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FALLOUT CONCENTRATIONS OF 12 RADIONUCLIDES IN AIR: 1962 THROUGH MID-1964

by

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Newly developed multidimensional gamma-ray spectrometric counting techniques permit the direct measurement of twelve fallout radionuclides on air filters. Observed concentrations of Be^{7}, Na^{22}, Mn^{54}, Co^{60}, Y^{88}, $Zr^{95}Nb^{95}$, Ru^{106}, Sb^{124}, Sb^{125}, Cs^{134}, Cs^{137} and Ce^{144} during the past two and a half years help explain the origin and fallout rates of the trace radionuclides in air.

The absolute and relative concentrations of all the various air-borne radionuclides are of interest in studying their fallout rates and origin as well as determining their availability for uptake by man. The tracing of some of the minor radionuclides in fallout as well as more precise measurements of the major radionuclides has recently been made possible by use of multidimensional gamma-ray spectrometric counting methods[1]. At the present time it is possible to make reasonably precise measurements of the 12 radionuclides Be^{7}, Na^{22}, Mn^{54}, Co^{60}, $Zr^{95}Nb^{95}$, Y^{88}, Ru^{106}, Sb^{124}, Sb^{125}, Cs^{134}, Cs^{137} and Ce^{144} by the direct counting of an air filter sample. Such measurements are especially valuable since they permit a direct comparison, on the same sample, of the relative concentrations of this large group of radionuclides and thus provide information on both their fallout rates and origin. The spectrometer system consists of a three-crystal NaI(Tl) gamma-ray detector system in conjunction with a 4096 channel multiparameter analyzer. Use is made of the individual gamma-ray decay characteristic of each radionuclide for its identification and measurement by counting the sample between two 4 inch thick by 6 inch diameter detectors and cancelling those events which are not totally adsorbed in these principal detectors with a anticoincidence annulus. Coincidence counts are stored according to the two photon energies responsible for the event while non-coincidence events are stored in the normal manner. Anticoincidence shielding for background reduction and the use of large, efficient detectors for viewing the sample in a nearly 4π arrangement

provides a very sensitive as well as a selective arrangement. The radionuclides Na^{22}, Co^{60}, Y^{88}, Ru^{106}, Sb^{124}, Cs^{134} and Ce^{144} are uniquely measured from their characteristic coincidence gamma-ray spectra while the remaining radionuclides are measured from their single photon gamma-ray spectra. The selective measurement of mono-gamma emitters is improved by use of a difference spectra, obtained from two measurements taken 3 to 6 weeks apart.

Air sampling[2] is presently performed with a continuously operating vacuum pump which pulls air at 100 cfm through a 5 micron pore size membrane filter. These membrane filters have been shown to be essentially absolute for fallout radionuclides[2]. The filters are composited on a monthly or semimonthly basis, pressed into a standard geometry of 1/2 inch thick by 1 inch diameter and counted on the multidimensional gamma-ray spectrometer. The air sampling location was at a point 15 ft. above the ground, 6 miles north of Richland, Washington. The annual precipitation in this area is only about 8 inches and it does not account for the major deposition of fallout. The observed air concentrations of the twelve radionuclides being studied are recorded in Figure I. A more detailed presentation of our work including results from radiochemical measurements of other fallout radionuclides will be published later[3]. The radionuclides listed in Figure 1 result from fission and neutron activation during atomic tests, and also from cosmic ray interactions with the atmosphere. The radionuclides $Zr^{95}Nb^{95}$, Ru^{106}, Sb^{125}, Cs^{137} and Ce^{144} are fission products and presumably this is their main source in the atmosphere. Mn^{54}, Co^{60}, Y^{88}, Sb^{124} and Cs^{134} are formed as neutron activation products during testing while Be^{7} and Na^{22} are produced continuously in the atmosphere by cosmic ray interactions. The fallout rates of many of the fission products are measured from sampling locations the world over[4] and perhaps their main value in this report is to serve as a base line in comparing the concentrations of less abundant radionuclides in the atmosphere.

The radionuclide Y^{88} (105 d) was recently reported to be present on samples of grass collected in August 1962 near the Euratom Nuclear Center at Ispra, Italy[5]. It was in the form of insoluble particles containing much larger amounts of Zr^{95}-Nb^{95} and although its source was not known it did not come from the Nuclear Center and presumably came from either nuclear bombs or an uncontrolled release from a nuclear establishment. Our present study has established that Y^{88} is present in world-wide fallout and was therefore produced during the past atomic test period.

During 1962 Y^{88} was detectable only in our November sample. This was probably due to the fact that during 1962 smaller air volumes were collected and also, considerable delay occurred before the multidimensional spectrum measurements were made in late 1963. Y^{88} has been easily measurable during 1963 and 1964. The radionuclide Na^{22} was first reported to be present in the atmosphere by Marquez[6] in 1957 who found it in rain water at Rio de Janeiro in a concentration of 0.017 d/m/liter. Its current concentration in air, foods and people is being reported by the authors[7]. It is produced naturally by cosmic-ray spallation of argon in the atmosphere but also results from the reaction Na^{23} (n,2n) Na^{22} during nuclear weapons testing. Its potential value as a tracer of atmospheric circulation has been recognized and recently reported measurements for this purpose by Bhandari and Rama[8] have shown air concentrations ranging from 1 d/m per 10^6 ft^3 below the tropopause to 460 d/m per 10^6 ft^3 above the tropopause. It is evident from Figure 1 that the Na^{22} concentrations in air were about a factor of two higher during 1963 and 1964 than in 1962. Also the 1963 - 1964 values are about an order of magnitude higher than those reported for troposphere measurements prior to the 1961 atomic tests[8]. This suggests that a large amount of Na^{22} was produced during the past test series.

The Cs^{134} appears to be produced in nuclear detonations mainly by an n, γ reaction on Cs^{133} but is also produced directly by fission(9). The ratio of Cs^{134} to Cs^{137} during the past two and a half years did not show a pronounced drop until this spring; however, our biological studies(8) have shown that the ratio of these radionuclides in Alaskan caribou flesh dropped by a factor of 4 during the period from just prior to the last test series (October 1961) through July 1963. This suggests that a major infection of Cs^{134} into the atmosphere occurred prior to this last test series.

The radionuclides Mn^{54}, Co^{60}, and Sb^{124} are formed by neutron activation of the cladding material or other inert material in or near nuclear bombs. Co^{60} and Mn^{54} were reported to be present in the atmosphere in 1958 by Marquez, et al(10,11) who found them in rain water collected between June 1957 and March 1958. Their presence was attributed to thermonuclear testing. It has been recently reported(4) that a number of activation products including Mn^{54}, Fe^{55} and Sb^{124} were produced in relatively high abundance in the high yield detonations carried out by the USSR at Novaya Zemlya in the Fall of 1961. These tests may be a major source of the Mn^{54}, Co^{60}, Y^{88}, Sb^{124} and some of the other activation products which are included in this present study.

The Be^{7} is produced by cosmic ray spallation reactions in the atmosphere and its presence was first reported in the atmosphere by Arnold and Al-Salik in 1955(10) who chemically separated it from rain water samples. Measurements of its concentration in the atmosphere at various altitudes and latitudes have been reported(8, 12,13). In this present chronological study its concentration has just been observed during 1964. It is interesting to note that it has not shown the large Spring concentration increase typical of the fallout radionuclides. A longer period of observation will be required to determine its fallout pattern relative to atomic weapons produced radionuclides.

It is extremely interesting and significant that the activation products Y^{88} (105 d) and Sb^{124} (60 d) are at concentrations this year which are comparable with those one year ago while the fission product Zr^{95}-Nb^{95}(65 d) is about a factor of 20 lower. Also, the fission product Cs^{137} (30 Y) is at a concentration comparable with last year's and the Ru^{106} (1 yr) and Ce^{144} (285 d) are at about half their last year's concentrations. Since there have been no above ground atomic detonations during 1963 and 1964[14] all of these radionuclides entered the atmosphere prior to 1963 and their reservoir in the atmosphere has decreased with their respective half-lives since that time. This points to an obvious difference in the source of Y^{88} and Sb^{124} relative to most of the $Zr^{95}Nb^{95}$, and the other fission products and also allows one to estimate the rate of fallout of the radioactive material which was generated at the time the Y^{88} and Sb^{124} were formed. A continuation of these measurements is expected to establish any other differences in the fallout rates and origin of the activation products.

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FIGURE 1

RADIONUCLIDE CONCENTRATIONS IN THE AIR
AT RICHLAND, WASHINGTON DURING 1962-1963
d/m per 10^6 scf

Date	Be^7	Na^{22}	Mn^{54}	Co^{60}	Y^{88}	Zr^{95}-Nb^{95}	Ru^{106}	Sb^{124}	Sb^{125}	Cs^{134}	Cs^{137}	Ce^{144}
1-62*		11.9	1 870	26.2		287 000	27 600		2 590	7.8	2 730	53 500
2-62*		41.6	1 690	83.3		349 000	49 000		4 220	28.2	4 260	90 300
3-62*		0.863	722	28.1		85 700	15 400		1 490	2.87	1 470	12 400
4-62		13.5	3 520	26.2		59 600	57 200		1 900	20.3	4 430	86 100
5-62		8.85	1 350	15.0		114 000	25 000		2 470	10.4	3 900	43 600
6-62		15.8	2 310	18.9		113 000	34 100		5 200	13.3	4 540	58 800
7-62		11.5	2 180	17.3		83 200	25 900		3 620	12.2	3 710	43 000
8-62		7.54	1 500	13.5		44 000	13 300		1 890	6.50	1 810	20 500
9-62		12.7	2 800	19.9		97 900	20 700		2 890	19.4	2 750	36 500
10-62		4.89	1 260	10.6		70 400	11 300		1 170	10.7	1 290	19 900
11-62		5.86	1 640	13.0	25.7	95 800	10 500		1 280	12.1	1 450	20 000
12-62		3.97	1 210	11.3		89 700	8 090		996	3.28	1 030	19 200
1-63A**		4.33	1 670	58.0	20.5	156 000	9 540	68	1 330	11.2	1 490	22 500
1-63B		10.8	5 070	61.9	40.2	436 000	32 200	399	4 340	24.6	4 410	69 800
2-63A		10.4	3 200	324	20.9	188 000	19 400	202	5 810	10.4	2 610	38 400
2-63B		4.96	2 320	15.0	17.0	105 000	14 200	74	1 700	5.32	1 730	27 300
3-63A		12.1	4 060	28.8	34.4	158 000	26 900	201	2 900	9.44	3 090	45 200
3-63B		9.36	4 620	24.8	22.6	142 000	26 200	135	2 920	8.53	2 960	45 700
4-63A		13.9	5 890	32.1	24.4	145 000	33 900	159	3 240	10.2	5 130	55 400
4-63B		22.7	8 550	44.1	35.4	202 000	45 700	159	5 350	13.7	5 080	75 700
5-63A		27.8	10 900	56.5	48.1	184 000	58 400	145	6 380	19.5	6 550	90 000
5-63B		55.0	25 700	107	99.1	307 000	104 000	422	12 300	49.3	13 400	162 000
6-63A		21.3	9 190	42.1	27.9	101 000	40 900	125	5 100	16.7	5 010	59 400
6-63B		27.8	13 500	58.6	53.4	134 000	52 400	167	8 420	23.7	7 390	78 000
7-63A		34.0	12 100	54.7	51.8	105 000	49 600	137	6 450	21.6	5 990	60 500
7-63B		39.5	13 500	59.9	67.0	106 000	60 500	185	6 890	23.6	6 910	86 000
8-63A		39.5	11 900	52.3	77.2	92 400	56 300	169	7 160	33.3	6 980	85 000
8-63B		24.7	8 050	35.6	57.8	56 900	39 100	110	4 820	15.4	5 030	56 200
9-63A		16.4	5 340	25.0	44.4	36 100	27 500	99	3 940	17.6	3 820	40 600
9-63B		15.2	4 890	77.2	45.7	33 100	25 200	84	4 540	11.8	3 890	27 700
10-63A		17.9	3 700	38.1	44.1	21 200	23 400	97	2 620	13.1	3 010	33 600
10-63B		5.80	1 850	10.1	18.2	9 690	11 000	30	1 480	6.57	1 620	16 500
11-63A		6.43	1 730	7.94	16.9	7 830	9 480	33	1 440	24.5	2 060	12 800
11-63B		6.17	2 190	11.4	23.4	9 240	12 400	52	1 620	10.2	2 170	16 300
12-63A		5.75	963	6.24	12.6	2 620	7 360	34	757	6.64	1 030	9 150
12-63B		5.82	1 490	6.88	17.5	5 130	8 090	38	1 580	9.24	1 700	9 810
1-64A	4 330	3.42	794	11.8	8.73	3 230	5 690	29	679	6.11	870	7 030
1-64B	2 620	4.01	866	4.83	9.31	2 450	5 940	26	854	4.91	1 020	7 080
2-64A	6 310	9.61	2 170	13.6	25.4	5 690	14 800	73	2 320	11.5	2 620	18 000
2-64B	4 760	9.48	1 840	38.9	20.0	4 460	11 800	67	1 780	9.69	2 140	16 300
3-64A	3 790	7.21	1 330	9.64	16.1	3 210	10 100	61	1 320	2.70	1 570	12 500
3-64B	5 980	11.6	1 970	14.7	25.3	4 360	14 700	88	2 070	15.1	2 640	18 800
4-64A	3 500	13.0	1 630	27.2	24.0	3 270	14 300	79	1 780	7.23	2 250	17 000
4-64B		14.0	2 190	20.2	34.3		19 800	109		10.6		23 700
5-64A		19.4	3 400	24.4	40.9		24 200	124		5.70		29 700
5-64B		34.2	5 130	136	52.3		28 800	179		15.0		42 800

* Only small samples were available and the Na^{22} and Cs^{134} values give only the order of magnitude.

** A and B signify approximately the first and second half of each month, respectively.

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