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Soil Survey

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Summary

Lawrence Livermore National Laboratory (LLNL) will sample and analyze soil in the Big Trees Park area in Livermore, California, to determine if the initial level of plutonium (Pu) in a soil sample taken by the U.S. Environmental Protection Agency (EPA) in September 1993 can be confirmed. Nineteen samples will be collected and analyzed: 4 in the area where the initial EPA sample was taken, 2 in the nearby Arroyo Seco, 12 in scattered uncovered soil areas in the park and nearby school, and 1 from the sandbox of a nearby apartment complex. Two quality control (QC) samples (field duplicates of the preceding samples) will also be collected and analyzed. This document briefly describes the purpose behind the sampling, the sampling rationale, and the methodology.

Purpose of Sampling

EPA evaluated three offsite locations as part of a study called Confirmatory Study of Plutonium in Soil from the Southeast Quadrant of the Lawrence Livermore National Laboratory (Cohen et al., 1994). EPA asked LLNL to verify the Pu found in one sampling location because it exceeds what might be considered expected fallout levels, though the level is well below any published threshold action limits for Pu in soil, and radiological assessments indicate no significant risk from Pu exposures calculated using health-protective conservative assumptions. LLNL submitted splits of the EPA samples to both an onsite laboratory [Radiation Analytical Sciences Section (RAS)] and an offsite (Lockheed) analytical laboratory for analyses. Those analyses are in reasonable agreement with the EPA results (Table 1). The Pu value reported by Lockheed (less than 0.046 pCi/g $^{239/240}$Pu) does not invalidate the EPA and RAS results.

Table 1. Results of EPA analyses.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Nuclide</th>
<th>EPA result (pCi/g) (Error ± 2 $\sigma$)</th>
<th>RAS result (pCi/g) (Error ± 2 $\sigma$)</th>
<th>Lockheed result (pCi/g) (Error ± 2 $\sigma$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SSS-Pu-208-1cm</td>
<td>$^{238}$Pu</td>
<td>0.014 (0.013)</td>
<td>0.0151 (0.00381)</td>
<td>&lt;0.14 (0.072)</td>
</tr>
<tr>
<td>SSS-Pu-208-1cm</td>
<td>$^{239/240}$Pu</td>
<td>0.075 (0.025)</td>
<td>0.108 (0.0119)</td>
<td>&lt;0.074 (0.049)</td>
</tr>
<tr>
<td>SSS-Pu-208-5cm</td>
<td>$^{238}$Pu</td>
<td>0.011 (0.015)</td>
<td>0.0119 (0.00321)</td>
<td>&lt;0.058 (0.026)</td>
</tr>
<tr>
<td>SSS-Pu-208-5cm</td>
<td>$^{239/240}$Pu</td>
<td>0.164 (0.039)</td>
<td>0.148 (0.0141)</td>
<td>&lt;0.046 (0.038)</td>
</tr>
</tbody>
</table>

The purpose of this additional sampling is to determine the average Pu concentration or activity in the area around the location sampled by the EPA and to survey other nearby soil areas at the park and school. If the levels found in the additional samples do not exceed published threshold action limits for Pu in soil and confirm no significant risk to human health, no further work is expected.
We will conduct three specific evaluations to:

- Determine if the initial Pu value reported by the EPA is representative of the soil in the area where the initial sample was taken or results from sampling or handling anomalies (i.e., can the sample value be replicated by subsequent resampling in the area of the initial sample).
- Determine whether this value represents Pu values found elsewhere in the park, school, and the newer eastern extension of the park referred to as “Big Trees Park East” in this plan (Fig. 3).
- Calculate and assess potential health risks associated with the detected Pu levels.

Sample Locations

We will sample as closely as possible to the original EPA sampling location to make the first evaluation. Information from EPA sampling personnel, following their review of their sampling logbook, indicates that the original sample was taken along and south of the fence between Arroyo Seco and the park, 50 to 70 ft from the intersection of Kathy and Charlotte Ways and south of the arroyo (see Figs. 1 and 2). In addition, EPA sampling personnel marked their best estimate of the sampling locations on photos of the area we recently sent them. Four samples will be taken in this area (Location 1) to assure that the original sample location is part of the region sampled. All samples collected in this survey (as discussed in the “Sampling Method” section) will be to depths of 5 centimeters (cm).

For the second evaluation, we will perform a survey of other areas of interest at the park, school, and a nearby apartment complex. Samples will be collected and evaluated for Pu content from 15 such areas. The first area of interest is Arroyo Seco (Figs. 1 and 3). The City of Livermore and EPA have requested that arroyo sediments be examined. Because the arroyo is concrete-lined all along the northern boundary of the park west of Charlotte Way, two samples will be collected in the arroyo (Locations 2 and 3) to the east of Charlotte Way, upstream of the beginning of the concrete liner. These samples will be collected from sediments expected to contain relatively large amounts of silt and/or sand-size particles from the arroyo channel.

The next areas of interest are the picnic areas under the eucalyptus trees near the intersection of Charlotte and Kathy Ways; soil in this area is compacted and appears to bear heavy foot traffic (Figs. 1 and 2). Picnic tables are present, and it is likely that families with children who play on the nearby swings would congregate here. The picnic area to the east is Location 4; the more westerly picnic area is Location 5.
The children’s playground and swings area (Location 6) is expected to contain mostly sand with soil mixed in it (Figs. 1 and 2). It is likely that the sand is periodically replaced or replenished. Although the depth of the sand is generally at least 10 cm, making resuspension of underlying soil unlikely, this area will be sampled due to the expectation that many children play here.

The next area of interest (Location 7) is the path that leads along the northern boundary of the park, along Arroyo Seco (Figs. 1 and 2). It is a likely area for people taking walks and is the area of the park closest to homes located across the arroyo.

Location 8 is adjacent to the recently disked area to the west of the baseball field (Figs. 1 and 2). The dishing process can increase the resuspension of soil particles into the air and can break up otherwise compacted soils, making this an area of interest.

The next area of interest is the exposed soil where the roses are planted near Kathy Way (Figs. 1 and 2). Lawn areas reduce the possibility of soil resuspension, but areas of bare soil exist in the wells of trees planted in the lawn and in other planted areas such as beneath the rose bushes. These areas are also close to homes across Kathy Way. The southernmost of these areas (Location 9) will be sampled.

Three samples will be collected from the school yard west of Big Trees Park to provide survey data (Fig. 1 and 2). The first such sample will be from the soil adjacent to the easternmost school building (Location 10). The sandbox, interior to the school grounds, will also be sampled (Location 11). A sample will also be collected from the exposed soil in the grassy open playing field (Location 12) behind the school.

The next area of interest is the “Big Trees Park East.” A sample (Location 13) will be collected from exposed soil in the vicinity of the picnic tables within the park (Figs. 1 and 3).

The dirt road and pathway along the northside of Arroyo Seco is used by cyclists and pedestrians (Figs. 1 and 3). Two samples (Locations 14 and 15) will be collected from that pathway.

A sample will also be collected from the sandbox where children play in a nearby apartment complex (Location 16; Figs. 1 and 3). Just as in the Big Trees Park area sand boxes, this sandbox is expected to contain mostly sand, but with soil mixed in it. Although the sand in the box is expected to be at least 10 cm thick, making resuspension of underlying soil unlikely, this area will be sampled due to the expectation that many children play here.
Sampling Method

All samples will be collected to a depth of 5 cm. Because it is very difficult to collect representative 1-cm-deep samples, especially over uneven terrain, and because the highest values in the EPA sampling were in the 5-cm-deep samples, no 1-cm-deep samples will be taken. The soil will be collected with a coring device as stated in EMS Procedure EMP-S-S. However, instead of sampling at the corners and centers of two 1-meter-square areas as stated in EMP-S-S, seven cores will be collected as close to a single point as possible in order to gather sufficient sample material for the sample splitting discussed below.

Four samples will be collected in the EPA sampling area, and one sample will be collected at each of the 15 additional sampling locations. The total number of samples to be collected is 21 [4 samples at one area, plus 1 sample at 15 other areas, plus a standard quality control (QC) sample load of two field duplicate samples taken at randomly chosen locations].

Standard procedures for chain-of-custody and sample handling, as well as those quality assurance procedures that are applicable, will be followed. These are discussed in LLNL's Environmental Monitoring Plan (Tate et al., 1994) and the Environmental Monitoring Section Quality Assurance Plan (Garcia and Failor, 1993).

Sample Splits and QC Samples

Split samples allow for multiple analyses of a sample taken by one sampling event. This provides for comparison of multiple analyses performed either by one laboratory or by several laboratories. LLNL will split all samples into fifths; one split each will be given to the EPA and Department of Health Services (DHS) at the time of collection, one will be sent to an off-site contract analytical laboratory for Pu analyses. One sample will be archived, and one will be held for possible submittal to an internal LLNL analytical laboratory.

For this survey, we will use the same sample splitting methodology as that employed by the EPA in their September 1993 sampling. When the previous samples were collected, the EPA homogenized each sample by hand within a plastic sample container and then split the sample into two approximately equal parts. For this survey, two 100-g portions will be split out and given to the EPA and DHS. The remaining material will undergo LLNL's standard soil sample preparation procedure (drying, grinding, and sieving prior to splitting).

LLNL will provide two QC samples for analysis. One sample will come from an archived site annual survey offsite soil sample. The other sample will consist of DOE Environmental Measurements Laboratory-provided soil, which has been analyzed by 57 laboratories. The activities of these two quality control samples bracket the values expected from the survey.
Analytical Methods

LLNL submitted splits of some of the samples taken in the initial EPA study to Lockheed Analytical Laboratory in Las Vegas, NV, a contract analytical laboratory, for confirmatory analysis. The results of these confirmatory analyses were found statistically to be generally comparable to the results reported by the EPA (Cohen et al., 1994). LLNL will submit the samples taken under this plan to the same contract analytical laboratory. The sample preparation and analysis method to be used is described in Lockheed’s procedure LAL-91-SOP-0108 (LAL, 1991). One to two grams of soil are processed by microwave digestion followed by precipitation of ferric hydroxide. The precipitate is dissolved and loaded onto an AGI-X8 anion exchange resin column for separation of the heavy metals. Pu is extracted by selective elution, precipitated on neodymium fluoride, collected on a 0.2-micron filter, and counted by alpha spectroscopy. Appropriate Pu-242 tracers will be used. Standard laboratory QC samples should be processed with these samples and their data reported to LLNL along with the sample data.

Data Analysis

QC sample data will be examined from field QC duplicates, known samples, and any laboratory QC samples. The first evaluation will be made by comparing these values with the results of the initial EPA value.

Results from the survey locations will be compared against one another to determine if they fall within the range of analytical uncertainty. The values will also be averaged and a standard deviation determined. The second evaluation will be based on the comparison of these values to those found in the EPA Sampling Area.

Historical LLNL surveillance monitoring results for Pu at a variety of locations throughout the Livermore Valley, as documented in the site annual environmental reports (e.g., Gallegos et al., 1993), are log normally distributed. The following will be performed using log transformed data.

1. Calculate the mean and standard deviations of the reported Pu concentrations of the four additional samples collected from Location 1.

2. Calculate a one-sided, 95% upper prediction interval using the values from the four new samples from Location 1. If the EPA value is inside the prediction interval, conclude that the EPA value is representative. The formula and a reference are given below.

3. Calculate a one-sided, 95% upper prediction interval using all other soil sample locations from the survey (excluding Location 1). If the EPA value is inside the prediction interval, conclude that the EPA value is representative.
An upper 95% prediction interval is given by
\[ x_u = \bar{x} + t_{0.95,n-1}(1 + \frac{1}{n})^{\frac{1}{2}}s \]

where \( \bar{x} \) is the sample mean, \( s \) is the sample standard deviation, \( n \) is the number of samples, and \( t_{0.95,n-1} \) is the 95th percentile of the t-distribution with \( n-1 \) degrees of freedom (Hahn and Meeker, 1993). Percentiles of the t-distribution are tabulated in most statistics books, including Table A.7 in Hahn and Meeker. The mean and standard deviation do not include the value that is to be compared with the interval.

Schedule

We expect sampling plan preparation, presentation to interested parties, and acceptance to be accomplished in January 1995. Sampling should be completed in January, weather permitting. Analytical results should be available by March and data reported by April 1995. The format for data reporting will be that used in the LLNL's Environmental Report. We will report the results of this survey as an LLNL report that will be made public.

Acknowledgments

Numerous Laboratory people provided input into the conversations, planning, writing, and editing that led to this survey plan. The individuals include: Paris Althouse, Donna Couture, Gretchen Gallegos, Marion Heaton, Bert Heffner, Kim Heyward, Don MacQueen, David S. Myers, Nan Prentice, Ellen Raber, Duke Ramsey, Maureen Ridley, Howard Sherman, and Kris A. Surano.

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Lockheed Analytical Laboratory (LAL) (1991), Actinide Sample Preparation, Nuclide Separation, and Analysis, Lockheed Analytical Laboratory, Las Vegas, NV (LAL-91-SOP-0108).
Tate, P., et al. (1994), LLNL Environmental Monitoring Plan, including associated procedures. Lawrence Livermore National Laboratory, Livermore, CA (UCRL-ID-106132, rev. 1).
Figure 1. Big Trees Park area.
Figure 2. Big Trees Park.
Figure 3. "Big Trees Park East".