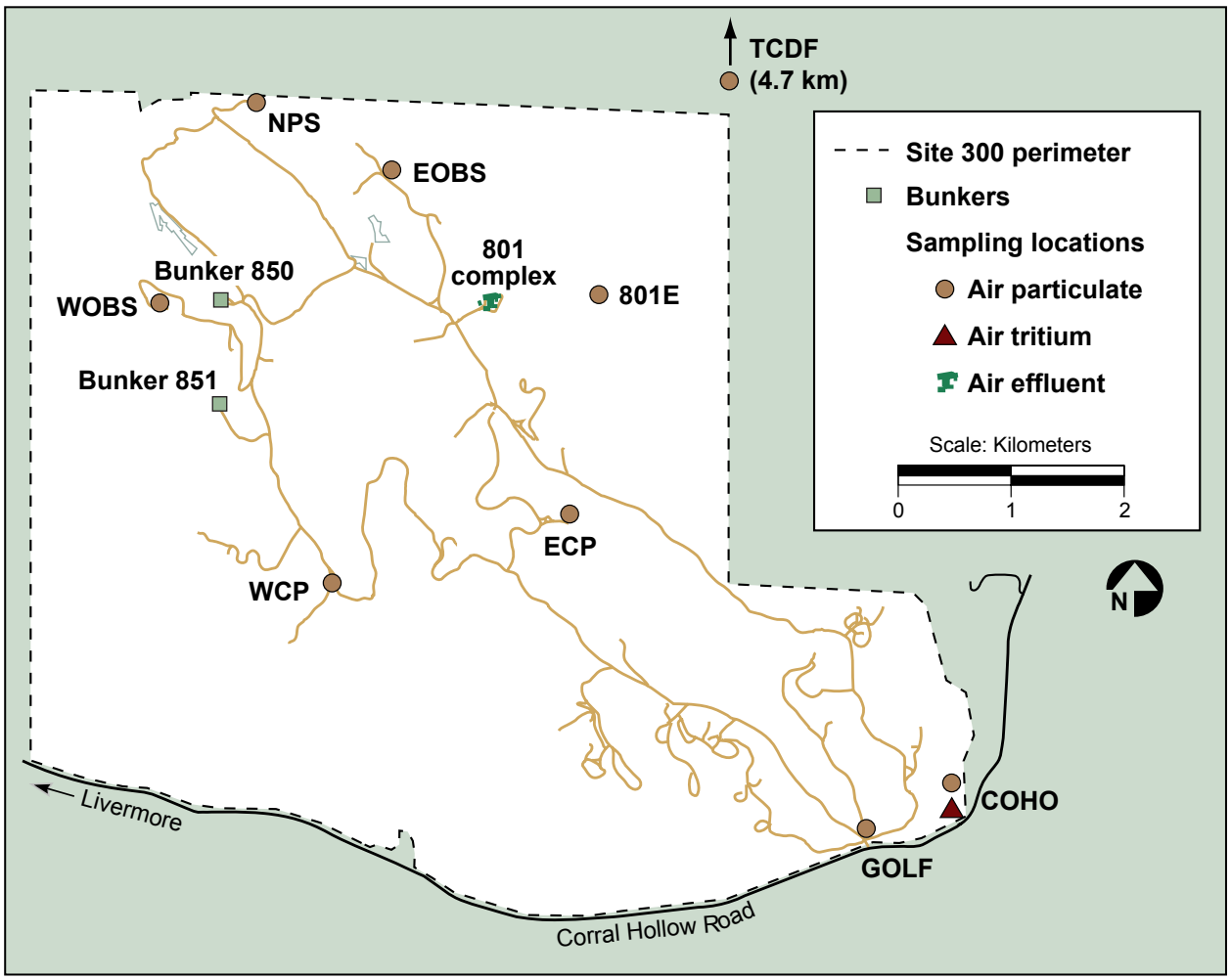


Figure 4-2. Site 300 air monitoring locations, 2005

To establish the background levels of gross alpha and beta activity that are used to determine if a particulate release has occurred from monitored stacks, LLNL operates three low-volume radiological air particulate samplers at locations HOSP and FCC in the Livermore Valley (see **Figure 4-3**) and NPS at Site 300 (see **Figure 4-2**). 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.

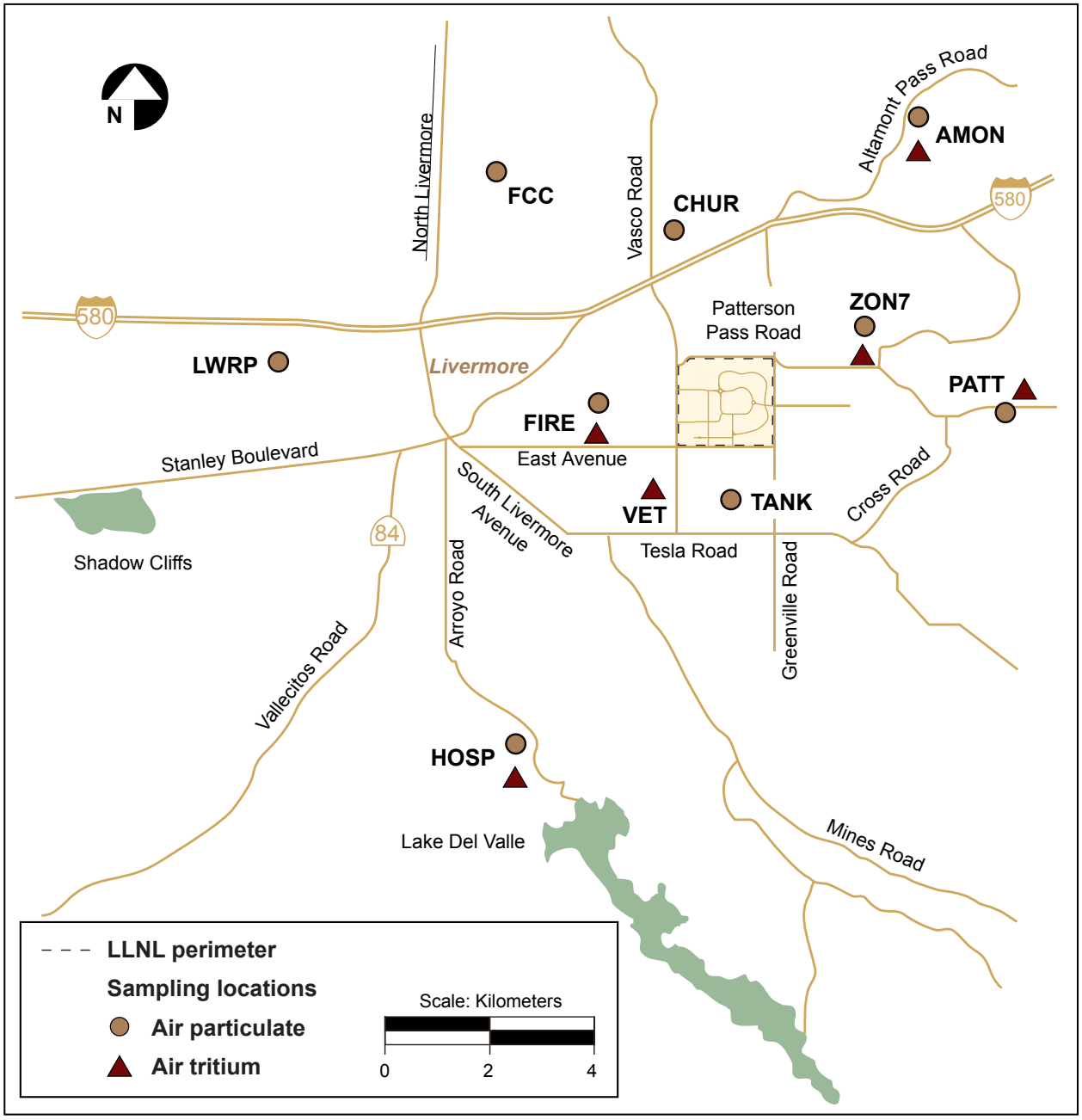
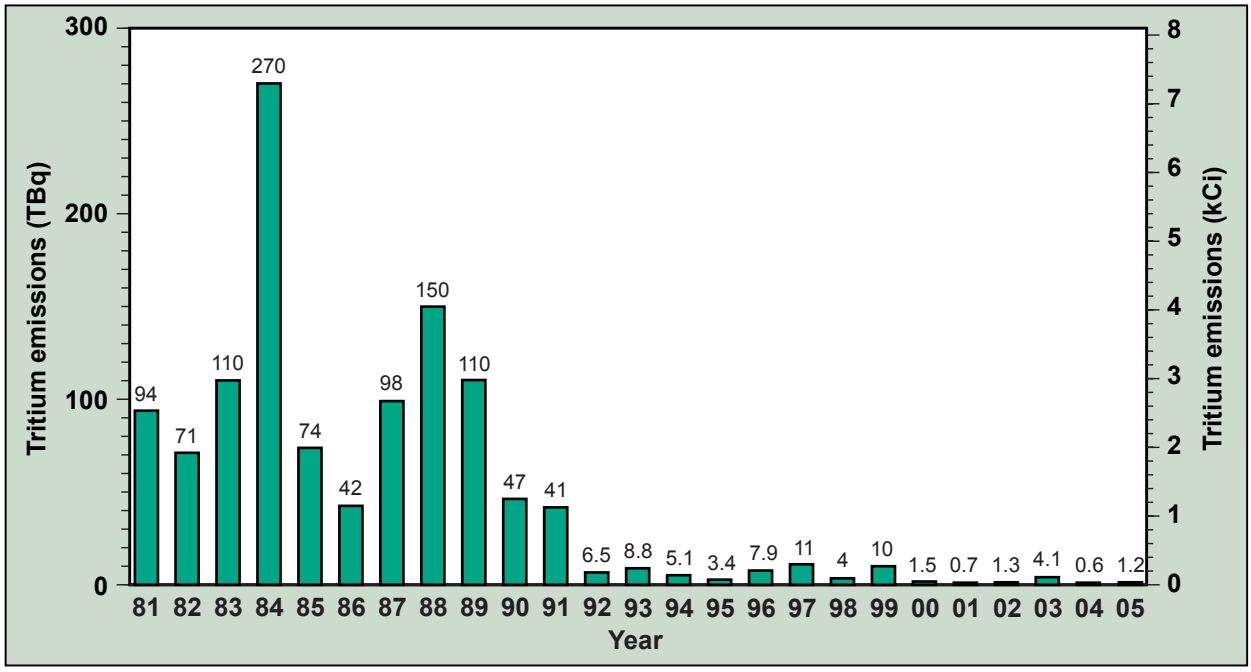


Figure 4-3. Air particulate and tritium sampling locations in the Livermore Valley, 2005

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 "Ch4 Air Effluent" included on the report CD.

Air Effluent Radiological Monitoring Results

In 2005, a total of 1.2 TBq (32 Ci) of tritium was released from the Tritium Facility (Building 331). Of this, approximately 1.1 TBq (30 Ci) were released as tritiated water vapor (HTO). The remaining tritium released, 0.074 TBq (2.0 Ci), was tritiated hydrogen gas (HT). The median emissions from the facility were 2000 Bq/m^3 (5.4×10^{-8} Ci/m^3) for HTO, and 110 Bq/m^3 (3.1×10^{-9} Ci/m^3) for HT. The highest single weekly stack emission from the facility was 0.18 TBq (4.9 Ci), of which more than 99% was HTO. Emissions from Building 331 for 2005 continued to remain considerably lower than those during the 1980s. **Figure 4-4** illustrates the combined HTO and HT emissions from the facility since 1981.



Note: Emissions from accidental releases in 1984, 1985, 1990, and 1991 contribute to total tritium released.

Figure 4-4. Tritium Facility combined HTO and HT emissions from 1981 through 2005

Monitoring for tritium emissions at the Decontamination and Waste Treatment Facility (Building 695) began in February 2005. A total of 0.085 TBq (2.3 Ci) of measured tritium was released with 0.081 TBq (2.2 Ci) as HTO and 3.3×10^{-3} TBq (0.088 Ci) as HT. Because monitoring did not begin at the first of the year, an additional emission of 0.017 TBq (0.47 Ci) was estimated by taking an average of measured emissions and applying it to the time period when monitoring was not in place. The total emission for 2005 (measured and estimated emissions combined) was 0.10 TBq (2.7Ci), of which 4.0×10^{-3} TBq (0.11 Ci) was HT. The tritium emissions from Building 695 are far below the level of regulatory concern, and monitoring is in place as part of a best management practice.

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-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 2005, a significant number of samples (7) collected throughout the year from one release emission point at Building 801A 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 801A, that involve the use of uranium and/or transuranic materials. The gross alpha and gross beta activity emissions for Building 801A were 1.6×10^{4} Bq/y (4.2×10^{-7} Ci/y) and 5.9×10^{4} Bq/y (1.6×10^{-6} 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 4-2** provides a summary of all 2005 radiological emissions as determined from continuous sampling of facility exhausts.

Table 4-2. Measured radiological air effluent emissions above the detection limit for Livermore site and Site 300, 2005

Building (Facility)	HT (Bq)	HTO (Bq)	Gross alpha (Bq)	Gross beta (Bq)
331 (Tritium Facility)	5.8×10^{10}	1.1×10^{12}	—	—
695 (Decontamination and Waste Treatment Facility)	4.1×10^{9}	9.6×10^{10}	—	—
801A (Contained Firing Facility)	—	—	1.6×10^{4}	5.9×10^{4}

Nonradiological Results

The Livermore site currently emits approximately 151 kg/day of regulated air pollutants as defined by the Clean Air Act, including nitrogen oxides, sulfur oxides, particulate matter (PM-10), carbon monoxide, and reactive organic gases/precursor organic compounds (ROGs/POCs) (see **Table 4-3**). The stationary emission sources that release the greatest amount of regulated pollutants at the Livermore site are natural gas fired boilers, internal combustion engines (such as diesel generators), solvent cleaning, and surface coating operations (such as painting). The ROGs/POCs emissions appear higher in 2005 than in 2004 because, in 2005, the ROGs/POCs emissions from permit-exempt adhesive and architectural paint sources were added to the "Estimated releases" for the Livermore site for consistency with the source tracking requirements of the site-wide Synthetic Minor Operating Permit (SMOP). Such permit-exempt sources represent an additional 9.5 kg/day of total ROGs/POCs in 2005, while the permit sources contributed 15.4 kg/day (as compared to 16.0 kg/day in 2004). Overall, the ROGs/POCs, nitrogen oxides, carbon monoxide, and PM-10 emissions decreased in 2005, and the sulfur oxide emissions increased slightly.

Table 4-3. Nonradioactive air emissions, Livermore site and Site 300, 2005

Pollutant	Estimated releases (kg/day)	
	Livermore site	Site 300
ROGs/POCs	24.9	0.41
Nitrogen oxides	68.6	0.52
Carbon monoxide	49.9	0.11
Particulates (PM-10)	5.6	0.28
Sulfur oxides	1.7	0.03

Note: In previous Environmental Reports, the ROGs/POCs pollutant category was titled "Organics/volatile organics."

LLNL air pollutant emissions are very low compared with daily releases of air pollutants from all sources in the entire Bay Area. For example, the total emissions of nitrogen oxides released in the Bay Area for 2005 were approximately 4.89×10^5 kg/day, compared with the estimated release from the Livermore site of 68.6 kg/day, which is 0.014% of total Bay Area source emissions for nitrogen oxides. The 2005 BAAQMD estimate for ROGs/POCs emissions was 3.63×10^5 kg/day, while the estimated releases for 2005 from the Livermore site were 24.9 kg/day, or 0.007% of the total Bay Area source emissions for ROGs/POCs.

The total estimated air pollutant emissions during 2005 from operations (permitted and exempt stationary sources) at Site 300 are presented in **Table 4-3**. The stationary emission sources that release the greatest amounts of regulated air pollutants at Site 300 include internal combustion engines (such as diesel generators), a gasoline-dispensing facility, paint spray booths, drying ovens, and soil vapor extraction equipment. Overall, the emissions for all pollutant categories at Site 300 decreased in 2005.

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) was 2.9×10^{-2} µSv/y (2.9×10^{-3} mrem/y) and the dose from Building 695 (modeling HT emissions as HTO) was 8.7×10^{-3} µSv/y (8.7×10^{-4} mrem/y). The dose from Building 801A was 1.1×10^{-4} µSv/y (1.1×10^{-5} mrem/y). Thus, the estimated radiological dose caused by measured air emissions from LLNL operations is minimal. See Chapter 7 for a discussion of doses.

Estimated nonradioactive air emissions are small compared with local air district emission criteria for the surrounding areas, and as such, have little impact on the environment or public health.

Ambient Air Monitoring

LLNL monitors ambient air to determine if radionuclides or beryllium are being released by Laboratory operations, what the concentrations are, and what the trends are in the environs. Beryllium is the only nonradiological emission from LLNL that is monitored in air. Normally for nonradiological emissions, LLNL obtains permits from local air districts (i.e., BAAQMD or SJVAPCD) that require monitoring of equipment usage, material usage, and record keeping during operations. The BAAQMD has exempted LLNL from the permitting process because LLNL can demonstrate that monthly average

beryllium concentrations in air are well below regulatory limits of 10,000 pg/m^3 at perimeter locations.

In 2003, the EPA approved use of 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 referred to in this chapter present the DCG and the percent of the DCG for the given isotope.

Sampling Locations

Sampling locations for each monitoring network are listed in **Table 4-4** and shown in **Figures 4-1**, **4-2** and **4-3**. Monitoring networks are established for air surveillance of radioactive particulates, tritiated water vapor, and beryllium metal. There are 7 air particulate samplers on the Livermore site, 9 in the Livermore Valley, 8 at Site 300, and 1 just west of the outskirts of Tracy There are 11 air tritium samplers at the Livermore site, 6 in the Livermore Valley, and 1 at Site 300. Beryllium is monitored at 6 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 3 locations onsite and at the location north of Site 300. All monitoring networks use continuously operating samplers.

Air sampling locations are grouped in the following categories: site perimeter, upwind, downwind, diffuse sources or areas of known contamination on site, and special interest locations. At the Livermore site, the mean air monitoring results for values greater than zero at locations CRED and VIS are used to calculate dose from minor sources to the SW-MEI for NESHAPs compliance; at Site 300, because resuspension of soil is the minor source of greatest interest, the mean concentrations of all on-site air samplers are used to calculate dose to the SW-MEI (see Chapter 7). Based on dispersion modeling using site-specific meteorological data, the ambient air samplers, particularly those on the site perimeters, have been placed to monitor locations where elevated air concentrations due to LLNL operations are expected. Before startup of a new operation, the need for a new sampling location is assessed.

Table 4-4. Sampling locations with 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	Historic 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	Offsite	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.

Sample Collection and Analysis

The air particulate networks use high-volume air sampling units, which collect airborne particulate on Whatman 41 cellulose filters. Air flows through the filters at a continuous rate of 0.42 m^3/min, and samples are collected weekly.

Tritium samplers, operating at a flow rate of 500 cm^3/min, draw air through sampling flasks containing silica gel that absorbs the air moisture. These flasks are changed every two weeks.

Throughout the year at varied 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 contaminant introduced during the sampling process. Ambient air samples were obtained in accordance with written standardized procedures summarized in the *Environmental Monitoring Plan* (Woods 2005).

An LLNL state-certified analytical laboratory performed all sample analyses. 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. Details about the analyses and the associated quality control are summarized in the *Environmental Monitoring Plan* (Woods 2005). Beryllium metal concentration is determined by inductively coupled plasma-mass spectrometry. See **Table 4-4** for the frequency of analysis at each location.

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

Emissions from uranium use at the Livermore site are 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, all sampling 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 indicators; nuclides known to be released from a facility, such as plutonium, uranium, and tritium at LLNL, must be analyzed for specifically. 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 are shown in the tables located in the file "Ch4 Ambient Air" included on the report CD.

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 4-5** 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 for gross alpha 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 resuspended 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 2005, the gross alpha activity (annual median value) for the Livermore site perimeter was 20 $\mu Bq/m^3$ (0.54 fCi/m^3); for the upwind and downwind Livermore Valley stations, the value was 16 $\mu Bq/m^3$ (0.43 fCi/m^3); and for Site 300, the value was 16 $\mu Bq/m^3$ (0.43 fCi/m^3). The annual gross beta median for all upwind and downwind locations was 271 $\mu Bq/m^3$ (7.3 fCi/m^3); for the Livermore site perimeter it was 287 $\mu Bq/m^3$ (7.7 fCi/m^3); and for Site 300 it was 323 $\mu Bq/m^3$ (8.7 fCi/m^3). These values are all typical annual average values. All ambient air analytical results are summarized in the file "Ch4 Ambient Air" included on the report CD.

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 resuspended soils. The pattern of activity as seen in **Figure 4-5** is very similar to the Livermore site.

On July 19, 2005, a grass fire burned more than 6200 acres including approximately 2100 acres at Site 300 on the west side of the site. During the fire, EPD/TAMM air sampling units were collecting particulate at eight locations on site. The filters were collected as soon as access to the sampling units was allowed. After being held for 4 days to allow for radon decay, the filters were screened for gross alpha and gross beta (GAB) activities. Concentrations of GAB were elevated compared with the weekly sampling data from the weeks leading up to the fire. The samples were also analyzed by

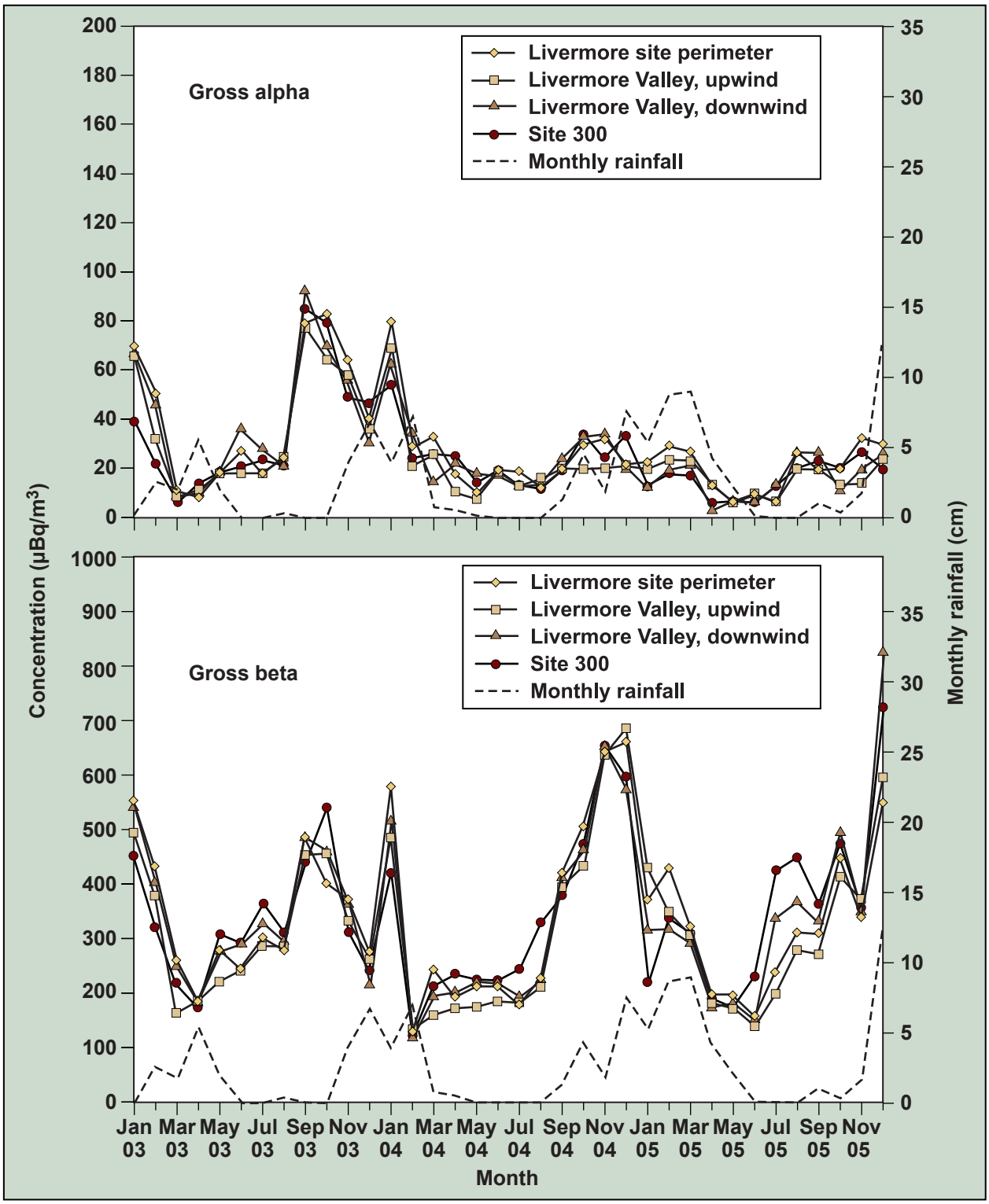


Figure 4-5. Three-year history of monthly median gross alpha and gross beta activities for all particulate samples grouped by area, along with corresponding monthly rainfall totals, 2003–2005

mass spectrometry for uranium isotopes; concentrations were slightly elevated over the monthly composite samples. Similar elevated concentrations of GAB and uranium are observed after controlled burns at Site 300.

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 can 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. Site composite samples are scanned for 47 isotopes, which contain over 350 different gamma ray energies. These include fission products, activation products, actinides, and naturally occurring products. Of these isotopes, beryllium-7 (cosmogenic), 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.

Plutonium Concentrations

Historical environmental plutonium-239+240 activity for the past 20 years is shown in **Figure 4-6**. Locations HOSP and VIS represent typical upwind and onsite sampling locations, respectively. 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.

Plutonium-239+240 was detected in 14 of the 204 samples tested from Livermore area air samples. Six of those positive samples came from on-site samplers. The highest recorded on-site plutonium-239+240 detection of 33 nBq/m^3 (0.89 aCi/m^3) was at the COW location and was 0.004% of the DCG, while the highest off-site plutonium value of 88 nBq/m^3 (2.4 aCi/m^3) was recorded at the LWRP location and was 0.012% of the DCG. Plutonium was detected in 3 of the 12 composite samples collected from Site 300 with the highest detection of 25 nBq/m^3 (0.68 aCi/m^3), which was 0.003% of the DCG, occurring in July.

Uranium Concentrations

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

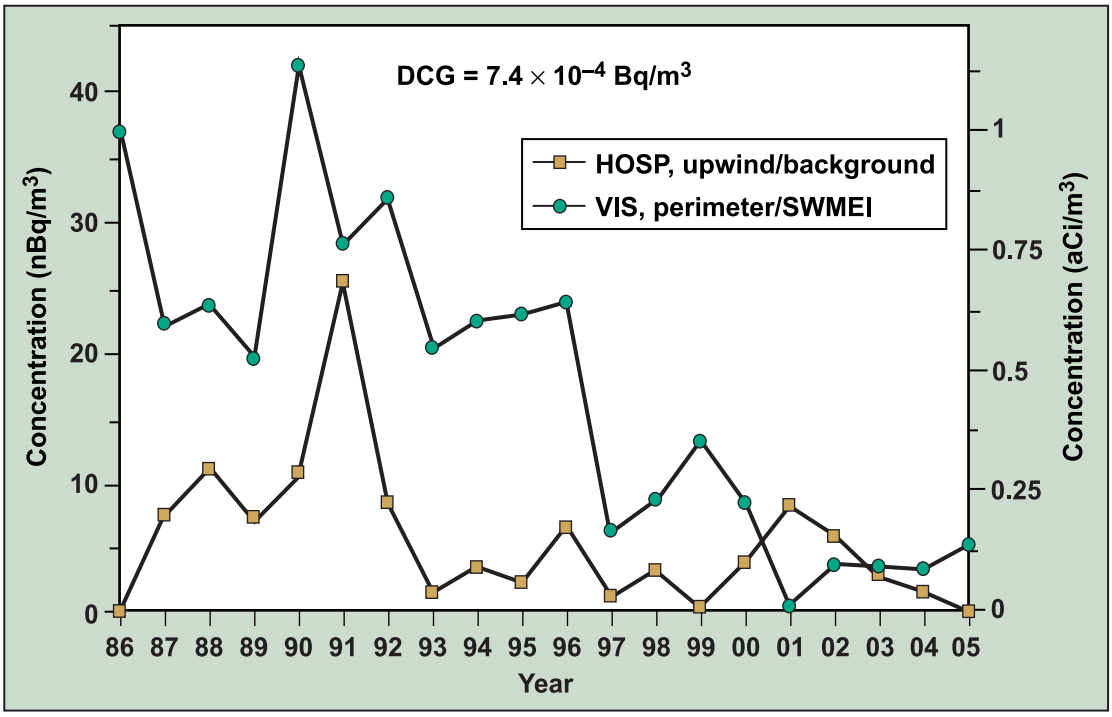


Figure 4-6. Calculated annual median concentrations of plutonium-239+240 for HOSP and VIS for the last 20 years

Uranium isotopes are naturally occurring. All but two of the uranium-235 analyses had positive detections, and all but one of the uranium-238 samples had a positive detection. The Livermore site monthly composites had a uranium-235 median concentration of 0.11 pg/m^3 and a uranium-238 median concentration of 15 pg/m^3. This results in a uranium-235/uranium-238 median ratio of 0.0073, which is considered natural uranium and typical of what has been recorded in the past.

The annual median uranium-235 concentration for all Site 300 locations was 0.17 pg/m^3 (or less than 0.0004% of the DCG) and the uranium-238 median concentration was 26 pg/m^3 (or less than 0.009% of the DCG). The annual median for the Site 300 uranium-235/uranium-238 ratio was 0.0063, which is indicative of the presence of some depleted uranium.

In 2005, a total of six depleted uranium shot experiments were conducted at Bunker 851. The two closest sample locations to the bunker were WOBS and WCP with annual median isotopic ratios of 0.0056 and 0.0042, respectively. The other sample locations at Site 300 had annual median isotopic ratios ranging from 0.0068 to 0.00729, which are more in line with natural uranium ratios.

Concentrations for both uranium-235 and uranium-238 were elevated at locations WCP and NPS after the grass fire in July (see Section "Gross Alpha and Gross Gamma Concentrations" earlier in this chapter for more information on the grass fire). The highest measured uranium-235 value for 2005 was 3.4 pg/m^3 (0.007% of the DCG) at WCP in July. The highest measured uranium-238 value was 952 pg/m^3 (0.3% of the DCG) at WCP also in July. Both WCP and NPS were downwind from the fire and near the fire's edge. The uranium-235/uranium-238 ratio for all locations at Site 300 for July ranged from 0.0035 to 0.0074, which is consistent with depleted and natural uranium ratios (0.002 and 0.0072, respectively). The elevated concentrations are attributed to increased mass loading of the filter due to resuspension of particulates from the fire.

Tritium Concentrations

Tritium data presented in **Table 4-5** summarize the biweekly tritium in air data provided in data tables (see file "Ch4 Ambient Air" on the report CD). Locations (see **Figures 4-1**, **4-2** and **4-3**) are grouped by expected concentrations of tritium. The highest concentrations of tritium are found near area (diffuse) sources monitored by the B331 and B624 samplers on the Livermore site. Area sources include stored containers of tritium waste or tritium-contaminated equipment from which HTO diffuses into the atmosphere. The annual mean and median concentrations for 2005 for the B331 and B624 air tritium samplers combined were essentially no different than the 2004 values. However, this was due to 2005 concentrations being higher at the B331 sampler and lower at the B624 sampler than in 2004. Because of operations at the Tritium Facility, the concentration of the B331 sample for December 1–15 was extremely high (23,700 mBq/m^3; see file "Ch4 Ambient Air" on the report CD). Concentrations this high were last observed in 1998. Samples from seven other locations exhibited their maximum concentrations for the year for the same sampling period. Because some of the samples (e.g., HOSP and COHO) were from locations that monitor background concentrations of tritium and some of the samples (e.g., VET,

Table 4-5. Tritium in air samples (mBq/m^3), 2005

Sampling locations	Detection frequency[a]	Mean	Median	IQR	Maximum	Median Percent of DCG[b]
Diffuse on-site sources	51 of 51	1590	444	740	23700	0.012
Livermore site perimeter[c]	182 of 222	67.3	45.1	60.5	1350	0.0012
Livermore Valley	51 of 147	8.27	8.18	18.9	126	0.00022
Site 300	8 of 25	6.33	7.29	22.2	40.0	0.002

a Rejected samples are not included in the statistics.
b DCG = Derived Concentration Guide of 3.7 x 10^6 mBq/m^3 for tritium in air.
c Locations COW, DWTF, MET, and POOL are not strictly on the perimeter of the site.

AMON, PATT) were from locations where detections are not expected, contamination of the analytical laboratory by the B331 sample was considered the likely cause of the unexpectedly high concentrations observed. As a result, all samples except for B331 were rejected for the sampling period as not being representative.

Air concentrations measured at samplers near the perimeter of the Livermore site are the next highest after those measured near diffuse sources, but the concentrations near the perimeter are, on average, less than 10% of those near the diffuse sources. Location DWTF, which is a sampler located downwind of the Decontamination and Waste Treatment Facility, exhibited the highest median annual concentration of the perimeter locations at just 0.0039% of the DCG. Median concentrations for 2005 for perimeter locations were on average about 60% higher for 2005 than for 2004. In 2005, releases from the Tritium Facility were about double those in 2004. The effect of this may be seen when the mean of all maximum concentrations for all perimeter locations for 2005 (331 mBq/m^3 [8.94 pCi/m^3]) is compared with that for 2004 (161 mBq/m^3 [4.35 pCi/m^3]).

Three samples (CAFE, VET, and POOL) exhibited maximum concentrations for the March 10–24 sampling period (see file "Ch4 Ambient Air" on the report CD). The highest value for all perimeter locations was 1350 mBq/m^3 (36.5 pCi/m^3) at POOL during this time. These high values are probably due to the much higher than average release from the Tritium Facility during the week of March 17–25.

In 2005, median concentrations at perimeter locations COW, MET, and MESQ were about 2.6 times greater than the concentrations observed in 2004. This increase is greater than expected from known release rates and is correlated with the presence of a transportainer that was located in the northwest area of the laboratory and contained tritiated waste from a building undergoing renovation.

All of the median concentrations in the Livermore Valley and at Site 300 (**Table 4-5**; see also file "Ch4 Ambient Air" on report CD for biweekly data) were below the detection limit in 2005. 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 Site 300 released tritium to the environment in 2005, 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 "Ch4 Ambient Air" on report CD for data.) The highest value at the Livermore site was 19 pg/m^3, which was recorded at location CAFE in October. This value is only 0.19% of the BAAQMD ambient concentration limit (ACL) for beryllium (10,000 pg/m^3). These data are similar to data collected from previous years.

Figure 4-7 is a plot of the median beryllium concentration at the Livermore site perimeter from 1975 through 2005. 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.

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 pg/m^3. The highest value for the Site 300 area sampling occurred at the offsite location TCDF in November with a value of 83 pg/m^3, which was just 0.83% of the ambient concentration limit.

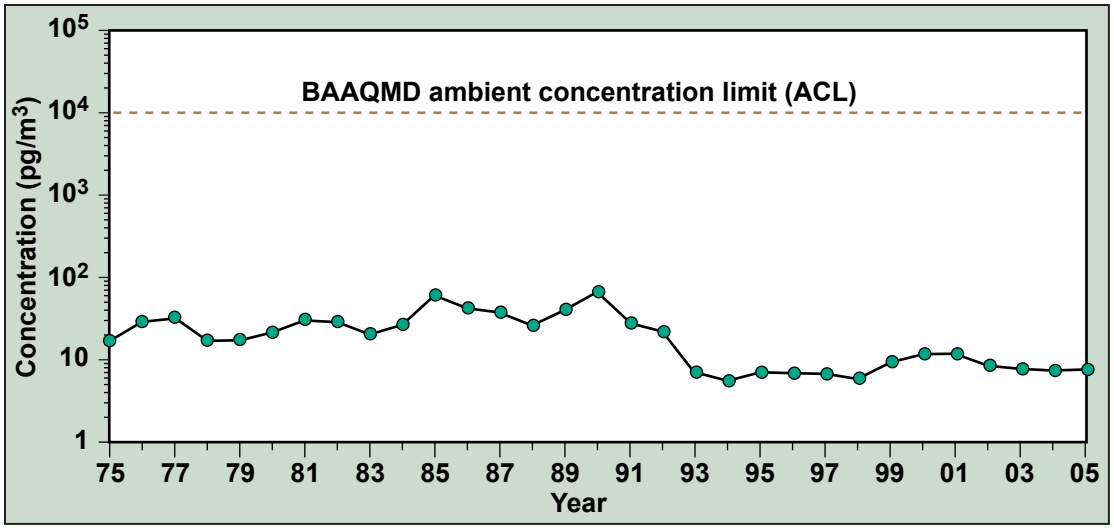


Figure 4-7. Median concentration of beryllium in air particulate samples taken at the Livermore site perimeter, 1975–2005

Environmental Impact of Ambient Air

LLNL operations involving radioactive materials had minimal impact on ambient air during 2005. 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 median and mean tritium concentrations for all Livermore site perimeter air tritium sampling locations in 2005 were about 60% and 30% higher, respectively, than in 2004 due to higher release rates from the Tritium Facility and its area source. 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 (0.5µrem/y) (i.e., the annual dose from inhaling air with a concentration at the detection limit of about 25 mBq/m^3 [0.675 pCi/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 (145 mBq/m^3) or VIS (107 mBq/m^3), the resulting doses would still be tiny (30.5 nSv/y [3.05 µrem/y] and 22.5 nSv/y [2.25 µrem/y], respectively). Put another way, the maximum concentration at CRED is just 0.3% of concentration limits for minor sources set by the U.S. EPA in Table 2, Appendix E to 40 CFR 61 (Harrach et al. 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 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 from LLNL operations.