5

Air Surveillance Monitoring

Paris E. Althouse
Paula J. Tate

Introduction

Lawrence Livermore National Laboratory performs air surveillance monitoring 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; DOE Order 5400.1, *General Environmental Protection Program* Chapter IV, paragraph 1.a. and paragraph 5; and DOE Order 5400.5, *Radiation Protection of the Public and the Environment,* Change 2, Chapter II, Paragraph 1 (except 1.a.3.c. and 1.c), and Chapter III. The *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (U.S. Department of Energy 1991) provides the guidance for implementing DOE Orders 5400.1 and 5400.5. In general, the constituents for which LLNL monitors are at levels far below the regulatory standards.

LLNL conducts surveillance monitoring of 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 air monitoring program, LLNL collects particles on filters and chemically traps vapors on a collection medium. Concentrations of various airborne radionuclides (including particles and tritiated water vapor) and beryllium 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 (Gallegos et al. 2000).

Methods

Several monitoring networks were established for surveillance of air particulates and tritium in the environs of LLNL and Site 300, as well as in the surrounding Livermore Valley and in the City of Tracy. The sampling locations for each monitoring network are listed in **Table 5-1** and shown on **Figures 5-1**, **5-2**, and **5-3**. All monitoring networks use

continuously operating samplers. The radiological high-volume sampling networks use glass-fiber filters; the beryllium high-volume networks use cellulose filters; and the low-volume network uses Millipore AW-19 filters. The collection medium for tritium is silica gel.

Table 5-1. Air sampling locations listed by monitoring network.

High-volume radiological (glass fiber filters)	High-volume beryllium (cellulose filters)	Low-volume gross alpha and beta (millipore filters)	Tritium (silica gel)
Livermore site sampling locations			
B531[a]	CAFE		B292[a]
CAFE	COW		B331[a]
COW	MESQ		B514[a]
CRED[a]	MET		B624[a]
MESQ	SALV		CAFE
MET	VIS		COW
SALV			MESQ
VIS			MET
			POOL
			SALV
			VIS
Livermore Valley sampling locations			
AMON		FCC	AMON
CHUR		HOSP	FIRE
FCC			HOSP
FIRE			VET
HOSP			XRDS
LWRP			ZON7
PATT			
TANK			
ZON7			
Site 300 sampling locations			
801E	801E		
ECP	EOBS		
EOBS	GOLF		
GOLF			
NPS			
WCP			
WOBS			
Site 300 off-site sampling locations			
PRIM	TFIR		PRIM
TFIR			

[a] These locations are in areas of diffuse sources and are monitored to fulfill NESHAPs requirements.

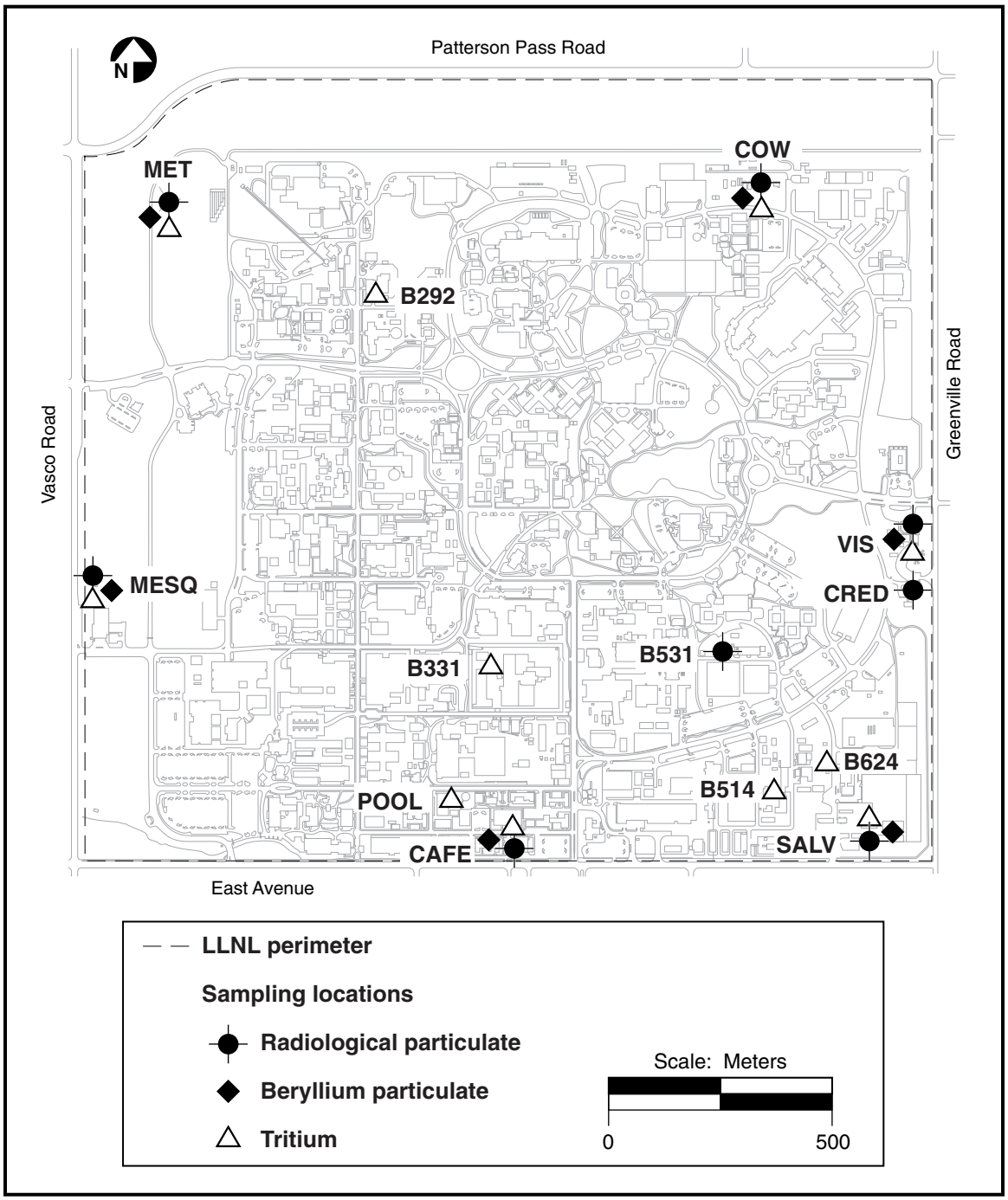

Figure 5-1. Air particulate and tritium sampling locations on the Livermore site, 1999.

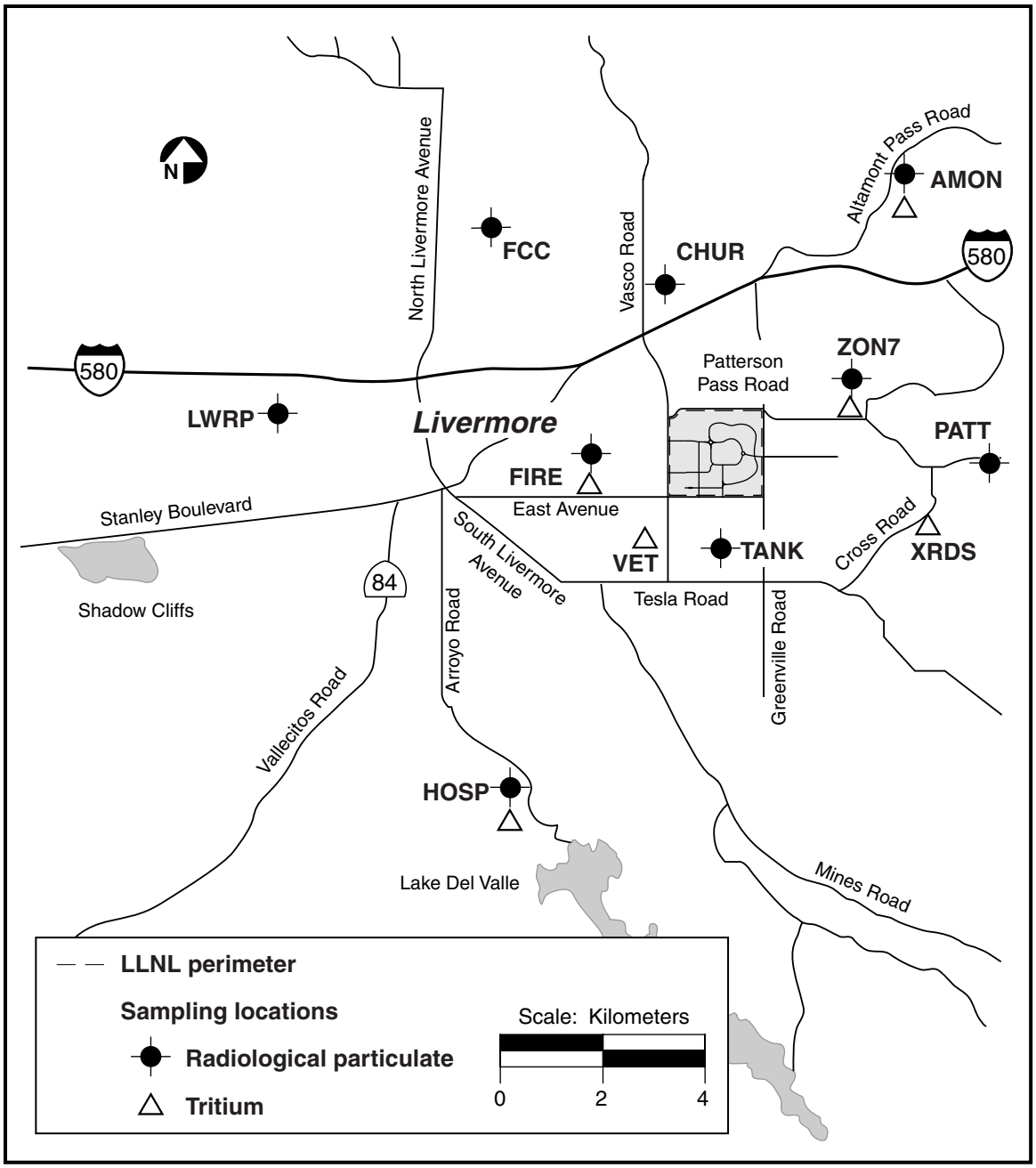

Figure 5-2. Air particulate and tritium sampling locations in the Livermore Valley, 1999.

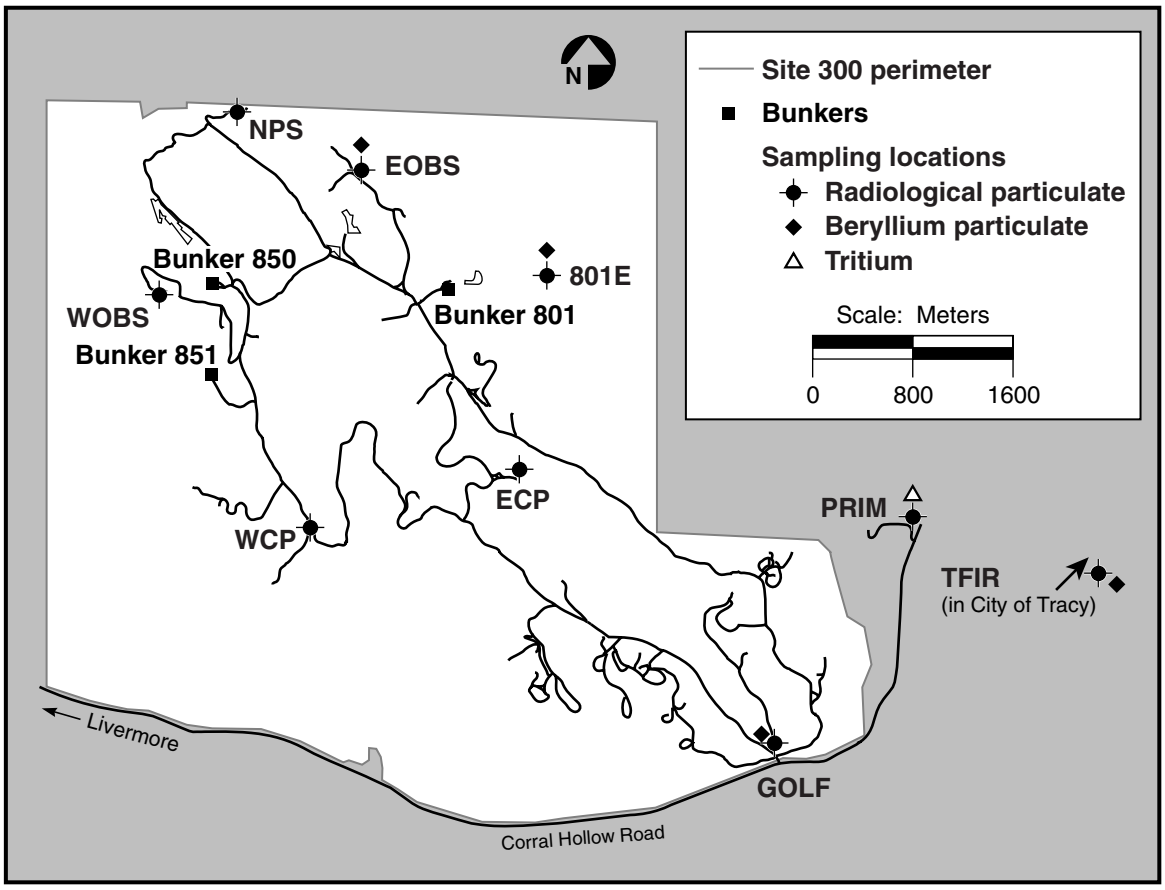

Figure 5-3. Air particulate and tritium sampling locations at Site 300 and off site, 1999.

All air samplers are positioned to provide reasonable probability that, if there were any significant concentration of radioactive or beryllium effluents from LLNL operations, it would be detected. The geographical details of the particulate sampling locations are described in the Environmental Protection Department's location database. Details for accessing the database are available in the Locations Database SOP Supplement EMP-QA-DM, *Sample and Data Management*.

Particulate filters are changed each week at all locations, and tritium samples are changed every two weeks. Duplicate quality control samplers are operated for two months each year in parallel with the permanent sampler at a given site, and these samples are analyzed to confirm results.

Air Particulate Sampling Locations

The Livermore site radiological air surveillance sampling network consists of six samplers at the perimeter; in addition, two areas of special interest (B531 and CRED shown in **Figure 5-1**) are monitored for plutonium only. These two locations in the southeast quadrant are areas of known plutonium contamination attributed to historic operations, which included the operation of solar evaporators for plutonium-containing liquid waste.

The Livermore Valley network (**Figure 5-2**) consists of air particulate samplers located in all compass directions from the Livermore site. For the purposes of data analysis, four samplers (FCC, FIRE, HOSP, and CHUR) located in the least prevalent wind directions are considered to be upwind or representative of background locations, and four samplers (PATT, ZON7, TANK, and AMON) located in the most prevalent downwind directions are considered most likely to be affected by Laboratory operations. An additional sampler is located in another area of special interest, the Livermore Water Reclamation Plant (LWRP), because of a 1967 and earlier plutonium releases to the sanitary sewer system with subsequent soil contamination and potential resuspension (see Chapter 10, Results, for a discussion of this).

Livermore site beryllium monitoring continued in 1999 at the six perimeter locations. To satisfy beryllium reporting requirements and determine the effects of the Laboratory's beryllium operations, LLNL conducted a technical assessment of the beryllium monitoring locations at Site 300 in 1997. Although there is no requirement to sample for beryllium at Site 300, as a best management practice, LLNL has decided to continue beryllium monitoring at three locations on site and at one location in the City of Tracy (TFIR).

Two sampling systems were added in July 1997 as part of the new low-volume radiological air surveillance sampling network. The samplers are situated at the FCC and HOSP locations, sites that are generally upwind of the Livermore site. The results are used to establish background levels of gross alpha and beta activity for direct comparison to results from the air effluent samplers (see Chapter 4). The sampling systems are very similar to the air-effluent samplers used in facilities, including sampling system design, sampler operation, filter media, sample tracking, sample analysis, and processing of results.

Tritium Sampling Locations

LLNL also maintains 11 continuously operating, airborne tritium samplers on the Livermore site (**Figure 5-1**), six samplers in the Livermore Valley (**Figure 5-2**), and one sampler near Site 300 (**Figure 5-3**) to assess historical and current activities that influence environmental impacts. Four of the Livermore site locations (B331, B292, B514, and B624) monitor diffuse tritium emissions.

Radiological Analysis

As outlined in *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (U.S. Department of Energy 1991), gross alpha and gross beta air filter results are used as trend indicators; specific radionuclide analysis is done for plutonium, uranium, and gamma emitters. Radiological analytical results are reported as a measured concentration per volume of air, or as less than the minimum detection concentration (MDC) when no activity is detected. In all cases, the MDC is adequate for demonstrating compliance with the pertinent regulatory requirements for radionuclides that may be or are present in the air sample and for evaluating LLNL-induced environmental impacts. Particle size distributions are not determined because the estimated effective dose equivalent to the maximally exposed individual is well below the 0.01-mSv (1-mrem) allowable limit as discussed in the above-mentioned environmental regulatory guide.

Gross alpha and gross beta activities are determined by gas-flow proportional counting, plutonium by alpha spectrometry, uranium by mass spectrometry, gamma emitters by gamma spectroscopy, and tritium by liquid scintillation. Further details of the surveillance monitoring methods are included in Chapter 5, Data Supplement.

Results

This section discusses the air monitoring results from all air surveillance locations at the Livermore site, Site 300, and all off-site surveillance locations.

In April 1997, the radiological air particulate sampling filter media were changed from cellulose to glass fiber; however, blank glass-fiber filters contain detectable amounts of some naturally occurring radiological isotopes (Althouse 1998) including uranium-235, uranium-238, potassium-40, radium-226, radium-228 and thorium-228. LLNL adjusts

the measured concentrations of these isotopes according to U.S. Environmental Protection Agency (EPA) procedures (Eadie and Bernhardt 1976) and subtracts the appropriate blank filter content from the gross analytical result to obtain a corrected net result.

Livermore Site

Airborne Radioactivity

Table 5-2 summarizes the monthly gross alpha and gross beta results for the LLNL perimeter, Livermore Valley, Site 300, and Site 300 off-site sampling locations. Detection frequencies, median concentrations, interquartile ranges (IQR), and maximum concentration values for each network are included. (See Data Supplement Tables 5-1, 5-2, and 5-3 for detailed location results for all high-volume networks for gross alpha and gross beta concentrations.)

Typical gross alpha activity (median value) for the LLNL perimeter is 4.5×10^{-5} Bq/m^3 (1.2×10^{-15} Ci/m^3); for the upwind Livermore Valley stations, the value is 3.8×10^{-5} Bq/m^3 (1.0×10^{-15} Ci/m^3); and for the downwind Livermore Valley stations, the value is 5.0×10^{-5} Bq/m^3 (1.3×10^{-15} Ci/m^3). Negative values occur when the activity of the analytical background filters is higher than the activity on the filters being analyzed. Typical gross beta activity (median value) for the LLNL perimeter is 3.2×10^{-4} Bq/m^3 (8.7×10^{-15} Ci/m^3); for the upwind Livermore Valley stations, the value is 3.0×10^{-4} Bq/m^3 (8.1×10^{-15} Ci/m^3); and for the downwind Livermore stations, the value is 3.4×10^{-4} Bq/m^3 (9.1×10^{-15} Ci/m^3). The primary sources of the alpha and beta activities are the naturally occurring radioisotopes of uranium and thorium, and any residual fallout from atmospheric weapons testing and the 1986 Chernobyl reactor accident. The values are slightly higher than those obtained from previous monitoring data during the past several years and are likely caused by a change in March in the analytical laboratory used to perform the gross alpha and gross beta analysis. Data were also elevated when the analytical laboratory was changed in 1993.

The monthly median gross alpha and gross beta concentrations are plotted in **Figures 5-4** and **5-5**, respectively. The gross beta results followed a pattern similar to previous years' data. The gradual increase in beta activity throughout the summer was most likely caused by an increase in resuspension of soils that occurs during the dry season.

Table 5-2. Gross alpha and gross beta concentration in air particulate samples summarized by month, 1999.

Month	Gross alpha (10^{-6} Bq/m^3)				Gross beta (10^{-6} Bq/m^3)			
	Detection frequency[a]	Median	IQR[b]	Maximum	Detection frequency	Median	IQR	Maximum
LLNL perimeter locations								
Jan	4/30	28.0	59.3	85.1	30/30	712	680	1460
Feb	1/24	9.78	21.8	68.0	20/24	203	100	478
Mar	21/24	45.9	20.9	72.9	24/24	230	72.8	327
Apr	22/29	50.3	49.3	98.8	29/29	269	148	844
May	7/24	18.6	18.1	74.4	24/24	315	90.1	394
Jun	11/23	27.3	37.0	130	23/23	185	116	525
Jul	22/30	43.1	39.9	125	30/30	245	108	659
Aug	13/24	35.1	64.6	96.9	24/24	331	357	696
Sep	23/23	71.4	36.4	120	23/23	648	179	932
Oct	28/30	228	168	385	30/30	1300	607	2660
Nov	20/24	115	77.2	214	24/24	882	276	1070
Dec	24/24	86.6	94.1	277	24/24	696	778	2530
Livermore Valley upwind locations								
Jan	2/20	16.1	64.8	84.6	20/20	651	822	1460
Feb	1/16	8.0	42.7	64.1	16/16	196	100	443
Mar	10/16	44.8	32.4	76.2	16/16	235	38.9	310
Apr	12/20	37.1	35.7	106	20/20	258	83.9	666
May	6/16	16.3	20.4	61.8	16/16	275	84.6	442
Jun	4/15	21.0	11.9	54.4	15/15	203	109	444
Jul	13/20	38.5	38.5	104	20/20	247	84.4	736
Aug	11/16	35.1	34.5	98.8	16/16	325	379	655
Sep	16/16	70.1	18.2	98.8	16/16	670	198	892
Oct	19/20	252	179	414	20/20	1270	566	2350
Nov	13/16	115	116	212	16/16	812	283	1120
Dec	15/16	90.8	87.0	259	16/16	605	832	2500

Table 5-2. Gross alpha and gross beta concentration in air particulate samples summarized by month, 1999 (concluded).

Month	Gross alpha (10^{-6} Bq/m^3)				Gross beta (10^{-6} Bq/m^3)			
	Detection frequency[a]	Median	IQR[b]	Maximum	Detection frequency	Median	IQR	Maximum
Livermore Valley downwind locations								
Jan	3/25	11.3	50.9	101	25/25	717	696	1480
Feb	1/20	14.4	33.5	64.0	20/20	198	129	376
Mar	17/20	53.7	32.4	85.5	20/20	248	47.2	349
Apr	20/25	51.4	45.5	128	25/25	302	84.4	725
May	7/20	15.7	28.7	102	20/20	268	52.1	472
Jun	12/20	30.2	26.9	62.5	20/20	201	80.4	403
Jul	12/25	24.7	48.6	104	25/25	253	75.9	740
Aug	18/20	48.1	35.0	102	20/20	375	447	673
Sep	20/20	70.7	38.9	120	20/20	710	130	903
Oct	22/25	231	180	492	25/25	1210	648	2680
Nov	19/20	119	89.7	182	20/20	821	353	1080
Dec	18/20	92.9	129	302	20/20	677	895	2450
Site 300[c] sampling locations								
Jan	2/35	20.8	42.2	110	34/35	551	651	1320
Feb	3/28	7.70	36.5	79.5	26/28	189	122	497
Mar	21/28	49.4	44.0	89.9	28/28	247	80	367
Apr	29/35	50.3	32.6	134	35/35	344	133	803
May	18/28	33.3	25.4	70.7	28/28	376	109	466
Jun	20/28	35.3	31.4	86.2	28/28	320	161	488
Jul	27/35	63.3	50.7	143	35/35	301	155	873
Aug	20/22	49.4	50.9	128	22/22	463	426	818
Sep	28/28	96.8	41.1	142	28/28	984	257	1210
Oct	32/35	221	152	352	35/35	1200	509	2330
Nov	26/28	116	93.8	214	28/28	849	369	1240
Dec	28/28	91.6	110	302	28/28	644	833	2510

a Detection frequency is the number of samples with results above the detection limit divided by the number of samples.

b IQR = Interquartile range.

c Results for Site 300 off-site locations TFIR and PRIM are given in the Data Supplement Table 5-15.

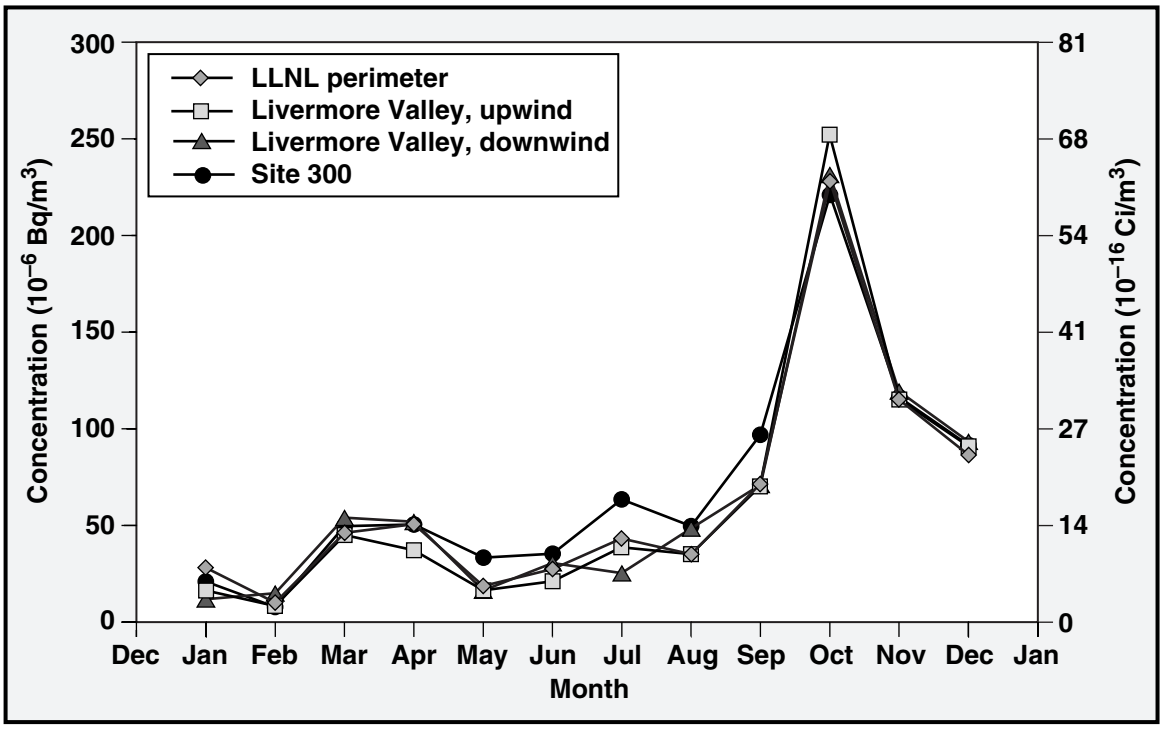

Figure 5-4. Monthly median gross alpha concentrations in particulate air samples from the LLNL perimeter, Livermore Valley, and Site 300 sampling locations, 1999.

Gamma-emitting radionuclide concentrations in air that contribute to the activity in the Livermore site perimeter samples are summarized in **Table 5-3**. (See Data Supplement Table 5-4 for monthly gamma activity data.) Of the nuclides identified, all were naturally occurring, with the exception of cesium-137. The primary source of cesium-137 is long-term global fallout and fallout resuspension.

By analyzing air samples for gamma-emitting radionuclides, LLNL verifies that there is no evidence of a release of the small inventories of mixed fission products and radiochemical tracers used at LLNL and also obtains baseline data on global fallout. The Derived Concentration Guides (DCGs) for these radionuclides are shown in **Table 5-3**. For air, DCGs specify the concentrations of radionuclides that could 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 (DOE Order 5400.5). (Chapter 13, Radiological Dose Assessment, provides an explanation of

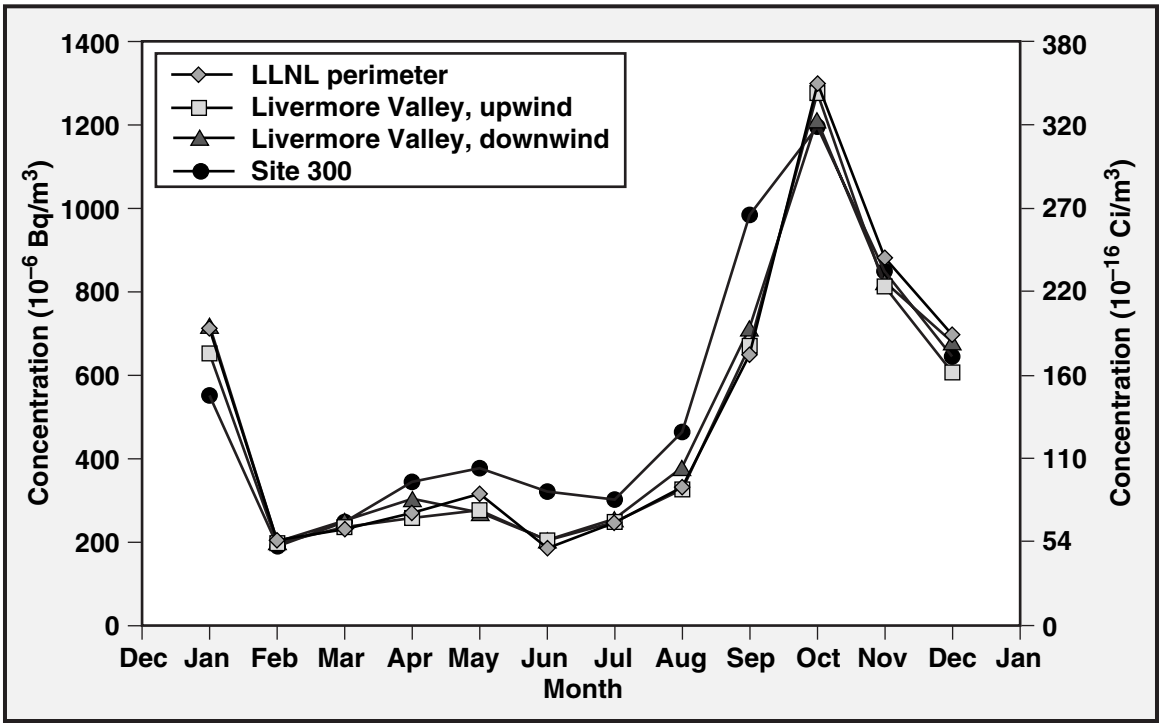

Figure 5-5. Monthly median gross beta concentrations in particulate air samples from the LLNL perimeter, Livermore Valley, and Site 300 sampling locations, 1999.

this and other units of dose.) **Table 5-3** also presents the percent of the DCGs, which demonstrates that the level of gamma activity present in air at the Livermore site perimeter was far below the DCGs.

Table 5-4 shows the concentrations of airborne plutonium-239+240 on air filters from the LLNL perimeter locations. (See Data Supplement Table 5-5 for the monthly data by location.) The highest concentration was registered at location VIS in May 1999; the concentration value is reported as 3.9×10^{-8} Bq/m^3 (1.1×10^{-18} Ci/m^3), which represents 0.005% of the DCG. The median concentration at location VIS is 1.3×10^{-8} Bq/m^3 (3.5×10^{-19} Ci/m^3), which is slightly lower than that for the previous year.

Table 5-4 also shows the detection frequency, median concentration, IQR, maximum concentration, and percent of DCG for the concentration of plutonium on air filter samples collected in the Livermore Valley. (See Data Supplement Table 5-6 for monthly data.) The highest off-site concentration of plutonium-239+240 occurred at PATT during October, which had a median observed value of 0.0004% of the DCG.

Table 5-3. Gamma activity in air particulate samples, Livermore site perimeter and Site 300, 1999.

	^{7}Be	^{40}K	^{137}Cs	^{22}Na	^{226}Ra	^{228}Ra	^{228}Th
	(10^{-3} Bq/m^3)	(10^{-6} Bq/m^3)					
Livermore perimeter locations							
Median	3.4	12	0.21	0.24	–0.011	0.33	0.29
Interquartile range	0.85	20	—[a]	—[a]	0.95	0.98	0.72
Maximum	7.8	40	0.57	0.57	0.96	3.7	1.8
Percent of DCG[b]	2.3×10^{-4}	3.7×10^{-5}	1.4×10^{-6}	6.4×10^{-7}	2.6×10^{-3}[c]	3.0×10^{-4}	0.019
Site 300 locations							
Median	3.8	13	0.20	0.43	0.17	0.36	–0.31
Interquartile range	2.3	41	—[a]	—[a]	2.0	1.5	1.2
Maximum	7.2	61	0.62	0.71	1.8	5.4	3.3
Percent of DCG	2.5×10^{-4}	3.8×10^{-5}	1.3×10^{-6}	1.1×10^{-6}	4.6×10^{-4}	3.2×10^{-4}	0.22[c]
DCG (Bq/m^3)	1.5×10^{3}	33	15	37	0.037	0.11	1.5×10^{-3}

[a] No measure of dispersion calculated; see Chapter 14, Quality Assurance.

[b] Derived Concentration Guide. Percent calculated from the median concentration.

[c] Percent of DCG calculated with maximum value because the median is negative.

Table 5-4 shows the median concentrations of airborne plutonium-239+240 at the two diffuse source locations (B531 and CRED). (See Data Supplement Table 5-7 for monthly data.) The median concentration of 2.7×10^{-8} Bq/m^3 (7.3×10^{-19} Ci/m^3) at location B531 is higher than the median concentration for any of the other air particulate sampling locations, but it is still only 0.004% of the DCG. The higher concentrations are attributed to historic waste management operations, which included the operation of solar evaporators for plutonium-containing liquid waste (Silver et al. 1974).

In October, the plutonium concentrations reported were above the minimum detectible level for all locations. While this is unusual, none of these values exceeded the action levels identified in the *Environmental Monitoring Plan* (Tate et al. 1999). However, because the concentrations were unusual, they were investigated at the analytical laboratory. The analytical laboratory found no source of error or contamination. A similar increase was detected in nearly all other particulate surveillance data (including low-volume data) in October. This increase is likely the result of increased particulate resuspension and subsequent filter loading that occurs during dry months. This anomaly did not occur in any other months in 1999.

Table 5-4. Plutonium-239+240 activity in air particulate samples (10^{-9} Bq/m^3), 1999.

Sampling location[a]	Detection frequency[b]	Median	Interquartile range	Maximum	Percent of DCG[c]
Livermore Valley downwind locations					
AMON	4/12	3.58	4.16	16.1	4.84×10^{-4}
PATT	4/12	2.68	4.57	25.2	3.62×10^{-4}
TANK	2/12	2.01	1.99	8.66	2.71×10^{-4}
ZON7	2/12	1.87	2.65	23.1	2.52×10^{-4}
Livermore Valley upwind locations					
CHUR	3/12	1.37	5.78	23.8	1.84×10^{-4}
FCC	1/12	0.103	2.85	11.1	1.39×10^{-5}
FIRE	2/12	2.10	3.12	11.1	2.84×10^{-4}
HOSP	1/12	0.264	1.52	6.40	3.56×10^{-5}
LLNL perimeter locations					
CAFE	8/12	5.51	7.01	25.1	7.45×10^{-4}
COW	3/12	4.64	2.22	28.7	6.28×10^{-4}
MESQ	1/12	3.85	3.19	22.0	5.21×10^{-4}
MET	2/12	2.52	2.83	7.99	3.41×10^{-4}
SALV	3/12	3.88	2.43	18.0	5.25×10^{-4}
VIS	7/12	13.0	20.1	38.9	1.76×10^{-3}
Diffuse on-site sources locations					
B531	12/12	26.9	50.2	94.0	3.64×10^{-3}
CRED	7/12	6.05	4.29	33.5	8.18×10^{-4}
Special interest location					
LWRP	5/12	5.06	3.24	21.1	6.83×10^{-4}
Site 300 on-site locations					
S300 composite	4/12	1.79	2.52	8.7	2.41×10^{-4}
Site 300 off-site locations					
PRIM	2/12	1.91	3.78	11.3	2.58×10^{-4}
TFIR	3/11	2.43	5.67	12.6	3.29×10^{-4}

[a] See **Figures 5-1**, **5-2**, and **5-3** for sampling locations.

[b] Detection frequency is the number of samples with results above the detection limit divided by the number of samples.

[c] DCG = Derived Concentration Guide of 7.4×10^{-4} Bq/m^3 (2×10^{-8} µCi/m^3) for ^{239}Pu activity in air. Percent calculated from the median concentration.

Figure 5-6 shows the annual median concentrations of plutonium-239+240 for locations SALV (on site) and FCC (off site) from 1982 to 1999. Location FCC represents a typical upwind background location, and SALV represents a typical perimeter location. The annual median concentration for FCC was 1.0×10^{-10} Bq/m^3 (2.7×10^{-21} Ci/m^3).

Figure 5-6 uses a log scale and, for the years in which a negative median concentration was calculated, the positive value closest to the median was plotted. The higher values in the past at SALV may be attributed to historical activities at LLNL. In 1993, clean top soil was laid over much of the area, reducing the potential for increased levels from soil resuspension. The sampler at SALV was moved to a nearby grassy knoll, possibly resulting in a decrease in the plutonium median for 1999. The downward trend at location FCC is the result of decreasing residual global fallout.

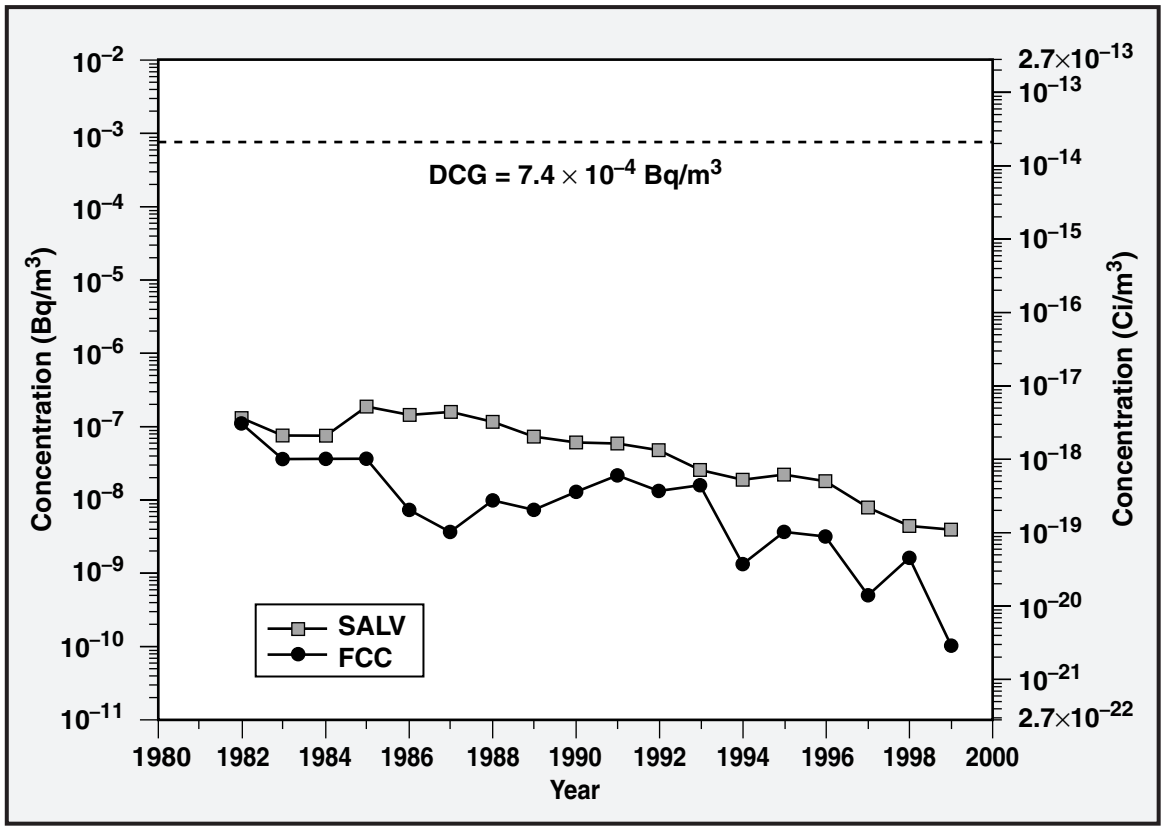

Figure 5-6. Annual median plutonium concentrations in air particulate samples at two locations, SALV and FCC, 1982–1999.

The ratio of uranium-235 to uranium-238 can be used to identify the source of the uranium. Both uranium-235 and uranium-238 occur naturally in the area, but only 0.7% of the naturally occurring uranium is uranium-235, and the remainder is almost entirely uranium-238. The median uranium-235 and uranium-238 mass concentrations in air samples from the Livermore site perimeter are shown in **Table 5-5**. (See Data Supplement Table 5-8 for monthly data.) The maximum measured concentration of uranium-238 (at location SALV during October) is less than 0.2% of the DCG. All uranium-235/uranium-238 median ratios are generally as expected for naturally occurring uranium; however, September monthly data in the Data Supplement show some unexpected uranium-235/uranium-238 ratios. The cause for these anomalous data is unknown; however, these ratios may be the result of increased variability in measured concentrations near the detection limit. No significant environmental impact stems from the observed ratios.

The low-volume radiological air sampling locations HOSP and FCC have typical gross alpha and gross beta activity of 4.7×10^{-5} Bq/m^3 (1.3×10^{-15} Ci/m^3) and 4.8×10^{-4} Bq/m^3 (1.3×10^{-14} Ci/m^3), respectively. (See Data Supplement Tables 5-9 and 5-10 for monthly median data.) These gross alpha values are slightly higher than those reported from the high-volume sampling systems at the same locations. The difference is probably caused by differences in the filter type. LLNL is conducting a study to determine the cause of the differences.

Table 5-6 shows the median concentrations of tritiated water vapor for the Livermore Valley sampling locations. (See Data Supplement Table 5-11 for biweekly data for each location.) The highest annual median concentration was observed at location ZON7. At approximately 3.9×10^{-2} Bq/m^3 (1.1×10^{-12} Ci/m^3), this concentration represents 0.001% of the DCG. The highest biweekly concentration was observed in February at ZON7. If it were a yearly average, this concentration, 0.27 Bq/m^3 (7.3×10^{-12} Ci/m^3), would be 0.007% of the DCG. The 1999 tritium values were slightly higher than those reported last year because of slightly elevated emissions from the Tritium Facility (Building 331) during January, February, and March.

Table 5-6 also shows the median concentrations of tritiated water vapor that were observed at the Livermore site perimeter sampling locations. (See Data Supplement Table 5-12 for biweekly data.) The highest annual median concentration was observed at location POOL, which was 0.14 Bq/m^3 (3.8×10^{-12} Ci/m^3), or 0.004% of the DCG.

Table 5-5. Uranium mass concentration in air particulate samples, 1999.

Sampling location[a]	^{235}U[b] (10^{-7} µg/m^3)	^{238}U[c] (10^{-5} µg/m^3)	$^{235}U/^{238}U$ [d] (10^{-3})
LLNL perimeter locations			
CAFE			
Median	4.29	6.57	6.70
Interquartile range	2.54	5.32	0.335
Maximum	11.3	17.9	NA[e]
Percent of DCG[f]	9.13×10^{-4}	2.19×10^{-2}	NA
COW			
Median	4.96	7.13	6.88
Interquartile range	3.74	5.96	0.363
Maximum	10.9	15.7	NA
Percent of DCG	1.05×10^{-3}	2.38×10^{-2}	NA
MESQ			
Median	3.90	6.43	6.97
Interquartile range	3.07	4.41	0.549
Maximum	8.88	12.4	NA
Percent of DCG	8.30×10^{-4}	2.14×10^{-2}	NA
MET			
Median	2.66	3.80	6.85
Interquartile range	1.26	2.09	0.419
Maximum	9.32	13.5	NA
Percent of DCG	5.67×10^{-4}	1.27×10^{-2}	NA
SALV			
Median	2.34	3.77	6.88
Interquartile range	1.62	2.39	0.931
Maximum	10.3	14.8	NA
Percent of DCG	4.97×10^{-4}	1.26×10^{-2}	NA
VIS			
Median	3.25	4.76	6.85
Interquartile range	2.72	5.3	0.410
Maximum	11.4	17.1	NA
Percent of DCG	6.92×10^{-4}	1.59×10^{-2}	NA

Table 5-5. Uranium mass concentration in air particulate samples, 1999 (concluded).

Sampling location[a]	^{235}U[b] (10^{-7} µg/m^3)	^{238}U[c] (10^{-5} µg/m^3)	$^{235}U/^{238}U$ [d] (10^{-3})
Site 300 on-site locations			
Site 300 composite			
Median	2.91	4.55	6.29
Interquartile range	3.73	6.16	0.919
Maximum	9.06	18.6	NA
Percent of DCG	6.20	1.52	NA
Site 300 off-site location			
PRIM			
Median	4.01	5.70	6.98
Interquartile range	4.01	5.56	0.604
Maximum	14.7	18.2	NA
Percent of DCG	8.53	1.90	NA

[a] See **Figures 5-1** and **5-3** for sampling locations.

[b] Derived Concentration Guide = 0.047 µg/m^3 for uranium-235 activity in air. Uranium-235 activities in Bq/m^3 can be determined by dividing the weight in µg/m^3 by 12.5, and pCi m^3 can be determined by dividing by 0.463.

[c] Derived Concentration Guide = 0.3 µg/m^3 for uranium-238 activity in air. Uranium-238 activities in Bq/m^3 can be determined by dividing the weight in µg/m^3 by 80.3, and pCi m^3 can be determined by dividing by 2.97.

[d] Naturally occurring uranium has a uranium-235/uranium-238 ratio of 7.1×10^{-3}.

[e] NA = Not applicable.

[f] DCG = Derived Concentration Guide. Percent calculated from the median concentration.

Diffuse sources of tritium on the Livermore site are monitored at air tritium sampling locations B292, B331, B514, and B624. **Table 5-6** shows the median concentrations of tritiated water vapor for these sampling locations. (See Data Supplement Table 5-13 for biweekly data.) The highest median concentration was observed at location B624. This concentration was 4.5 Bq/m^3 (1.2×10^{-10} Ci/m^3) and represents 0.1% of the DCG. The highest biweekly tritium concentration, 13.8 Bq/m^3 (3.7×10^{-10} Ci/m^3), was observed in April at location B331. If it were a yearly average, this concentration would represent 0.4% of the DCG.

The B331 location is near the Tritium Facility (Building 331), where LLNL personnel have reduced operations in recent years and performed significant inventory reduction and cleanup activities. During this process, tritium-contaminated equipment slated for disposal is stored in an area outside B331 before being sent to Hazardous Waste Management facilities. During 1999, outgassing from such waste processing released an estimated 2.7×10^{11} Bq (7.3 Ci) of tritium to the atmosphere outside Building 331.

Table 5-6. Tritium in air samples (10^{-3} Bq/m^3), 1999.

Sampling location[a]	Detection frequency[b]	Median	IQR[c]	Maximum	Percent of DCG[d]	Median dose (mSv)[e]
Livermore Valley locations						
AMON	13/25	20.0	25.6	175	5.4×10^{-4}	4.2×10^{-6}
FIRE	10/24	13.7	24.2	147	3.7×10^{-4}	2.8×10^{-6}
HOSP	5/23	3.27	10.7	87.0	8.8×10^{-5}	6.8×10^{-7}
VET	10/24	15.3	34.2	242	4.1×10^{-4}	3.2×10^{-6}
XRDS	9/25	10.2	24.9	93.2	2.8×10^{-4}	2.1×10^{-6}
ZON7	19/25	39.2	44.4	268	1.1×10^{-3}	8.1×10^{-6}
Livermore perimeter locations						
CAFE	23/26	65.0	85.5	1,890	1.8×10^{-3}	1.3×10^{-5}
COW	25/25	52.5	95.8	688	1.4×10^{-3}	1.1×10^{-5}
MESQ	16/25	45.9	68.1	357	1.2×10^{-3}	9.5×10^{-6}
MET	19/26	35.8	48.6	236	9.7×10^{-4}	7.4×10^{-6}
POOL	25/26	139	269	1,400	3.8×10^{-3}	2.9×10^{-5}
SALV	20/25	50.7	71.3	485	1.4×10^{-3}	1.1×10^{-5}
VIS	25/25	89.9	84.3	622	2.4×10^{-3}	1.9×10^{-5}
Diffuse on-site sources locations						
B292	26/26	182	228	540	4.9×10^{-3}	3.8×10^{-5}
B331	22/22	2,530	7,930	13,800	6.8×10^{-2}	5.2×10^{-4}
B514	26/26	1,650	1,230	2,950	4.5×10^{-2}	3.4×10^{-4}
B624	26/26	4,520	2,410	9,180	1.2×10^{-1}	9.4×10^{-4}
Site 300 off site location						
PRIM	2/25	4.11	15.0	27.4	1.1×10^{-4}	8.5×10^{-7}

[a] See **Figures 5-1**, **5-2**, and **5-3** for sample locations.

[b] Detection frequency is the number of samples with results above the detection limit divided by the number of samples.

[c] IQR = Interquartile range.

[d] DCG = Derived Concentration Guide of 3.7×10^3 Bq/m^3. Percent calculated from the median concentration.

[e] 1 mSv = 100 mrem.

The B624 location is situated in the Building 612 yard, which is dedicated to hazardous waste, radioactive waste, and mixed-waste management activities. The yard has several areas where waste containers that are outgassing tritium are stored outdoors.

The B514 sampling location is in a hazardous waste management area where tritium-contaminated waste is treated, and the B292 location is near an underground retention tank that had previously leaked. The concentrations in air at the B514 sampling location are variable because of the changing concentrations of tritium in the waste stream. The 1999 median concentrations at B292 and B514 are similar to the median concentrations in 1998.

Beryllium in Air

The median concentrations of airborne beryllium for the Livermore site perimeter sampling locations are shown in **Table 5-7**. (See Data Supplement Table 5-14 for monthly data.) The highest value of 37.8 pg/m^3 was found in the October composite at location COW and was most likely attributed to resuspension and mass loading of particulates containing naturally occurring beryllium from construction activities and a lack of rainfall. The median concentration for this location is 0.11% of the monthly ambient concentration limit (ACL) of 10,000 pg/m^3 established by the Bay Area Air Quality Management District (BAAQMD) and the EPA.

Table 5-7. Beryllium[a] in air particulate samples (pg/m^3), Livermore site perimeter and Site 300 locations, 1999.

Sampling location[b]	Detection frequency[c]	Median	Interquartile range	Maximum
Livermore perimeter locations				
CAFE	11/12	11.1	7.40	23.8
COW	12/12	10.8	9.10	37.8
MESQ	11/12	11.0	7.50	36.4
MET	11/12	7.83	4.28	23.9
SALV	10/12	7.47	6.31	25.7
VIS	12/12	10.3	8.02	33.2
Site 300 locations				
801E	12/12	11.8	9.32	30.9
EOBS	10/12	5.53	4.77	18.2
GOLF	12/12	10.2	14.3	26.4
TFIR	11/11	13.4	6.24	32.3

[a] The state ambient concentration limit is 10,000 pg/m^3.

[b] See **Figures 5-1** and **5-3** for sampling locations.

[c] Detection frequency is the number of samples with results above the detection limit divided by the number of samples.

Figure 5-7 is a plot of the median beryllium concentration at the Livermore site perimeter from 1974 through 1999. The decrease in median concentration in 1993 and the increase in 1999 were the result of a change in the analytical laboratory used to perform the analysis. The overall median concentration from 1974 through 1999 was calculated to be 0.2% of the ACL. Unless there is a change in LLNL's operations, the beryllium levels are expected to remain unchanged.

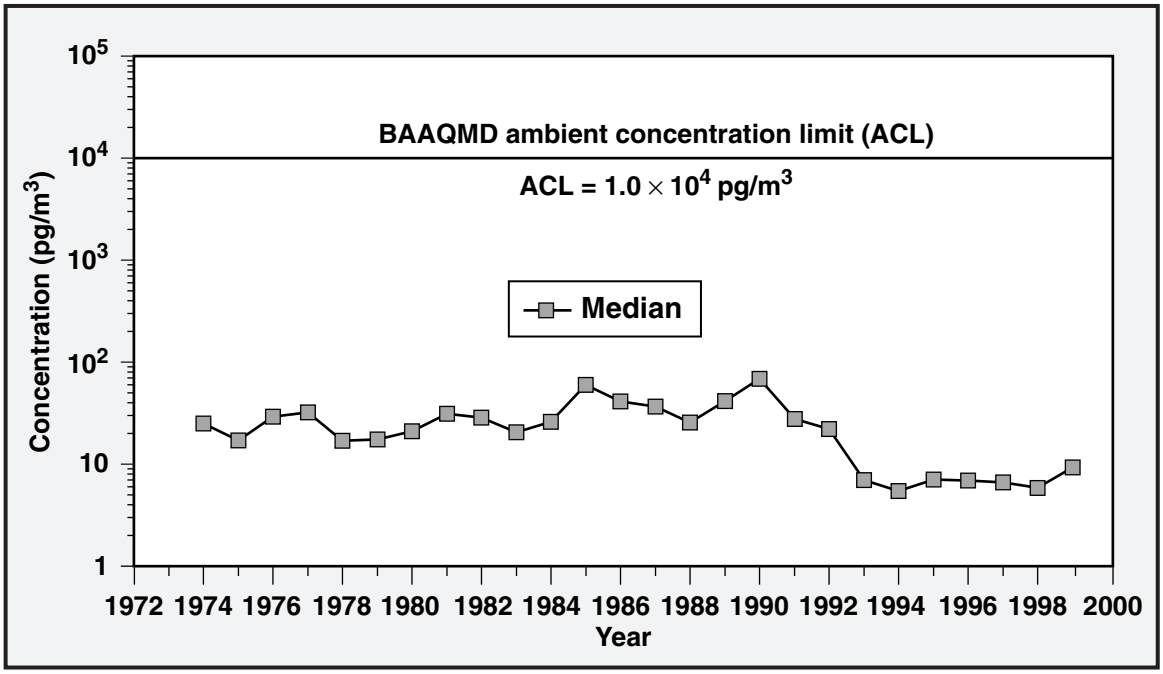

Figure 5-7. Median concentration of beryllium in air particulate samples taken at the Livermore site perimeter, 1974–1999.

Site 300

Airborne Radioactivity

Table 5-2 shows the detection frequency and the monthly gross alpha and gross beta median, IQR, and maximum for sampling locations at Site 300. (See Data Supplement Table 5-15 for monthly data.) The monthly median gross alpha and gross beta concentrations are shown in **Figures 5-4** and **5-5**. The Site 300 gross alpha and gross beta results show a similar pattern to those found at the Livermore site. Typical gross alpha activity is 5.0×10^{-5} Bq/m^3 (1.3×10^{-15} Ci/m^3). Typical gross beta activity is

4.2×10^{-4} Bq/m^3 (1.1×10^{-14} Ci/m^3). These values are slightly higher than those obtained from previous monitoring data during the past several years and were likely caused by the variations in how the different analytical laboratories performed the analyses. (As mentioned above, LLNL selected a new analytical laboratory to perform gross alpha and gross beta analysis in March 1999).

The primary sources of observed gross alpha and gross beta activity are naturally occurring radioisotopes of uranium and thorium, their decay products, and any residual fallout from atmospheric weapons testing and the 1986 Chernobyl reactor accident.

Table 5-3 lists the annual median activities, detection frequencies, IQR, maximum, the percent of the DCG, as well as the DCG, of gamma-emitting radionuclides in samples from Site 300. (See Data Supplement Table 5-16 for monthly data.) All these radionuclides were measured at concentrations significantly below the DCGs. Of the nuclides identified, all are naturally occurring, with the exception of cesium-137. The primary source of ^{137}Cs normally is long-term global fallout and resuspension.

Table 5-4 shows the median concentration of plutonium-239+240 on air-filter samples collected from Site 300. (See Data Supplement Table 5-17 for monthly data.) The highest concentration of plutonium-239 was recorded in the September composite at a level of 8.7×10^{-9} Bq/m^3 (2.3×10^{-19} Ci/m^3), or 0.001% of the DCG.

Table 5-5 shows the median concentration of uranium-235, uranium-238, and the uranium-235/uranium-238 ratio on air samples from Site 300 and vicinity. (See Data Supplement Table 5-18 for monthly data.) The highest concentration of uranium-238 was observed in the September composite at a level of 1.9×10^{-4} µg/m^3. The highest uranium-235 concentration was recorded at PRIM during March at a level of 1.5×10^{-6} µg/m^3.

As previously discussed in the Livermore Site Results section, the ratio of uranium-235 to uranium-238 is used to identify the source of the uranium. Because Site 300 operations use depleted uranium that contains very little uranium-235, it follows that if the ratio remains constant and near 0.7% (within the limit of sampling and analytical error), then the uranium-238 measured is from natural sources. The uranium-235/uranium-238 ratios in the September and October Site 300 composite (and in August and September at location PRIM) are less than expected for natural sources, which indicates some impact from operations at Site 300. These data are supported by Site 300 activities from B851 published in the NESHAPs Report (Gallegos et al. 2000). The median concentration of uranium-238 for 1999, however, is only 0.02% of the DCG.

The uranium-235/uranium-238 median ratios for PRIM (off site) are generally as expected for naturally occurring uranium; however, because of the higher value recorded for the uranium-235 during March, this ratio of 2.21×10^{-2} indicates other than natural uranium at this site. This sample was recounted at the analytical laboratory, and the values were consistent with the original sample. While no significant environmental impact stems from the observed uranium-235 value (0.003% of the DCG), it is highly unusual and is not corroborated with the Site 300 composite sample or seen in other months. Its cause is unknown; however, similar anomalous data have appeared in the past. The overall levels were essentially the same as those reported in previous years.

Table 5-6 shows the median concentration of tritiated water vapor that was observed at the new sampling location (PRIM) near Site 300. (See Data Supplement Table 5-19 for biweekly data.) The annual median concentration is 4.1×10^{-3} Bq/m^3 (1.1×10^{-13} Ci/m^3), or 0.0001% of the DCG.

Beryllium in Air

The detection frequency, median concentration, IQR, and maximum concentrations of airborne beryllium for the Site 300 sampling locations are shown in **Table 5-7**. (See Data Supplement Table 5-20 for monthly data.) The highest beryllium concentration of 32.3 pg/m^3 occurred in October at location TFIR. The median concentration for this location is 0.13% of the federal and state ambient concentration limit, which is 10,000 pg/m^3.

Environmental Impact

The environmental impacts from both radioactive and nonradioactive effluents are described in this section.

Radioactive Materials

LLNL operations involving radioactive materials had little impact on radionuclide concentrations in ambient air during 1999. Radionuclide concentrations in air at the Livermore site and in the Livermore Valley were well below the levels that would cause concern to the environment or public health according to existing regulatory standards.

The diffuse tritium sources at B292, B331, B514, and B624 had a localized effect; typically, tritium concentrations detected at the site perimeter or off site were not from diffuse sources.

The concentrations of radionuclides measured around Site 300 and in the City of Tracy were well below all standards and, except for uranium isotopes, reflected background or naturally occurring levels of these materials. (See Chapter 13, Radiological Dose Assessment, for a discussion of estimated dose from these data.) The uranium-235/uranium-238 ratios in August and September were less than the ratio of naturally occurring concentrations of these isotopes, which suggested the presence of depleted uranium in those Site 300 air samples. This depleted uranium resulted from current testing as substantiated by Site 300 explosive experiments during those months (Gallegos et al. 2000). Nevertheless, the detected levels remain far below regulatory standards.

Nonradioactive Materials

The concentrations of beryllium at both sites 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 $\mu g/m^3$ of particulates. Using a value of 50 $\mu g/m^3$ 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 annual medians for the Livermore site and Site 300 are 9.3 pg/m^3 and 8.6 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.