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Livermore Big Trees Park: 1998 Summary Results

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1. Introduction

This report summarizes work conducted in 1998 by the Lawrence Livermore National Laboratory (LLNL) to determine the extent and origin of plutonium at concentrations above background levels at Big Trees Park in the city of Livermore. This summary includes the project background and sections that explain the sampling, radiochemical and data analysis, and data interpretation. This report is a summary report only and is not intended as a rigorous technical or statistical analysis of the data.

1.1. Background

Big Trees Park is a 4.23-acre public park in the city of Livermore located about one-half mile west of LLNL (see Figure 1). Plutonium was discovered at higher-than-expected concentrations in Big Trees Park in 1993 during a U.S. Environmental Protection Agency (EPA) check of background plutonium values in the vicinity of LLNL. In 1995, LLNL, in collaboration with EPA, state regulators, and the public, collected additional soil samples from Big Trees Park to verify the 1993 finding and evaluate any potential hazards to the public.

After this 1995 sampling, the EPA and state regulators concluded that the plutonium in soil at Big Trees Park was below the residential preliminary remediation goal (PRG; 2.5 picocuries/gram), presented no health hazard, and required no further action.

Early in 1998, a health consultation prepared by the California Department of Health Services-Environmental Health Investigations Branch (CDHS-EHIB), under contract with the federal Agency for Toxic Substances and Disease Registry (ATSDR), suggested further sampling. LLNL volunteered to conduct additional sampling and analysis and to work with the regulatory agencies to ensure public concerns were met. In August and September 1998, additional soil samples were collected at Big Trees Park.

The 1998 sampling project addressed Recommendation 1 of the health consultation by determining the vertical and lateral extent of plutonium-239+240 in the soil. It also evaluated the likelihood of each of three potential pathways—water-borne/arroyo distribution, plutonium-contaminated sewage sludge used as a soil amendment, and aerial distribution—proposed in the health consultation as ways plutonium could have reached the park. Finally, special sampling was carried out at (1) the disked area, which is an area annually disked for weed abatement and an area of special concern to some local residents; (2) the playing field adjacent to the school; (3) the eastern extension of

Big Trees Park; and (4) the locations previously identified in 1995 as exhibiting concentrations of plutonium in soil above background levels.

A chronology of events, including the development of Big Trees Park and the various sampling efforts at the park, is presented in Table 1.

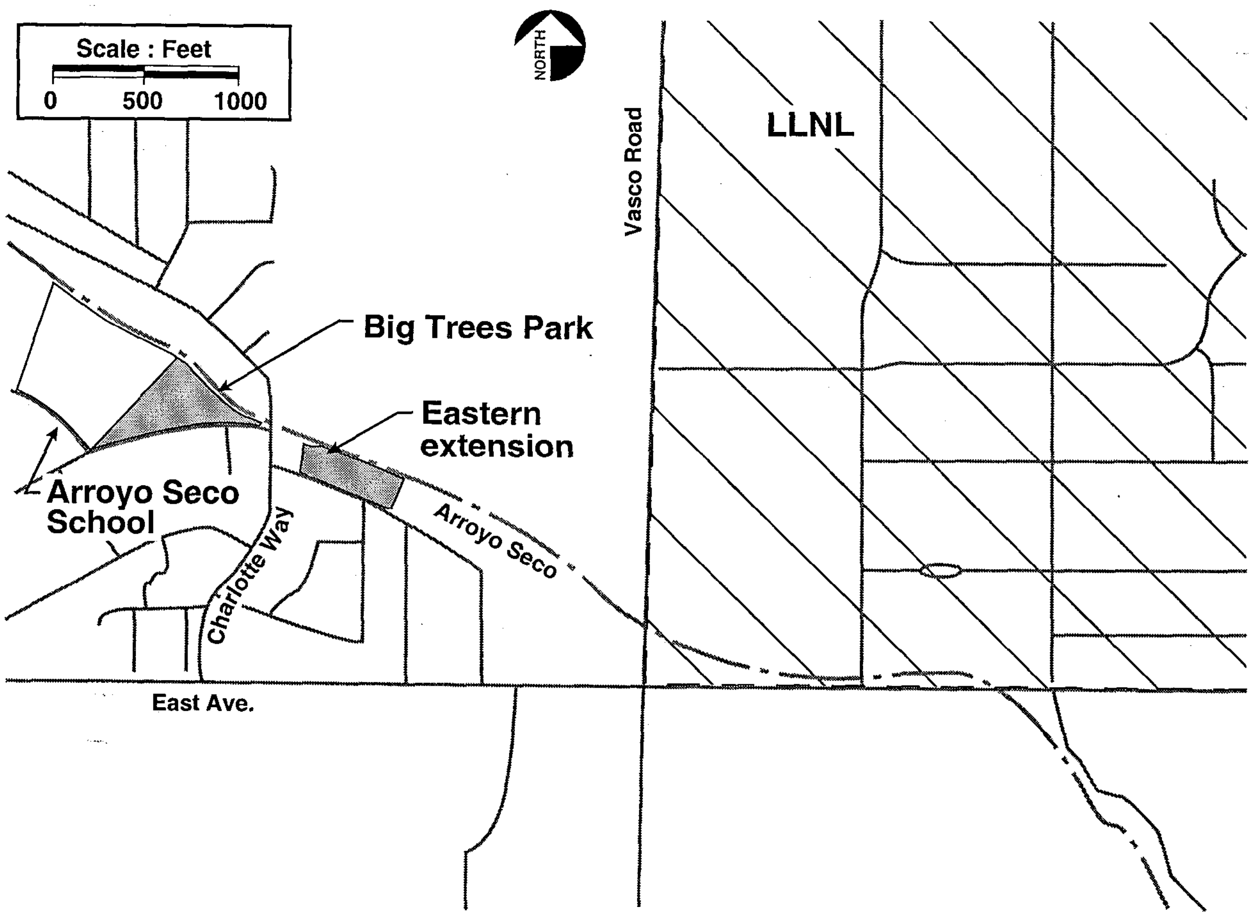

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Figure 1. Big Trees Park and its location with respect to LLNL, Arroyo Seco, and Arroyo Seco School.

Table 1. Chronology of events at Big Trees Park.

Date	Activity	Source
July 17, 1969	Property, which becomes Big Trees Park, deeded to City of Livermore.	County Tax Assessor's map and Metroscan[a]
1970	New Arroyo Seco channel excavated and concrete lined from just east of Charlotte Way to connect the already concrete-lined arroyo west of Big Trees Park.	Kaufman & Broad 1969, 1970
1970	Excavation of new concrete-lined arroyo generates approximately 9500 cubic yards of excess soil.	R. M. Galloway & Associates, 1970
1970	Excess soil generated from preparing roadbeds for street construction and lots for foundations was stockpiled in the area that would become the park.	R. M. Galloway & Associates, 1970
1969–1970	Subdivision graded per city specifications.	Kaufman & Broad 1969, 1970
Mar 23, 1971	Concrete-lined portion of arroyo deeded to Alameda County Flood Control and Water Conservation District Zone 7.	County Tax Assessor's map and Metroscan
April and June 1971	Big Trees Park constructed per the terms of the LARPD for the installation of the irrigation system and turf. No imported fill or soils other than fertilizer were to be used in installing the irrigation system and turf.	LARPD, 1971
1972	No trees are apparent adjacent to the south side of the concrete-lined portion of the arroyo.	Aerial photograph, 1972
1970–1972	Arroyo Seco Elementary School constructed.	Aerial photos of the area 1970 and 1972
1975	Trees are apparent adjacent to the south side of the concrete-lined portion of the Arroyo Seco channel.	Aerial photos of the area 1975
1986	Play area and picnic tables upgraded. LARPD indicates that sand was brought in and gravel and soil from the older, smaller play area (dimensions not defined) was removed to make room for the larger play area.	Ingledue, 1997, 1998 LARPD drawing, 1988
1986	Big Trees Park eastern extension was constructed.	Ingledue, 1997, 1998 Aerial photograph, 1985
1988	Asphalt added to improve Big Trees Park paths.	LARPD drawing, 1988
1993	EPA collects a background sample from Big Trees Park that exceeds plutonium-239+240 global fallout background levels for this area.	NAREL, 1994
Oct–Dec 1994	LLNL meets and develops sampling plan with representatives of homeowners association near the park, City of Livermore, Livermore schools, LARPD, EPA, CDHS-RHB, and others.	EPA, 1995; McConachie, 1998

[a] Commercial online service for determination of property ownership.

Table 1. Chronology of events at Big Trees Park (continued)

Date	Activity	Source
Jan 1995	LLNL samples Big Trees Park, Big Trees eastern extension, schoolyard, and vicinity. EPA and CDHS-RHB collect split samples of the soil for independent analyses.	MacQueen, 1995
July 1995	LLNL report published and distributed. Pathway for plutonium from LLNL to park not definitive. All plutonium concentrations less than residential guideline.	MacQueen, 1995
Sept 1995	EPA fact sheet on plutonium published—"The levels of plutonium detected off site do not pose an unacceptable risk to local residents."	EPA, 1995
Feb 1998	Regulatory agencies recommend that LLNL sample deeper and investigate pathways.	CDHS/ATSDR; 1998/1999
	In response to regulator recommendations, LLNL develops a sampling plan with the cognizant regulatory agencies and stakeholder input.	LLNL, 1998; Liddle, 1998
Aug and Sept 1998	Samples collected at Big Trees Park in accordance with negotiated sampling plan.	LLNL, 1998
Feb 1999	EPA finds "no unacceptable risk" from plutonium levels at Big Trees Park.	Heffner, 1999
May 1999	CDHS/ATSDR release final health consultation	CDHS/ATSDR; 1998/1999

2. Sampling

The sampling strategy was based on choosing sampling locations and analytes that would provide data to (1) determine the vertical and lateral extent of plutonium in soil at Big Trees Park, (2) pick the most likely plutonium distribution pathway, and (3) add data at locations previously identified as exhibiting plutonium concentrations in soil that were above background concentrations. A summary of sampling locations, numbers of samples collected, sampling depths, and materials sampled for (analytes) is presented in Table 2. Sampling locations within Big Trees Park are shown in Figure 2.

Samples were collected, handled, and analyzed as specified in the sampling plan (LLNL, 1998; Liddle, 1998). After the results were received, the EPA, ATSDR, and LLNL analyzed the quality control data and concluded that the data were appropriate for determining the levels of plutonium in soil at the park and for making decisions about public health and safety.

In summarizing the data for this report, the following conventions were followed. A detectable quantity was defined as a measured result larger than the radiological counting uncertainty (two standard deviations), even if the measured result was below the analytical minimum detectable amount. Where samples taken at any given location were split up, an average of the values of the splits, whether analyzed by the same or different analytical laboratories, was used to represent the location. Average values include both detectable and non-detectable quantities; that is, in calculating averages for any data set, all values, including values identified as detectable and non-detectable quantities, were used (DOE, 1991). The radiological counting uncertainty associated with individual results or averages has not been reported. The uncertainty among analytical laboratories was similar; no one laboratory was noticeably more precise; and consideration of analytical uncertainty does not change any of the conclusions reached in the report. An upper limit for global fallout background concentrations was defined as 0.012 picocuries/gram (pCi/g), which is the 80% upper confidence level on the 95th percentile of annual surveillance data upwind of the LLNL Livermore site (LLNL, 1998, Appendix D). The words "above background" have been used as a shorthand notation for "above global fallout background." As stated previously, this report is a summary report only and is not intended as a rigorous technical or statistical analysis of the data.

Table 2. Big Trees Park sampling plan.

Sample set	Analytes[a]	Potential pathways	Depths (cm)	Number of locations	Number of samples
Current arroyo channel	Pu	Water	0–5 0–25	2 LLNL 1 SNL 2 Near park 2 Downstream	7
Old arroyo channel	Pu	Water	0–15	3	3
Ornamental trees	Am Pu Metals	Air Water Sludge	0–45 45–90 90–135	10 Pairs (20)	60
Grid	Pu Am (0–5 cm) Metals (3 locations)	Air Water Sludge	0–5 5–10 10–20 20–30 30–40	30	150
Special sampling at 1995 Locations 1, 7, and 8	Pu Am (Location 1 at 0–5 cm) Metals (Locations 7 & 8)	Air Water Sludge	0–5 5–10 10–20 20–30 30–40 40–85	8 (Location 1)[b] 1 (Location 7) 1 (Location 8)	60
Special sampling of disked area	Pu Metals (1 location)	Water Air	0–15	4	4
Special sampling of playing field	Pu	Air Water	0–5 5–10 10–20 20–30 30–40	2	10
Special sampling of Big Trees eastern extension	Pu	Air Water	0–5 5–10 10–20 20–30 30–40	3	15
				Total	**309[c]**

[a] Analytes are plutonium (Pu-239+240 and Pu-238), americium-241 (Am-241), and metals (chromium, copper, lead, nickel, and zinc).

[b] Samples could not be collected at 20–30 cm, 30–40 cm, or 40–85 cm at one of the locations due to lack of sample integrity.

[c] Ten percent of the total number of sample locations had an associated collocated sample for quality control. In addition, 9 locations were selected for field splits.

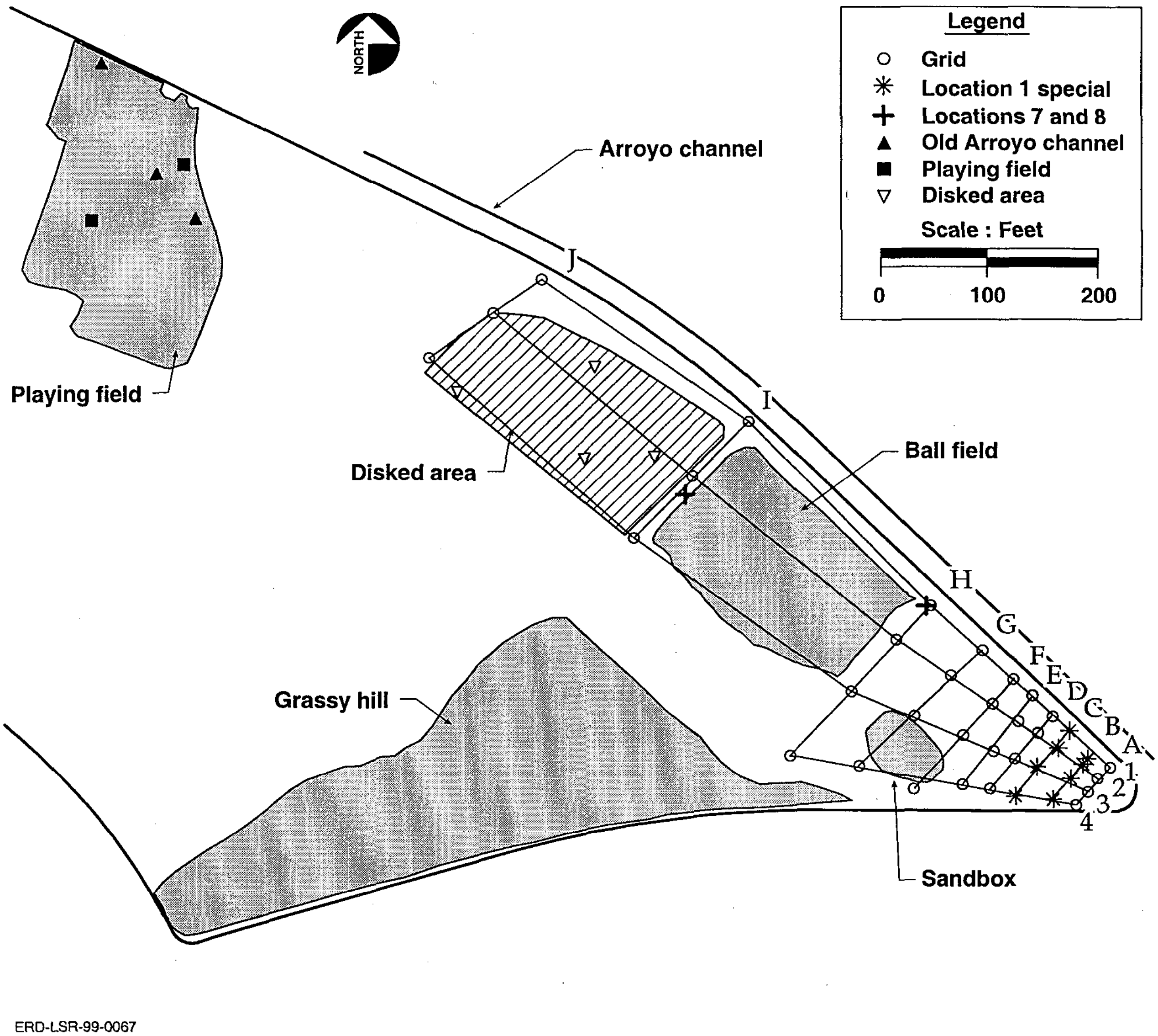

Figure 2. Map of Big Trees Park showing the 1998 sampling locations within the park, the school, and the sampling grid.

2.1. Extent of Plutonium at Big Trees Park

Sampling to determine the extent of plutonium at levels above background was conducted on a grid consisting of 4 radial lines (1–4) and 10 perpendicular cross lines (A–J). Radial 1 was cast parallel to and next to the concrete-lined arroyo channel at the northern edge of the park (see Figure 2). The cross lines were drawn perpendicular to Radial 1 and the channel where possible. Physical obstructions sometimes dictated the sample location. Perpendiculars were placed at gradually increasing intervals with the result that sampling was densest in the area around 1995 Location 1. The grid was mapped in this way to encompass 1995 sampling locations 1, 7, and 8, which had the highest plutonium concentrations in the 1995 study, and to include the ornamental trees, which were important in testing the "sludge pathway hypothesis" (see Section 2.2.2.). For ease of discussion in this document and to underscore that Location 1 special sampling is intended to be part of the grid sampling, these locations have been assigned a simple, unified numbering system rather than the location naming system used in the sampling plan.

2.1.1. Data Analysis and Interpretation

Plutonium activity data are displayed in Table 3 in entries that correspond to the place on the sample grid where the sample was collected. These data fall into four categories: nondetects (italics), detections at background levels (regular typeface), detections above the 0.012 pCi/g background determined from annual surveillance data (boldface), and detections of the same order of magnitude as the original 1993 EPA sample result for the park of 0.16 pCi/ g (shaded entries). Values in the shaded entries are the highest measured activities, and are the most likely to be evidence of plutonium concentrations that are above background. The data patterns in Table 3 indicate the vertical and lateral distribution of plutonium within the sample grid.

2.1.1.1. Depth

Radiochemical analysis showed that only three samples taken on the grid (B1, E1, and H2) yielded Pu-239+240 concentrations above background levels (i.e., greater than 0.012 pCi/g) at depths below 10 cm (see Table 3). Figure 3, which shows average plutonium concentrations plotted for the different depth intervals sampled, shows that plutonium levels dropped steeply at depths greater than 10 cm. Consequently, the above-background plutonium levels are confirmed to be at or near the surface. Because above-background plutonium values were found only rarely and not found in any pattern at depth, the values discussed in interpreting the lateral extent of plutonium concentrations are the averages of the samples collected at 0–5 cm and 5–10 cm.

2.1.1.2. Lateral

The data show a distinctly higher level of plutonium along Radial 1 (see Table 3). Bar graphs of relative radionuclide concentrations drawn on the grid clearly depict the higher concentrations along Radial 1 (Figure 4). Figure 4 also shows elevated Pu-239+240 concentrations along grid perpendicular H, especially at H1 and H4.

Table 3. Plutonium-239+240 concentration values for samples taken in 1998 on a grid at Big Trees Park.[a] Values in shaded boxes are the highest measured activities. Values in boldface type are above 0.012 pCi/g background; those in regular typeface are detections at background level; and those in italic are nondetections. (See Section 2.1.1 for discussion.)

Radial	Depth interval (cm)	Plutonium-239+240 concentration (pCi/g)									
		Perpendicular[b]									
		J	I	H	G	F	E	D	C	B	A
1	0-5	**0.042**	**0.12**	**0.28**	**0.32**	**0.64**	**0.59**	**0.30**	**0.020**	0.012	**0.14**
	5-10	*–0.00028*	**0.17**	**0.17**	**0.24**	**0.27**	**0.77**	*0.00048*	*0.00023*	**0.035**	*–0.00090*
	10-20	*0.0012*	0.0086	0.012	0.012	0.010	**0.025**	*0.00075*	*0.00081*	0.0057	*–0.0011*
	20-30	*–0.00014*	*0.0035*	*0.0051*	*0.0025*	*0.0014*	*–0.00014*	*–0.00027*	*–0.000063*	*0.0011*	*0.0003*
	30-40	*–0.00014*	*0.0031*	*0.0012*	*0.0022*	*0.0011*	*–0.00085*	*–0.00047*	*–0.000067*	**0.013**	*0.0015*
2	0-5	*0.0016*	0.0084	**0.023**	0.0014	**0.028**	**0.015**	0.0057	0.010	0.0078	0.0053
	5-10	*0.0011*	0.012	**0.32**	0.012	*–0.000047*	*0.000094*	0.0040	*–0.000059*	0.0068	0.0026
	10-20	*0.0010*	0.010	**0.12**	*0.00080*	*0.000041*	*0.0010*	0.0023	0.0016	0.0048	*–0.000093*
	20-30	0.0026	*0.00030*	0.0031	*0.00071*	*0.00084*	*–0.000046*	*0*	*0.00025*	0.0025	*0.00044*
	30-40	*0.00030*	*0.00062*	*0.00091*	*0.00072*	*0.000090*	0.0023	*0.00097*	*–0.00053*	*0.00024*	*0.00081*
3	0-5	0.001	0.004	**0.031**	0.0028	**0.027**	0.0092	0.0037	0.011	**0.037**	*–0.00081*
	5-10	*0.0018*	0.0028	*0.00059*	**0.015**	*–0.00051*	0.01	0.0033	0.0041	*–0.00041*	*–0.0015*
	10-20	*–0.00050*	*–0.0016*	*–0.00052*	0.0097	*–0.000050*	*0.000065*	0.0076	*0.00045*	*0.00026*	*–0.0015*
	20-30	0.0018	*0.0010*	*–0.0014*	0.0024	0.0052	*–0.00058*	0.0050	*0.00022*	c	*–0.00030*
	30-40	*0.00085*	*0.00023*	*–0.00018*	*0*	0.0050	*–0.00067*	*0.00019*	*–0.00036*	c	*–0.0015*
4	0-5	d	d	**0.21**	**0.029**	*0.0077*	0.0048	*0.00068*	0.0028	**0.022**	0.0053
	5-10	d	d	**0.090**	*0.0025*	0.0049	0.0093	*0.00093*	0.0034	0.0067	0.0055
	10-20	d	d	0.0082	0.0067	*0.00066*	*–0.00089*	*0.00053*	*0.00082*	*0.000053*	*0.00055*
	20-30	d	d	*0.00097*	0.0031	0.0046	*0.00055*	*0.000019*	0.0019	*0.00073*	*0.0021*
	30-40	d	d	0.0086	*0*	0.0050	*0.0036*	*0.00079*	*0.00039*	*–0.000072*	*–0.00063*

Note: Sampling plan locations G0101–G0401 are assigned 1A–4A; L1S01–L1S04 are assigned 1B–4B; L1S05–L1S08 are assigned 1C–4C; G0102–G0402 are assigned 1D–4D; G0103–G0403 are assigned 1E–4E; G0104–G0404 are assigned 1F–4F; G0105–G0405 are assigned 1G–4G; G0106–G0406 are assigned 1H–4H; G0107–G0307 are assigned 1I–3I; and G0108–G0308 are assigned 1J–3J

a See Figure 2 for a map of sample locations.

b Perpendiculars are listed in reverse alphabetical order to correspond with the physical pattern of the sample locations at the park. See Figure 2.

c Lack of core integrity prevented a sample from being obtained.

d No sample planned at this location.

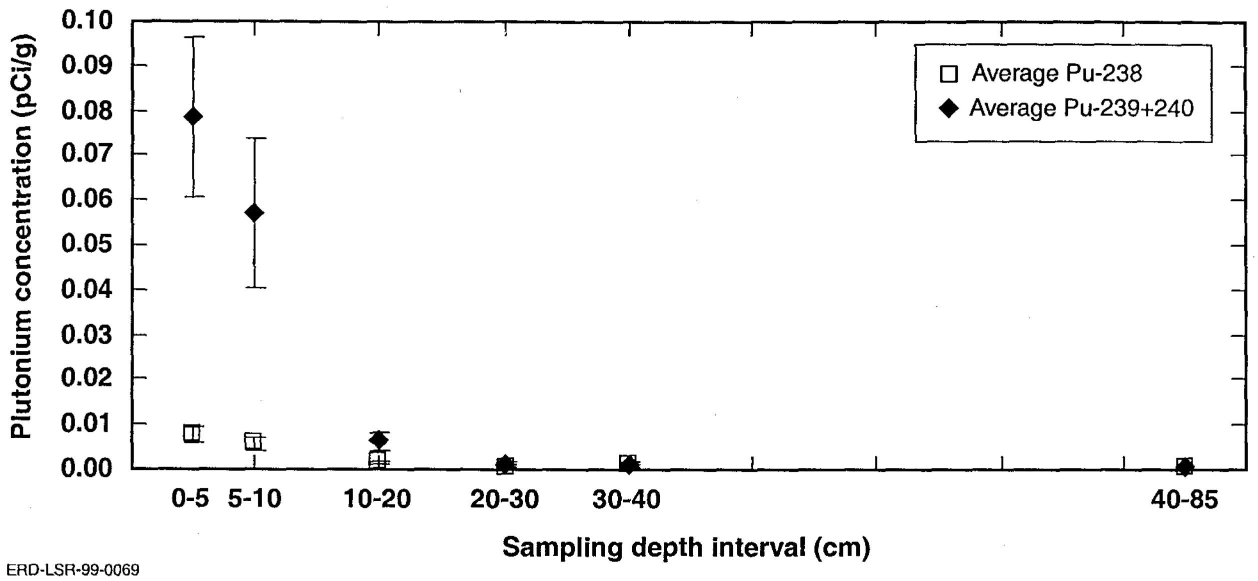

Figure 3. Average plutonium concentrations (pCi/g) at various depth intervals at Big Trees Park. (Error bars are standard error of the mean.)

The Pu-238 data show the same pattern as the Pu-239+240 data, but the Pu-238 activity values are approximately 10% of those for Pu-239+240. Since the Pu-238:Pu-239+240 activity ratio for weapons grade or fallout plutonium is approximately 3 to 6% (Perkins and Thomas, 1980; Battelle, 1981), this 10% activity ratio indicates that neither fallout nor weapons-grade plutonium alone are responsible for the elevated plutonium levels present at Big Trees Park. The unusual Pu-238:Pu-239+240 activity ratio suggests that the source of plutonium is not related to one particular operation, but is more likely a mixture of sources, such as the final liquid waste stream from LLNL. In addition, mass spectrometric analysis of several samples from Big Trees Park yielded excess and variable Pu-242, compared to weapons-grade plutonium compositions; these results also strongly indicate that the elevated plutonium levels are from several sources or waste streams (Lougheed and Moody, 1999; Velsko, 1995; Kelly, 1995).

Americium-241 (Am-241) was measured only for samples collected at the 0–5 cm depth interval at grid locations. Concentrations of americium-241 along the grid radials show the same pattern as plutonium (Figure 4).

Results for these grid locations show that elevated concentrations of plutonium and americium are generally confined to the northern border of the park, near the ornamental trees.

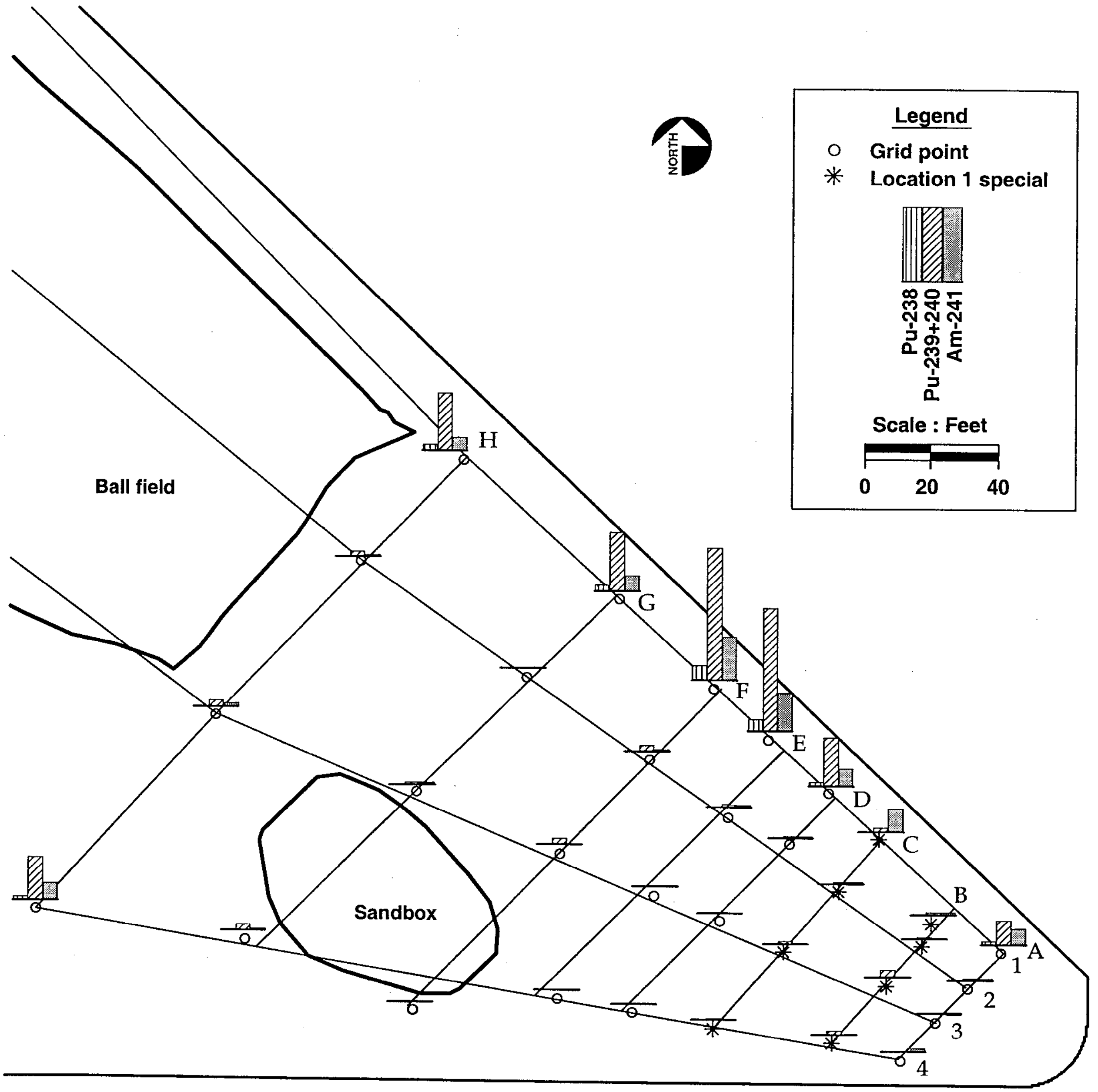

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Figure 4. Bar graphs of relative radionuclide concentrations at grid locations. Plutonium results are the averages for the 0–10 cm depth interval, and americium results are averages for the 0–5 cm depth interval.

2.2. Distribution Pathways

One of the three goals of the 1998 sampling was to investigate the likelihood of the three proposed pathways— waterborne transport via Arroyo Seco, sludge, or aerial deposition—by which plutonium could have reached Big Trees Park.

2.2.1. Waterborne/Arroyo Pathway

The authors of the draft health consultation hypothesized that traces of plutonium might have reached Big Trees Park in water-borne sediments carried from LLNL down the Arroyo Seco, which cuts across the southwestern corner of the LLNL site and flows past Big Trees Park on its northern boundary. Low-level plutonium contamination in the southeastern portion of the Livermore site was first documented by LLNL in 1974 (Silver et al., 1974). The activities responsible for the contamination at the southeastern portion of the Livermore site occurred between 1962 and 1976. Until 1965, part of the southeastern Livermore site did drain to the Arroyo Seco. However, since that time the drainage has been directed to the center of the site and then to the Arroyo Las Positas.

Because the Arroyo Seco at Big Trees Park was rechanneled in 1970, samples were collected in both the current arroyo channel and in the former location of the channel (old arroyo channel) as identified from maps. Three locations (Figure 5) were sampled from the old arroyo channel, because the old arroyo would be the only place where a historic release of plutonium would not have been washed away during the intervening years. The current channel was sampled at seven locations, which are shown in Figure 5.

Upstream (east) of Vasco Road, sediment samples in the current arroyo were collected at the 0–5 cm depth interval, consistent with 1995 sampling and recent LLNL surveillance monitoring (1993–present). At the two locations downstream of Big Trees Park and the two locations near the eastern extension of the park, sediment samples were collected from the 0–25 cm depth interval to determine if older plutonium-containing sediments were present. The fill/sediment interface of the old arroyo channel was identified for the three planned samples, and the samples were collected from the first 0–15 cm below the interface at 305-, 295-, and 285-cm deep. All arroyo samples were analyzed for Pu-238 and Pu-239+240 to determine if this pathway could explain the presence of above-background plutonium levels along Radial 1.

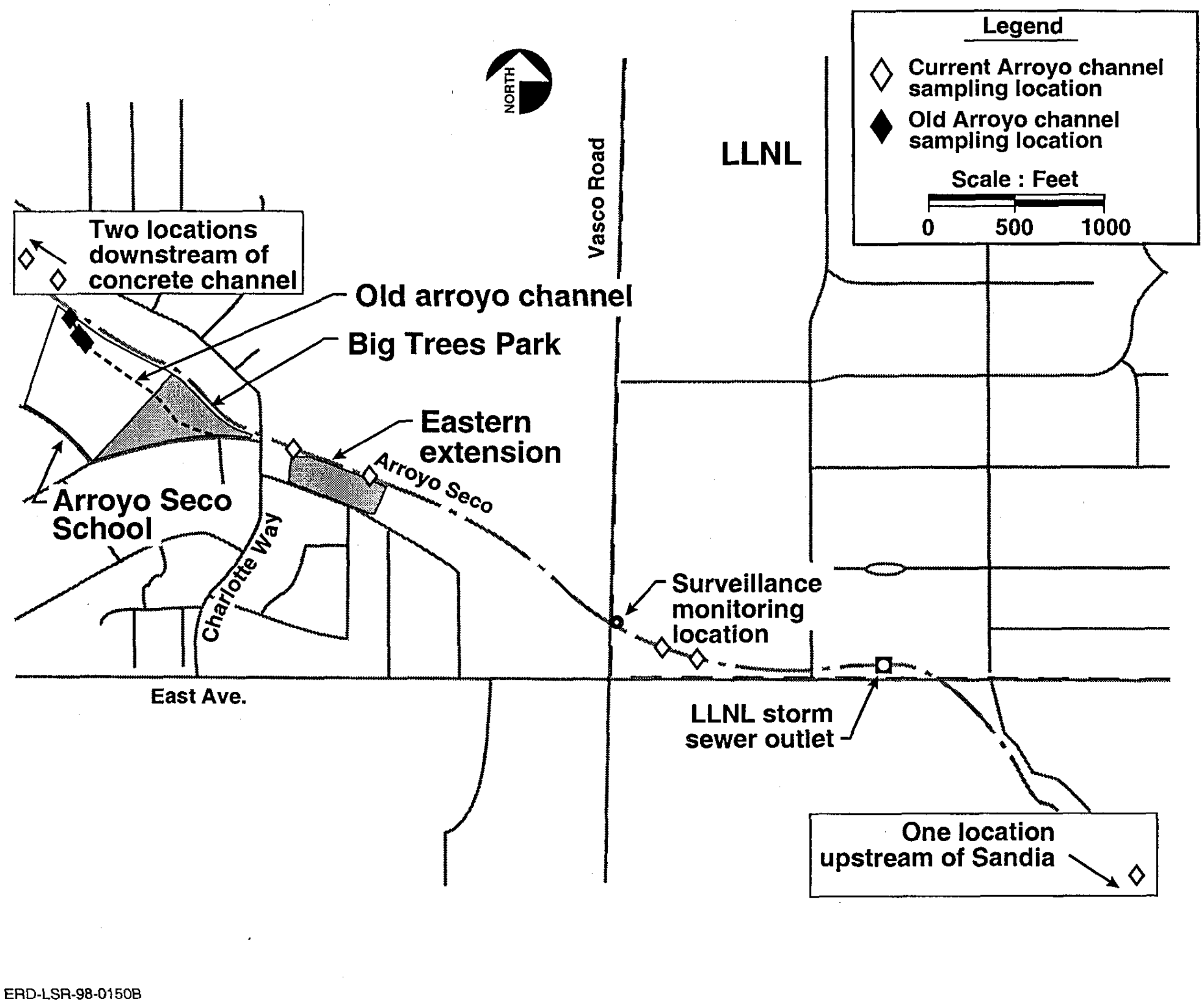

Figure 5. Arroyo Seco sampling locations, 1998.

2.2.1.1. *Data Analysis and Interpretation*

Plutonium concentrations in the arroyo samples were nearly all below the detection level. However, one field-duplicate and one field-split sample collected at each of the two locations downstream of the concrete channel (Fig. 5) contained Pu-239+240 at 0.04 pCi/g. If the samples had been of material similar to that found along Radial 1, it would have been expected that Pu-238 would have been detected at approximately 0.004 pCi/g. However, the Pu-238 results were well below 0.004 pCi/g; in fact, the Pu-238 values were non-detects, indicating that fallout was the source of the material found in the arroyo. Moreover, the primary sample associated with each of these samples did not contain Pu-239+240 above background levels.

The average of the Pu-239+240 concentrations for current arroyo sediment was 0.0033 pCi/g. The average is within the range of background data, and is well below the 1995 Location 1 concentrations (0.164 to 1 pCi/g). The data collected from the former arroyo channel were all below the detectable quantities. The average for the Pu-239+240 concentrations for "old arroyo" sediment was not detectable, with reported values averaging 0.00049 pCi/g. Because the Pu-239+240 concentrations are well below the 1995 Location 1 range, and the average concentration is within the range of background concentrations, the waterborne/arroyo channel hypothesis is considered to be unlikely. In addition, annual surveillance monitoring of the arroyo shows no residue of past releases, nor evidence of recent releases.

Moreover, the distribution of radionuclides on the grid is also inconsistent with an arroyo source, because movement of arroyo soil out of the arroyo to the park surface by earthmoving equipment would most likely not be confined to the top 10 cm; nor would it likely be confined to Radial 1 and Perpendicular H.

2.2.2. Sludge Pathway

This pathway hypothesis suggests that sewage sludge from the Livermore Water Reclamation Plant (LWRP) containing Pu-239+240 was a component of a soil amendment used when ornamental trees were planted in Big Trees Park along the concrete-lined portion of the Arroyo Seco. The probable source of plutonium in the sludge is releases to the LLNL sanitary sewer, with the largest single release occurring in 1967. Processed sewage sludge was made available to the public and municipal agencies for use as a soil amendment from the early 1960s to the mid-1970s. According to LWRP staff, 1 to 4 years passed from the time effluent entered the sewage treatment system until the sludge derived from it was available to the public. LLNL effluent reaching the LWRP prior to the 1967 release may likely have contained plutonium, but isotopic analyses were not conducted at that time. All releases to the LWRP were below the applicable regulatory limits of the time.

The sludge pathway was investigated by sampling the wells of 10 trees likely to have been planted when sludge was available as a soil amendment. Radial 1 runs along the line of these trees, including trees at 1995 Location 1. A 1975 aerial photograph shows that trees were present adjacent to the arroyo and in the vicinity of 1995 Location 1.

In sampling tree wells, each tree well location was paired with another location at least 1 meter beyond the irrigation berm that surrounds each tree. The second location was chosen far enough away from the tree well to likely not have been affected by soil amendment that might have been introduced to the subsurface during tree planting.

At each location (both inside and outside the tree well), samples were collected at three depths: 0–45 cm, 45–90 cm, and 90–135 cm. The first two intervals were estimates of the depths of the holes that were dug to plant the trees. The deepest interval, 90–135 cm was intended to provide a sample of soil beneath the depth of the tree well.

Samples were analyzed for Pu-238 and Pu-239+240, Am-241, and five metals commonly present in sewer sludge. It was believed that if the source of the plutonium at Big Trees Park was sewage sludge, metal constituents of the sludge might be detectable in the tree wells in quantities greater than the companion samples away from the trees. Samples were also analyzed for Am-241 because it was reported to have been a constituent of some LLNL sewer releases, and because it is a decay product of plutonium-241, which is present in fallout and in weapons-grade plutonium.

2.2.2.1. Data Analysis and Interpretation

Plutonium-239+240

Soils from beneath all 10 trees that were sampled exhibited detectable quantities of Pu-239+240 inside the tree well at the 0–45 cm depth internal, and 7 trees had detectable quantities outside the tree well at 0–45 cm.

At the deeper depths, four inside tree-well samples and three outside tree-well samples had detectable quantities of Pu-239+240.

The average Pu-239+240 values for the tree samples are presented in Table 4. From these averages, it can be seen that the Pu-239+240 values are substantially higher inside the tree wells at 0–45 cm than outside the tree wells at 0–45 cm. If a comparison is made of each pair of tree samples, only one tree does not exhibit this characteristic.

The averages also show that Pu-239+240 is more prevalent at the shallower 0–45 depth interval than at the deeper ones. If the sludge were used as a soil amendment when the trees were planted, it is likely that Pu-239+240 values at the 45–90 cm depth would be less than those found at 0–45 cm, but greater than the Pu-239+240 values found at 90–135 cm.

The results actually show much higher values at 0–45 cm inside the tree wells than in any other tree well samples. The 45–90 cm samples obtained from inside the tree wells did not yield results at an intermediate range, as would have been expected if the sludge had been place in the tree wells when the trees were planted. However, the Pu-239+240 activity actually measured is consistent with sludge being applied in the upper part of the tree well as a fertilizer or top-dressing after the trees were planted.

Table 4. Results of sampling inside and outside of ornamental tree wells. Italicized values are nondetects.

Sample set	Depths (cm)	Number of locations	Number of samples	Number of Pu-239+240 detections	Average Pu-239+240 results (pCi/g)	Average Pu-238 results (pCi/g)
Inside tree wells	0–45	10	10	10	0.046	0.0048
	45–90		10	2	0.00063	0.00095
	90–135		10	2	0.0016	*0.00024*
Outside tree wells	0–45	10	10	7	0.0024	0.00066
	45–90		10	1	*0.00021*	0.00072
	90–135		10	2	*0.00029*	0.00085

Further support for the top-dressing sludge hypothesis can be found by comparing the Pu-239+240 values in grid samples to values in tree-well samples. If all depths (0–40 cm) of the grid samples at each location are averaged and those averages are compared to 0–45 cm deep tree-well samples, the Radial 1 samples are of the same order of magnitude as the samples collected inside the tree wells (see Figure 6), and the other grid samples (Radials 2–4) are of the same order of magnitude as those taken outside the tree wells.

Plutonium-238

The Pu-238 data also support the top-dressing sludge hypothesis. Although Pu-238 is present at lower levels, eight of the inside tree-well samples at 0–45 cm had detectable quantities of Pu-238. In comparison, all other depths, both inside or outside the tree well, had at most three detections. The averages for Pu-238 values also show that Pu-238 is substantially higher inside the tree wells at 0–45 cm than inside the tree wells at deeper depths or outside the tree wells.

Americium-241

The Am-241 data cannot be interpreted as straightforwardly as the plutonium isotope data because of the presence of two outlier values in samples taken outside the tree wells. If these values are excluded from the analysis, the Am-241 data follow the same pattern as the plutonium. In other words, the mean values of the data for samples inside the tree wells at 0–45 cm are significantly higher than the data at deeper depths inside the tree wells or at any depth outside the tree wells.

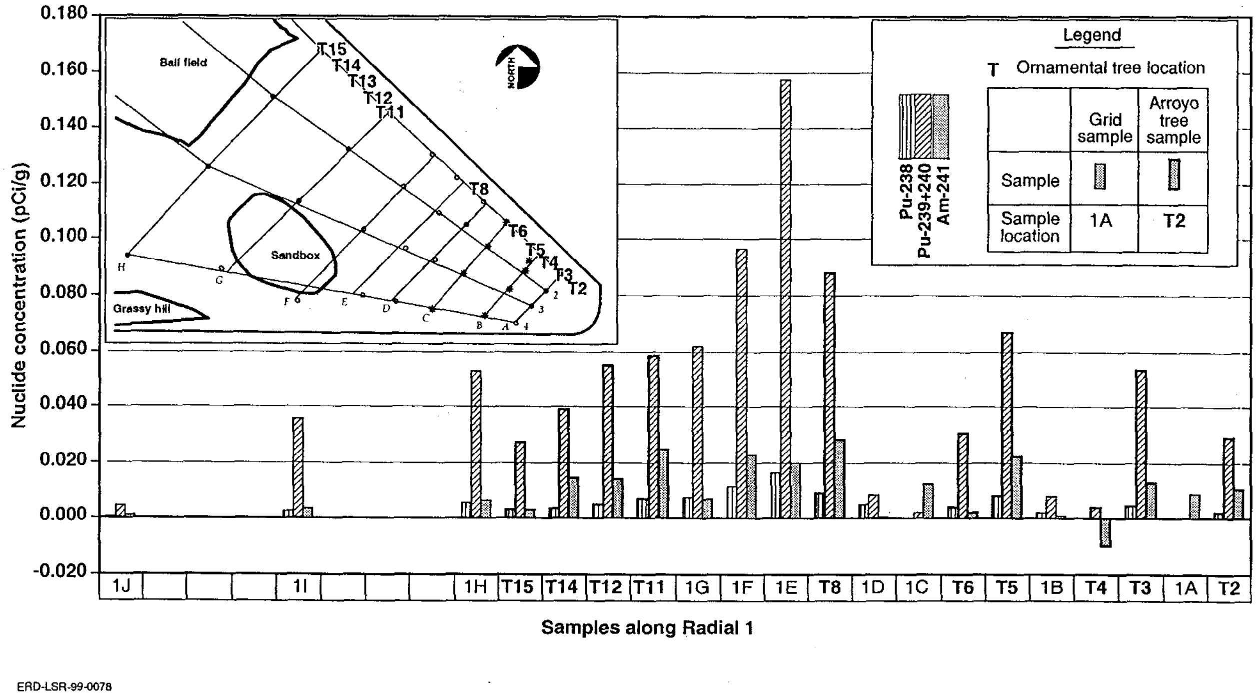

Figure 6. Plutonium and americium concentrations measured at grid locations on Radial 1 and in tree wells. Tree samples were collected at 0–45 cm depth intervals and grid samples were normalized to the same depth interval. Tree locations are depicted in bold for comparison with grid locations.

Metals

The presence of five metals (chromium, copper, lead, nickel, and zinc) was also evaluated inside and outside the tree wells at the three depths. The metals were not found at concentrations typical of sewage sludge, probably because of weathering or leaching of these materials after application of the sludge as a soil conditioner. Weathering of metals in sewage sludge applied to soil is a well-documented, but not completely understood, phenomenon (McBride et al., 1997).

Nonetheless, statistically significantly higher concentrations of all metals were found at the 0–45 cm depth when comparing the results from inside the tree wells to outside the tree wells. Although the metals at the concentrations found could be attributed to the application of other fertilizers or pest-control treatments, the metals data remain consistent with the top-dressing sludge application hypothesis.

2.2.3. Aerial Distribution Pathway

Another pathway that has been suggested for transmission of plutonium to Big Trees Park is aerial distribution. The grid radials show a definite increase above background at the first grid radial. The pattern of distribution illustrated in Figure 4 is not consistent with aerial distribution because aerial distribution would show randomly distributed levels of plutonium above background throughout the grid.

Additional data available about air dispersion of plutonium also do not support an air route of transport of plutonium to Big Trees Park from LLNL. One potential source could be the Plutonium Facility (Building 332). However, all plutonium-handling operations are triple-HEPA filtered. Sampling systems at the Plutonium Facility consistently show no emissions from that building, except in 1980 when a release occurred. Air modeling of the release, assuming the wind was blowing directly from the Plutonium Facility to the park (which it was not), indicates that the amount of plutonium at the park would be too small to measure. Similarly, modeling of resuspension of plutonium found on the southeast quadrant of the Livermore site, another potential source, also cannot explain the pattern of plutonium distribution at Big Trees Park. In addition, the meteorological data show that Big Trees Park is downwind of the LLNL Livermore site only about 5% of the year. Most significantly, air surveillance data collected throughout the Livermore Valley do not yield any results that support the suggestion that air deposition is the cause of the plutonium levels measured at the park.

2.2.4. Special Sampling

As a result of public comments and discussions among DOE/LLNL and the regulatory agencies, samples were also collected in the disked area; the playing field; the park's eastern extension; and 1995 sampling Locations 1, 7, and 8 (Figure 2).

2.2.4.1. Disked Area

The disked area is an open field on the northeast corner of the school property, next to the park. This area is disked annually, for weed abatement and fire control, but otherwise appears to be unmaintained. A nearby resident attending a DOE/LLNL-hosted presentation of the draft sampling plan reported that children play in this area and suggested that the area be sampled.

Four samples were collected at locations randomly selected within the disked area. After several years of disking, it can be reasonably assumed that soil has been thoroughly mixed to depths of at least 0–15 cm. Because this is the first sampling of this area, the primary question is whether or not there are levels of plutonium above background in this area. The average Pu-239+240 concentration in the disked area is 0.0018 pCi/g, which is well within expected background concentrations.

2.2.4.2. *Playing Field*

The playing field is an open field behind the school grounds proper. It is now turfed, but was a dirt field at the time of sampling. It did not appear to be used by the school as a play area during recess, but it was accessible to children as they travel to and from school. Location 12 from 1995 was in this area, and, for the 1995 samples, all three analytical laboratories reported values well within the range of background concentrations.

Two samples were collected in 1998 from locations within this area. The average concentration at 0–10 cm is 0.0013 pCi/g, again, well within expected background concentrations.

2.2.4.3. *Special Sampling of the Big Trees Park Eastern Extension*

The eastern extension of Big Trees Park was constructed in about 1986, approximately 15 years after the development of the main grounds of Big Trees Park. It is adjacent to the arroyo, just upstream of where the arroyo enters the concrete channel east of Charlotte Way.

Location 13 from the 1995 sampling was in this eastern extension. Two of three laboratories reported results well within the range of fallout. The third laboratory reported a result above background, but with a high degree of uncertainty (MacQueen, 1995).

Three samples were collected in 1998 within this area, equally spaced along the length of this section of the park. The average Pu-239+240 results were 0.0025 pCi/g at the 0–10 cm depth. This value is well within the range of background. As for one of the arroyo samples, field duplicates of two samples at Big Trees Park Eastern Extension contained Pu-239+240 at the 0.04 pCi/g level. If these samples had been of material similar to that found in the first grid radial or the tree wells, Pu-238 would have been detected at approximately 0.004 pCi/g. However, the Pu-238 was well below that level; the Pu-238 values were non-detects, indicating that fallout was the likely source of the material. Moreover, the primary sample associated with each of these duplicate samples did not contain Pu-239+240 above background levels.

2.2.4.4. *Special Sampling of 1995 Sampling Locations 1, 7, and 8*

The highest concentrations measured in 1995 were at Location 1 (0.164–1 pCi/g), Location 7 (0.05 pCi/g), and Location 8 (0.02 pCi/g). In the 1998 sampling plan, 1995 Location 1 was close to the grid locations B1 and C1. The Location 7 sample was near grid location H1, and Location 8 was sampled at the end of the ball field, near grid location I2.

The highest Pu-239+240 concentration of the Location 1 special sampling on the grid was 0.037 pCi/g found at B3. All samples collected on Location 1 special grid points had Pu-239+240 results well below the highest value found in 1995 of 1.0 pCi/g.

The resampling at Location 7 yielded a value of 0.06 pCi/g at 0–5 cm, confirming the previous result. Interestingly, Location 7 is almost on Radial 1, which is closest to the ornamental trees. Consequently, the higher values at Location 7 are explainable by the soil amendment hypothesis.

The resampling at Location 8 yielded a value of 0.005 pCi/g at 0–5 cm and 0.009 pCi/g at 5–10 cm. These results do not confirm the 1995 0.02 pCi/g result. However, the sample collected at the 40-85 cm depth had a value of 0.04 pCi/g. The value for the 10–20 cm sample was 0.005 pCi/g, and the values for the 20–30-cm and 30–40-cm samples were non-detects. The 0.04 pCi/g value for the deepest sample is most likely the result of sample inhomogeneity. This sample was taken in extremely hard clay, and the field technician experienced great difficulty in homogenizing the sample. The maximum value for all other locations sampled from this depth was 0.002 pCi/g; the minimum value was 0.0001 pCi/g, a non-detect. In any case, the results for Location 8 are well below the residential PRG of 2.5 pCi/g, and well below any level of health concern.

3. Conclusion

The extensive soil sampling conducted in 1998 in Big Trees Park provides an ample and accurate set of data on which to base a number of conclusions. First, the data show that there is virtually no above-background plutonium at depths below 10 cm. Second, the data can be used to gain an understanding of the lateral extent of above-background plutonium at the park. The data clearly show the plutonium to be primarily associated with grid Radial 1, with a secondary tendency along grid Perpendicular H. Third, the data can be used to deduce which of the three proposed pathways to the park is the most likely. The sewage sludge pathway is the most likely way for plutonium to have reached the park because it is the only pathway hypothesis that is consistent with the data.

Finally, and most importantly, this large body of data provides substantial evidence that the levels of plutonium at the park are well below the U.S. EPA Region IX residential preliminary remediation goal of 2.5 pCi/g. This has led the U.S. EPA to determine that there is no cause for health concern, and there is no need for cleanup at the park.

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