Preliminary Radiological Results of Samples from Potential Pathways from the Los Alamos National Laboratory (LANL) into the Public Domain.

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Introduction

In response to public concerns, Concerned Citizens for Nuclear Safety (CCNS) and The Radioactivist Campaign (TRAC) began technical exploration of LANL's possible radiological effects on public lands in June 2002. Based on that introduction, TRAC and CCNS collaborated on a raft trip, sampling down the Rio Grande, past LANL, in October 2002. Preliminary results of samples collected from the public domain next to LANL are reported here.

Methods Summary

Samples were collected and analyzed for radionuclides in an integrated "survey" procedure. TRAC developed this procedure to identify artificial radioactivity "above background" levels in candidate pathways from nuclear facilities, over the last 20 years. This integrated survey procedure differs from ordinary (EPA) procedures in important ways that substantially affect interpretation of reported results.

TRAC's survey procedure addresses public concerns for radioactivity entering public neighborhoods around nuclear facilities. Such concerns span a range of questions from whether "any" artificial radioactivity occurs offsite to possible environmental and public health effects. In consideration of such concerns, TRAC preferentially samples vegetation that often bioaccumulates radioactivity from identifiable surface water, groundwater, and airborne pathways from nuclear facilities. Sample selection and analysis steps optimize identification of the widest range of likely radionuclides, at the lowest detection levels, while minimizing false positive results.

TRAC employs no toxics nor artificial radionuclides in sample preparation, employs only archived and public-domain radionuclide reference standards, and produces no toxic nor radioactive wastes inconsistent with the original "environmental sample" materials, in accord with the spirit of public-interest science conducted in the public domain (without government licensing). Preparation consists of drying samples either in a conventional or microwave oven and then homogenizing the samples into a 0.27 specific gravity matrix in 130mL PET counting bottles. Samples are analyzed in a single, 23-hour pass through a broadband (10-4000KeV), highly stabilized, deep-well NaI, photon spectrometer that allows true spectral subtractions. (The high precision of HPGe detectors introduces offset errors from simple spectral subtractions.)

Subtraction of background and reference spectra allows detection of x-ray, gamma, and pure beta emitting radionuclides. Pure beta emitters are detected by multi-point spectral matching, including effects of bremsstrahlung, compton scattering, and other interactions with the detector. All spectra are acquired on 8000 energy channels and then transformed to constant photopeak width (FWHM) 3 channels, yielding 165 transformed channels.

This transformation and true spectral subtractions of appropriate reference spectra allows exceptionally low detection levels, subject to safeguards against false-positive, secondary effects. One safeguard involves counting only in “negative” regions of the processed energy spectra. Negative bias of this safeguard precludes meaningful reporting of nil and negative analytical results.

Reportability of analytical results is usually limited by quality of background reference materials.

Reliability of reported results relies on independent reproducibility. TRAC retains field notes and analytical records, and analyzed samples are archived and released for independent analyses. Sample locations are within the public domain, and sample media are reported along with the results, allowing completely independent sampling and confirming analyses. TRAC provides opportunities for government agencies and other parties to sample jointly and to compare radiological results.

Background

The public has expressed concerns that LANL wastes might be leaching along groundwater pathways into the Rio Grande. Responding to these concerns, TRAC began collecting vegetation samples from surface drainages and from springs entering into the river. Other samples have been collected for reference and other purposes, including “background,” and analyzed.

Surface and groundwater pathways from LANL can be partly understood by evaluation of beryllium-7 in water and in biological samples that (always) contain water. Beryllium-7 (Be-7), with a halflife of 53.3 days, provides a useful clock for the age-since-rainfall of the water in such samples. That is, Be-7 provides a measure of the fraction of (superficial) water that derives from recent rainfall in a sample, in comparison to the fraction of older surface and/or groundwater in a sample.

The element beryllium is taken up by vegetation because it mimics the biologically-essential, chemical element magnesium. Magnesium is incorporated into chlorophyll in green plants and is a vital trace mineral in the human diet. Magnesium is incorporated into bones and is necessary for proper function of nerves and muscles, including the heart. Beryllium is thus accumulated by biota in the place of magnesium, but the mimicry is imperfect, and beryllium is toxic.

The radioactive isotope Be-7 is a useful indictor of “age” of water in samples of vegetation samples because of this bioaccumulation, which depends on the species, availability of competitive magnesium, biological retention time of water, etcetera.

Beryllium-7 is primarily of cosmogenic origin. Cosmic rays enter the Earth’s upper atmosphere, strike nuclei of nitrogen and oxygen atoms in air, and chip fragments off these atoms. Some of the nuclear fragments are unstable and decay.

The most abundant, radioactive fragments are atoms of tritium (H-3), Be-7, and carbon-14 (C-14); see Table 1.

Table 1. Cosmogenic Radioactivity in the Atmosphere

Radionuclide	Halflife [years]	Activity [pCi/m^3]
H-3	12.3	4.5 pCi/m^3
Be-7	0.145	0.5 (varies with altitude)
C-14	5760.	1.8

Reference: R.L. Kathren, RADIOACTIVITY IN THE ENVIRONMENT: Sources, Distribution, and Surveillance, Harwood Academic Publishers, Chur, Switzerland (1984) Table 2-5.

Be-7 is produced in the atmosphere at the rate of 800 atoms/m^2 per second. This cosmogenic Be-7 falls to earth in rain, both in dissolved and in particulate fractions.

The 53.3 day halflife of Be-7 is useful for evaluation of the contribution of recent rainwater in either a water sample or a biological sample in which beryllium has accumulated. Relatively high activities of Be-7 indicate a large fraction of rainwater in a sample; whereas, relatively low activities of Be-7 indicate a greater fraction of non-rainwater, such as “regional groundwater” or water stored in snowpack or reservoirs.

Be-7 is easily detected by emission of a photon of 477.6 KeV during decay by electron capture, yielding a stable atom of lithium-7. Beryllium-7 ordinarily migrates in groundwaters that carry divalent cations. Beryllium and magnesium belong to Group 11A of the Periodic Table of the Elements, along with calcium, strontium, barium, and radium.

The interest in Be-7 as an indicator of age (since time of rainout) of water both in water samples and in vegetation invites common units of measurement for both water and vegetation samples. Yet radioactivity in water samples is ordinarily reported as activity per liter; while radioactivity in vegetation is usually reported as activity per gram, dry weight.

Noting that a liter of water has a mass of one kilogram, the unit of radioactivity used throughout this preliminary report is:

picocuries/kilogram wet weight [pCi/Kg(wet)]
--where 1 pCi = one nuclear disintegration per 27 seconds.

To obtain activities on a per gram(dry) basis, multiply the tabulated activity by the listed wet/dry ratio for a sample and divide by 1000. The biological concentration factor (CF) for Be-7 in vegetation is the ratio of tabulated Be-7 divided by the sum of dissolved and particulate Be-7 in co-collected water.

Other radionuclides reported here are strontium-90 (Sr-90) and cesium-137 (Cs-137). These two radionuclides have halflives of almost 30 years and are produced by nuclear fission in reactors and in nuclear explosions. Both Sr-90 and Cs-137 are still detected worldwide from historic atmospheric testing of nuclear weapons, and both radionuclides are produced by LANL processes.

Guidelines for surface water quality			
Be-7	6000.	pCi/Kg	(EPA-570/9-76-003)
Sr-90	8.	"	(40 CFR 141)
Cs-137	200.	"	(EPA-570/9-76-003)

Radioactive Sr-90 is ordinarily identified by beta analysis after strontium is isolated chemically. During decay, Sr-90 emits one electron at 546 KeV and then another electron at 2186 KeV. These energetic electrons interact with atoms in the material containing the decaying Sr-90 and with various photon detectors, providing a spectral signature that is identifiable. Strontium-90 ordinarily migrates in groundwaters that carry divalent cations. Strontium mimics biologically essential calcium. Strontium and calcium are Group 11A elements, along with beryllium and magnesium, already mentioned.

Radioactive Cs-137 is identified by its 661.7 KeV photon emission accompanying beta decay to stable barium-137. Cesium-137 ordinarily migrates in groundwaters that carry monovalent cations. Cesium is listed in Group 1A of the Periodic Table of the Elements, along with sodium and potassium, both of which are biologically essential elements.

Results

Table 2, below, is arranged in a geographical order, with results from samples collected farther downstream along the LANL shoreline of the Rio Grande lower in the table. Results for two samples from the LANL plateau appear at the bottom of Table 2, below the dashed line: "-----".

Table 2. Preliminary Radiological Data

Sample				picocuries/kilogram (wet)		
Location	Setting	Medium	Wet/Dry	Be-7	Sr-90	Cs-137
Mortandad	stream	willow leaves	4.62	260.	--	--
CCNS	spring	unidentified*	2.83	1300.	--	--
#3	spring	willow leaves	2.53	2300.	Bkg.	Bkg.
#4A	spring	aquatic moss	6.85	30.	[150.]	<MDA
below #4A	stream	aquatic moss**	6.70	1200.	<MDA	10.
"	"	water, dissolved	--	0.48	--	--
"	"	water, particulate	--	0.56	--	--
"	"	sediment <2mm**	1.00	160.	<MDA	50.
#5	spring	willow leaves	2.59	2500.	--	--
Water Canyon	stream	willow leaves	2.67	3300.	--	--
Ancho conflu.	river	aquatic moss**	10.2	20.	[90.]	--
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culvert	wash	willow leaves	2.94	1400.	--	--
Met. Station	plateau	sage leaves	2.64	1400.	1100.	--

Be-7 - activity corrected to time of sample collection.

* leaves from unidentified yellow composite flower, nonconforming medium.

Bkg. - sample used as background for "willow leaves" medium in data set. Reportable values for this medium are "above Bkg."

** - sample rinsed in source water to remove suspended material. Other samples were not cleaned.

[ ] - reportable result fails one or more quality assurance criteria. The two data did not have adequate "background" reference material of the "aquatic moss" media.

<MDA - unreportably low, positive result is below minimum detectable activity.

-- - nil or negative result is not reportable by this analytical procedure.

Sample locations and rationale are listed in Table 3, on the next page.

Table 3 Sample Locations

Location	Medium	North 35°	West 106°	Number*	Rationale
Mortandad	willow leaves	49.695’	10.261’	2x1011	Most upstream candidate drainage location.
CCNS	unidentified	49.356’	10.649’	2x1012	Newly discovered spring, possibly of new series.
#3	willow leaves	49.179’	10.729’	2x1013	Sample Series 3 spring.
#4A	aquatic moss	48.244’	11.827’	261608	Pathway: spring close to plateau, with large discharge into river.
below #4A	aquatic moss	48.143’	11.725’	2x1015	Compare Spring #4A
“	water	“	“	2x1017	with increasing stream
“	sediment <2mm	“	“	2x1016	below spring. Break out constituents.
#5	willow leaves	47.283’	11.864’	2x1110	Sample Series 5 spring.
Water Canyon	willow leaves	47.121’	12.162’	2x1111	Sample large drainage.
Ancho conflu.	aquatic moss	46.207’	13.142’	2x1112	Sample confluence of Ancho stream with Rio Grande.
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culvert	willow leaves	49.764’	14.609’	2x1313	Sample drainage from northwest side of LANL for comparison.
Met. Station	sage leaves	52.520’	15.271’	2x1312	Pathway: biological sample downwind of LANSCE accelerator complex.

* Sample Number digits are: Year Month Day Day Hour Hour. Month is numeric, with October as “x”, November as “y”, and December as “z”. Hours count to 24, as military time. “2x1312” is 2002, October 13, 12:00 noon.

On the basis of the results reported in Table 2, TRAC followed-up sampling along the Rio Grande from April 30 through May 02, 2003, along with CCNS and other interested parties.

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