10. Soil and Sediment Monitoring

Gretchen M. Gallegos
Donald H. MacQueen

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 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, surface and subsurface sediment sampling helps support the goal of the LLNL Ground Water Protection Management Program (Chapter 9).

Since 1971, surface soil sampling in the vicinity of the 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 high-explosives tests at Site 300. The inclusion of other gamma-emitting naturally occurring nuclides (^{40}K, ^{232}Th, and ^{235}U) and the long-lived fission product ^{137}Cs provides background information and baseline data on global fallout from historical above-ground 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 1995 are provided in **Figures 10-1** through **10-3**. The locations were selected to represent background concentrations (distant locations unlikely to be affected by LLNL operations) as well as 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

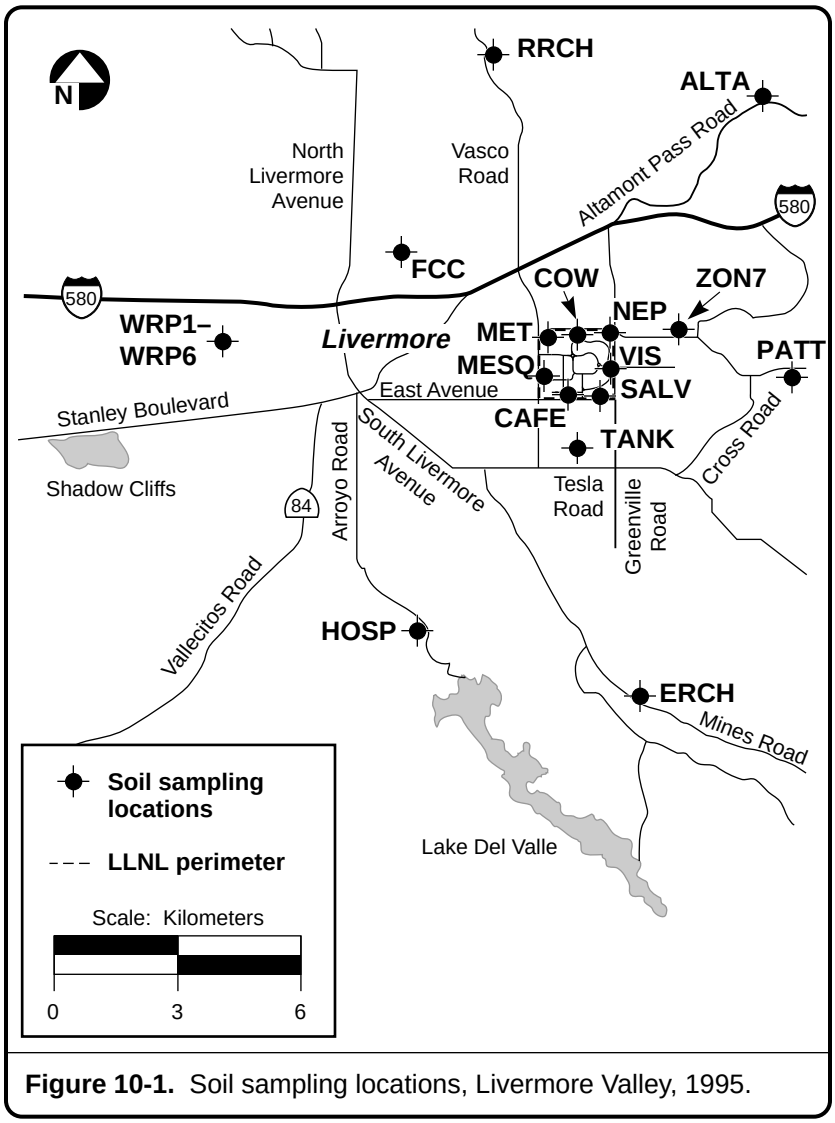

Figure 10-1. Soil sampling locations, Livermore Valley, 1995.

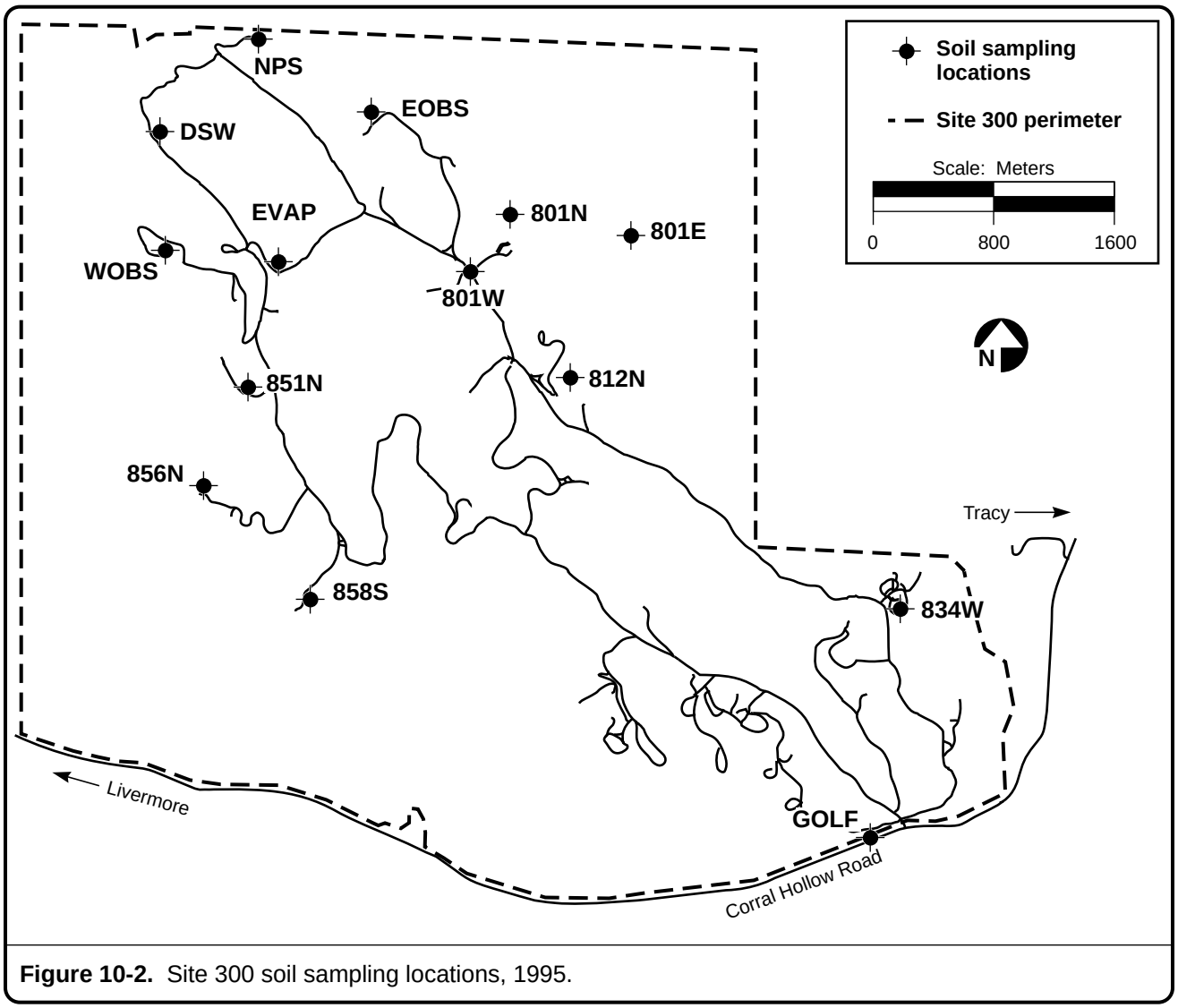

Figure 10-2. Site 300 soil sampling locations, 1995.

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.

Methods

Soil and sediment sampling is conducted according to written, standardized procedures (Tate et al. 1995, Appendix A). 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

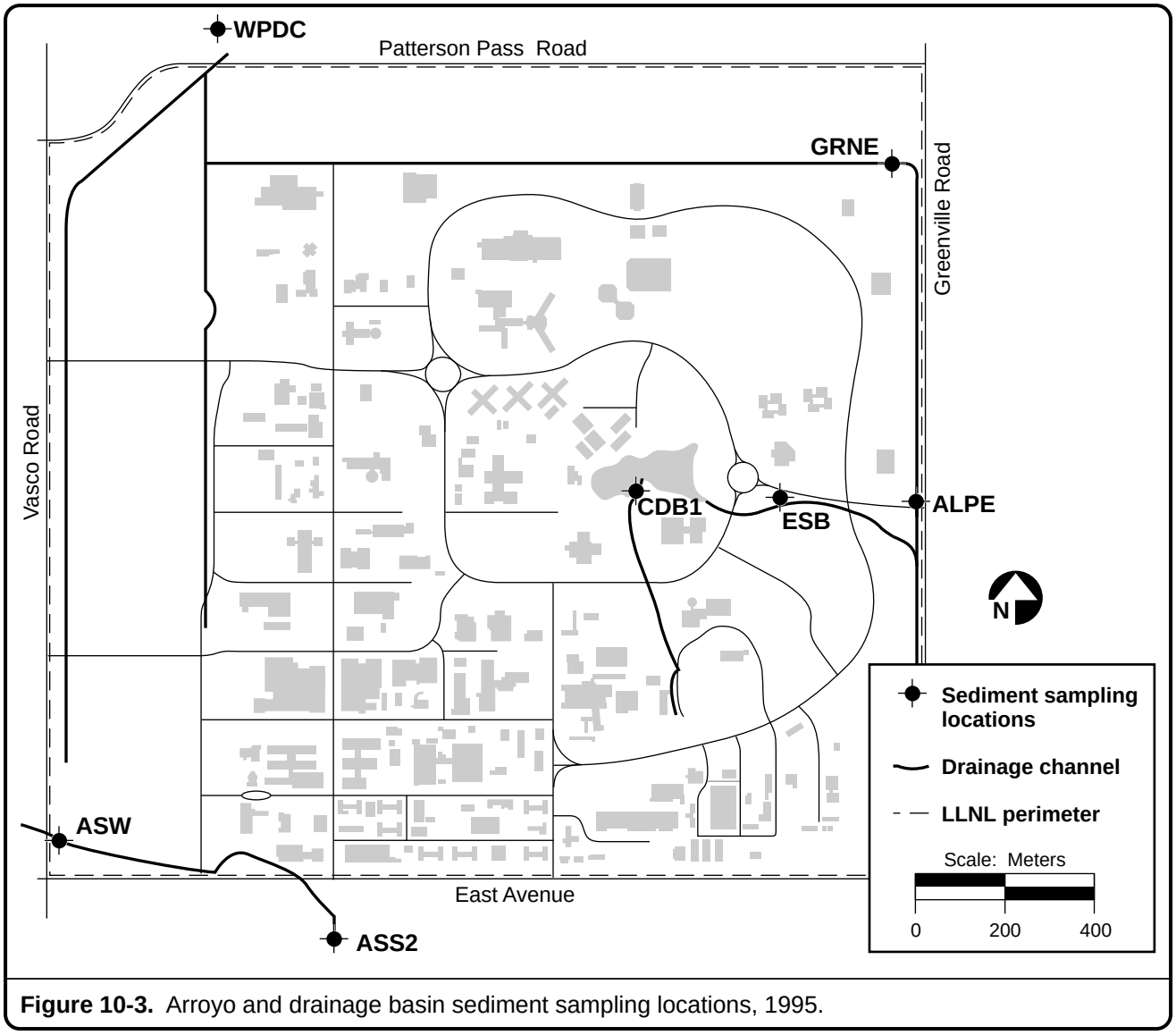

Figure 10-3. Arroyo and drainage basin sediment sampling locations, 1995.

the air is the primary pathway for potential contamination. Quality control duplicate samples are submitted with each batch of soil samples. At locations chosen for this sampling, two identical samples are collected.

Samples of recent sediment are collected annually from drainages at and around the Livermore site after the cessation of spring runoff. For 1995, samples at the Livermore site were analyzed for radionuclides and samples for Site 300 were analyzed for radionuclides and beryllium. During 1996, additional subsurface sediment sampling will support the LLNL Ground Water Protection Management Program (Chapter 9).

Soils and sediment samples are delivered to LLNL's Chemistry and Materials Science's 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 (Hall and Edwards 1994). 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 1994). The 10-g subsamples for beryllium analyses are sent to a contract analytical laboratory and are analyzed by graphite-furnace atomic absorption spectroscopy. For 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 10-1 presents summary data on the concentrations of $^{239+240}$Pu, ^{40}K, ^{60}Co, ^{137}Cs, ^{232}Th, ^{235}U, and ^{238}U in surface soils from the Livermore Valley sampling locations. The complete data for 1995 soils and sediment sampling is presented in Table 10-1, Volume 2, of this report. The concentrations and distributions of all observed radionuclides in soil for 1995 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 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 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 and 1994, $^{239+240}$Pu was detected at background levels—0.22×10^{-3} Bq / g (6.1×10^{-3} pCi / g)—at location ZON7. 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 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 first observed in soils near LWRP during the early 1970s, again were detected at LWRP sampling locations. As in 1990 through 1992, ^{241}Am was detected in an LWRP sample; it is most likely caused by the natural decay of the trace concentrations of ^{241}Pu that were present in the release.

Table 10-1. Summary of soils and sediment analytical data, 1995.

Analyte and location	Detection frequency	Median	IQR[a]	Maximum
$^{239+240}$Pu (10^{-3} Bq/dry g)				
Livermore Valley soils	15/15	0.09	0.19	1.1
LWRP[b] soils	6/6	3.5	6.4	25
Livermore site sediments	7/7	0.07	1.2	3.3
Site 300 soils	14/14	0.15	0.12	0.20
^{137}Cs (10^{-3} Bq/dry g)				
Livermore Valley soils	15/15	2.6	3.1	8.1
LWRP soils	6/6	1.8	4.5	7.4
Livermore site sediments	6/7	0.7	0.5	1.4
Site 300 soils	14/14	5.4	4.8	7.4
^{40}K (Bq/dry g)				
Livermore Valley soils	15/15	0.474	0.089	0.640
LWRP soils	6/6	0.386	0.065	0.459
Livermore site sediments	7/7	0.488	0.011	0.529
Site 300 soils	14/14	0.477	0.156	0.629
^{232}Th (μg/dry g)[c]				
Livermore Valley soils	15/15	6.2	1.3	10
LWRP soils	6/6	6.1	1.3	7.2
Livermore site sediments	7/7	5.4	2.7	8.2
Site 300 soils	14/14	9.4	1.3	13
^{235}U (μg/dry g)[d]				
Livermore Valley soils	14/15	0.021	0.005	0.025
LWRP soils	6/6	0.022	0.004	0.029
Livermore site sediments	5/7	0.019	—[e]	0.024
Site 300 soils[f]	13/16	<0.025	—[e]	0.34
^{238}U (μg/dry g)[g]				
Livermore Valley soils	14/15	1.8	0.3	2.8
LWRP soils	6/6	2.1	0.5	2.9
Livermore site sediments	6/7	1.6	0.4	3.0
Site 300 soils[f]	14/16	<2.7	—[e]	140.0
^{3}H (Bq/L extracted water)[h]				
Livermore site sediments	7/7	6.3	6.3	20
^{241}Am (10^{-3} Bq/dry g)[i])				
LWRP soils	1/6	<2	—[e]	8

...concluded on next page

Table 10-1. Summary of soils and sediment analytical data, 1995 (concluded).

Analyte and location	Detection frequency	Median	IQR[a]	Maximum
^{60}Co (10^{-3} Bq/dry g)[i]				
LWRP soils	2/6	<0.1	—[e]	0.5
Be (mg/kg)[j]				
Site 300 soils[f]	16/16	0.9	0.5	5.7

Note: Detection frequency means the number of samples of all samples taken having measured values above the detection limit.

a IQR = interquartile range.

b LWRP = Livermore Water Reclamation Plant.

c 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.

d 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.

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

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

g 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.

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

i Cobalt-60 and Americium-241 are only detected in LWRP soil samples.

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

Historical plots of average $^{239+240}$Pu concentrations in soil in the Livermore Valley, at Site 300, and at LWRP are shown in **Figure 10-4**. Livermore Valley and Site 300 concentrations have remained relatively constant over the past 10 years and generally are indicative of worldwide fallout (locations on site and ZON7 show activities greater than background). Greater variability in $^{239+240}$Pu is seen at LWRP. However, six samples are evaluated to determine the median at LWRP. Moreover, 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.

Low levels of ^{60}Co were detected at the LWRP. While ^{60}Co is in use at the Livermore site, it is only present in gram quantities in three facilities (Buildings 151, 194, and 514) or in sealed sources. Low levels of ^{60}Co, on the order of 0.0037 Bq/g (0.1 pCi/g), have also been detected intermittently in sewage sludge samples. If the Livermore site were the source of ^{60}Co, this activity of ^{60}Co in the sludge would translate into about 1.5×10^{-6} Bq/mL (40×10^{-6} pCi/mL0 in the effluent leaving the site, which is below the detection limits of current analytical methods. This level is also well below the DOE effluent limit of 0.925 Bq/mL (25 pCi/mL). The reader should note that LLNL is

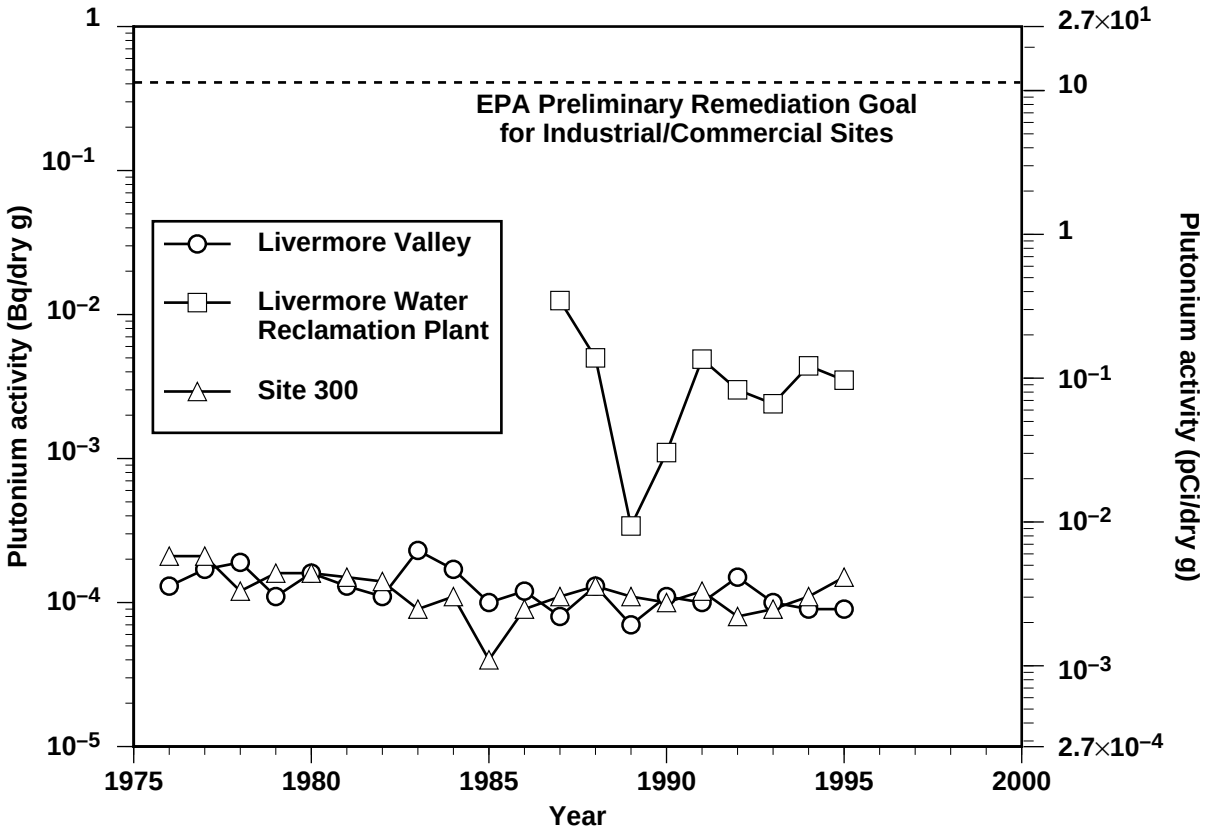

Figure 10-4. Median $^{239+240}$Pu activities in surface soils, 1976 to 1995.

not the only contributor to the waste stream that arrives at the LWRP and that ^{60}Co is used in a variety of medical, technical, and research applications. It is not possible to determine if LLNL is the source of ^{60}Co at LWRP. However, it can be concluded that LLNL controls on the release of ^{60}Co are sufficient to ensure that LLNL activities do not adversely affect LWRP applications.

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 10-1 presents summary data on radionuclides detected in the sediment samples; a complete presentation of 1995 sediment data is found in Table 10-1, Volume 2, of this report. The levels of $^{239+240}$Pu were generally at background concentrations, reflective of worldwide fallout. The slightly higher values at

ALPE 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 1994: ^{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 below those reported from 1988 through 1992, but above those for 1993 and 1994. Median tritium values are shown in **Figure 10-5** and show a general decline since measurement began. In 1993, the sediment sampling procedure was revised so that samples for gamma analysis were collected at the surface (5 cm deep). However, the depth for taking samples for tritium analysis was retained at 15 cm. Moreover, sampling was not later in the year than usual, so there would not have been additional evaporative losses due to sampling delays. Tritium in sediments will continue to be evaluated.

Site 300 Results

Table 10-1 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 1995 soils data for Site 300 is found in Table 10-1, Volume 2, of this report. The concentrations and distributions of all observed radionuclides in

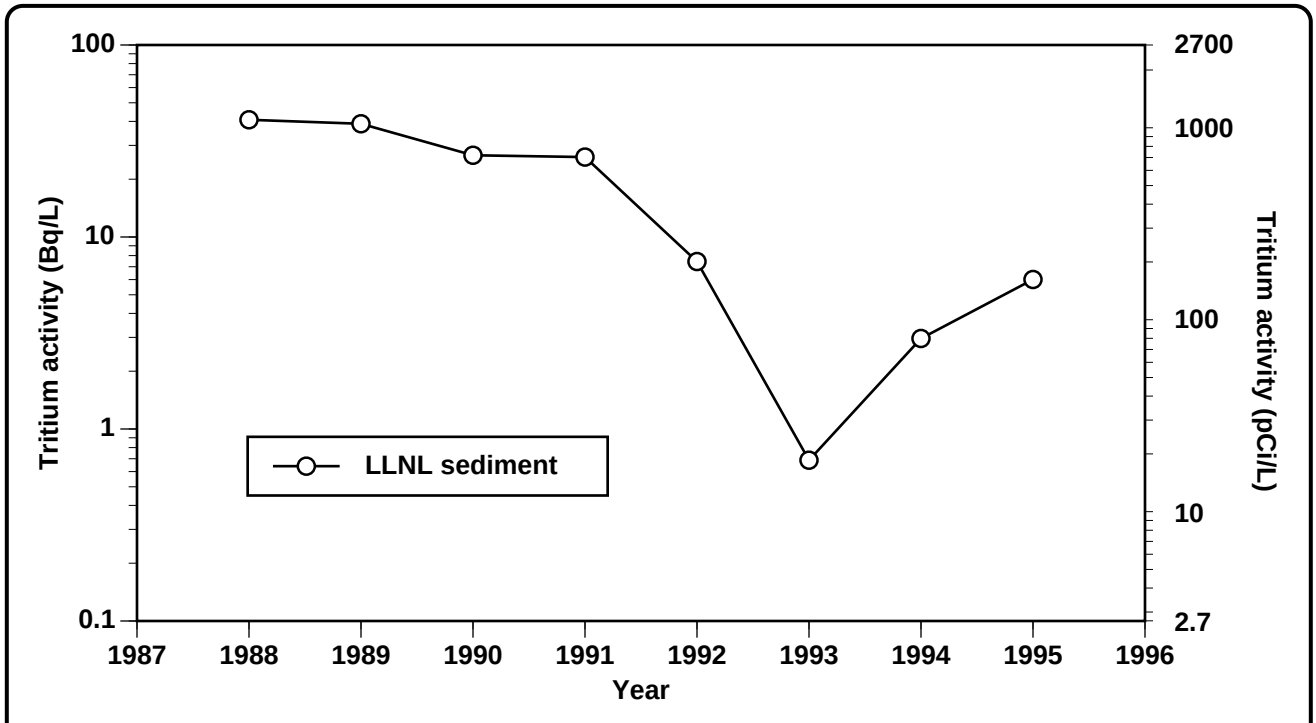

Figure 10-5. Median tritium concentrations in sediments (Bq/L of recovered water), 1988 to 1995.

Site 300 soil for 1995 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 10-6**. Median values have remained relatively constant for both places. The highest values at Site 300 are caused by the use of depleted uranium in high-explosive tests.

During 1995, one sample from a region near a firing table (834W) had substantially higher than background concentrations of ^{238}U and beryllium. The $^{235}U / ^{238}U$ ratios, at 0.2%, confirm the presence of depleted uranium; the ratio in naturally occurring material is 0.7%. To investigate the elevated ^{238}U and beryllium result at 834W, LLNL personnel reanalyzed the original soil sample and resampled and analyzed the original sampling location. The high value of 136 μg / g of ^{238}U in the original sample was confirmed by reanalysis. However,

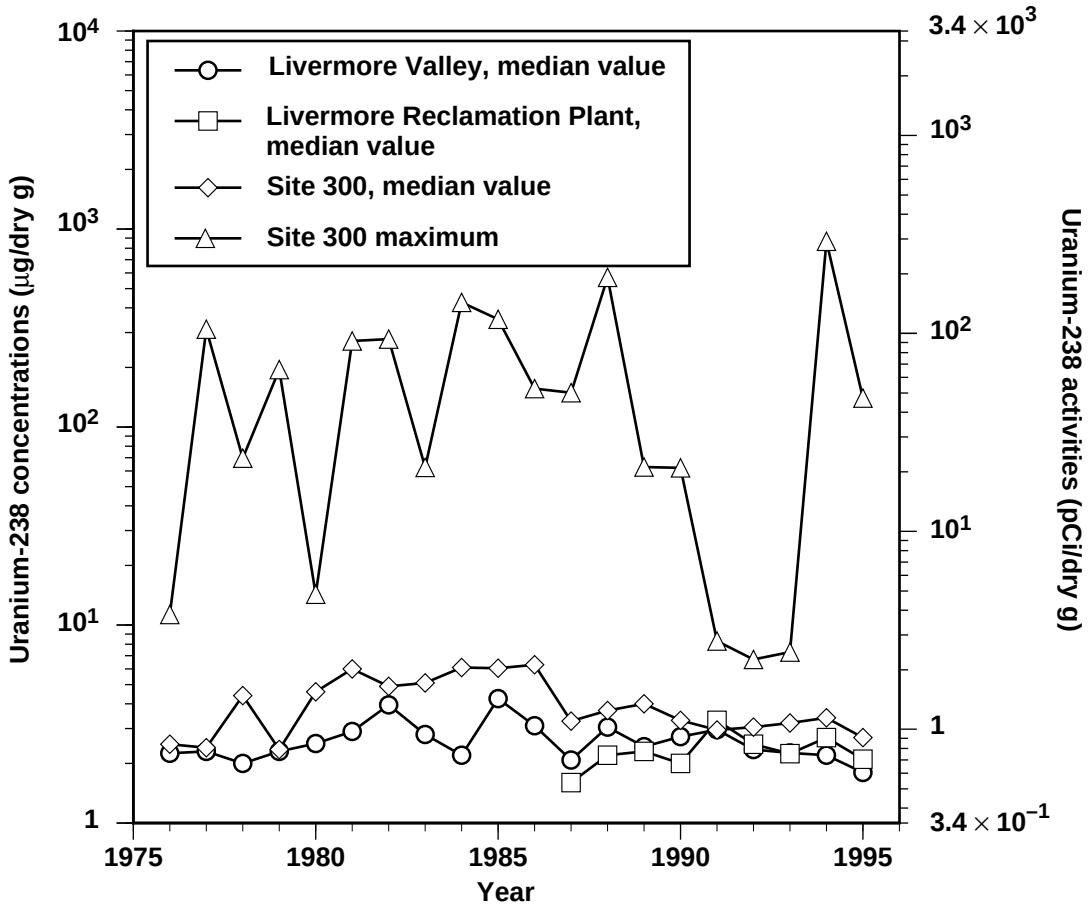

Figure 10-6. Uranium-238 concentrations in surface soils, 1976 to 1995.

the result of the resample of the location yielded <2.1 μg/g of ^{238}U, well below the original sample value; this disparity in sampling results was to be expected considering the heterogeneous nature of the contamination. In contrast, the finding of beryllium above background was not confirmed by reanalysis. An additional sampling location, 801N, yielded results having a $^{235}U/^{238}U$ ratio of 0.2%. This sampling location is near an active test area, so it is expected that results might show the presence of depleted uranium.

Environmental Impact

This section discusses the environmental impacts at the 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 1995 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 25×10^{-3} Bq/g (0.68 pCi/g) for $^{239+240}Pu$ measured at LWRP during 1995 represents 6.8% of the EPA preliminary remediation goal for commercial or industrial sites of 0.37 Bq/g (10 pCi/g) (EPA 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 proposed EPA remediation goal, which is shown in **Figure 10-4** 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 834W and 801N, and beryllium at location 834W, 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, and measured values for samples from this location have generally not exhibited elevated levels of ^{238}U and beryllium. The elevated results for ^{238}U and beryllium indicate that areas outside the firing table may be contaminated by firing table debris. The investigation planned as part of the Site 300 CERCLA restoration efforts will clarify the nature and extent of the contamination in this area.

10. Soil and Sediment Monitoring

Special Studies

In 1995, LLNL conducted an on-site soil cleanup action in response to the results from a special study of soil: Plutonium in the Soil in the Southeast Quadrant of Livermore Site. LLNL also completed its study of plutonium in soil at Big Trees Park, Livermore, CA.

Plutonium in Soil, Southeast Quadrant of Livermore Site

From 1962 to 1976, solar evaporation trays were located in the southeast quadrant of LLNL. The trays were approximately 6 m × 6 m × 3 m deep, constructed of concrete coated with polyamide epoxy paint and lined with polyvinylchloride or polyethylene liners. Plutonium-containing liquid waste was put in these trays to reduce by evaporation the total volume of disposable waste (Buerer 1983).

In 1991, in response to a Tiger Team comment, 195 surface soil samples from the southeast quadrant of LLNL were collected and analyzed for plutonium. The highest level detected was 0.11 Bq/g (3 pCi/g). In 1993, EPA decided to resample the areas with levels above those expected from global fallout for further confirmation of LLNL's sampling results and to sample locations to the west of the 1991 sampling locations to assure that the boundary of the area of interest had been appropriately set. Only one location, designated LL01-064, was identified in the EPA's 1993 sampling as containing more than the EPA industrial preliminary remediation goal (PRG) of 0.37 Bq/g (10 pCi/g); the location contained up to 11.5 pCi/g of $^{239/240}$Pu.

The soil containing plutonium at the location LL-01-064 near the northwest corner of T5475 was excavated on October 14, 1995, by LLNL personnel after appropriate safety reviews. Approximately 0.45 m^3 (1.5 m^2 area × 0.3 m deep) of soil was removed. Nine samples and one duplicate were collected after the soil was excavated to verify the removal of the soil over the PRG. All nine sample locations contained concentrations well below the industrial preliminary remediation goal.

The excavated area was backfilled with clean fill and seeded with new grass. The excavated soil was placed in four 55-gallon drums and collected for disposal in accordance with the appropriate regulations. This action complies with current EPA direction for the cleanup of soils.

Plutonium in Soil, Big Trees Park, Livermore

During the 1993 EPA investigation of plutonium in soils in the southeast quadrant of the Livermore site, 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 EPA, LLNL, and the California Department of Health Services (DHS) in 1995.

As reported in MacQueen (1995), samples from 13 of 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 EPA's risk-based preliminary remediation goal for plutonium for residential areas 0.09 Bq/L (2.5 pCi/g) (EPA 1991). Background values were found in all sandboxes, school grounds, picnic areas, and under the large eucalyptus trees for which the park was named. Samples from two locations adjacent to the ballfield had plutonium concentrations slightly above background levels, but still 2/100 to 1/100 of the EPA's risk-based preliminary remediation goal for plutonium for residential areas.

Four samples taken in the area near the original EPA sample area had plutonium concentrations that were above the EPA's initial sample's concentration, but even the highest concentration detection was two-fifths of the EPA's risk-based preliminary remediation goal for plutonium for residential areas. These data confirm the initial EPA result for this small area of soil found under trees along the fence separating the Arroyo Seco from the park. The EPA and California Department of Health Services (DHS) analytical results agree with those from LLNL. Both agencies 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}Pu in the soil samples, and that there is no unacceptable risk to human health or the environment. For more detailed information, see the report *Livermore Big Trees Park, January 1995 Soil Survey Results* (MacQueen 1995).