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Lawrence Radiation Laboratory
UNIVERSITY OF CALIFORNIA
LIVERMORE
94550

Bio-Medical Division

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**RADIONUCLIDES IN SURFACE AIR
AT LIVERMORE, CALIFORNIA**

Keith O. Hamby

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RADIONUCLIDES IN SURFACE AIR AT LIVERMORE, CALIFORNIA

Abstract

Analyses of three sets of air filters exposed in the fall and winter of 1970-1971 on the grounds of the Lawrence Radiation Laboratory, Livermore, California, show the presence of a number of radionuclides usually attributed to worldwide fallout. These include ^{95}Zr , ^{103}Ru , ^{106}Ru , ^{137}Cs , ^{141}Ce , ^{144}Ce ,

^{125}Sb , and ^{155}Eu . Also observed was the cosmic-ray-induced ^7Be .

Relatively large volumes of surface air were sampled over three different periods ranging from two to eight weeks in duration. Radioactive components of the samples were determined by gamma spectroscopy using Ge(Li) detectors.

Introduction

The inventory of radionuclides in the atmosphere has been of continuing interest to many segments of the world's population since the discovery of radioactivity. Much work has been done, and considerable data have been reported over a period of many years^{1,2}; the effort continues, since the inventory is not a stable one. The injection of man-made radioactivity and the discovery of cosmic-ray-induced nuclides are the most recent perturbations. Concern over the long-

term effects on human life of low levels of radioactivity indicates the need to continue monitoring the nature and quantity of radionuclides in the atmosphere.

This report describes a system for sampling large volumes of air and for determining radionuclide content by low-level, high-resolution gamma spectroscopy. Results are given for three separate sampling runs made at Livermore, California, between September 1970 and March 1971.

Instrumentation

The sequential air sampler described by Holladay, *et al.*³ consists of several sampling heads that can be operated sequentially in time (in series) or simultaneously (in parallel). In the case of samples 1 and 2 reported here, each sample consists of three filters exposed

simultaneously. Sample 3 is a single filter only. The filter used is made of convoluted fiberglass (Mine Safety Appliance Co., MSA-77227) with an efficiency of more than 99% for particles down to 0.025 μ in diameter.^{4,5}

In this apparatus the volume of air

filtered is determined by monitoring the pressure drop across the filter, as described in Ref. 3. The flow rate is approximately 1.9 cfm per inch of equivalent water-pressure drop across the filter. Mean air flow during the sampling periods was 10 cfm for the eight-week exposure and 12 cfm for the two- and three-week exposures.

The exposed filters are prepared for counting by compressing them to a thickness of 1/8 in. and sealing them in 5-mm-thick, 200-cc aluminum cans under a

polystyrene spacer. In the case of samples 1 and 2, all three filters were compressed into a single can.

Gamma analysis of samples 1 and 2 was done on a simple Ge(Li) detector system (12-cc active volume). Sample 3 was analyzed by a Compton-suppressed Ge(Li) system (18-cc active volume). Both systems have been described by Phelps.⁶

Data accumulated on magnetic tape were reduced and analyzed on one of the LRL CDC-6600 computers.

Results and Discussion

Data pertaining to sample collection are shown in Table 1. Air volumes for samples 1 and 2 are only approximate due to incomplete pressure-drop data and, in the case of sample 2, the failure of one of the blowers shortly before the end of the sampling period.

Figure 1 shows the first 500 keV of a typical gamma spectrum, that of sample 1 as determined on the 12-cc Ge(Li) diode detector system. Figure 2 shows the second 500 keV of this spectrum. Distinct photopeaks are observed for ⁷Be, ⁹⁵Zr, ¹⁰³Ru, ¹⁰⁶Ru, ¹³⁷Cs, ¹⁴¹Ce, and ¹⁴⁴Ce. In addition, traces are seen of ¹⁵⁵Eu and

¹²⁵Sb. Background peaks due to the prominent members of the thorium and uranium natural decay chains are due to the detector background. Beyond 1000 keV, only a weak ⁴⁰K peak was observed. This peak is also due to the system background.

Table 2 shows the total amount of each radionuclide observed in our three samples. Uncertainties shown are those due to counting statistics only. Radioactive decay corrections were made from counting time to the mid-time of the sampling period.

Table 3 shows the specific activity of each radionuclide in terms of

Table 1. Sample filtering data.

	Sample 1	Sample 2	Sample 3
Length of filtering period (days)	21	56	16
Mid-time of filtering period	11 Sep 1970	15 Nov 1970	1 Mar 1971
Number of filters ^a	3	3	1
Approximate volume (10 ⁵ m ³)	31	68	7.8

^aThe three filters of samples 1 and 2 were canned together and counted as a single sample.

Fig. 1. The gamma spectrum of sample 1 from 0 to 500 keV as determined by the 12-cc Ge(Li) diode detector system.

Table 2. Total activity in picocuries for each radionuclide in each sample.

	Half-life	Sample 1	Sample 2	Sample 3
^{144}Ce	285 d	1780 ± 23	1657 ± 26	133 ± 9
^{141}Ce	32 d	46 ± 5	367 ± 12	39 ± 3
^7Be	53 d	2528 ± 101	5961 ± 186	454 ± 30
^{103}Ru	40 d	73 ± 10	327 ± 21	45 ± 4
^{106}Ru	365 d	874 ± 50	826 ± 50	45 ± 17
^{125}Sb	2.7 y	85 ± 19	99 ± 19	10 ± 4
^{137}Cs	30 y	142 ± 10	148 ± 10	13 ± 3
^{95}Zr	65 d	418 ± 11	370 ± 10	54 ± 3
^{155}Eu	5 y	14 ± 2	14 ± 3	Trace

Fig. 2. The gamma spectrum of sample 1 from 500 to 1,000 keV as determined by the 12-cc Ge(Li) diode detector system.

picocuries per thousand cubic meters of air.

The presence in samples representing large volumes of air of long-lived fission products ^{106}Ru , ^{137}Cs , ^{144}Ce , ^{125}Sb , and ^{155}Eu is not unexpected. The presence of radionuclides with half-lives of from one to two months, ^{103}Ru , ^{141}Ce , and ^{95}Zr , can be attributed to continuing atmospheric weapons tests, e.g., the Chinese event of about 3 Mt at the Lop Nor test site on October 14, 1970.⁷

The creation of radionuclides in the stratosphere through interactions of cosmic radiation and atmospheric components was predicted by Libby in 1946.⁸

Table 3. Concentrations of observed radionuclides in picocuries per thousand cubic meters of air.

	Sample 1	Sample 2	Sample 3
^{144}Ce	57.4	24.4	17.1
^{141}Ce	1.5	5.4	5.0
^7Be	81.5	87.7	58.2
^{103}Ru	2.4	4.8	5.8
^{106}Ru	28.2	12.1	5.8
^{125}Sb	2.7	1.5	1.3
^{137}Cs	4.6	2.2	1.7
^{95}Zr	13.5	5.4	6.9
^{155}Eu	0.5	0.2	Trace

In 1955 Arnold and Al-Salih reported the discovery of cosmic-ray-induced ^7Be .⁹ Quantitative determinations of the ^7Be content of surface air have been reported by Lindholm¹⁰ in Sweden and by Parker¹¹ in England. Parker reports a range of

values from 2 to 8×10^{-14} Ci of ^7Be per kilogram of air. The average value for our three samples is about 85 pCi per 10^3 m^3 of air. This value converts to about 7×10^{-14} Ci per kilogram of air and is in good agreement with the previous results of Parker.

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NOTES

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