Radiological Results of Initial Samples from Some Potential Pathways from the Lawrence Livermore National Laboratory (LLNL) into the Surroundings --Part 1 (Rev.3)

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Introduction and Purpose

The Lawrence Livermore National Laboratory (LLNL) has provided innovative design and engineering to support the Nation's nuclear weapons program since 1952. The RadioActivist Campaign (TRAC) initiated sampling in the public domain around LLNL in December 2003. This initial sampling seeks to establish a technical foundation to independently assess candidate ***pathways*** of radionuclides from this *world premier scientific center* into its neighborhood. In consideration of LLNL's key research-and-development role in the Department of Energy's (DOE's) nuclear weapons complex, this study has been designed to reveal artificial radionuclides with halflives shorter than one week.

TRAC's main concerns are for airborne and waterborne pathways of artificial radionuclides from LLNL into the surrounding neighborhood. Areas of focal interest are *downwind of* LLNL, which is to the northeast, and *downstream of* LLNL, which is Arroyo Seco to the west and Arroyo Las Positas to the northwest of LLNL.

TRAC plans follow-up sampling in May 2004.

TRAC will base its radiological assessment on the results of these two sampling trips and on inputs from public-interest groups, from concerned citizens, from LLNL, and from published information.

Sampling Narrative

TRAC staff arrived in Livermore on 13 December 2003. Rainfall a few days before had left a mud puddle near the east side of Greenville Road, northeast of LLNL. Eleven liters (=11 kilograms wet = "11 kgwet") of brown water were collected from the undisturbed puddle. This water was later allowed to settle at TRAC's laboratory and split into an unfiltered fraction (Sample 1) and settled sediment (Sample 2). --Sample Numbers are *contextual* rather than *chronological*. Sample Numbers appear in the headers in Table 1 of the Results.

Following heavy rainfall during the pre-dawn hours of 14 December, TRAC collected samples from the bed of Arroyo Seco, below the west (downstream) side of the South Vasco Road bridge. At the time of this sampling, storm run-off water was augmented by flow from LLNL's A1 Groundwater Treatment facility on the east side of the bridge. 21 kgwet of clear flowing, unfiltered surface water were collected (Sample 8). 0.4 kgwet of young sorrel leaves were collected from this location (Sample 9).

TRAC then proceeded to one of the upstream drainages into Arroyo Las Positas, on the east side of LLNL. This *upstream* location is *downwind* of LLNL. TRAC collected 20 kgwet of clear flowing, unfiltered surface water from the ditch on the south side of the power substation that is on the east side of Greenville Road (Sample 4). This sampling location was upstream of most drains from the substation. TRAC opted to wait for new grass to grow before sampling grass at this location.

TRAC staff observed fog in the uplands to the northeast (downwind) of LLNL. On 16 December, TRAC checked the roadsides between Altamont and Patterson Passes for suitable sampling locations and sample media. TRAC picked 0.3 kgwet of new grass growing below pastureland and above the north shoulder of South Flynn Road, close to the intersection with North Flynn Road (Sample 3).

On 17 December, TRAC drove along Corral Hollow Road, east of LLNL to LLNL's Site 300. TRAC checked roadside vegetation with a Geiger counter, downslope and downgradient of Pit 6 along Corral Hollow Road. TRAC picked 0.3 kgwet of leaves from a tree incorrectly identified as mountain ash, from the south side of Corral Hollow Road, next to the Carnegie S.V.R.A. and opposite a secondary entrance to Site 300. This Sample 12 was apparently downgradient (in the groundwater flow direction) from Pit 6. A sample of 0.3 kgwet of leaves was then collected from an unidentified tree on the north side of Corral Hollow Road, in a wash below a berm near Gate PER-SW05, below Pit 6 (Sample 11).

Later on the 17th, TRAC staff accessed Arroyo Las Positas, northwest (downstream) of LLNL, on the east side of the South Vasco Road bridge. Arroyo Las Positas was free flowing with water from LLNL. 0.3 kgwet of reed grass was sampled (Sample 6). TRAC used a Geiger counter to select sediment in the arroyo bed as Sample 7.

On 18 December, TRAC staff walked the perimeter of LLNL, checking for "hot spots" with a Geiger counter. An anthill outside the southeast corner of the LLNL fenceline exhibited twice background radioactivity. This anthill, located at the northwest corner of the East Avenue and Greenville Road intersection, was about twenty meters from disturbed grounds within the LLNL perimeter fence. 0.05 kgwet of anthill was sampled. The radioactivity of this Sample 10 decreased to background by the time it was re-checked at TRAC's lab. That decrease suggested the initial radioactivity in the anthill might have originated from natural radon gases permeating into the anthill passages underground.

Finally, TRAC picked 0.4 kgwet of young grass (Sample 5) from the same upstream location in Arroyo Las Positas as surface water had been sampled on 14 December (Sample 4).

Methods Summary

Sample selection and collection, narrated above, were the front end of an integrated process through a single-pass, radiological analysis in TRAC's lab, leading to post-analysis and ending in this data report.

Water samples were quiescently evaporated, by microwaves, to paste on plastic film. All other samples were oven dried <100C.

Samples were counted for intervals of 23 hours in a multiply stabilized, well-type, sodium-iodide detector with an energy window from 3 to 3000 KeV. The 8,000-channel, highly nonlinear, acquired spectra were transformed to 165-channel spectra of constant photo-peak width of 3 channels (FWHM). Sample analyses then began with sequential, *true* subtractions of background and reference (standard) spectra.

Each prepared sample was counted as soon as feasible to allow detection of artificial radionuclides with halflives less than one week. Samples were then recounted, and the initial spectrum minus one or more subsequent spectra provided "short-lived decay spectra." Natural thorium and uranium decay chain spectra were matched to sample spectra (—initial spectra, short-lived decay spectra, and final spectra—) and subtracted to minimize their short-lived and long-lived contributions to the sample spectra.

Other than natural thorium and uranium decay chain imbalances, the prevalent short-lived radionuclide in the samples was beryllium-7 (Be-7), with a halflife of 53 days. Be-7 is produced naturally in the upper atmosphere by cosmic ray spallation of nitrogen and oxygen atoms. This *cosmogenic* Be-7 falls to earth in rain. Be-7 is also produced by artificial nuclear reactions. Be-7 results do not seem to warrant reporting with the artificial radionuclide results in Table 1.

Cesium-137 (Cs-137) is a routine TRAC laboratory analysis, after thorium and uranium interferences have been subtracted. Likewise, americium-241 (Am-241), with its x-ray peak for confirmation, is a routine analysis. Iron-59 (Fe-59) is not a routine analysis for TRAC. This radionuclide was counted on its clean peak at 1099 KeV with the 1292 KeV peak as confirmation, and then reconfirmation by re-recounting to check the halflife of 45 days. Uncertainties of the Fe-59, Cs-137, and Am-241 analyses are reported as "±" one standard deviation counting error, as generated by Canberra G2K software.

Strontium-90 (Sr-90) is analyzed by four-point matching a sample spectrum against a standard Sr-90 spectrum, after all radionuclides through Cs-137 have been subtracted from the sample spectrum. This analysis depends on the peculiar shape of the Sr-90 spectrum, with bremsstrahlung features from direct 546 KeV beta decay and subsequent 2186 KeV beta decay, from Compton scattering into the sodium-iodide scintillation detector, and from a characteristic x-ray interaction at about 32 KeV. The standard deviation of an Sr-90 reports is the standard deviation of the repeated results of replicate counts with their separate analyses.

Analysis for unspecified short-lived radionuclides presents challenges: There are potentially so many candidate radionuclides, some unidentified phenomena can easily result in some photopeak(s) being incorrectly attributed to some radionuclide(s) not present in the sample. Such *false positive* results are anti-conservative and improperly raise public concern. On the other hand, if many procedural hurdles are imposed before any short-lived radionuclide is reported, there are excessive *false negatives*, and the environment and public health are inadequately protected. Finally, short-lived radionuclides may disappear before analyses can be replicated independently. This loss of replicability unavoidably compromises the scientific validity of reports of short-lived radionuclides.

Note that some radionuclides of concern at LLNL do not yield substantial photon activity above 3 KeV. Tritium (H-3) and plutonium (Pu-239, Pu-240) are examples. Such radionuclides are not screened well by TRAC's single-pass procedure. On the other hand, Am-241, the decay daughter of Pu-241, usually accompanies plutonium and so may flag the presence of plutonium in a sample.

Results

Preliminary radiological results appear in Table 1, on the next two pages. Before a result is reported here, it must pass through a "detect" screen to avoid a false positive report. Analyses failing to pass this screen are indicated in Table 1 by "--", meaning "not detected."

Sample Numbers in Table 1 are ordered as follows: Samples 1, 2, and 3 are from downwind, northeast of LLNL. Samples 4 and 5 are from downwind but upstream, to the east of LLNL. Samples 6 and 7 are downstream of Samples 4 and 5 and are upwind (northwest) of LLNL. Samples 8 and 9 are downstream, west of LLNL. Sample 10 is anecdotal from the fenceline of LLNL. Samples 11 and 12 are downslope and (hydrologically) downgradient of LLNL's Site 300's Pit 6.

Radioactivity is reported as "pCi" = picocurie. All sample radioactivities are reported on a wet weight basis ("kgwet" = kilogram wet) for easy comparison to drinking water standards based on

one liter = 1 kgwet

Uniform reporting in units of "kgwet" has the added advantage of easy calculation of bio-accumulation factors, in cases where the same radionuclide is reported in both water and vegetation collected from the water. One pCi/kgwet is one nuclear disintegration per minute, in a liquid pound (one pint). To convert radioactivity results to dry weight basis, multiply the radioactivity by the "Wet/Dry Weight Ratio" in Table 1.

"n/a" means "not applicable". "Wet/Dry Weight Ratio" does not exist for water.

To convert radioactivity to becquerels (Bq), multiply by 0.037.

Sample locations are given by North Latitude and by West Longitude, based on WGS 84 datum, with degrees on the side of Table 1 and minutes tabulated.

"Sample Identifier" is the sample tracking number, which is the year, month, day, and hour of sample collection. For Sample 1, the Sample Identifier is 3z1316, where: the leading "3" = 2003; "z" = December; "13" = 13th day of December; and "16" = 16:00 hours = 4:00 PM.

Table 1. Preliminary Radiological Results from First sampling.

Sample Number. Setting:	1. Mud Puddle	2. Mud Puddle	3. Flynn Pass
Sample Direction from LLNL:	near northeast	near northeast	far northeast
Location:	east side of Greenville Rd, near Hawthorne	as Sample 1.	north side of S. Flynn Rd, west of N. Flynn Rd.
Medium (Material):	unfiltered water	settled sediment	young fine grass
Wet/Dry Weight Ratio:	n/a: 11.22kgwet	984.*	6.8
Photon **Radioactivity** [pCi/kgwet]			
Iron-59 (Fe-59):	--	--	--
Strontium-90 (Sr-90):	--	--	190.±160.
Cesium-137 (Cs-137):	0.02±0.03	0.25±0.06	--
Americium-241 (Am-241):	--	--	--
Latitude: North 37º + minutes:	42.653’	42.653’	42.342’
Longitude: West 121º + minutes:	41.908’	41.908’	38.696’
Sample Identifier:	3z1316	3z1316s	3z1611

* From 11.22 kg puddle water, sediment settled and was dried to 11.4 g.

Sample No. Setting:	4. Positas East	5. Positas East	6. Positas North
Sample Direction from LLNL:	near east	near east	near northwest
Location:	Arroyo Las Positas bed, southeast of substation, east side of Greenville Rd.	as Sample 4.	Arroyo Las Positas bed, east side of S. Vasco Rd, southeast of train station
Medium (Material):	unfiltered water	young fine grass	reed grass
Wet/Dry Weight Ratio:	n/a: 20.38kgwet	6.2	5.3
Photon **Radioactivity** [pCi/kgwet]			
Iron-59 (Fe-59):	--	--	290±90
Strontium-90 (Sr-90):	--	--	--
Cesium-137 (Cs-137):	0.03±0.016	2.9±1.6	8.7±2.1
Americium-241 (Am-241):	--	--	4.5±2.0
Latitude: North 37º + minutes:	41.560’	41.561’	41.810’
Longitude: West 121º + minutes:	41.736’	41.735’	43.008’
Sample Identifier:	3z1411	3z1810	3z1713

Table 1, Completed. Preliminary Radiological Results from First sampling.

Sample No. Setting:	7. Positas North	8. Seco West	9. Seco West
Sample Direction from LLNL:	near northwest	near west	near west
Location:	near Sample 6	Arroyo Seco bed, west side of S. Vasco Rd. bridge	as Sample 8
Medium (Material):	sediment	water	sorrel
Wet/Dry Weight Ratio:	1.0 nominal*	n/a: 20.66kgwet	8.1
Photon **Radioactivity** [pCi/kgwet]			
Iron-59 (Fe-59):	--	--	--
Strontium-90 (Sr-90):	--	1.3±0.4	--
Cesium-137 (Cs-137):	--	--	--
Americium-241 (Am-241):	--	--	--
Latitude: North 37° + minutes:	41.807’	40.875’	40.875’
Longitude: West 121° + minutes:	43.023’	43.131’	43.131’
Sample Identifier:	3z1714	3z1409	3z1410

* The dried weight is taken as the wet weight.

Sample No. Setting:	10. fence SE	11. 300 South	12. 300 South
Sample Direction from LLNL:	@ SE corner	South of Pit 6	SE of Pit 6
Location:	NW corner of Greenville Rd. and East Ave.	North side of Corral Hollow Rd., by Gate PER-SW05	South side of Corral Hollow Rd., opposite Site 300 access
Medium (Material):	anthill	tree leaves	tree leaves
Wet/Dry Weight Ratio:	1.0 nominal*	7.9	3.4
Photon **Radioactivity** [pCi/kgwet]			
Iron-59 (Fe-59):	--	--	--
Strontium-90 (Sr-90):	--	--	--
Cesium-137 (Cs-137):	--	--	--
Americium-241 (Am-241):	--	--	--
Latitude: North 37° + minutes:	40.792’	38.105’	38.005’
Longitude: West 121° + minutes:	41.813’	32.851’	32.542’
Sample Identifier:	3z1811	3z1712	3z1711

* The dried weight is taken as the wet weight.

Discussion

Although patterns of the artificial radionuclides reported in Table 1 might be inferred from the descriptions of the sample locations, TRAC awaits the second sampling with its results and completion of this study before drawing specific technical conclusions.

General references for relative comparison of the Results in Table 1 appear in Table 2:

Table 2, Comparison Radioactivities for Results in Table 1.

Federal guidelines for surface water quality			
Fe-59	???	pCi/kgwet	???
Sr-90	8.	"	(40 CFR 141)
Cs-137	200.	"	(EPA-570/9-76-003)
Am-241	15.	"	gross alpha (40 CFR 141)

The only radionuclide report that exceeds its comparison reference value is Sr-90 in Sample 3. The report of 190±160 pCi/kgwet greatly exceeds the reference value of 8 pCi/kgwet, but with a low level of confidence. This result invites follow-up sampling during TRAC's second field trip in May 2004. [Sample 5 also measured positive for Sr-90 (at 240 pCi/kgwet), but this measurement failed a form-fit test for detection and so is not reported.]

General description of each of these radionuclides in the LLNL context follows:

Fe-59: Iron-59 is a short-lived radionuclide, with a halflife of 45 days. Fe-59 is produced by neutron bombardment of steel, for example stainless steel in reactor cooling water pipes. Fe-59 can then released into circulating water by processes of corrosion or erosion.

Iron is an essential element in trace quantities and has a bio-accumulation factor up to 30,000.

Sr-90: Strontium-90 is a long-lived radionuclide, with a halflife of 29 years. Sr-90 is a main product of nuclear fission. Sr-90 remains from worldwide fallout from testing nuclear weapons in the earth's atmosphere in the 1950s and 60s.

Sr-90 is a main component of liquid waste streams from inadequately managed nuclear reactors, for example into River Techa from the notorious Mayak facilities and into the River Tom from the Seversk reactors in Siberian Russia. Ordinarily, substantial Sr-90 is only released into the atmosphere from industrial-scale nuclear operations in the event of fire. A fire at the Chernobyl Nuclear Power Station in Russia in April 1986 lofted half as much Sr-90 as it lofted its companion fission product Cs-137.

Strontium is in Group 2 of the periodic table of the elements, along with calcium. Sr-90 mimics calcium which is an element essential to cellular control processes. In calcium-poor areas, Sr-90 is concentrated in the food chain, along with calcium.

Preferential biological uptake of Sr-90 and other natural processes tend to remove Sr-90 fairly quickly from interactions in the biosphere.

The tendency of Sr-90 to mimic essential calcium earns Sr-90 the unusually low guideline value of 8 pCi/kgwet in Table 2.

Cs-137: Cesium-137 is a long-lived radionuclide, with a halflife of 30 years. Cs-137 remains from worldwide fallout from testing nuclear weapons in the earth's atmosphere in the 1950s and 60s.

Six percent of nuclear fissions yield the inert gas xenon-137, with a halflife of four minutes. Xenon-137 in a main gaseous release from stacks of industrial-scale nuclear facilities that retain waste gases for less than half an hour. The released xenon-137 decays to long-lived Cs-137 within a few minutes, and the Cs-137 falls to earth or is rained out, downwind of the release point.

Cesium is a Group 1 chemical element, along with potassium. Cesium binds so strongly to clay particles in soils that uptake through plant roots is quickly minimized. Cs-137 most often enters rooted plants, such as grasses, by absorption of fallout into foliage.

Although cesium plays no ordinary biological role, in potassium-poor environs, cesium is taken up as a substitute for potassium. Natural potassium contains 0.012% of the radioactive isotope K-40, with a halflife of 1.27 billion years. K-40 contributes most of the radioactive burden in the average human body. There is thus some reason to believe that evolutionary processes that might provide some bodily protection against radioactive K-40 might also protect against analogous harms from Cs-137. Cs-137 has the relatively high reference guideline of 200 pCi/kgwet.

In most cases, elevated Cs-137 provides a public warning of the presence of radioactive fission products in the environment. In 2003, TRAC reported traces of Cs-137 seeping into the Rio Grande from Los Alamos National Laboratory, as an "early warning." TRAC also reported Cs-137 from fallout in 2003, at a level of "public health concern," downwind of the Department of Energy's Savannah River Site (SRS) in South Carolina. Downwind of SRS, Cs-137 was at least a factor of ten higher than reported here, downwind of LLNL.

Am-241: Americium-241 is a long-lived radionuclide, with a halflife of 433 years. Am-241 is a byproduct of production of artificial plutonium by neutron bombardment of natural uranium-238. Am-241 exhibits a distinctive photopeak at 59.5 KeV, making Am-241 a readily detectible fingerprint of plutonium.

Americium, plutonium, and other alpha-particle-emitting *actinides* warrant special attention because of their radioactive toxicity. The actinides concentrate and remain in bones, kidney, and liver tissues, where their alpha radioactivity is carcinogenic.

Radiological studies in 1996 and 1997 —after the world's largest underground nuclear explosion, 5 megaton yield "Canninkin" on 6 November 1971, under Amchitka Island in Alaska's Aleutians— reported Am-241 at one pCi/kgwet in aquatic vegetation. That evidence of leakage of artificial actinides from U.S. nuclear weapons testing into the aquatic environment has prompted responsive governmental actions that continue. Although the reported Am-241 content of Sample 6 of the present study is greater than the radioactivity of the Amchitka samples, the Am-241 content of Sample 6 is far below the official guideline of concern, 15 pCi/kgwet for gross alpha radioactivity.

For more information:

For a comprehensive background to the subject of radioactivity in the environment, see Merril Eisenbud's **Environmental Radioactivity from Natural, Industrial, and Military Sources**, published by Academic Press.

Check out <www.radioactivist.org> to see how this study compares to other TRAC projects. Go to <www.resolve.org> for information about other studies funded by the MTA Fund.

For information about LLNL's Environmental Community Relations program and environmental monitoring around LLNL by government agencies, go to <www-envirinfo.llnl.gov>.

To learn of citizens' existing concerns for pollution from LLNL, see Tri-Valley CAREs' website at <www.trivalleycares.org>.

To see how LLNL's national security mission fits into the bigger picture of our society and its democratic institutions, visit the Western States Legal Foundation's website at <www.wslfweb.org>.

Appreciation

TRAC thanks Bert Heffner and LLNL staff for technical and logistic interfacing and background for this study, Marylia Kelley of Tri-Valley CAREs for a community perspective, the Alameda County Plutonium Action Taskforce for a pre-existing reference point of community concerns, and those individuals who expressed their own concerns.

This study is supported by a grant from the Citizens' Monitoring and Technical Assessment Fund (MTA Fund).

Please send your comments or questions regarding this data report to the author. Your feedback will help TRAC provide the most useful information in Rev.2 of this report and in subsequent outreach materials. Thank you.

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