9

Soil and Sediment Monitoring

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Introduction

Soil is weathered material, mainly composed of disintegrated rock and organic material, that will sustain growing plants. Soil can contain pollutants originally released directly to the ground, to the air, or through liquid effluents. DOE guidance for environmental monitoring (U.S. Department of Energy 1991) states that soil should be sampled to determine if there is measurable long-term buildup of radionuclides in the terrestrial environment and to estimate environmental radionuclide inventories. The guidance recommends that radionuclides specific to a particular operation or facility as well as those that occur naturally should be monitored. Particulate radionuclides are of major interest in the LLNL soil monitoring program because airborne particulate releases are the most likely pathway for LLNL-induced soil contamination.

Sediments are defined, for the purposes of this chapter, as finely divided solid materials that have settled out of a liquid stream or standing water. To evaluate current conditions, LLNL samples recent sediments in storm drainage channels and the two arroyos on site. The accumulation of radioactive materials in sediment could lead to exposure of humans through ingestion of aquatic species, through sediment resuspension into drinking water supplies, or as an external radiation source (U.S. Department of Energy 1991). Note, however, that the LLNL Livermore site and Site 300 do not have habitats for aquatic species that are consumed by people, nor do they have surface drainage that directly feeds drinking water supplies. In addition, subsurface sediment sampling is conducted to support the LLNL Ground Water Protection Management Program (Chapter 8).

Since 1971, surface soil sampling in the vicinity of the LLNL Livermore site and Site 300 has been part of a continuing LLNL monitoring program designed to measure any changes in environmental levels of radioactivity and to evaluate any increase in radioactivity that might have resulted from LLNL operations. These samples have been analyzed for plutonium and gamma-emitting radionuclides, such as depleted uranium, which is used in some explosive tests at Site 300. The inclusion of other gamma-emitting naturally occurring nuclides (^{40}K and ^{232}Th) and the long-lived fission product ^{137}Cs provides background information and baseline data on global fallout from historical aboveground nuclear weapons testing.

Similarly, sediment samples have been collected from selected arroyos and other drainage areas at and around the Livermore site since 1988; these locations largely coincide with selected storm water sampling locations (see Chapter 7). The number of sediment sampling locations was reduced in 1994 to correspond to reductions in storm water sampling locations. In addition, in 1991, LLNL began analyzing surface soil samples for beryllium, a potentially toxic metal used at both the Livermore site and Site 300. However, analysis for beryllium was discontinued at the Livermore site in 1995, because beryllium was not ever measured above background values.

Location maps for soil and sediment sampling conducted during 1997 are provided in **Figures 9-1** through **9-3**. The locations were selected to represent background concentrations (distant locations unlikely to be affected by LLNL operations) as well as

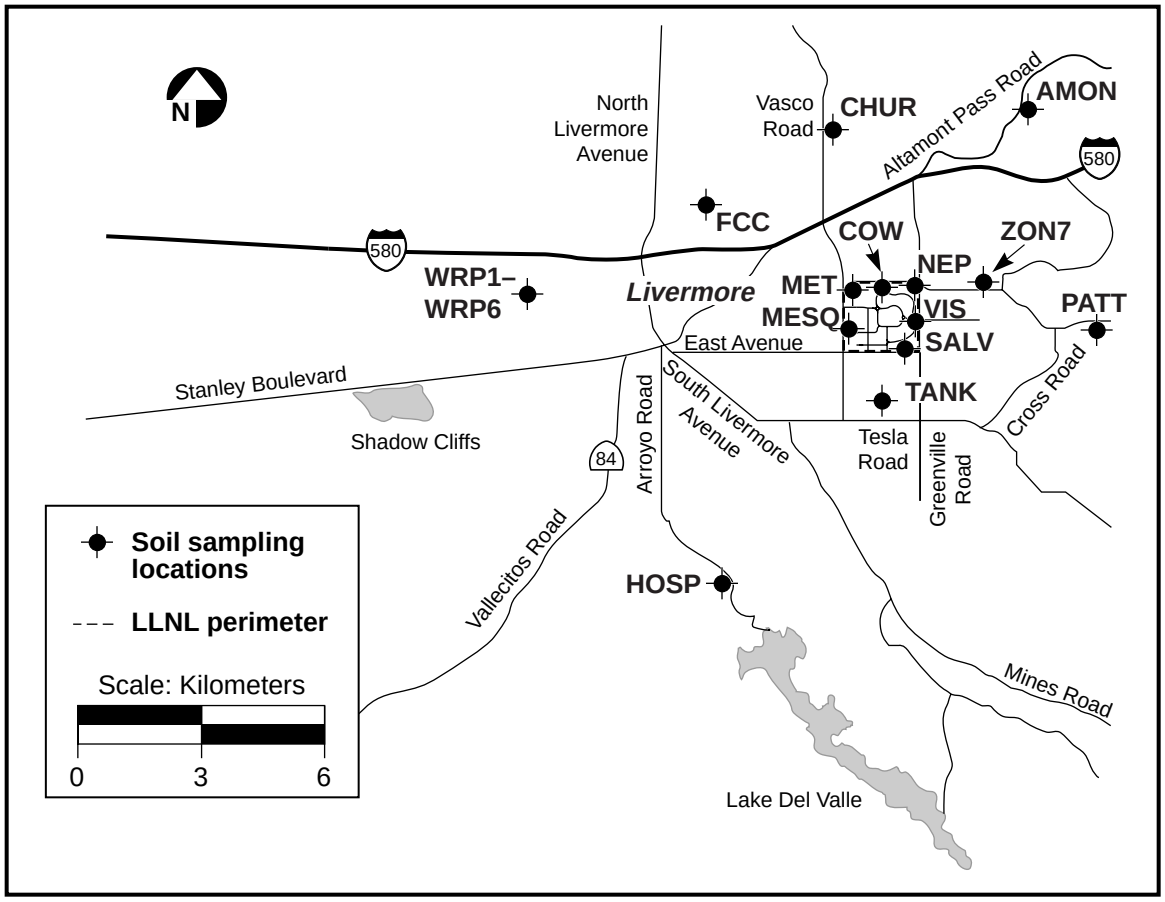

Figure 9-1. Soil sampling locations, Livermore Valley, 1997.

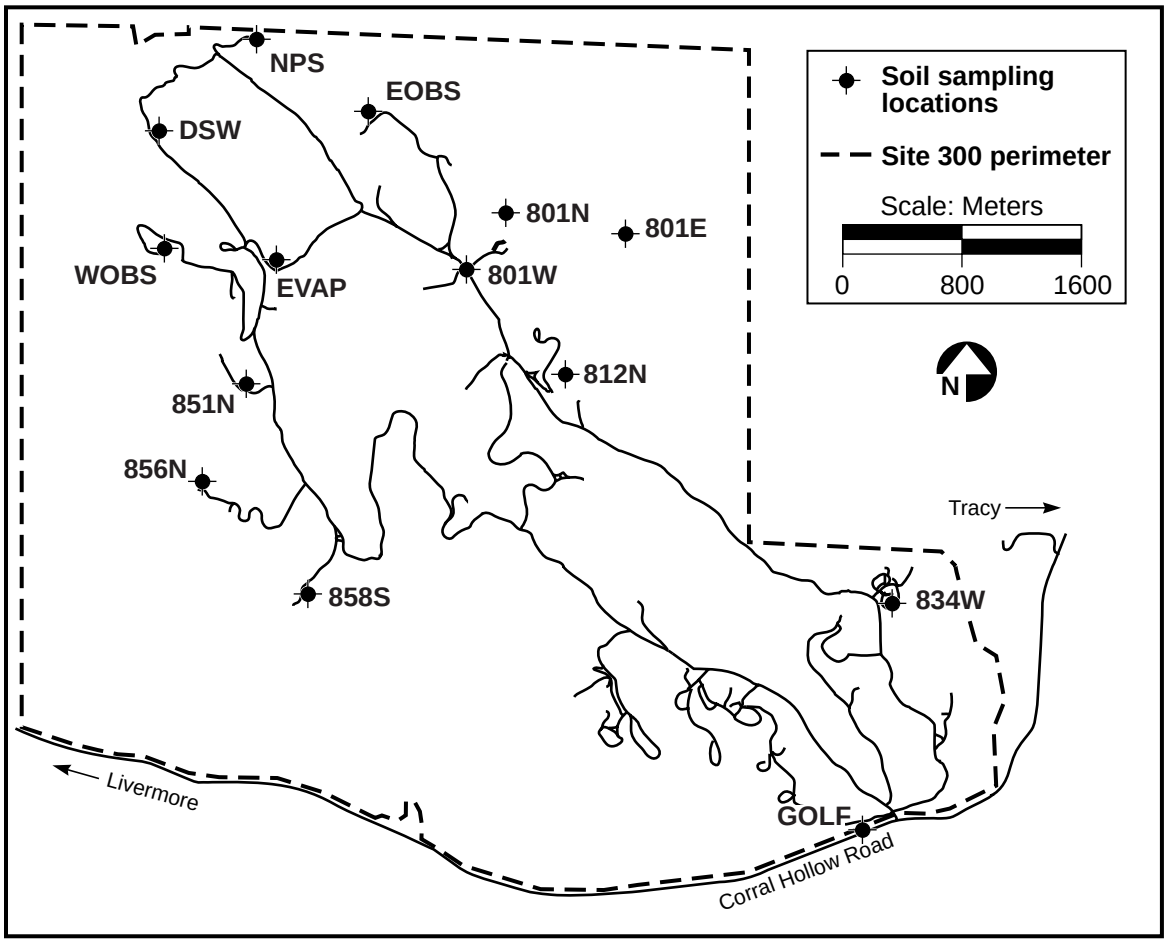

Figure 9-2. Site 300 soil sampling locations, 1997.

areas where there is the potential to be affected by LLNL operations. Areas with known contaminants, such as the Livermore Water Reclamation Plant (LWRP), are also sampled. In general, Site 300 soil sampling locations were established around firing tables and other areas of potential soil contamination. Arroyo and drainage channel sediment sampling locations were chosen to coincide with major Livermore site storm water drainages. All soil and sediment sampling locations have permanent location markers for reference.

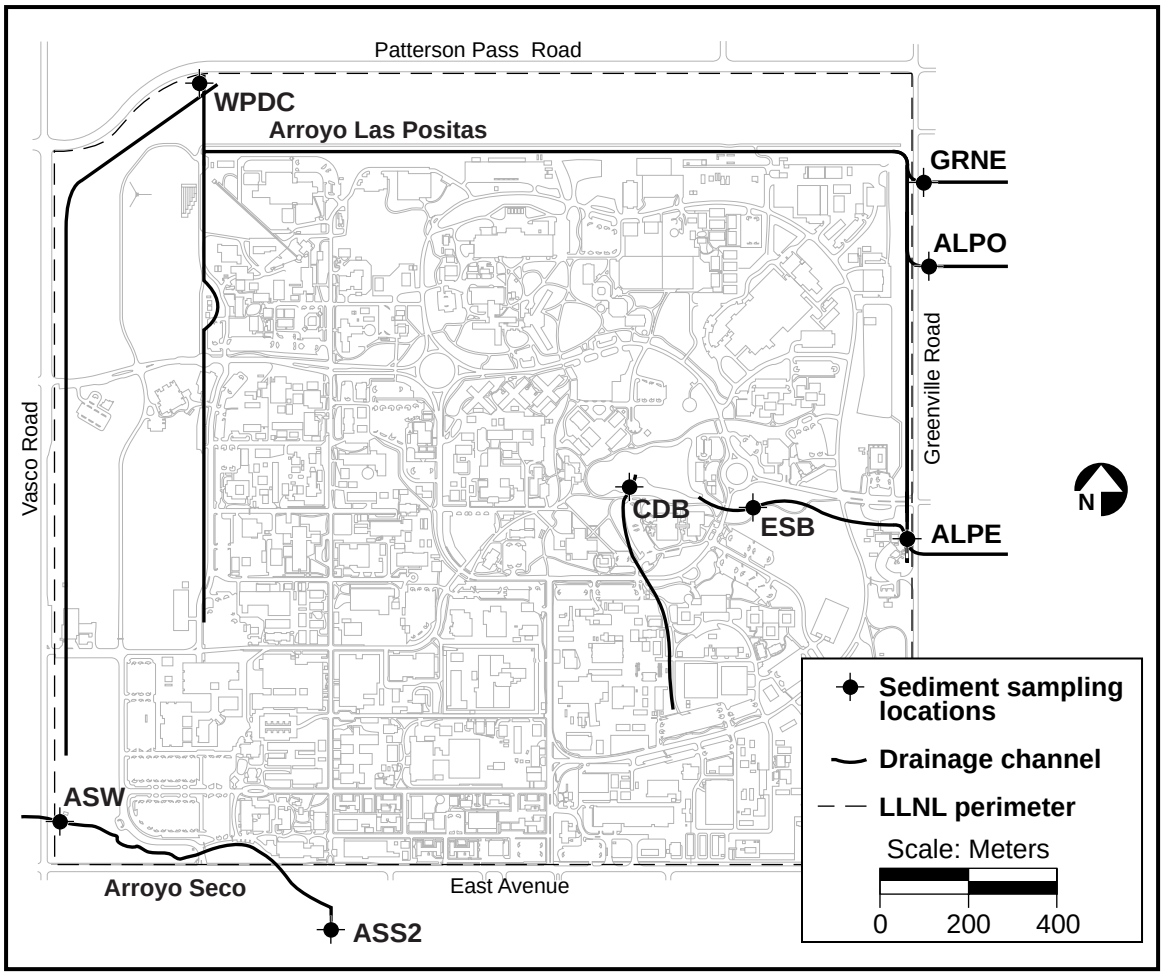

Figure 9-3. Arroyo and drainage basin sediment sampling locations, 1997.

Methods

Soil and sediment sampling is conducted according to written, standardized procedures (Tate et al. 1995). Soil samples are collected from undisturbed areas near the permanent sampling location marker. These areas generally are level, free of rocks, and are unsheltered by trees or buildings. All samples are collected from the top 5 cm of soil because surface deposition from the air is the primary pathway for potential contamination and resuspension of materials from the surface into the air is the primary exposure pathway to nearby human populations.

Soil sampling location CAFE was removed from the sampling program because the location did not meet the requirement of being unsheltered by trees or buildings; it also

was near a heavily travelled area. Soil sampling locations RRCH, ALTA, and ERCH were also removed from the sampling program due to problems with accessibiliy stemming from the private ownership of the property where the samples were taken. Soil sampling locations CHUR and AMON are replacement locations for RRCH and ALTA. Soil sampling location ERCH was not replaced; it was a background location, as was RRCH (which was replaced), and sufficient background samples are obtained from the other locations. Approximately 10% of samples are sampled in duplicate; two identical samples were collected at each location chosen for this sampling.

Samples of recent sediment are collected annually from drainages at and around the LLNL Livermore site after the cessation of spring runoff. Although added as a new sediment sampling location in 1997, ALPO was not sampled in 1997 because the location was constantly under water from releases upstream of the Livermore site. For 1997, samples at the Livermore site were analyzed for radionuclides, and samples for Site 300 were analyzed for gamma-emitting radionuclides and beryllium. Analysis of Site 300 soil samples for plutonium was discontinued in 1997 because plutonium has not been used at the site and sample results have continuously been at background levels since sampling was begun in 1972. During 1997, additional subsurface sediment sampling supported the LLNL Ground Water Protection Management Program (Chapter 8).

Soil and sediment samples are delivered to LLNL's Chemistry and Materials Science Environmental Services (CES) laboratory for analyses. Soil samples are dried, ground, sieved, and blended. The plutonium content of a sample aliquot is determined by alpha spectroscopy. Other sample aliquots (300 g) are analyzed for more than 150 radionuclides by gamma spectroscopy using a high-purity germanium (HPGe) detector (Hall and Edwards 1994a, b, and c). The 10 g subsamples for beryllium analyses are sent to a contract analytical laboratory and are analyzed by graphite-furnace atomic absorption spectroscopy. For sediment samples collected for tritium analyses, CES uses freeze-drying techniques to recover water from the samples and determines the tritium content of the water by liquid-scintillation counting. Chain-of-custody procedures are followed throughout the sampling, delivery, and analytical processes.

Livermore Valley Results

Table 9-1 presents summary data on the concentrations of $^{239+240}$Pu, ^{241}Am, ^{137}Cs, ^{40}K, ^{232}Th, ^{235}U, and ^{238}U in surface soils from the Livermore Valley sampling locations. The complete data for 1997 soil and sediment sampling is presented in Table 9-1 of the Data Supplement. The concentrations and distributions of all observed radionuclides

Table 9-1. Summary of soil and sediment analytical data, 1997.

Analyte and location	Detection frequency[a]	Median	IQR[b]	Maximum
^{238}Pu (μBq/dry g)				
Livermore Valley soils	10/13	6.8	9.7	38.5
LWRP[c] soils	6/6	222	183	389
Livermore site sediments	5/7	6.4	27.8	210
$^{239+240}$Pu (μBq/dry g)				
Livermore Valley soils	13/13	64	106	559
LWRP[c] soils	6/6	4000	2577	8070
Livermore site sediments	7/7	20	364	1930
^{137}Cs (10^{-3} Bq/dry g)				
Livermore Valley soils	12/13	1.98	1.59	5.55
LWRP soils	6/6	2.33	2.29	4.14
Livermore site sediments	6/7	0.37	0.57	1.24
Site 300 soils	14/14	2.10	2.21	7.25
^{40}K (Bq/dry g)				
Livermore Valley soils	13/13	0.488	0.122	0.596
LWRP soils	6/6	0.409	0.027	0.451
Livermore site sediments	7/7	0.451	0.033	0.503
Site 300 soils	14/14	0.437	0.075	0.607
^{232}Th (μg/dry g)[d]				
Livermore Valley soils	13/13	6.9	1.3	8.1
LWRP soils	6/6	6.9	0.5	7.2
Livermore site sediments	7/7	5.2	1.4	7.7
Site 300 soils	14/14	8.8	1.9	11.9
^{235}U (μg/dry g)[e]				
Livermore Valley soils	11/13	0.020	—[f]	0.024
LWRP soils	6/6	0.019	0.004	0.025
Livermore site sediments	5/7	0.018	—[f]	0.024
Site 300 soils[g]	16/16	0.025	0.009	0.185
^{238}U (μg/dry g)[h]				
Livermore Valley soils	13/13	2.1	0.5	3.1
LWRP soils	6/6	2.1	0.2	2.8
Livermore site sediments	7/7	2.0	0.5	2.3
Site 300 soils[g]	16/16	4.1	3.5	71.3

Table 9-1. Summary of soil and sediment analytical data, 1997 (concluded).

Analyte and location	Detection frequency[a]	Median	IQR[b]	Maximum
^{3}H (Bq/L extracted water)[i]				
Livermore site sediments	5/7	12.5	21.7	61.1
^{241}Am (10^{-3} Bq/dry g)[j]				
LWRP soils	2/6	<2.6	—[f]	5.3
Be (mg/kg)[k]				
Site 300 soils	14/14	1.2	0.5	5

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

b IQR = Interquartile range.

c LWRP = Livermore Water Reclamation Plant.

d Thorium-232 activities in Bq/dry g can be determined by dividing the weight in µg/dry g by 247.3, and pCi/dry g can be determined by dividing by 9.15.

e Uranium-235 activities in Bq/dry g can be determined by dividing the weight in µg/dry g by 12.5, and pCi/dry g can be determined by dividing by 0.463.

f Insufficient number of detections to calculate IQR. (See Site 300 results for discussion.)

g Includes results from reanalysis of original sample and analysis of resample.

h Uranium-238 activities in Bq/dry g can be determined by dividing the weight in µg/dry g by 80.3, and pCi/dry g can be determined by dividing by 2.97.

i Tritium (^{3}H) analysis is only conducted on sediment samples.

j Americium-241 is only detected in LWRP soil samples.

k Beryllium analysis is only conducted on soils sampled at Site 300; the analysis is a chemical, not a radiochemical analysis.

in soil for 1997 are within the ranges reported in previous years and generally reflect worldwide fallout and naturally occurring concentrations. The ratio of ^{235}U to ^{238}U generally reflects the natural ratio of 0.7%; however, there is uncertainty in the $^{235}U/^{238}U$ ratio because of the difficulty in measuring small quantities of ^{238}U by gamma spectroscopy.

Plutonium has, in the past, been detected at levels above background at ZON7, the off-site soils sampling location near the LLNL Livermore site and in the prevailing downwind direction. Because of the high level of variability inherent in the measurement of soils, we do not always find plutonium above background levels at this location. As in 1991, 1994, 1995, and 1996, $^{239+240}$Pu was detected at background levels—151 $\mu Bq/g$ (4.1×10^{-3} pCi/g)—at location ZON7 in 1997. Since 1973, soil samples in this area have generally shown $^{239+240}$Pu values that are higher than background. The slightly higher values near the Livermore site have been attributed to historic operations, which included the operation of solar evaporators for

plutonium-containing liquid waste in the southeast quadrant (Silver et al. 1974). LLNL no longer operates the solar evaporators or engages in any other open-air treatment of plutonium-containing waste. Nonetheless, $^{239+240}$Pu from historic operations is carried off site by resuspension of soil by wind. Similarly, elevated levels of $^{239+240}$Pu, resulting from an estimated 1.2×10^9 Bq (32 mCi) plutonium release to the sewer in 1967 and earlier releases, first observed in soils near LWRP during the early 1970s, again were detected at LWRP sampling locations. As in 1990 through 1992 and 1996, ^{241}Am was detected in LWRP samples; it is most likely caused by the natural decay of the trace concentrations of ^{241}Pu that were present in the release.

Historical plots of median $^{239+240}$Pu concentrations in soil in the Livermore Valley upwind and downwind of the center of the LLNL Livermore site, at Site 300, and at LWRP are shown in **Figure 9-4**. Livermore Valley upwind and Site 300 concentrations have remained relatively constant since monitoring began and generally are indicative

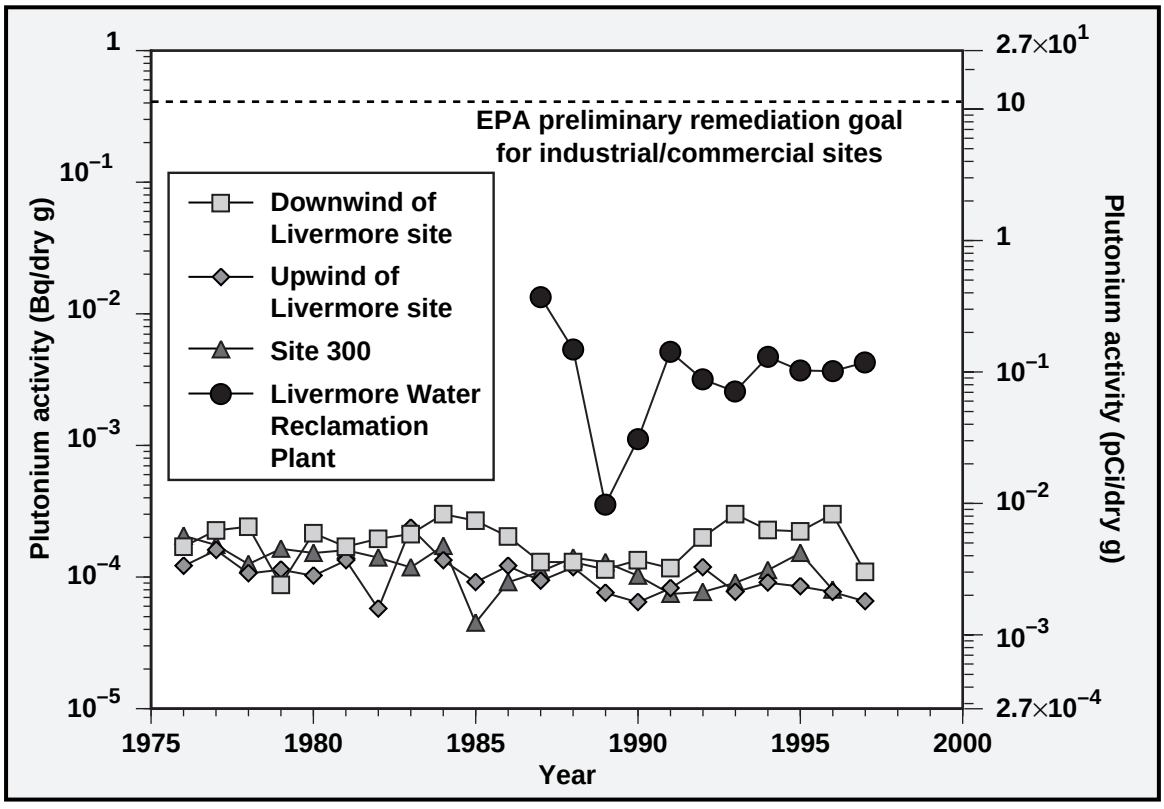

Figure 9-4. Median $^{239+240}$Pu activities in surface soils, 1976 to 1997. Upwind and downwind designations are relative to the center of the Livermore site.

of worldwide fallout. Increased variability can be noted in the downwind concentrations, which in 1997 included sampling locations VIS, PATT, NEP, COW, and ZON7; the concentrations of plutonium at these locations reflect resuspension of low-level plutonium contamination in soils in the southeast quadrant of the Livermore site. Greater variability in $^{239+240}$Pu is seen in samples from LWRP. However, only six samples are evaluated to determine the median at LWRP. In addition, the $^{239+240}$Pu is likely to be present in discrete particles, so the random presence or absence of the particles will dominate the measured $^{239+240}$Pu in any given sample.

Beryllium analysis for Livermore Valley soils was discontinued in 1995. The few LLNL operations that use beryllium are high-efficiency particulate air (HEPA) filtered. In addition, sampling data to date have shown no evidence of beryllium contamination in the Livermore Valley (Tate et al. 1995). Should beryllium usage change, LLNL's environmental monitoring staff would reevaluate the need for beryllium monitoring in soils.

Table 9-1 presents summary data on radionuclides detected in the sediment samples; a complete presentation of 1997 sediment data is found in Table 9-1 of the Data Supplement. The levels of $^{239+240}$Pu were generally at background concentrations, reflective of worldwide fallout. The moderately higher values at sampling locations (see **Figure 9-3**) CDB and ESB may be attributed to historic activities in the southeast quadrant at LLNL; these locations are both in drainages for that area. Most other radionuclides were detected at levels similar to those reported from 1988 through 1996: ^{137}Cs, a fission product, was found at worldwide background concentrations; and ^{40}K, ^{232}Th, ^{235}U, and ^{238}U—naturally occurring radionuclides—were detected at background concentrations. Tritium concentrations were within the range of previous data. The median tritium value for 1997, 12.5 Bq/L (338 pCi/L), is slightly higher than the median tritium value for 1996, 9.5 Bq/L (257 pCi/L). This slight increase can be explained by the increase in tritium emissions from the Tritium Facility (see Chapter 4, Air Monitoring). Tritium in sediments was evaluated for differences upwind and downwind of the Livermore site. A statistically significant difference was found using the Tukey-Kramer honestly significant difference (HSD) test, with the downwind sediment samples having higher measured concentrations than the upwind sediment samples. Tritium in sediments will continue to be evaluated.

Site 300 Results

Table 9-1 presents summary data on the concentrations of $^{239+240}$Pu, ^{40}K, ^{137}Cs, ^{232}Th, ^{235}U, and ^{238}U in soil from the Site 300 sampling locations; a complete presentation of

1997 soils data for Site 300 is found in Table 9-1 of the Data Supplement. The concentrations and distributions of all observed radionuclides in Site 300 soil for 1997 lie within the ranges reported in all years since monitoring began and, with the exceptions discussed below, reflect naturally occurring concentrations. The ratio of ^{235}U to ^{238}U generally reflects the natural ratio of 0.7%. Historical trends of ^{238}U concentrations from both the Livermore Valley and Site 300 are shown in **Figure 9-5**. Median values have remained relatively constant for both places. The highest values at Site 300 result from the use in past years of depleted uranium in high-explosive tests, and are generally found at sampling location 812N. The reader may notice that the plot of maximum values differs in this report as compared to similar plots in previous reports. In previous reports, the maximum values were erroneously plotted for the years 1976, 1978, 1980, 1983, 1987, 1989, 1991, and 1992. The cause of this error has not been determined. In all cases where an incorrect data point was used, the second highest data point was plotted.

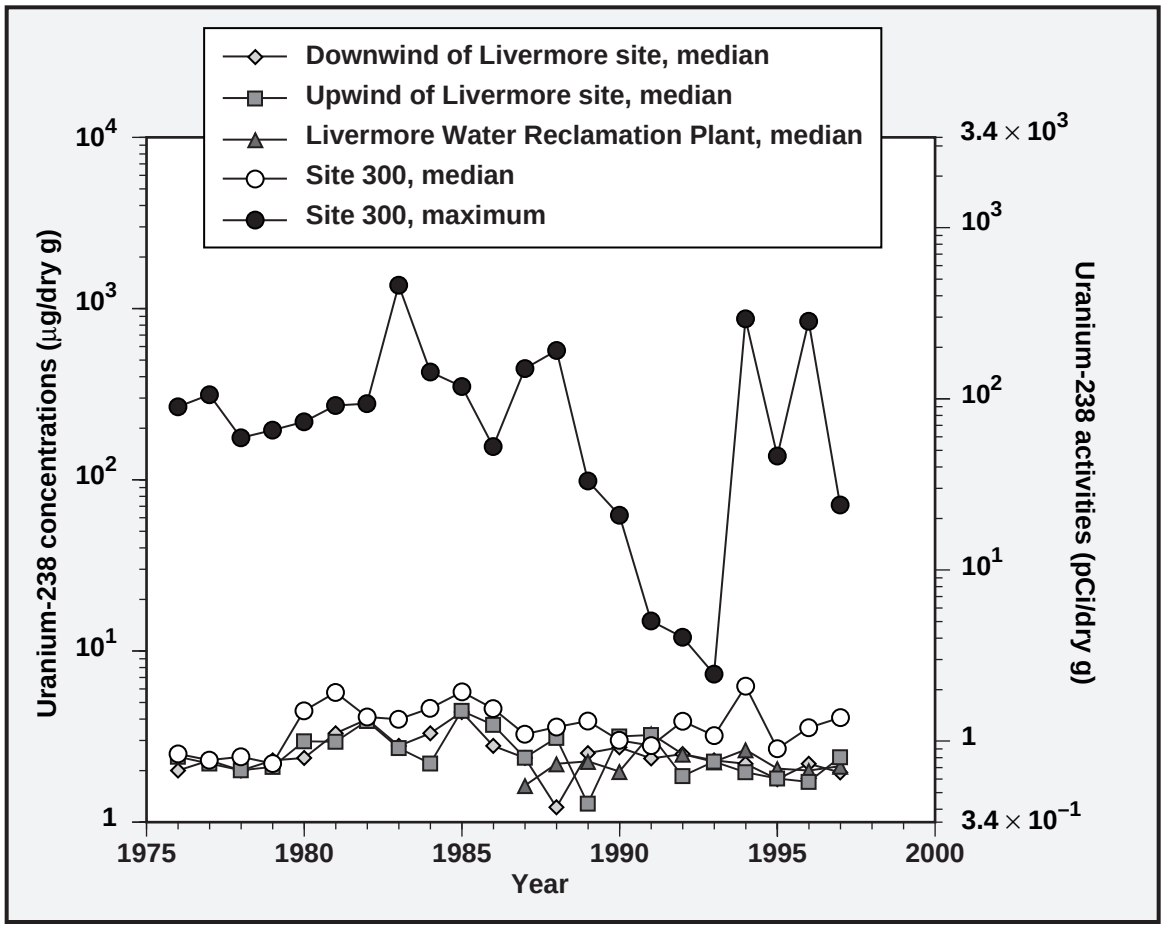

Figure 9-5. Uranium-238 concentrations in surface soils, 1976 to 1997.

During 1997, samples taken near firing tables 812N and 851N were found to contain ^{238}U at concentrations higher than background. The $^{235}U/^{238}U$ ratios in these samples were less than the ratio in naturally occurring uranium, indicating the presence of depleted uranium. Resampling and analysis of soils at 812N confirmed the presence of elevated concentrations of depleted uranium, whereas resampling and analysis of soils at the 851N did not confirm the presence of elevated concentrations of depleted uranium (see Table 9-1 in the Data Supplement). This disparity in results was not unexpected considering that the contamination is not uniformly dispersed over the soil.

Environmental Impact

This section discusses the environmental impacts at the LLNL Livermore site and Site 300 inferred from soil and sediment monitoring.

Livermore Site

Routine soil and sediment sample analyses indicate that the impact of LLNL operations on these media in 1997 has not changed from previous years and remains insignificant. Most analytes of interest or concern were detected at background concentrations, in trace amounts, or could not be measured above detection limits.

The highest value of 8.1 mBq/g (0.22 pCi/g) for $^{239+240}Pu$ measured at LWRP during 1997 represents 2.2% of the EPA preliminary remediation goal for commercial or industrial sites of 0.37 Bq/g (10 pCi/g) (U.S. Environmental Protection Agency 1991). Statistical analysis shows that all LWRP $^{239+240}Pu$ soils data are lognormally distributed, and at LWRP there is no general increase or decrease in $^{239+240}Pu$ values with time. Moreover, all measured concentrations, regardless of location and year, have been a small fraction of the EPA preliminary remediation goal, which is shown in **Figure 9-4** for comparison. Sampling of soils for radiological materials will continue on an annual basis.

Site 300

With the exception of elevated concentrations of ^{238}U at locations 812N and 851N, the concentrations of radionuclides and beryllium observed in soil samples collected at Site 300 are representative of background or naturally occurring levels. In 1988, contaminated gravel from the firing table at Building 812 was removed to on-site

landfills; however, elevated levels of ^{238}U are still measured at this location. The investigation planned as part of the Site 300 CERCLA restoration efforts will clarify the nature and extent of the contamination in this area. The firing table at Building 851N is an active firing table, and a small fraction of the operations at the firing table disperse depleted uranium.

Big Trees Park

During the 1993 U.S. EPA investigation of plutonium in soils present in the southeast quadrant of the LLNL Livermore site, U.S. EPA personnel collected a soil sample at Big Trees Park in Livermore to obtain a background sample. This soil sample showed plutonium at a concentration higher than what is expected from global fallout for this region. The park was resampled by the U.S. EPA, LLNL, and the California Department of Health Services (DHS) in 1995.

As reported in MacQueen (1995), samples from 13 of the 16 locations sampled at the park had plutonium concentrations consistent with background levels found throughout the Bay Area. These levels were 1/600 to 1/10,000 of the U.S. EPA's risk-based preliminary remediation goal for plutonium for residential areas of 0.09 Bq/g (2.5 pCi/g) (U.S. Environmental Protection Agency 1991). Background values were found in all sandboxes, school grounds, picnic areas, and under the large eucalyptus trees for which the park is named. Samples from two locations adjacent to the ballfield had plutonium concentrations slightly above background levels, but still 1% to 2% of the U.S. EPA's risk-based preliminary remediation goal for plutonium for residential areas.

Four samples taken in the area near the original U.S. EPA sample area had plutonium concentrations that were above the initial U.S. EPA sample concentration, but even the highest concentration was 40% of the U.S. EPA's risk-based preliminary remediation goal for plutonium for residential areas. Both the U.S. EPA and the California DHS concur that there is no regulatory concern from any of the sample results, that there is no significant lifetime cancer risk resulting from the low concentrations of $^{239+240}Pu$ in the soil samples, and that there is no unacceptable risk to human health or the environment.

In 1997, the Agency for Toxic Substances Disease Registry (ATSDR), which had contracted with California DHS to conduct a health consultation for plutonium, held a public meeting on the subject of plutonium at Big Trees Park. At this meeting, the agencies restated that although the levels of plutonium at Big Trees Park were not a health concern, they were interested in knowing how the plutonium got to the park, and

that this question warranted further investigation. The report issued by ATSDR on this subject was issued in draft in 1998 (see Chapter 3).

The process for obtaining additional samples to evaluate the potential pathways for plutonium to be present at the park is currently underway. It is anticipated that additional sampling will be completed in 1998. Over the years, LLNL has frequently investigated the presence of radionuclides in local soils. Several of the studies are listed in **Table 9-2.**

Table 9-2. Special soil studies.

Year	Subject	Reference
1971–1972	Radionuclides in Livermore Valley soil	Gudiksen et al. 1972; Gudiksen et al. 1973
1973	Radionuclides in San Joaquin Valley soil	Silver et al. 1974
1974	Soil study of southeast quadrant of Livermore site	Silver et al. 1975
1977	Sediments from LLNL to the San Francisco Bay	Silver et al. 1978
1980	Plutonium in soils downwind of the Livermore site	Toy et al. 1981
1990	195 samples taken in southeast quadrant for study	Gallegos et al. 1992
1991	Drainage channels and storm drains studied	Gallegos et al. 1991
1993	EPA studies southeast quadrant	Gallegos et al. 1994
1993	Historic data reviewed	Gallegos 1993
1995	LLNL, EPA, and DHS sample soils at Big Trees Park	MacQueen 1995