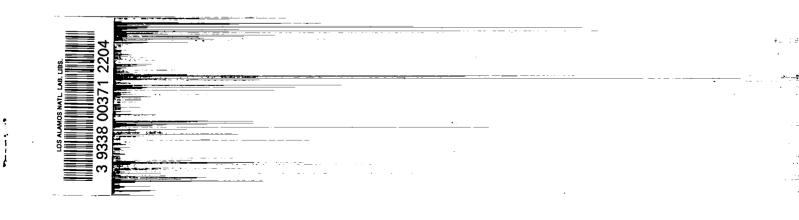


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LOS ALAMOS SCIENTIFIC LABORATORY OF THE UNIVERSITY OF CALIFORNIA \circ LOS ALAMOS NEW MEXICO

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

^{*}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.¹ 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,

^{**}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 acid¹ 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 Geiger^2 and Boni.³

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 | 1 962, 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 | 32 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.

•

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 22 | 54 | 808 |
| May | 2913 | 92 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. | 3 359 | 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

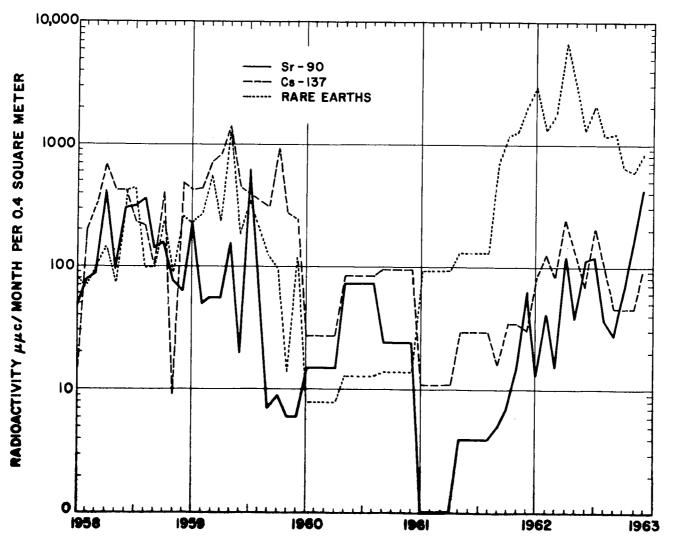
| 1963 Month | Sr-90, μμc/month | Cs-137, µµc/month | Rare Earths, µµc/month |
|---------------|---------------------|----------------------|---------------------------|
| Jan. | 618 | 445 | 23538 |
| Feb. | 2131 | 910 | 31218 |
| March | 596 | 763 | 36509 |
| April | 2804 | 1458 | 56425 |
| May | 1378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

TABLE 8

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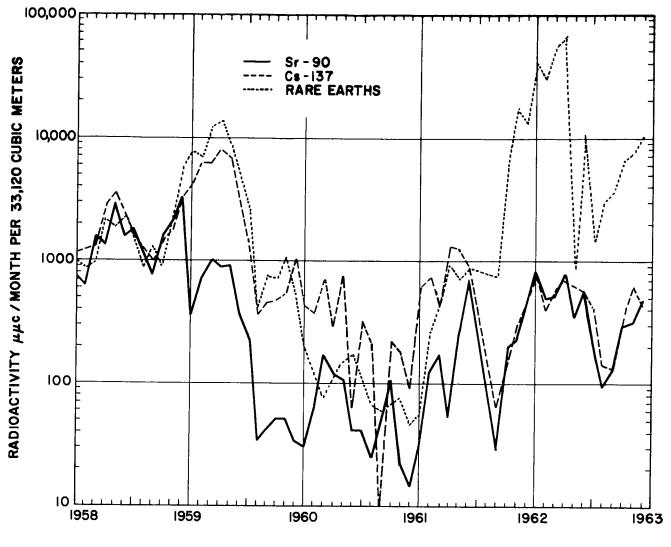
Sr-90, Cs-137, AND RARE EARTH ACTIVITY OF MONTHLY RAIN-COLLECTOR SAMPLES COLLECTED AT LOS ALAMOS, NEW MEXICO

| 1963 Month | Sr-90, μμc/month | Cs-137, µµc/month | Rar 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 |



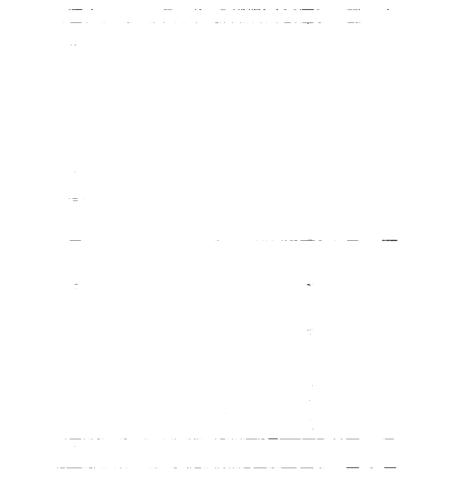


Radioactivity of Monthly Rain-Collector Samples Collected at Los Alamos, New Mexico





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



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