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Produce and Fish Sampling Program of Los Alamos National Laboratory's Environmental Surveillance Group

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PRODUCE AND FISH SAMPLING PROGRAM OF LOS ALAMOS NATIONAL LABORATORY'S ENVIRONMENTAL SURVEILLANCE GROUP

by

John G. Salazar

ABSTRACT

This report describes produce and fish sampling procedures of the Environmental Surveillance Group at the Los Alamos National Laboratory. The program monitors foodstuffs and fish for possible radioactive contamination from Laboratory operations. Data gathered in this program on radionuclide concentrations help to estimate radiation doses to Laboratory personnel and the public.

I. INTRODUCTION

As part of the routine environmental surveillance program at Los Alamos National Laboratory, radiation doses from ingestion of produce and fish are estimated. The sampling of produce and fish also helps to determine background levels of potential environmental contaminants and to detect any above-background concentrations caused by Laboratory activities.

II. PRODUCE AND FISH SAMPLING PROGRAMS

A. Sampling Program Design

1. Fruit, Vegetable, and Fish Samples. Fruit, vegetable, and fish samples are collected to monitor possible radioactive contamination from Laboratory operations. Produce samples are collected during the fall harvest in the Los Alamos area and above and below points where intermittent streams that cross the Laboratory join the Rio Grande. Fish samples are collected during the spring, summer, and fall at Cochiti, El Vado, Heron, and Abiquiu Reservoirs.

2. Sampling Locations. Produce samples are categorized in three groups (background, offsite, and onsite). Background samples are collected from various farmers in the Española vicinity (Fig. 1). The Rio Grande and Rio Chama are the main sources of irrigation for this area. These locations are suitable for background samples because they are upstream from the Laboratory and not affected by Laboratory operations. Offsite locations are in the communities of Cochiti, White Rock, Pajarito Acres, and Los Alamos (Fig. 1). Cochiti is dependent on the Rio Grande for irrigation, whereas the others use water from deep wells in the Los Alamos Community water system. Onsite locations are all within the Laboratory's boundaries. Produce harvested from onsite locations is all fruit and its water source is from precipitation.

There are two groups of fish samples: background and offsite. Abiquiu, El Vado, and Heron Reservoirs are used as background sampling locations. They are located on the Rio Chama/Willow Creek drainage and are far enough upstream from the Laboratory that airborne emissions do not affect them (Fig. 1).

The offsite location is Cochiti, which is on the Rio Grande downstream from the Laboratory. There are no

Fig. 1. Northern New Mexico region.

onsite sampling locations, because there are no bodies of water that are large enough for fish.

3. Sampling Frequency and Type. Produce and fish samples are collected once a year. Harvest for produce begins in early August and extends through September. Produce collected are garden-variety squash, cucumbers, chile, corn, carrots, melons, tomatoes, peaches, apricots and apples (Fig. 2).

Fish samples are collected between May and September. Fish types include trout, salmon, bass, walleye, catfish, suckers, and carp (Fig. 3).

4. Sampling Methods and Sample Preparation. During the harvest, produce is bought directly from local farms (Fig. 4). Maps are drawn to show sampling locations within the gardens. This documentation is essential if above-background concentrations of radioactivity are found. The following steps are used for produce samples.

Fig. 2. Fruits and garden-variety vegetables are collected.

Fig. 3. All types of fish caught are used in the fish sampling program.

Fig. 4. Fruits and vegetables are purchased from area farmers.

- a. Samples are placed in plastic bags and labeled.
- b. In the laboratory, produce samples are washed and towel dried, as they would be before being eaten.
- c. Samples are diced and put into a tared 1000 mL beaker.
- d. Wet weights are determined.
- e. Five milliliters of moisture is distilled from produce samples for tritium analysis.
- f. Samples are oven dried at 100°C (~48 h).
- g. Oven-dried weights are determined.
- h. Samples are ash dried at 400°C (~72 to 96 h).
- i. Ash weights are determined.
- j. Samples are submitted for analysis.

There are two groups of fish samples: higher-level feeder (game fish) and bottom-level feeder (nongame fish). The fish are dissected and the entrails and muscle tissues kept in separate beakers (Fig. 5). Steps c, d, f, g, h, i, and j from the produce sampling apply.

5. Radionuclides Sampled. Both produce and fish samples are analyzed for $^{238,239}\text{Pu}$, U, ^{137}Cs , and ^{90}Sr concentrations. Only produce samples are analyzed for tritiated water (HTO or T_2O) (Fig. 6). These radionuclides are most likely to be released into the environment by Laboratory activities.¹

B. Sampling Equipment

The Environmental Surveillance Group (HSE-8) has been granted a fish collecting permit from the New Mexico Department of Game and Fish. This permit is

Fig. 5. Dissected fish are placed into beakers.

issued yearly and must be in possession of the permittee during fish collecting trips.

HSE-8 currently uses a 5 meter (17 ft), modified V-hull boat with an 80 hp motor for fish sampling. Gill nets are used to catch the fish [(1-1/2 mesh, 30 x 1.3 meter (100 ft by 6 ft)] (Fig. 7). Tubs, meat cleavers, knives, and wood-cutting blocks are used in the produce and fish sampling procedures.

Fig. 6. Vegetable samples are heated for tritium analysis.

C. Analytical Methods¹

1. **Tritium.** Only produce samples are analyzed for tritium. Water (H_2O , HTO , T_2O) is distilled (4 h at 120°C) from produce samples and collected. A 5 mL aliquot of the collected water is then counted by liquid scintillation. Fifteen milliliters of scintillation liquid is added and the solution vigorously shaken. The sample is counted in a liquid scintillation counter for 50 min or 10 000 counts, whichever comes first. Quality control standards and blanks are counted in conjunction with each set of samples.

2. **Plutonium 238 and 239.** Produce and fish samples are burned to ash in high-temperature ovens and then leached with HF-HNO_3 . All samples are spiked with standardized ^{242}Pu and ^{243}Am during dissolution to serve as a chemical recovery tracer.

Dissolved samples are thoroughly digested in 7.2 M HNO_3 , and 1M NaNO_2 is added to ensure that plutonium is in the tetravalent state. The solution is passed through a preconditioned anion-exchange column. The initial eluate and the first 20 mL of a 7.2 M HNO_3 wash is saved for ^{241}Am analysis. The column is then washed with 7.2 M HNO_3 and 8 M HCl . Plutonium is eluted with a freshly prepared solution of 1 g/L NHI in 1M HCl . The

Fig. 7. Gill nets are used to catch fish.

eluate is appropriately conditioned, and plutonium is electrodeposited from a 4% solution of $(\text{NH}_4)_2\text{C}_2\text{O}_4$. The plated plutonium is counted on an alpha spectrometer. Values reported for ^{239}Pu are the sum of ^{239}Pu and ^{240}Pu , because both have identical alpha energies.

3. **Uranium.** Samples are analyzed for total uranium by delayed neutron activation. The samples are irradiated in a thermal neutron port at Los Alamos National Laboratory's Omega West Reactor and pneumatically transferred to a neutron counter where the delayed neutrons produced by the fission of ^{235}U are measured. Total uranium is calculated assuming the natural $^{235}\text{U}/^{238}\text{U}$ ratio of 0.0072.

4. Cesium 137. One hundred grams of ash are weighed into 500 mL polyethylene bottles and filled with 7.2 mL HNO₃. The amount of ¹³⁷Cs is determined by counting on a Ge(Li) detector coupled to a multichannel analyzer. The activity is calculated by direct comparison with standards prepared in the same geometrical configuration as the samples. Gross gamma activity is measured by counting in an NaI(Tl) well counter, which accommodates the 500 mL bottles. A single-channel analyzer adjusted to register gamma radiation between 0 and 2 MeV is interfaced to the detector. Gross gamma determinations are reported as net counts per unit time and unit weight.

5. Strontium 90. Sample preparation and dissolutions are similar to those described in the section on plutonium. After dissolution, the residue is dissolved in HCl, the pH is adjusted to 2, and yttrium is separated from strontium by extraction into 20% HDEHP in toluene. The isolated ⁹⁰Sr is left undisturbed for 2 weeks to allow the daughter ⁹⁰Y to attain radioactive equilibrium. After that period, inactive yttrium carrier is added and ⁹⁰Y is again extracted from ⁹⁰Sr by solvent extraction into 5% HDEHP in toluene. Yttrium is back extracted into 3 M HNO₃ and precipitated as the hydroxide. Yttrium hydroxide is redissolved and the oxalate is precipitated. This precipitate is oven fired to the oxide and then filtered and weighed to determine the chemical yield. Yttrium oxide precipitate is counted on a gas proportional counter to measure the activity. Samples are recounted after 3 days to verify the separation of ⁹⁰Y from other beta-emitting nuclides.

D. Data Analysis

1. Statistical Treatment of Data. There are three groups of produce samples: background (Rio Chama and upper Rio Grande at Española), offsite (Cochiti, Los Alamos, White Rock, and Pajarito Acres), and onsite (any fruit bearing trees on Laboratory property) (Table I). The two groups (game and nongame fish) of fish samples are subcategorized into bottom-level feeders and higher-level feeders (Table II).²

2. Variance, t Test, Degrees of Freedom.³ The variances are calculated by using the following equation:

$$S^2 = \frac{\sum_{i=1}^N (C_i - \bar{C})^2}{n - 1}$$

where

C_i = radionuclide concentration in sample i ,
 $\bar{C}$ = mean radionuclide concentration of the n of samples,
 n = number of samples, and
 S^2 = variance of radionuclide concentrations in a group.

A Student's t test³ is used to determine if there are any statistically significant differences among group means:

$$t = \frac{\bar{C}_1 - \bar{C}_2}{\sqrt{\frac{S_1^2}{n_1} + \frac{S_2^2}{n_2}}}$$

where

$\bar{C}_1$ = mean radionuclide concentration of group 1,
 $\bar{C}_2$ = mean radionuclide concentration of group 2,
 S_1^2 = variance of group 1,
 S_2^2 = variance of group 2,
 n_1 = number of samples in group 1, and
 n_2 = number of samples in group 2.

The degrees of freedom (df) are calculated as follows.

$$df = \frac{\left(\frac{S_1^2}{n_1} + \frac{S_2^2}{n_2}\right)^2}{\left(\frac{S_1^2}{n_1}\right)^2 + \left(\frac{S_2^2}{n_2}\right)^2} - 2$$

The two-sided critical region at level of significance $\alpha = 0.05$ (that is, 5%) is calculated as shown.

$$t \leq t_{\alpha/2}, t \geq t_{1 - (\alpha/2)}$$

where

t = calculated Student's t percentile, and

$t_{\alpha/2}, t_{1 - (\alpha/2)}$ = percentiles of Student's t distribution at $\alpha/2$ level of significance (two-tailed test).

3. Data Handling. Currently, analytical results from produce and fish samples are stored in the Environmental Surveillance Group's minicomputer with information such as sample number, sample type, location, weights,

TABLE I
RADIONUCLIDE CONTENT OF FRUITS AND VEGETABLES COLLECTED DURING 1983

Location: Water Source:	Background		Offsite			Onsite		
	Española Rio Chama	Española Rio Grande	Cochiti Rio Grande	Los Alamos Community System	White Rock/ Pajarito Acres Community System	TA 3	TA 21	TA 34
Radionuclide:								
²³⁸Pu (pCi/g dry weight)								
No. of Samples	5	10	15	3	10	3	2	1
Maximum ^a	0.00075 ± 0.00050	0.00043 ± 0.00043	0.00024 ± 0.0014	0.00031 ± 0.00027	0.00083 ± 0.00067	0.00071 ± 0.00024	0.00049 ± 0.00081	...
Minimum ^a	-0.00012 ± 0.00035	-0.00030 ± 0.00030	0.00044 ± 0.00044	-0.00006 ± 0.00019	-0.00011 ± 0.00004	0.00013 ± 0.00020	0.00010 ± 0.00015	...
$\bar{x} \pm s$	0.00020 ± 0.00033	0.00005 ± 0.00022	0.00027 ± 0.00071	0.00009 ± 0.00019	0.00029 ± 0.00028	0.00032 ± 0.00033	0.00030 ± 0.00028	0.00020 ± 0.00039
^{239,240}Pu (pCi/g dry weight)								
No. of Samples	5	10	15	3	10	3	2	1
Maximum ^a	0.00033 ± 0.00010	0.0012 ± 0.00092	0.0033 ± 0.00100	0.00027 ± 0.00011	0.0033 ± 0.0010	0.00051 ± 0.00020	0.00049 ± 0.00049	...
Minimum ^a	-0.00035 ± 0.00059	-0.00083 ± 0.00050	0.00008 ± 0.00010	-0.00031 ± 0.00016	0.00014 ± 0.00029	0.00020 ± 0.00013	0.00005 ± 0.00010	...
$\bar{x} \pm s$	0.00009 ± 0.00026	0.00016 ± 0.00053	0.00044 ± 0.00081	0.00005 ± 0.00031	0.00069 ± 0.00096	0.00037 ± 0.00016	0.00027 ± 0.00031	-0.00020 ± 0.00039
Uranium (µg/g dry weight)								
No. of Samples	5	9	15	3	10	3	2	1
Maximum ^a	0.022 ± 0.005	0.012 ± 0.0033	0.024 ± 0.0048	0.011 ± 0.0021	0.15 ± 0.027	0.059 ± 0.0083	0.042 ± 0.006	...
Minimum ^a	0.0005 ± 0.0006	0.0000 ± 0.0009	0.00083 ± 0.00058	0.0010 ± 0.0008	0.00008 ± 0.00072	0.0088 ± 0.0030	0.0045 ± 0.0024	...
$\bar{x} \pm s$	0.0084 ± 0.0085	0.0057 ± 0.0048	0.0076 ± 0.0068	0.0047 ± 0.0053	0.0018 ± 0.047	0.040 ± 0.028	0.023 ± 0.026	0.0045 ± 0.0018
¹³⁷Cs (pCi/g dry weight)								
No. of Samples	5	10	15	3	10	3	2	1
Maximum ^a	0.97 ± 1.46	1.83 ± 0.92	1.24 ± 1.21	0.46 ± 0.63	2.07 ± 0.90	0.41 ± 0.19	0.37 ± 0.31	...
Minimum ^a	-1.39 ± 1.68	-1.11 ± 0.65	1.07 ± 1.02	-0.63 ± 1.01	-1.16 ± 0.82	-0.18 ± 0.13	0.18 ± 0.23	...
$\bar{x} \pm s$	-0.082 ± 0.91	0.067 ± 0.72	-0.053 ± 0.60	0.05 ± 0.59	0.39 ± 1.06	0.17 ± 0.31	0.28 ± 0.13	-0.44 ± 0.41
⁹⁰Sr (pCi/g dry weight)								
No. of Samples	5	10	11	1	8	2	1	1
Maximum ^a	0.052 ± 0.0071	0.10 ± 0.011	0.29 ± 0.020	...	0.076 ± 0.0071	0.070 ± 0.0031	...	...
Minimum ^a	-0.0016 ± 0.0048	0.0012 ± 0.0012	0.0063 ± 0.0013	...	0.0022 ± 0.0015	0.011 ± 0.0035	...	...
$\bar{x} \pm s$	0.025 ± 0.025	0.029 ± 0.034	0.056 ± 0.081	0.0089 ± 0.0089	0.027 ± 0.023	0.040 ± 0.042	0.0031 ± 0.0031	0.028 ± 0.0073

^aCounting uncertainty.

TABLE II
RADIONUCLIDE CONTENT OF FISH COLLECTED DURING 1983

Location	Abiquiu, El Vado, and Heron Reservoirs				Cochiti Reservoir			
	Bottom Feeder ^a	Bottom Feeder (gut) ^b	Higher Level ^a	Higher Level (gut) ^b	Bottom Feeder ^a	Bottom Feeder (gut) ^b	Higher Level ^a	Higher Level (gut) ^b
Radionuclide:								
²³⁹Pu (pCi/g dry weight)								
No. of Samples	10	8	8	6	13	10	8	5
Maximum ^c	0.00004 ± 0.00011	0.0017 ± 0.0007	0.00006 ± 0.00003	0.00058 ± 0.00033	0.00056 ± 0.00022	0.0064 ± 0.0038	0.00006 ± 0.00000	0.00028 ± 0.0000
Minimum ^c	-0.00002 ± 0.00004	-0.00063 ± 0.00019	0.00006 ± 0.00005	0.00009 ± 0.00027	0.00005 ± 0.00001	-0.00029 ± 0.00078	0.00014 ± 0.00007	-0.00004 ± 0.00009
$\bar{x} \pm s$	0.00001 ± 0.00002	0.00029 ± 0.00075	0.00000 ± 0.00005	0.00016 ± 0.00023	0.00007 ± 0.00015	0.00085 ± 0.00017	-0.00003 ± 0.00008	0.00013 ± 0.00012
^{239,240}Pu (pCi/g dry weight)								
No. of Samples	10	8	8	6	13	10	8	5
Maximum ^c	0.00011 ± 0.00006	0.0044 ± 0.0010	0.00006 ± 0.00005	0.001 ± 0.0003	0.00078 ± 0.00022	0.0059 ± 0.0011	0.00021 ± 0.00000	0.0023 ± 0.0020
Minimum ^c	-0.00009 ± 0.00010	0.00011 ± 0.00011	0.00012 ± 0.00005	0.00007 ± 0.00008	0.00018 ± 0.00002	0.00058 ± 0.00068	-0.00011 ± 0.00009	0.00002 ± 0.00005
$\bar{x} \pm s$	0.00003 ± 0.00006	0.0011 ± 0.0014	0.00002 ± 0.00006	0.00041 ± 0.00040	0.00011 ± 0.00029	0.0020 ± 0.0019	0.00006 ± 0.00009	0.00052 ± 0.00102
Uranium (μg/g dry weight)								
No. of Samples	10	9	8	6	13	10	8	5
Maximum ^c	0.0507 ± 0.0051	0.33 ± 0.03	0.0061 ± 0.0010	0.084 ± 0.008	0.056 ± 0.057	1.35 ± 0.14	0.021 ± 0.005	0.54 ± 0.007
Minimum ^c	0.0079 ± 0.0010	0.058 ± 0.006	0.0000 ± 0.0019	0.0046 ± 0.0007	0.0079 ± 0.0001	0.075 ± 0.0075	0.0053 ± 0.0018	0.007 ± 0.0007
$\bar{x} \pm s$	0.020 ± 0.014	0.18 ± 0.11	0.0018 ± 0.0024	0.028 ± 0.031	0.027 ± 0.014	0.49 ± 0.38	0.0088 ± 0.0051	0.12 ± 0.24
¹³⁷Cs (pCi/g dry weight)								
No. of Samples	10	9	8	5	13	10	8	5
Maximum ^c	0.39 ± 0.28	0.45 ± 0.14	0.10 ± 0.06	0.73 ± 0.28	0.17 ± 0.10	0.58 ± 0.66	0.15 ± 0.07	0.43 ± 0.22
Minimum ^c	-1.4 ± 1.2	-1.4 ± 2.5	0.061 ± 0.050	1.3 ± 0.5	0.19 ± 0.076	0.16 ± 0.15	-0.58 ± 0.54	-0.022 ± 0.11
$\bar{x} \pm s$	-0.076 ± 0.49	0.0002 ± 0.57	0.038 ± 0.055	0.23 ± 0.76	0.043 ± 0.11	0.20 ± 0.24	0.063 ± 0.25	0.21 ± 0.22
⁹⁰Sr (pCi/g dry weight)								
No. of Samples	10	8	5	4	12	10	6	2
Maximum ^c	0.23 ± 0.009	0.082 ± 0.006	0.11 ± 0.01	0.056 ± 0.005	0.11 ± 0.005	0.36 ± 0.02	0.18 ± 0.007	0.034 ± 0.002
Minimum ^c	0.069 ± 0.004	0.0044 ± 0.0044	0.0098 ± 0.0006	-0.0011 ± 0.007	0.035 ± 0.003	0.0018 ± 0.0018	0.049 ± 0.002	0.032 ± 0.002
$\bar{x} \pm s$	0.14 ± 0.050	0.027 ± 0.024	0.043 ± 0.040	0.023 ± 0.026	0.076 ± 0.025	0.05 ± 0.11	0.093 ± 0.046	0.033 ± 0.001

^aSamples consisted of fish less digestive system (gut).

^bSamples consisted of gut only.

^cCounting uncertainty.

and analytical results. Information can be extracted from the data bases to generate summary reports (Tables I and II).

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REFERENCES

1. "Environmental Surveillance at Los Alamos During 1980," Los Alamos National Laboratory report LA-8810-ENV (April 1981).
2. "Environmental Surveillance at Los Alamos During 1983," Los Alamos National Laboratory report LA-10100-ENV (April 1984).
3. M. G. Natrella, *Experimental Statistics*, National Bureau of Standards Handbook 91 (August 1963).

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