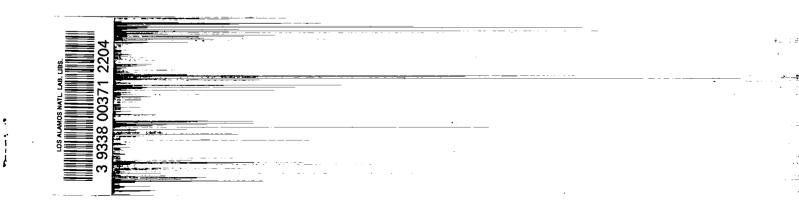


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# LOS ALAMOS SCIENTIFIC LABORATORY OF THE UNIVERSITY OF CALIFORNIA $\circ$ LOS ALAMOS NEW MEXICO

STRONTIUM-90, CESIUM-137, AND RADIOACTIVE RARE EARTHS IN ENVIRONMENTAL RAIN AND AIR AT LOS ALAMOS, NEW MEXICO 1958 - June 1963



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# LOS ALAMOS SCIENTIFIC LABORATORY OF THE UNIVERSITY OF CALIFORNIA LOS ALAMOS NEW MEXICO

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> STRONTIUM-90, CESIUM-137, AND RADIOACTIVE RARE EARTHS IN ENVIRONMENTAL RAIN AND AIR AT LOS ALAMOS, NEW MEXICO 1958 - June 1963

> > by

E. R. Graham



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## ABSTRACT

A study was made of the strontium-90, cesium-137, and the radioactive rare earths in environmental rain and air at Los Alamos, New Mexico. Samples of rain and air were collected continuously from January 1958 through June 1963.

The results of the study showed the activity of the strontium-90 to be the highest during the summer of 1958; the activity then decreased slowly reaching the lowest level during September of 1961. Starting in October of 1961 the activity started to rise, however, never reaching the level of 1958.

The observed results of the environmental concentration of cesium-137 and rare earth radioactivity were similar to those for strontium-90. The results of the activity measured in the air and that in the rain followed the same monthly distribution. The results of the study would indicate that the rain-held activity is just a wash out of the activity present in the Los Alamos air.

## INTRODUCTION

As a part of the Environmental Health Program at the Los Alamos Scientific Laboratory, samples of rain and samples of airborne radioactive particles are collected at regular intervals to determine the gross beta-gamma radioactivity. The rain samples are collected with an oversized rain gauge; the airborne particles are collected in air To arrive at the concentration of long-life filters. fission products in the rain and the air samples, a study was made of the methods suitable for sample preparation and the determination of the activity of strontium-90, cesium-137, and the radioactive rare earths. Since the samples were collected over a period of several years, the changes in contamination of the air of the Los Alamos area by fallout and, possibly, industrial waste fission products may be This information, along with future studies, ascertained. will allow for an evaluation of the environmental air radionuclide contamination of the Los Alamos area.

It was the objective of this study to determine the

amount of strontium-90, cesium-137, and radioactive rare earths in the rain and air samples collected by the Health Division at Los Alamos.

#### PROCEDURE

## Rain Samples

The rain samples were collected in an oversized rain collector with a cross section of approximately 0.4 square meter. This area was such that a rainfall of 0.1 inch would deliver 1 liter of rain water. During dry periods, the rain gauge was washed down with a liter of distilled water daily. The wash samples as well as the rain samples were reduced in volume and plated dry on 1-inch-diameter, stainless steel planchets to determine the beta-gamma activity. The gross beta-gamma activity results have been reported in LAMS reports numbers 2397, 2499, 2702, 2870, and 3071.

The planchets prepared during one month\* were composited for a sample and placed in a 400 milliliter beaker and 200 milliliters of 0.01 M  $Na_2CO_3$  was added. The beaker containing the sample was placed in an ultrasonic cleaner and subjected to ultrasonic vibrations for 30 minutes. The

<sup>\*</sup>During 1960 and the first 8 months of 1961, a composite of 4 months represents one sample.

resulting supernatant liquid and a distilled water wash of the planchets was transferred to a large platinum evaporating dish and the contents reduced to dryness. The residue was digested with nitric-perchloric acid until clear, then dried again. The silicon was removed from the residue by treating with a mixture of concentrated nitric and hydrofluoric acid.<sup>1</sup> After the removal of the silicon, the resulting residue was soluble in dilute nitric acid. The soluble sample was transferred to a volumetric flask and set aside for future analysis.

## Air Filter Samples

Airborne radioactive particles were collected by filtration through 4-inch-diameter filters (MSA catalog number CR-17651). The sampling rate was 46 cubic meters per hour. Air samples were collected for 24-hour intervals during the work week; the week-end samples were for a 72-hour period. The gross beta-gamma activity was determined by counting in a thin-window proportional counter. The results have been reported in LAMS reports numbers 2397, 2499, 2702, 2870, and 3071. A collection of samples for one month\*\* was placed in a liter beaker and digested with concentrated nitric acid,

<sup>\*\*</sup>One month's sample would equal approximately 33,120 cubic meters of filtered air.

which required 48 hours or more. After this period 72 per cent perchloric acid was added and the digestion continued until the samples were clear. The sample was then transferred to large platinum dishes and reduced to dryness. A mixture of concentrated nitric and hydrofluoric acid<sup>1</sup> was added, and the silicon was removed by two treatments of the nitric-hydrofluoric acid mixture. After the removal of the silicon, the sample was soluble. It was then transferred to a volumetric flask and set aside for analysis.

Aliquots of the dissolved samples were analyzed for strontium-90, cesium-137, and radioactive rare earths by the methods outlined by  $\text{Geiger}^2$  and Boni.<sup>3</sup>

## DATES OF ANALYSIS

#### Rain-Collected Samples

The 1958 and 1959 samples were analyzed during the summer of 1960; the 1960 samples were analyzed during the summer of 1961; and the 1961 and January through May, 1962, samples were analyzed during the summer of 1962. The balance of 1962 samples and the January through June samples of 1963 were analyzed during the summer of 1963.

## Filter-Collected Air Samples

The 1958 and 1959 samples were analyzed during the

summer of 1960; the 1960 and January through June samples of 1961 were analyzed during the summer of 1961; and the July through December, 1961, and January through April, 1962, samples were analyzed during the summer of 1962. The balance of 1962 samples and the January through June samples of 1963 were analyzed during the summer of 1963.

## **RESULTS AND DISCUSSION**

The results of the strontium-90, cesium-137, and rare earth radioactivity analyses of the monthly composite of rain-collected and filter-collected air samples at Los Alamos are presented in Tables 1 through 8 and shown graphically in Figures 1 and 2.

## Rain Collected Activity

The activity of strontium-90 expressed as micromicrocuries per month was high during 1958; this was especially so during the summer months. The level decreased each year until September of 1961, at which time it started to increase every month. The level of strontium-90 reached in the first 5 months of 1962 did not exceed that found in 1958.

The results obtained for cesium-137 activity were similar to those for strontium-90 with the exception that the rise of activity in the fall of 1961 was not as rapid as

the strontium-90 or the rare earth activity.

The activity of rare earths was highest during the fall of 1961 and early 1962. If corrections for half life were made, this probably would not be the case since the 1958 samples were analyzed 2 years after collection while the late 1961 and 1962 samples were analyzed during 1962.

## Filter Collected Air Samples

The activity collected in filters from air at Los Alamos shows almost the same pattern of monthly distribution as the activity collected in the rain collector. It is apparent that at Los Alamos the environmental air may be sampled either by collecting samples of rain or by filtering the air.

#### REFERENCES

- 1. Holt, B. D., "Determination of Silicon by Distillation-Colorimetric Method," Anal. Chem. 32, 124-28 (1960).
- 2. Geiger, E. L., "Analysis of Fission Product Mixtures," Anal. Chem. 31, 806-09 (1959).
- 3. Boni, A. L., "Quantitative Analysis of Radionuclides in Process and Environmental Samples," Anal. Chem. <u>32</u>, 599-604 (1960).

# STRONTIUM-90 ACTIVITY OF MONTHLY RAIN-COLLECTOR SAMPLES COLLECTED AT LOS ALAMOS, NEW MEXICO

Month	1958, µµc/month	1959, µµc/month	1960, μμc/month	1961, µµc/month	1 <b>962,</b> μμc/month
Jan.	44	238	15*	1*	13
Feb.	76	49	15*	1*	42
March	88	56	15*	1*	15
April	428	56	15*	1*	120
May	96	162	73*	4*	38
June	304	20	73*	4*	110
July	316	633	73*	4*	118
Aug.	368	49	73*	4*	36
Sept.	140	7	24*	5	27
Oct.	160	9	24*	7	64
Nov.	76	6	24*	15	154
Dec.	60	6	24*	64	436

\*1/4 of the activity in a 4-month composite sample.

Rain-collector = 0.4 square meter in cross section.

# CESIUM-137 ACTIVITY OF MONTHLY RAIN-COLLECTOR SAMPLES COLLECTED AT LOS ALAMOS, NEW MEXICO

Month	1958, μμc/month	1959, µµc/month	1960, µµc/month	1961, µµc/month	1962, μμc/month
Jan.	9	426	27*	11*	82
Feb.	198	435	27*	11*	127
March	<b>32</b> 0	708	27*	11*	82
April	670	817	27*	11*	245
May	426	1319	84*	29*	136
June	426	471	84*	29*	68
July	237	397	84*	29*	205
Aug.	218	349	84*	29*	110
Sept.	102	313	95*	16	45
Oct.	388	926	95*	34	45
Nov.	9	275	95*	34	45
Dec.	471	246	95*	30	90

\*1/4 of the activity in a 4-month composite sample.

Rain-collector = 0.4 square meter in cross section.

# RARE EARTH RADIOACTIVITY OF MONTHLY RAIN-COLLECTOR SAMPLES COLLECTED AT LOS ALAMOS, NEW MEXICO

Month	1958, µµc/month	1959, µµc/month	1960, µµc/month	1961, µµc/month	1962, µµc/month
Jan.	84	225	8*	94*	2974
Feb.	69	269	8*	94*	1298
March	100	524	8*	94*	1745
April	146	240	8*	94*	6860
May	75	1383	13*	132*	3054
June	426	182	13*	132*	1291
July	442	342	13*	132*	2073
Aug.	98	218	13*	132*	1173
Sept.	98	124	14*	665	1209
Oct.	231	95	14*	1141	645
Nov.	93	14	14*	1250	600
Dec.	262	125	14*	2021	845

 $^*$ 1/4 of the activity in a 4-month composite sample.

Rain-collector = 0.4 square meter in cross section.

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# STRONTIUM-90 IN RADIOACTIVE PARTICLES FILTER-COLLECTED FROM AIR AT LOS ALAMOS, NEW MEXICO

Month	1958, µµc/month	1959, µµc/month	1960, μμc/month	1961, µµc/month	1962, μμc/month
Jan.	743	359	31	33	825
Feb.	620	718	63	127	498
March	1618	1011	175	178	531
April	1357	895	1 <b>22</b>	54	808
May	2913	<b>92</b> 0	108	276	354
June	1547	368	42	709	596
July	1811	227	42	-	201
Aug.	1145	34	25	-	98
Sept.	760	43	-	29	134
Oct.	1654	52	115	204	309
Nov.	2118	52	22	236	320
Dec.	<b>3</b> 359	34	14	440	472

One month's sample would equal approximately 33,120 cubic meters of filtered air.

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# CESIUM-137 IN RADIOACTIVE PARTICLES FILTER-COLLECTED FROM AIR AT LOS ALAMOS, NEW MEXICO

Month	1958, µµc/month	1959, µµc/month	1960, µµc/month	1961, µµc/month	1962, µµc/month
Jan.	1156	4227	436	609	797
Feb.	1263	6322	381	745	398
March	1322	6306	709	436	596
April	2777	8068	300	1336	704
May	3579	6922	818	1254	646
June	2400	2840	64	937	572
July	1643	1447	336	-	418
Aug.	1277	366	218	-	145
Sept.	993	459	9	64	136
Oct.	1477	481	227	154	354
Nov.	1784	554	172	300	163
Dec.	3237	1059	91	463	436

One month's sample would equal approximately 33,120 cubic meters of filtered air.

# RARE EARTH RADIOACTIVE PARTICLES FILTER-COLLECTED FROM AIR AT LOS ALAMOS, NEW MEXICO

Month	1958, µµc/month	1959, µµc/month	1960, μμc/month	1961, μμc/month	1962, µµc/month
Jan.	990	7704	196	58	40923
Feb.	863	6954	124	265	31032
March	977	12136	76	436	55989
April	2147	13772	113	912	67160
May	1884	9000	153	705	873
June	2227	5090	174	880	10967
July	1754	2727	105	-	1473
Aug.	890	418	69	-	3036
Sept.	1336	763	62	720	3836
Oct.	881	727	69	6230	6610
Nov.	2202	1102	80	17145	7523
Dec.	5560	525	47	13065	10927

One month's sample would equal approximately 33,120 cubic meters of filtered air.

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# Sr-90, Cs-137, AND RARE EARTH ACTIVITY IN RADIOACTIVE PARTICLES FILTER-COLLECTED FROM AIR AT LOS ALAMOS, NEW MEXICO

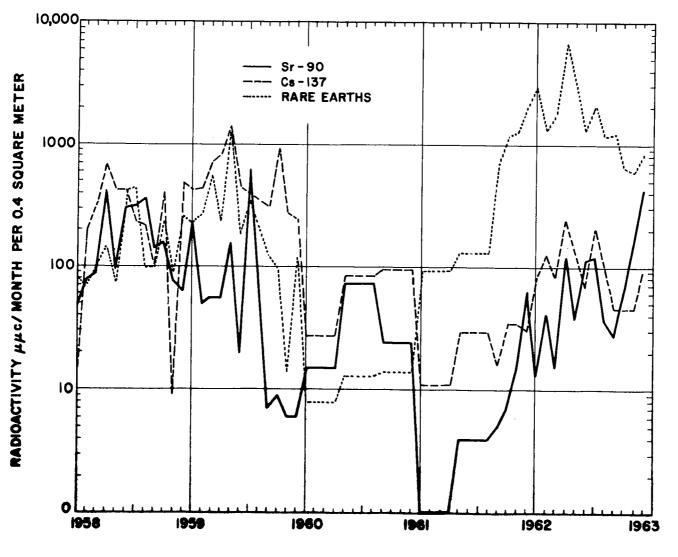
1963 Month	Sr-90, μμc/month	Cs-137, µµc/month	Rare Earths, µµc/month
Jan.	618	445	23538
Feb.	2131	910	31218
March	596	763	36509
April	2804	1458	56425
May	1378	1400	54607
June	1098	1861	60429

## TABLE 8

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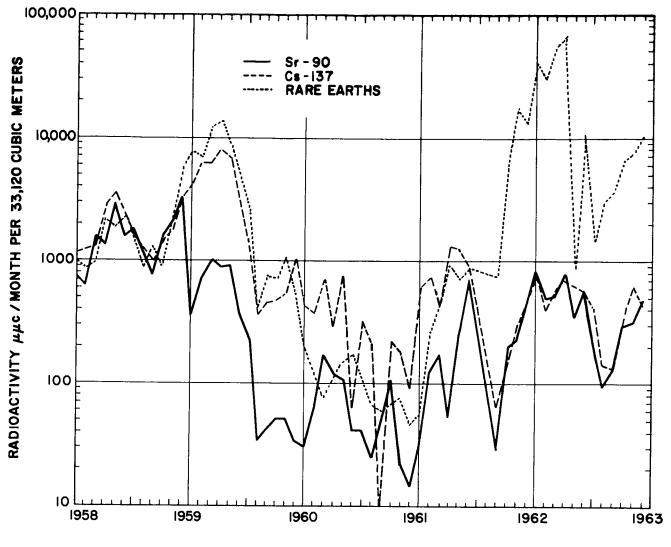
# Sr-90, Cs-137, AND RARE EARTH ACTIVITY OF MONTHLY RAIN-COLLECTOR SAMPLES COLLECTED AT LOS ALAMOS, NEW MEXICO

1963 Month	Sr-90, μμc/month	Cs-137, µµc/month	<b>Rar</b> e Earths, μμc/month
Jan.	68	152	1501
Feb.	363	115	2345
March	173	198	4001
April	250	213	4638
May	798	797	7137
June	475	377	44041



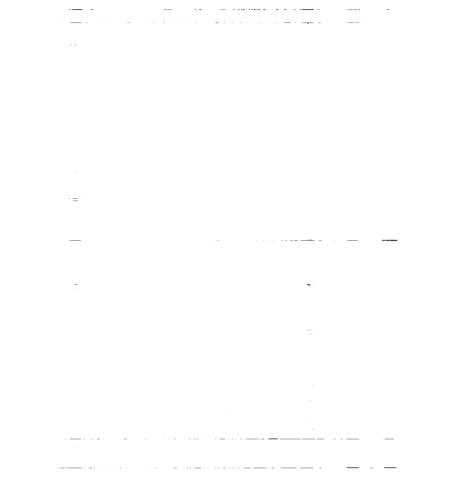


Radioactivity of Monthly Rain-Collector Samples Collected at Los Alamos, New Mexico





Radioactive Particles Filter-Collected From Air at Los Alamos, New Mexico



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