Radiological Results of Samples from Some Potential Pathways from the Lawrence Livermore National Laboratory (LLNL) into the Surroundings --Part 2 (Rev.1)

by Norm Buske, Director, The RadioActivist Campaign
<search@igc.org> July 30, 2004

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. That initial sampling established a basis to independently assess candidate ***pathways*** of radionuclides from this *world premier scientific center* into its neighborhood. See Part 1 (Rev.3) for a description of the December radiological study and its results. (Numbering of pages and tables continues here from Part 1.)

TRAC collected follow-up samples from around LLNL in May 2004. Those radiological results are reported here. TRAC plans to publish a radiological assessment of the results from both samplings, in September 2004.

TRAC's main concerns are for airborne and waterborne pathways that carry readily detectable, 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.

Sampling Narrative

TRAC staff returned to Livermore on 6 May 2004. The last measurable rain had fallen on 19 April. Vegetation was mature and drying, in comparison to young and green when TRAC had last sampled in December 2003.

After TRAC reported the radiological results of its December 2003 sampling, a question arose: Might those positive results be attributed to downwind fallout from the Vallecitos Atomic Lab, 10 miles southwest of LLNL. TRAC collected 0.77 kg*wet* (Sample 19) of mixed, mature grasses from the second drainage wash east of the atomic lab, crossing Highway 84 from the north side. (Grass sample size was increased after December 2003 to improve detection.)

TRAC then replicated Sample 3, young grass from Flynn Pass collected in December 2003 from northeast of LLNL, by collecting 0.91 kg*wet* mature grass Sample 13. Sample 13 was collected from the south side of S. Flynn Road, instead of the north side, because of greater abundance of grass.

TRAC had sampled flowing surface water and sorrel in Arroyo Seco, on the west side of S. Vasco Road, downstream of LLNL, in December 2003. On 7 May, TRAC collected aquatic moss from that same flow. The aquatic moss was repeatedly washed in the flow in the arroyo from LLNL's A1 Groundwater Treatment facility, on the east side of the road. Sample 17 was hand pressed to remove water, leaving 0.98 kg*wet* of wet moss.

Sample 6 of reed grass collected in December 2003 from Arroyo Las Positas, downstream of LLNL, on the east side of S. Vasco Road had yielded positive results for iron-59, cesium-137 (Cs-137), and Americium-241 (Am-241). TRAC focused on this location for follow-up sampling on 8 May 2004. Sample 14 was 0.88 kg*wet* of mature grasses picked from the arroyo bed. Sample 15 was 21.2 kg*wet* (= liters) of free flowing, clear surface water drawn unfiltered and without preservative. TRAC collected terrestrial moss with some algae, worms, and other biota from the concrete channel under the bridge. The moss sample was repeatedly washed in arroyo water on a 2 mm stainless steel screen and then hand pressed to remove excess water. Sample 450810 was 0.48 kg*wet*, primarily moss.

In December 2003, TRAC had collected two samples of tree leaves down gradient, relative to the groundwater flow, from Pit 6 at Site 300. Radiological results were negative. On 8 May 2004, TRAC collected leaves from a willow tree growing in Corral Hollow Creek, east of Site 300. Sample 18 was 0.81 kg*wet* of leaves with their stems, picked from the north side of the bridge deck on Corral Hollow Road. This sample location was both downwind and downstream of Site 300.

On 8 May, TRAC also collected 0.85 kg*wet* of mixed, mature grasses from the west side of Camino Tassajara, north of the county line. This Sample 20 was collected about 9 miles northwest of LLNL to provide a definitive background for the other grass samples in the study.

Methods Summary

See Part 1 of this report for description of general methods. Note that TRAC analyzes only for radioactivity releasing intense photons above 3 KeV energy. Tritium (H-3) and plutonium (Pu-239, Pu-240) are practically undetectable by this method. Such radionuclides are not screened well by TRAC's single-pass procedure. TRAC's single pass procedure can detect about a hundred different, artificial radionuclides, at possible environmental levels. Furthermore, Am-241, the decay daughter of Pu-241, usually accompanies plutonium and so may flag the presence of plutonium in a sample.

Also note that the overall analysis is biased slightly negative, for the express purpose of avoiding false positive reports that might raise unfounded concerns.

Moss samples were repeatedly washed in the surface water from which they were collected and then hand pressed to remove excess water. All samples were screened for radioactivity at lab entry. The water sample was quiescently evaporated, by microwaves, to paste on plastic film. All vegetation samples were oven dried <100C. Dried grass and other samples were then charred <250C to improve detection by a factor of two.

Cs-137 is a routine TRAC laboratory analysis, after thorium and uranium interferences have been subtracted. Likewise, Am-241, with its x-ray peak for confirmation, is a routine analysis. Iron-59 (Fe-59) is not a routine analysis for TRAC and was not detected in this set of samples from around LLNL. Uncertainties of the Cs-137 and Am-241 analyses are reported as "±" one standard deviation counting error, as generated by Canberra G2K software.

In Part 1, Table 1, Cs-137 was reported in young grass from Arroyo Las Positas, collected in December 2003 (Sample 6: 8.7±2.1 pCi/Kgwet). To compare the Cs-137 activity in that grass after it had matured, resampling in May 2004 A numerical report was forced for this sample that was known to contain Cs-1327, instead of allowing the routine report of "No Detect" (Table 3, below, Sample 14: –0.2±1.3 pCi/kg*wet*).

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. Sr-90 was not detected in any of the samples collected in May 2004. To compare Sr-90 in the mature grass from Flynn Pass that yielded a positive report for young grass (Part 1, Table 1, Sample 3: 190.±160. pCi/kg*wet*), a numerical report (Table 3, below, Sample 13: –100.±110 pCi/kg*wet*) was forced for the mature matured grass that had Sr-90 detected in the young grass, instead of allowing a routine report of "No Detect". The reported uncertainty of this result is one standard deviation of four analytical results.

No short-lived, artificial radioactivity was detected in any of the samples TRAC collected in May 2004.

Results

Preliminary radiological results appear in Table 3, on the next page, with negative results completed on the following page.

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 3 by "--" , meaning "Not Detected." Reports were forced for Sr-90 in one sample (Sample 13) and for Cs-137 in one sample (Sample 14) to obtain values for comparison to previous results."

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 kg*wet*

Uniform reporting in units of "kg*wet*" 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 3.

"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 3 and minutes tabulated.

"Sample Identifier" is the sample tracking number, which is the year, month, day, and hour of sample collection. For Sample 13, the Sample Identifier is 450610, where: the leading "4" = 2004; "5" = May; "06" = 6th day of May; and "10" = 10:00 hours = 10:00 AM, on a 24 hour basis.

Table 3. Preliminary Radiological Results from Second sampling.

Sample Number. Setting:	13. Flynn Pass	14. Positas North	15. Positas North
Sample Direction from LLNL:	far northeast	near northwest	near northwest
Location:	south side of S. Flynn Rd, west of N. Flynn Rd.	Arroyo Las Positas bed, east side of S. Vasco Rd, southeast of train station	as Sample 14
Medium (Material):	mature grasses	mature grasses	unfiltered water
Wet/Dry(Wet/Char) Weight Ratios:	2.8 (5.7)	3.9 (7.8)	21.16 kg*wet**
Photon **Radioactivity** [pCi/kg*wet*]			
Strontium-90 (Sr-90):	–100.±110.	--	--
Cesium-137 (Cs-137):	--	–0.2±1.3	0.027±0.011
Americium-241 (Am-241):	--	3.5±1.1	--
Latitude: North 37° + minutes:	42.337'	41.807'	41.807'
Longitude: West 121° + minutes:	38.696'	43.031'	43.031'
Sample Identifier:	450610	450816	450809

* Wet/dry ratio is not applicable to water. Mass of water is given instead.

Sample No. Setting:	16. Positas North	17. Seco West	18. 300 East
Sample Direction from LLNL:	near northwest	near west	east of Site 300
Location:	as Sample 14	Arroyo Seco bed, west side of S. Vasco Rd. bridge	Corral Hollow Creek, from Rd. bridge
Medium (Material):	washed moss	aquatic moss	willow leaves
Wet/Dry(Wet/Char) Weight Ratios:	5.1	8.9	2.4(5.0)
Artificial **Radioactivity**:	--	--	--
Latitude: North 37° + minutes:	41.807'	40.872'	39.393'
Longitude: West 121° + minutes:	43.031'	43.134'	29.204'
Sample Identifier:	450810	450717	450813

Table 3 concluded on the next page.

Table 3 Concluding, Preliminary Radiological Results from Second sampling.

Sample No. Setting:	19. Vallecitos	20. Tassahara
Sample Direction from LLNL:	far southwest	far northwest
Location:	wash E. of Vallecitos Atomic Lab, N. side of Hwy. 84.	Camino Tassajara, N. of county line, W. side of roadway
Medium (Material):	mature grasses	mature grasses
Wet/Dry(Wet/Char) Weight Ratios:	4.0(8.3)	2.5(5.2)
Artificial **Radioactivity**:	--	--
Latitude: North 37° + minutes:	36.839'	44.622'
Longitude: West 121° + minutes:	49.128'	52.307'
Sample Identifier:	450610	450815

Discussion

Although patterns of the artificial radionuclides reported in Table 3, above, and in Table 1 of Part 1 can be inferred from the descriptions of the sample locations, TRAC awaits completion of this study before drawing specific technical conclusions.

With the exception of Am-241 in grass in Arroyo Las Positas, artificial radioactivity in samples collected around LLNL in December 2003 had almost disappeared by May 2004. The Am-241 content of 4.5±2.0 pCi/kg*wet* (Sample 6) replicated with 3.5±1.1 pCi/kg*wet* (Sample 14). (The agreement was less close, on a dry weight basis. Uncertainty was decreased in Sample 14 in comparison to Sample 6 by analysis of charred grass instead of dried grass.)

Cs-137 in Sample 6 decreased from 8.7±2.1 pCi/kg*wet* in December 2003 to –0.2±1.3 pCi/kg*wet* in replicate Sample 14 in May 2004. Short-lived iron-59 had also disappeared from the grasses sampled from Arroyo Las Positas, downstream of LLNL.

Samples of grass (Sample 4) and water (Sample 5) that had been collected in December 2003 from Arroyo Las Positas east of LLNL had 2.9±1.6 and 0.03±0.016 pCi/kg*wet* of Cs-137 respectively. The ratio of Cs-137 in grass/water is one measure of the bioaccumulation factor for cesium [BF(Cs-137)] in Arroyo Las Positas grass. Based on the stated uncertainties, the range of BFs for cesium is:

$$28 < BF(Cs\text{-}137) < 250$$

The same BF range calculation can be made for Cs-137 in grass Sample 14 (–0.2±1.3 pCi/kg*wet*) and water Sample 15 (0.027±0.011 pCi/kg*wet*) for May 2004, disallowing a negative BF value:

$$0 < BF(Cs\text{-}137) < 69$$

The overlap of these two calculations of BF is:

$$28 < BF(Cs\text{-}137) < 69$$

The calculated BF ranges overlap enough to be considered consistent for Cs-137 in grasses growing in arroyos around Livermore.

If the BFs are somewhat consistent, that consistency suggests some connection between Cs-137 in running surface waters around LLNL and in grasses growing in those waters. That is, the Cs-137 activity in Arroyo Las Positas grass of 8.7±2.1 pCi/kg*wet* (Sample 6) just downstream of LLNL in comparison to 2.9±1.6 pCi/kg*wet* (Sample 5) just upstream of LLNL, suggests roughly tripling of Cs-137 in arroyo grass, *attending* surface water passage through LLNL. This is evidence of a source of Cs-137 in into arroyo water passing through LLNL, in December 2003.

Suppose a middle value of BF=50 is used to estimate the Cs-137 activity in Arroyo Las Positas water corresponding to 8.7±2.1 pCi/kg*wet* in arroyo grass in Sample 6, collected in December 2004. The Cs-137 activity in the arroyo water at that time would have been 0.17±0.04 pCi/kg*wet*. (8.7/50=0.17.) That compares to the activity of 0.027±0.011 pCi/kg*wet* measured in the arroyo water in May (Sample 15). That is to estimate the Cs-137 activity in downstream arroyo water to have **decreased six-fold** in the 5 months between December and May. (0.17/0.027 = 6.3.)

There is no reason for cesium in growing grass to be in instantaneous equilibrium with the cesium in stream water. The low activity of Cs-137 (<1.1 pCi/kg*wet*) in Sample 14 might be better explained by dilution of the 8.7±2.1 pCi/kg*wet* in the young grass of Sample 6 in December by the season's growth practically without additional Cs-137 contamination.

Another way of making the same point is to notice that 8.7±2.1 pCi/kg*wet* of Cs-137 in December grass Sample 6 had grown into <1.1 pCi/kg*wet* in the mature grass of Sample 14. That is to say, the Cs-137 content of the Arroyo Las Positas grass had **decreased at least six-fold** in the 5 month growing season. [(8.7-2.1)/(<1.1) > 6.]

Previous results for Samples 4 and 5 collected to the east (upstream and downwind) of LLNL can be compared to Samples 6 and 7 collected northwest (downstream and upwind) of LLNL; see Table 1 of Part 1. Such a comparison suggests that grass collected from Arroyo Las Positas, just down stream of LLNL (at Sample 14 location), is an advantageous sample medium and location for radiological monitoring of airborne and waterborne emissions from the nuclear weapons facility.

Merril Eisenbud [and T. Gesell, Environmental Radioactivity, 4th ed. Academic Press, San Diego (1997) 90-107] describes chemical processes that tightly bind metal atoms, like cesium-137, to clay particles in soils. Therefore, almost all radioactivity remaining around Livermore, from atmospheric weapons tests in the 1950s and 60s, would remain tightly bound to soil particle surfaces; or else it would have washed away from the LLNL landscape, decades ago because it had *not* been tightly bound. Thus, the artificial radioactivity tabulated in water and vegetation samples around LLNL in Part 1 of this report cannot be attributed to remains from worldwide fallout.

The near disappearance of detectable, artificial radioactivity from the environment around LLNL in the five months from December to May indicates that radioactivity was *superficial*. Fairly consistent BF~50 values suggest the transient superficiality of long-lived, artificial radioactivity around Livermore might be due to dilution by uncontaminated rainwater, storm water, and pumped-and-treated water discharged from LLNL.

In the five months between sampling in December 2003 and re-sampling in May 2004, the Cs-137 activity in grass in Arroyo Las Positas decreased by a factor of about six. The effective "halflife" of Cs-137 in this grass, attributable to dilution by nearly uncontaminated rainwater, storm water, and/or pumped-and-treated water is thus calculated to be about two months. ("Halflife" is the time during which the radioactivity in a sample decreases to half its initial value. Halflife usually refers to decrease in radioactivity due to radioactive decay. The term is here applied to the sum of all processes causing reduced radioactivity in a particular sample medium.)

Observation of the 0.57 inch rainstorm of 14 December 2004 suggested there might often be little storm water runoff after less than one-quarter inch of rain. That is, rainfalls at least 0.25 inches might correspond to enough storm water runoff to wash superficial radioactivity off the ground, plant leaves, buildings, and other surfaces.

For the purpose of this study a "rainstorm" in Livermore is evidenced by daily rainfall of at least one-quarter inch. Weather Service data for rainstorms before and during this study are summarized in Table 4.

Table 4. Rainstorms in Livermore, Before and Between Sampling Periods.

Code: (day number) Rainstorm = Rainfall ≥0.25 inches   sampling period

Month	Total Rainfall Inches						
JUL '03	0.00	no rainstorms in month					
AUG	0.29	no rainstorms in month					
SEP	0.00	no rainstorms in month					
OCT	0.02	no rainstorms in month					
NOV	2.02	(03) 0.27	(08) 0.28	(09) 1.08			
DEC	3.57	(07) 0.35	(10) 0.27	(14) 0.57 (18)		(25) 0.25	(29) 0.72
JAN '04	2.19	(01) 1.62				(24) 0.28	
FEB	4.01	(02) 0.72	(16) 0.46	(18) 0.69	(20) 0.37	(25) 0.60	(26) 0.28
MAR	0.39	no rainstorms in month					
APR	0.18	no rainstorms in month					
MAY	0.11	(06)	(08)	no rainstorms in month			

There were 5 rainstorms in Livermore, in the months preceding sampling in December 2003. Those all occurred in the month and a half just before sampling. TRAC sampled the rainstorm of 14 December. Then there were 10 rainstorms between the first sampling period and the second. Those 10 rainstorms all occurred in the first two and a half months of the 5 month interval between sampling periods.

These rainstorm data are consistent with a *dilution halflife* of two months (see above) provided the source of Cs-137 to the Livermore surroundings ceased before the end of December 2003.

At the public meeting at Tri-Valley CAREs, on 6 May 2004, a concerned citizen complained that as soon as TRAC finished monitoring in May, LLNL would start "releasing" again. The data in Tables 1 and 3 are consistent with that hypothesis. The data are also consistent with "releases" of measurable artificial radioactivity —not tritium— from LLNL virtually ceasing for the interval of TRAC's monitoring program.

TRAC informed LLNL that the simplest explanation of these data is that "LLNL virtually ceased occasional and continuous releases of gamma radioactivity to its main environmental pathways between late December 2003 and early May 2004." TRAC invited LLNL to reply "for the record" to this anticipated conclusion. LLNL's Bert Heffner has replied that "'for the record', it does not seem appropriate for me to comment until [the] report is published."

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>.

See Part 1 of this report for a general description of the radionuclides mentioned here in Part 2. Also see Part 1 to see where to go for "Appreciation" for people and organizations who are contributing to this study.

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

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