NRL Report 6305

Filter Pack Technique for Classifying Radioactive Aerosols by Particle Size

Part 3 - The Size Distribution of Airborne Fission Products During 1963 and 1964

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ABSTRACT

Some 60-odd collections of radioactive particulate matter dispersed in the atmosphere have been made during the past two years by use of a filter pack technique utilizing four superimposed filters. The distribution of radioactivity among the filters has been analyzed mathematically by means of four simultaneous linear algebraic equations which relate the particle size of the retained material to the filter characteristics so as to permit the assignment of radioactivity in each collection to four major size groupings: 1.1μ, 0.6μ, 0.3μ, and 0.15μ particle diameter.

The fission-product radioactivity collected at ground level is associated with particles having an "average" diameter between 0.3 and 1.1μ (assuming spherical particles of 1.8 g/cc density). The conclusion is reached that the extremely small radioactive particulates characteristic of nuclear bomb debris which have resided for an extended period in the stratosphere become attached to the larger nonradioactive aerosols typical of troposphere air by the time they arrive at ground level.

A comparison of the relative amounts of various long-lived fission products (Ce^{144}, Pm^{147}, Sr^{90}, Cs^{137}) and of the natural isotope RaD (Pb^{210}) in month-long collections by four-filter packs shows no significant evidence of fractionation effects among the long-lived fission products. This confirms the importance of the attachment process to the size of radioactive fission product conglomerates detected at ground level. Fresh debris from the Chinese nuclear test, however, exhibited a different isotopic distribution with size than did the older bomb debris. Proportionately more of the shorter-lived radioactive fission products collected during transit of debris from the Chinese test appeared in the largest particle size grouping.

PROBLEM STATUS

This is an interim report; work on this problem is continuing.

AUTHORIZATION

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FILTER PACK TECHNIQUE FOR CLASSIFYING RADIOACTIVE AEROSOLS BY PARTICLE SIZE

PART 3 - THE SIZE DISTRIBUTION OF AIRBORNE FISSION PRODUCTS DURING 1963 AND 1964

INTRODUCTION

Previous work (1-3) at this Laboratory has demonstrated the ability of a pack of superimposed filters of different retentivities to effect some classification of airborne radioactive particles by size on passage of an air stream through them in sequence. This earlier work has also indicated that quite large, random variations in the apparent particle size distribution of airborne fission products do occur and that these variations are related to meteorological phenomena rather than to seasonal effects or to the residence time of nuclear debris in the upper atmosphere.

Radiochemical analyses have demonstrated the absence of any significant degree of fractionation of various fission product radionuclides relative to one another among the different particle size groupings. This has led to the suggestion that radioactive debris from nuclear explosions that have resided in the upper atmosphere for several months become attached to the larger, nonradioactive atmospheric aerosols on their downward migration through the troposphere to ground level. Similar conclusions have been reached by workers at the Northeastern Radiological Health Laboratory, Winchester, Massachusetts, using a variation of this technique (4).

The large quantity of data collected at Washington, D.C., during the past two years by the NRL filter pack technique utilizing four superimposed filters has been subjected to mathematical analysis, so that quantitative estimates can now be given of the contributions of four different size groupings to the total airborne radioactivity.

EXPERIMENTAL PROCEDURES

Collections of long-lived, airborne radioactive particles have been made at ground level using four superimposed filters whose retention properties as a function of particle size and air velocities have been characterized (1,5). The moving air stream initially is intercepted by two pieces of IPC 1478 paper (Institute of Paper Chemistry), then transits a single piece of Type 5G paper (Hollingsworth and Vose), and finally deposits its remaining particulate matter on a sheet of Type 6 "absolute" filter material, as described previously (1). The distribution of particles among the filters of this pack under the collection conditions is shown in Fig. 1 as a function of particle diameter (assuming spherical particles of 1.8 g/cc density). This figure is consistent with the physical characterization of the individual filters (1) and the mathematical analysis of the radioactivity distribution actually found.

Radioactivity determinations have been made by counting comparable areas in the centers of the 4-in.-diameter filters in sequence on the same counter so as to avoid problems associated with counter geometry, changing counter characteristics, or radioactive decay. Samples were counted for β activity using conventional equipment after a minimum delay of one week following collection to permit decay of the natural radioactive component

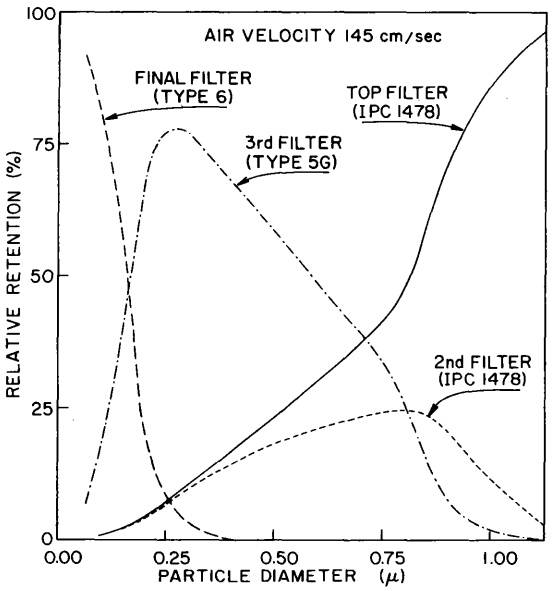

Fig. 1 - Effect of size on the distribution of particles among filters in a four-filter pack

due to thoron (ThB, 10.6-hr half-life). Long counting times (up to 18 hr) were employed to give good statistical accuracy. Sampling periods of 2 to 4 days were generally sufficient to give reasonable radioactivity collections; however, during the latter part of 1964, the statistical accuracy did suffer because of low ambient fission-product concentrations. The few natural radioactivity measurements employed shorter collection periods (30 min) followed by simultaneous counting of the short-lived radon daughter products on intercalibrated β counters.

Pertinent information on the 59 fission-product collections made during the period January 1963 through January 1965 is contained in Table 1. The overall correction factor for converting counts/min to disintegrations/min was 26.8 based on the area of filter counted (31.6%) and the counter efficiency toward 1.0-MeV β particles (11.8%). Supplementary information obtained on other equipment during October and November 1964 is presented in Table 2 to show the transient effect of debris from the Chinese atomic explosion of October 16, 1964, on atmospheric radioactivity in the Washington, D.C., area.

In addition to the above collections, large samples of airborne radioactivity were collected weekly on packs of three superimposed filters (initial filter, IPC 1478; second filter, Type 5G; final filter, Type 6 or Whatman GF/A) each 13 in. by 17 in., through which air was drawn at the rate of approximately 152 cfm (4.34×10^4 m^3 per week) or at an average linear velocity of 61 cm/sec, as described previously (2). These week-long collections were combined to give month-long samples characteristic of the months of February, May, August, and November, 1964. These four composited samples were subjected to radiochemical analysis for long-lived isotopes (Ce^{144}, Pm^{147}, Sr^{90}, Cs^{137}, Pb^{210}) using established procedures to determine the extent of isotopic fractionation as a function of particle size. A 5-day collection made in late October during passage of the Chinese bomb debris through the Washington, D.C., area was also subjected to radiochemical analysis. The radiochemical results are given in Table 3.

MATHEMATICAL ANALYSIS OF RADIOACTIVITY DISTRIBUTIONS

The first attempts to correlate the size distributions of airborne radioactive particulate matter with the relative amounts of radioactivity deposited on filters in three- or four-filter packs through use of simultaneous linear algebraic equations were not too

Table 1
Filter Pack Collections of Airborne Fission Products

Collection Date	Time (hr)	Precipitation (in.)	Air Flow (cfm)	Air Flow (cm/sec)	Volume Sample (m^3)	Gross β Activity (counts/min)*	Gross β Activity (pCi/m^3)	Radioactivity Distribution (%) IPC (1)	IPC (2)	Type 5G (3)	Type 6(4)
1963											
Jan. 18-21	63.5	0.42	15.9	134	1717	679.9 ± 3.5	4.78	51.34 ± 0.47	8.68 ± 0.10	36.22 ± 0.38	3.75 ± 0.05
Mar. 1-4	68	0.04	17.6	139	2035	1424 ± 4	8.45	33.60 ± 0.17	9.01 ± 0.08	52.98 ± 0.23	4.41 ± 0.03
Apr. 19-22	64	trace	17.2	136	1872	3040 ± 7	19.6	27.80 ± 0.14	7.80 ± 0.07	57.05 ± 0.21	7.35 ± 0.03
Sept. 20-23	68	0.12	18.25	144	2110	334.4 ± 1.5	1.91	36.06 ± 0.40	11.42 ± 0.18	49.43 ± 0.29	3.08 ± 0.09
Sept. 27-30	64	1.98	18.3	144	1991	568.5 ± 1.5	3.45	27.32 ± 0.18	11.29 ± 0.08	58.10 ± 0.23	3.29 ± 0.06
Oct. 4-7	68.5	0.0	19.5	148	2271	467.8 ± 1.7	2.49	26.25 ± 0.22	11.46 ± 0.08	58.59 ± 0.35	3.70 ± 0.07
Oct. 18-21	68	0.0	18.3	144	2115	380.3 ± 1.3	2.17	45.96 ± 0.29	11.33 ± 0.11	41.47 ± 0.26	1.24 ± 0.08
Oct. 25-28	72	trace	18.35	144	2246	173.7 ± 1.0	0.93	38.74 ± 0.41	11.92 ± 0.19	46.40 ± 0.49	2.94 ± 0.18
Nov. 1-4	66.5	0.71	18.7	147	2114	143.9 ± 0.9	0.82	34.82 ± 0.41	13.97 ± 0.29	48.30 ± 0.46	2.92 ± 0.21
Nov. 8-12	91.5	0.0	18.35	144	2854	160.9 ± 1.0	0.68	36.36 ± 0.52	12.37 ± 0.19	48.35 ± 0.48	2.92 ± 0.19
Nov. 15-18	71	0.0	18.35	144	2215	218.4 ± 1.0	1.19	32.23 ± 0.31	12.64 ± 0.15	53.11 ± 0.40	2.01 ± 0.14
Nov. 19-22	72.5	0.04	18.3	144	2256	446.1 ± 1.3	2.39	41.72 ± 0.22	11.95 ± 0.10	44.97 ± 0.23	1.37 ± 0.07
Nov. 22-26	96	1.00	18.5	146	3019	428.0 ± 1.2	1.71	35.05 ± 0.21	12.41 ± 0.10	50.54 ± 0.22	2.01 ± 0.07
Dec. 13-16	72	0.27	18.65	147	2283	155.9 ± 0.8	0.82	39.70 ± 0.38	13.86 ± 0.21	44.52 ± 0.40	1.92 ± 0.19
Dec. 20-24	92.5	0.64	18.35	144	2885	114.2 ± 0.8	0.48	43.87 ± 0.47	12.43 ± 0.28	41.68 ± 0.53	2.01 ± 0.26
Dec. 27-31	96	0.0	18.15	143	2962	235.5 ± 0.7	0.96	42.21 ± 0.22	11.63 ± 0.17	44.33 ± 0.22	1.83 ± 0.13
1964											
Jan. 15-17	54	0.0	18.75	148	1721	205.7 ± 0.9	1.44	41.57 ± 0.35	14.20 ± 0.15	43.07 ± 0.27	1.17 ± 0.20
Jan. 17-21	96	0.30	18.4	145	3003	235.9 ± 1.0	0.95	50.95 ± 0.37	12.80 ± 0.14	34.72 ± 0.30	1.53 ± 0.13
Jan. 24-27	74	0.62	18.85	148	2371	350.9 ± 1.3	1.79	31.35 ± 0.21	12.77 ± 0.13	53.15 ± 0.30	2.74 ± 0.09
Feb. 10-14	99.5	0.86	18.25	144	3087	416.4 ± 1.2	1.63	47.98 ± 0.26	11.48 ± 0.10	39.22 ± 0.19	1.32 ± 0.07
Feb. 17-20	76.5	1.00	18.55	146	2412	238.8 ± 1.1	1.19	42.67 ± 0.34	13.53 ± 0.18	41.54 ± 0.32	2.26 ± 0.13
Feb. 24-28	98.5	0.0	18.05	142	3023	333.8 ± 1.0	1.33	44.13 ± 0.25	11.71 ± 0.09	42.96 ± 0.20	1.20 ± 0.09
Mar. 6-9	78	0.26	18.55	146	2460	337.5 ± 1.2	1.66	43.32 ± 0.26	14.34 ± 0.10	40.39 ± 0.28	1.96 ± 0.09
Mar. 13-16	72	0.39	18.5	146	2264	330.2 ± 1.2	1.76	43.70 ± 0.29	15.29 ± 0.13	38.52 ± 0.26	2.48 ± 0.09
Mar. 20-23	73	1.05	18.6	146	2308	274.2 ± 1.2	1.43	45.44 ± 0.36	13.60 ± 0.16	39.20 ± 0.28	1.75 ± 0.11
Mar. 24-27	71	0.01	18.45	145	2227	311.3 ± 1.2	1.69	42.63 ± 0.31	10.60 ± 0.10	44.11 ± 0.28	2.67 ± 0.10
Apr. 3-7	73	0.86	18.5	146	2296	456.6 ± 1.1	2.40	39.03 ± 0.18	13.05 ± 0.10	45.75 ± 0.19	2.17 ± 0.07
Apr. 7-10	72	0.96	18.1	143	2215	273.0 ± 0.9	1.49	33.30 ± 0.25	12.78 ± 0.12	52.01 ± 0.24	1.90 ± 0.11
Apr. 10-13	72	0.01	18.3	144	2240	489.6 ± 1.2	2.64	35.33 ± 0.22	11.85 ± 0.09	50.69 ± 0.16	2.12 ± 0.06
May 1-4	72	trace	18.7	147	2289	408.2 ± 1.1	2.15	37.90 ± 0.20	11.42 ± 0.08	47.52 ± 0.22	3.16 ± 0.08
May 8-11	67.5	0.0	18.9	149	2169	515.4 ± 1.4	2.87	38.42 ± 0.19	9.93 ± 0.10	47.67 ± 0.22	3.98 ± 0.08
May 22-25	64	trace	18.95	149	2062	471.5 ± 1.2	2.76	41.02 ± 0.18	11.06 ± 0.09	45.22 ± 0.21	2.71 ± 0.07
June 19-22	65	0.61	18.85	148	2083	129.3 ± 0.7	0.75	46.33 ± 0.46	12.37 ± 0.25	38.82 ± 0.31	2.47 ± 0.23
June 26-29	67	0.03	18.95	149	2158	426.2 ± 1.4	2.38	41.60 ± 0.24	11.19 ± 0.10	44.42 ± 0.28	2.79 ± 0.07
July 2-6	96	0.20	18.65	147	3043	374.3 ± 1.1	1.48	50.28 ± 0.24	10.55 ± 0.09	37.19 ± 0.18	1.98 ± 0.08
July 10-16	144	0.38	18.35	144	4492	363.0 ± 1.2	0.98	48.73 ± 0.25	10.41 ± 0.09	38.90 ± 0.28	1.96 ± 0.09
July 23-27	95.5	trace	18.9	149	3069	41.6 ± 0.7	0.16	59.6 ± 1.4	13.0 ± 0.8	26.0 ± 0.9	1.4 ± 0.8
July 31-Aug.3	71.5	1.25	18.8	148	2285	215.7 ± 0.9	1.14	46.73 ± 0.34	12.19 ± 0.15	38.53 ± 0.29	2.55 ± 0.14
Aug. 7-10	71.5	trace	18.6	146	2261	161.2 ± 0.9	0.86	46.03 ± 0.40	12.34 ± 0.20	39.02 ± 0.38	2.61 ± 0.19
Aug. 14-17	72	0.07	18.75	148	2295	113.1 ± 0.8	0.59	51.99 ± 0.57	10.34 ± 0.28	34.84 ± 0.43	2.83 ± 0.27
Aug. 21-24	72	trace	18.55	146	2271	98.1 ± 0.7	0.52	48.32 ± 0.52	11.31 ± 0.31	37.41 ± 0.49	2.96 ± 0.31
Aug. 28-31	72	0.57	18.4	145	2252	49.2 ± 0.7	0.26	55.5 ± 1.1	11.8 ± 0.6	29.7 ± 0.7	3.0 ± 0.6
Sept. 4-8	96.5	0.0	18.8	148	3084	167.5 ± 0.9	0.66	52.60 ± 0.46	8.48 ± 0.19	36.24 ± 0.36	2.69 ± 0.18
Sept. 11-14	71.5	0.34	19.1	150	2322	78.8 ± 0.8	0.41	56.47 ± 0.85	8.50 ± 0.39	33.00 ± 0.72	2.03 ± 0.38
Sept. 18-21	72	0.62	19.0	150	2326	75.7 ± 0.8	0.39	56.14 ± 0.89	12.15 ± 0.41	30.78 ± 0.62	0.93 ± 0.40
Sept. 25-28	72.5	0.28	18.65	147	2298	66.9 ± 0.8	0.35	52.77 ± 0.98	10.31 ± 0.47	34.83 ± 0.73	2.09 ± 0.45
Oct. 2-5	72	0.04	18.8	148	2301	71.7 ± 0.8	0.37	49.51 ± 0.89	11.16 ± 0.44	36.54 ± 0.81	2.79 ± 0.42
Oct. 16-19	72	0.70	18.75	148	2295	41.2 ± 0.8	0.22	52.9 ± 1.6	11.9 ± 0.8	34.0 ± 1.2	1.2 ± 0.7
Oct. 23-26	65.5	0.0	18.6	146	2071	85.8 ± 0.9	0.50	68.65 ± 1.00	7.34 ± 0.36	22.49 ± 0.52	1.52 ± 0.35
Oct. 29-30	29.5	0.0	18.45	145	925	142.5 ± 0.9	1.86	64.07 ± 0.58	7.23 ± 0.26	27.65 ± 0.33	1.05 ± 0.22
Oct. 30-Nov.2	72	0.0	18.8	148	2301	110.4 ± 0.9	0.58	64.04 ± 0.76	7.70 ± 0.28	26.81 ± 0.42	1.45 ± 0.27
Nov. 2-4	48	0.0	18.6	146	1518	69.0 ± 0.8	0.55	65.80 ± 1.05	7.97 ± 0.45	25.22 ± 0.65	1.01 ± 0.44
Nov. 4-6	48	0.0	18.95	148	1546	57.8 ± 0.8	0.45	63.49 ± 1.07	7.61 ± 0.55	28.55 ± 0.80	0.35 ± 0.52
Nov. 6-9	64.5	0.0	18.4	145	2018	66.3 ± 0.8	0.40	56.71 ± 0.91	9.95 ± 0.47	30.92 ± 0.84	2.42 ± 0.45
Nov. 13-16	64.5	0.01	18.4	145	2018	60.6 ± 0.7	0.36	46.53 ± 0.85	9.74 ± 0.51	40.43 ± 0.81	3.30 ± 0.49
Nov. 20-23	64.3	0.0	18.6	146	2033	67.2 ± 0.7	0.40	51.49 ± 0.80	10.27 ± 0.46	36.01 ± 0.71	2.23 ± 0.45
Nov. 27-30	68.5	trace	18.6	146	2166	71.2 ± 0.7	0.40	51.26 ± 0.76	10.81 ± 0.44	33.43 ± 0.65	4.50 ± 0.43
Dec. 4-7	64.5	0.41	18.65	147	2045	24.1 ± 0.7	0.14	45.2 ± 2.1	12.0 ± 1.3	34.9 ± 1.9	7.9 ± 1.3
1965											
Jan. 8-11	65.5	1.14 (snow)	18.4	145	2049	57.0 ± 0.7	0.34	54.21 ± 0.97	11.75 ± 0.55	30.18 ± 0.80	3.86 ± 0.53

*Total count rate for four filters; overall counting efficiency for 4-in.-diameter filters was 3.73%.
Indicated uncertainties are the standard deviations (σ) based on counting statistics alone.

Table 2
Detection of Radioactive Debris from the Chinese Atomic Explosion of October 16, 1964, by Air Monitor Equipment at NRL, Washington, D.C.

Collection Period*	Duration (hr)	Air Flow (cfm)	Volume Sampled (m^3)	Fission Product Radioactivity		Activity Concentration (pCi/m^3)
				(counts/min)†	(dis/min)‡	
Oct. 20-21	24	27.0	1102	37	313	0.13
21-22	24	31.3	1277	87	737	0.26
22-23	24	25.4	1036	83	703	0.31
23-26	72	16.5	2020	213	1804	0.40
26-27	24	23.6	963	227	1923	0.90
27-28	24	26.5	1081	1908	16160	6.73
28-29	24	28.9	1179	2543	21540	8.22
29-30	24	24.5	1000	806	6830	3.07
Oct. 30-Nov. 2	65	17.9	1978	274	2320	0.53
Nov. 2-4	55	18.6	1739	218	1846	0.48
4-6	48	19.7	1608	202	1711	0.48
6-9	65	16.9	1867	217	1838	0.44
9-10	24	19.9	812	105	889	0.49
10-12	No collection					
12-13	24	31.3	1277	184	1558	0.55
13-16	No collection					
16-17	24	27.9	1138	92	779	0.31
17-18	24	21.9	894	127	1076	0.54
18-19	24	25.0	1020	103	872	0.38
19-20	24	32.0	1306	50	424	0.15

*Collections ended at 1600 EST except for Nov. 2 when collection was stopped at 0900.
†Fission product count rate determined from ratio of 6th to 16th hour count rates.
‡Based on counting efficiency of 11.8% for 1.0 MeV β particles.

successful (1). The occurrence of negative values of the contributions from certain size groups required readjustment of the coefficient matrix of the equations. A more satisfactory set of coefficients was developed through the efforts of the Applied Mathematics Staff at NRL; this work is contained in another report in this series (6).

From a suggested extreme range of possible values for each element obtained from the measured efficiencies of these filters toward H_2SO_4 aerosols (spherical particles of 1.8 g/cc density), and from a series of 40 measurements of the distribution of radioactivity among filters in a four-filter pack, a four-by-four coefficient matrix was developed which led to size distributions which were realistic in that they contained no negative values. Later, an additional group of 22 collections was evaluated by using this same matrix. In the entire group of 62 collections made over a 2-year period and which included three collections of radon daughter activity having a distinctly different size distribution, only a single negative component was found; moreover, it was so slight that it could be attributed to the statistical error in counting.

The system of equations exhibiting this coefficient matrix and which applies to aerosol collections made on the described four-filter system operated at a linear velocity of 145 cm/sec is as follows:

Table 3
Results of Radiochemical Analyses of Filter Pack Collections at NRL During 1964

Collection Date	Sample Volume (m^3)	Filter	Radioactivity Distribution (%) on Filters								Selected Activity Ratios			
			Sr^{89}	Sr^{90}	Y^{91}	Cs^{137}	Ce^{141}	Ce^{144}	Pm^{147}	Pb^{210}	Ce^{144}/Pm^{147}	Ce^{144}/Sr^{90}	Cs^{137}/Sr^{90}	Pb^{210}/Sr^{90}
February (Jan. 31-Feb. 28)	1.74×10^5	(1) IPC 1478	(15.8)	23.0	19.9	22.0	*	20.1	20.5	9.0	3.3	12.8	1.37	0.13
		(2) Type 5G	(76.8)	72.6	76.4	72.7	*	75.7	74.7	72.1	3.4	15.3	1.43	0.32
		(3) Type 6	(7.4)	4.4	3.7	5.3	*	4.2	4.8	18.9	2.9	13.9	1.73	1.39
	Total Activity† (10^3 dis/min)		(8.18)	10.00	6.49	14.30	*	146.6	44.3	3.23	3.3	14.7	1.43	0.32
May (May 1-28)	1.67×10^5	(1) IPC 1478	*	21.5	22.6	22.4	*	20.9	22.3	10.2	2.0	9.3	1.42	0.06
		(2) Type 5G	*	68.2	68.2	67.6	*	69.7	68.7	70.9	2.2	9.9	1.35	0.14
		(3) Type 6	*	10.3	9.2	10.0	*	9.4	9.0	18.9	2.3	8.8	1.33	0.25
	Total Activity† (10^3 dis/min)		*	31.7	6.94	43.2	*	305.4	139.5	4.26	2.2	9.6	1.36	0.13
August (July 31-Aug. 28)	1.74×10^5	(1) IPC 1478	*	37.3	34.9	29.6	*	33.1	32.3	14.8	2.0	7.4	0.88	0.12
		(2) Type 5G	*	56.8	57.5	64.6	*	60.1	61.2	56.8	1.9	8.9	1.26	0.31
		(3) Type 6	*	5.9	7.6	5.8	*	6.8	6.5	28.4	2.1	9.8	1.08	1.48
	Total Activity† (10^3 dis/min)		*	10.2	0.92	11.26	*	85.6	43.9	3.13	1.9	8.4	1.11	0.31
October ‡ Oct. 29-Nov. 2	3.10×10^4	(1) IPC 1478	28.4	39.1	61.5	49.1	63.0	51.0	50.0	16.6	1.8	9.3	2.32	0.13
		(2) Type 5G	55.0	48.4	35.0	41.3	34.1	45.3	46.2	57.8	1.7	6.7	1.58	0.36
		(3) Type 6	16.6	12.5	3.5	9.6	2.9	3.7	3.8	25.6	1.8	2.1	1.40	0.61
	Total Activity† (10^3 dis/min)		14.4	1.32	9.02	2.44	20.33	9.45	5.32	0.403	1.8	7.2	1.85	0.31
November Nov. 2 - Dec. 1	1.80×10^5	(1) IPC 1478	34.5	45.6	56.7	49.4	60.7	45.2	46.2	16.0	1.7	6.9	1.82	0.54
		(2) Type 5G	57.2	50.9	40.0	46.8	36.6	50.8	49.9	66.9	1.8	7.0	1.55	2.01
		(3) Type 6	8.3	3.5	3.3	3.8	2.7	4.0	3.9	17.1	1.8	8.1	1.83	7.6
	Total Activity† (10^3 dis/min)		10.8	5.37	7.15	9.03	9.89	37.6	21.6	8.23	1.7	7.0	1.68	1.53

*Not detectable above background of other activity; ( ) uncertain values.
†Activity corrected for decay to midpoint of sampling period.
‡Collection made during transit of radioactive debris from the Chinese nuclear test of Oct. 16, 1964.

$$A_1 = 95.00\,x_1 + 30.00\,x_2 + 10.00\,x_3 + 2.00\,x_4$$

$$A_2 = 4.75\,x_1 + 21.00\,x_2 + 9.00\,x_3 + 1.96\,x_4$$

$$A_3 = 0.25\,x_1 + 48.80\,x_2 + 77.80\,x_3 + 38.00\,x_4$$

$$A_4 = 0.00\,x_1 + 0.20\,x_2 + 3.20\,x_3 + 58.04\,x_4\ .$$

In this system A_1, A_2, A_3, and A_4 are the measured activity distributions (in percent of the total) on the four filters (IPC 1478 through Type 6, respectively), and x_1, x_2, x_3, and x_4 are the relative amounts of radioactivity associated with particles in the four size groupings, from largest to smallest size. Solutions to these equations are readily obtainable by use of any of a number of computers, most of which already have programs available.

Comparison of the coefficients of the x terms in the aforementioned matrix with the particle size distributions shown in Fig. 1 indicates that the x_1 group corresponds to an average particle size of 1.1μ (diameter); x_2 corresponds to 0.6μ; x_3, to 0.3μ and x_4, to 0.15μ. These sizes are really particle size distributions of unknown form centered about the indicated particle size.

Six hypothetical cases covering the range of observed fission product distributions have been evaluated by solving the resulting systems of simultaneous linear equations to determine the magnitude of the variations in the calculated size distributions which result from statistical errors in counting. These results are described in the Appendix. To

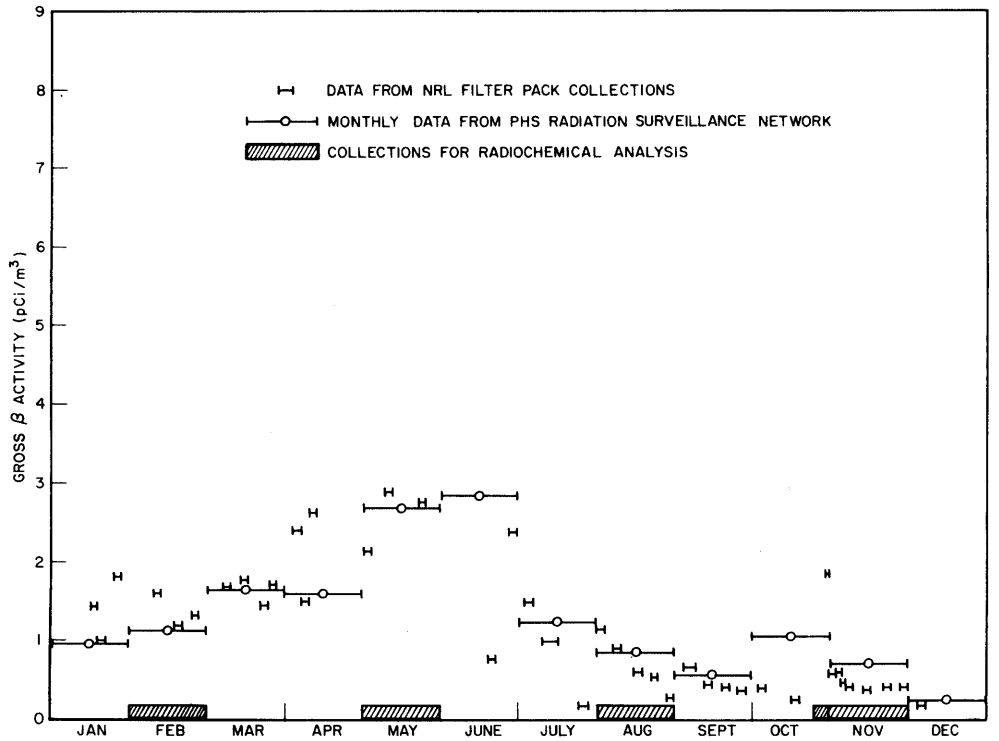

Fig. 2 - Concentrations of fission-product β activity in the ground-level air at Washington, D.C., during 1964

summarize here, each particle size grouping (x term) is found to be most strongly influenced by statistical variations (or errors) in the measured relative radioactivity of the corresponding filter (A term). There is some crossover or feedback of errors between A_2 and x_1, A_1 and x_2, A_3 and x_2, and A_2 and x_3. Since x_4 is generally quite small in the case of fission products, the percentage error in this grouping may be quite large even though the absolute error is small.

The differences in the distributions of radioactivity with particle size obtained for the various collection periods are far greater than can be attributed to errors in counting; the observed differences must represent real changes in the radioactive component of atmospheric dusts.

RESULTS AND DISCUSSION

Gross Fission-Product β Activity in the Air

The average fission-product concentrations in the air at ground level have been calculated for each of the filter pack collections made during 1963 and 1964 (Table 1). The 1964 air concentrations in picocuries per cubic meter (pCi/m^3) are shown in Fig. 2 as a function of time, together with the monthly average radioactivity levels reported by the Public Health Service Radiation Surveillance Network for the Washington area. The fission product concentrations have decreased considerably since the spring of 1963 (2), when new records were established; moreover, the day-to-day variability is also considerably less, possibly because of a more uniform stratospheric source. The decrease in activity levels between 1963 and 1964 (a factor of 2 to 3), however, was not nearly so great as in the 1959 to 1960 period when a 20-fold decrease in corresponding spring peaks was

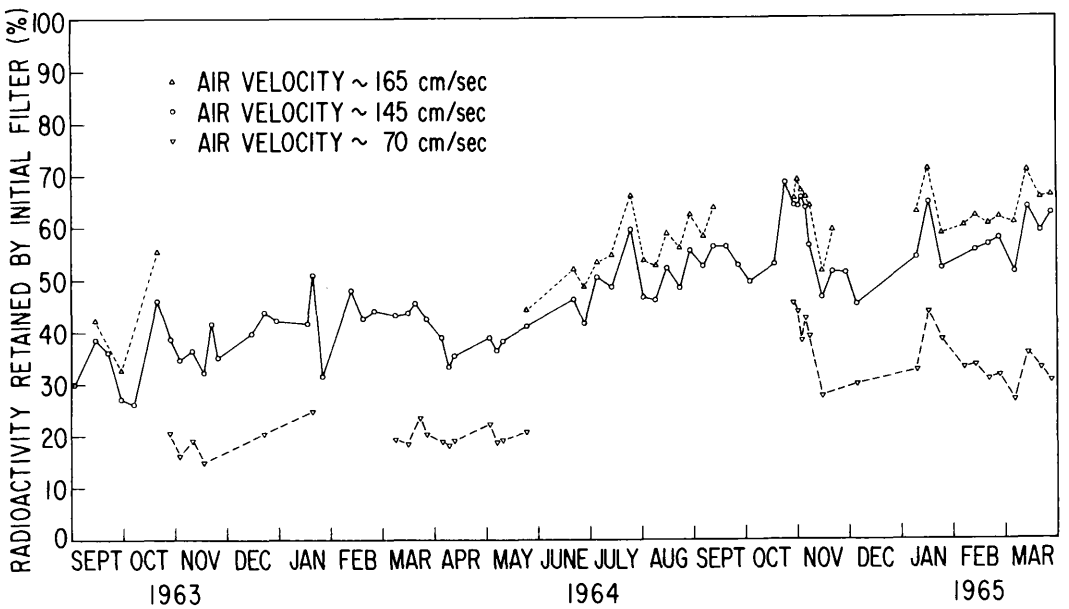

Fig. 3 - Temporal changes in the retention of gross fission-product radioactivity by the initial IPC 1478 filter in a pack

observed (7). This slower disappearance of radioactivity at ground level during the 1963-64 period is probably directly related to the larger size of the nuclear explosions carried out in 1961 and 1962 and the higher altitude of stabilization of the radioactive clouds.

Temporal Changes in the Largest Component of Atmospheric Fission Products

The fraction of the total airborne fission-product radioactivity retained by the poorest filter in the multifilter system is a qualitative measure of the contribution of the larger radioactive aerosols to the total radioactivity; the variation of this fraction with time is indicative of gross changes taking place in this component. In previous work it was shown that the relative contribution of radioactivity from this component decreased rather markedly following cessation of nuclear testing and then appeared to undergo random changes with time (1-3).

The percentages of the gross fission-product radioactivity retained by the initial IPC filter in the extended series of filter pack collections made during the period September 1963 through March 1965 are shown in Fig. 3 as a function of the collection date. Corresponding results obtained from other pack collections made at different air velocities (and a resulting different retention efficiency for the larger aerosols) indicate that the variability of the large-particle component is a characteristic of the atmospheric aerosol distribution and not an artifact introduced by the sampling or analytical procedure. No explanation can be offered for the observed general increase in the relative number of large radioactive aerosols observed during the latter half of 1964. However, the peak in the large-aerosol component during the latter part of October 1964 coincides with the arrival of debris from the Chinese atomic test of October 16. The transitory nature of this event is evident here and also in the data presented in Table 2.

Particle Size Distribution of Airborne Fission Products

The distribution of radioactivity among the filters in the four-filter packs (Table 1) has been analyzed by means of the system of four simultaneous equations described in an earlier section; the resulting radioactivity distributions by size groups are presented in Table 4. The most striking feature is the generally negligible contribution of the smallest particles (nominally 0.15μ diameter) to the total fission-product conglomerate. In contrast, analyses of the distribution of the radon daughters, RaB plus RaC, on identical filter packs indicate this radioactivity to be associated largely with particles less than 0.3μ in diameter (Table 5).

Temporal changes in the distribution of fission-product radioactivity with particle size during a two-year period are depicted in Fig. 4, based on the radioactivity distribution on the four-filter pack. The lower curve, which is the quantitative assignment to the larger particle size grouping (1.1μ), shows an increase in relative radioactivity during 1964 with a peak during the period of transit of the Chinese bomb debris. The sums of the two largest size groupings (1.1μ and 0.6μ) are rather erratic and do not show this progressive change with time. These groupings together have, since mid-October 1963, included between 50 and 90% of the gross long-lived β activity. Most of the remaining activity has been in the 0.3μ grouping with the generally small remainder falling into the 0.15μ range. The three areas into which the graph is divided by the two curves thus represent roughly the average radioactivity in the 0.3μ, 0.6μ, and 1.1μ groups, respectively, from top to bottom.

The overall averages of radioactivity assigned to the four size groups are presented by quarters and half-years in Table 6. The causes of the observed changes are not apparent, but there is a real trend toward the concentration of activity in the 1.1μ group at the expense of the smaller groups. Radioactivity collected during the two-week period around the end of October, however, displayed a quite different size distribution, with a shift of activity primarily from the 0.6μ to the 1.1μ grouping.

Aside from the radioactivity from the Chinese atomic test, all airborne fission products were produced prior to the end of 1962 and, after mid-1963 at the latest, must have come from the stratosphere. Thus, their size on arrival at the ground level collection site must be related to interactions taking place between the small stratospheric aerosols and various components of the troposphere. Seasonal variations in tropospheric aerosol types and concentrations are known to exist and the concentrations, moreover, are strongly influenced by weather. Thus, seasonal variations in the size distribution of the fission-product aerosol found at ground level would not be surprising, nor would random changes occurring because of the variable past history of the air mass. None of these obvious solutions can account for the observed trend of the increasing association of radioactive fission products with the largest aerosol size group.

Radiochemical Results

Radiochemical analyses were performed on composite collections of 4 or 5 weeks' duration made during the months of February, May, August, and November 1964 and on a 5-day collection made during the period of transit of the Chinese atomic bomb debris. The relationship of these collections to the gross fission-product activity levels in the Washington area is shown in Fig. 2. The first three collections cover the period of increase and decrease of radioactivity resulting from the spring maximum in stratospheric deposition, while the last two document the atmospheric radioactivity concentrations resulting from the Chinese test. The results of the radiochemical analyses are shown in Table 3.

Table 4
Distribution of Fission Product Radioactivity Among
Four Particle Size Groupings, 1963-1964

Collection Date	Gross Activity* (pCi/m³)	Radioactivity Distribution (%) by Size Groups			
		1.1μ	0.6μ	0.3μ	0.15μ
1963					
Jan. 18-21	4.78	45.29	16.03	34.16	4.52
Mar. 1-4	8.45	25.45	11.65	58.41	4.49
Apr. 19-22	19.6	21.14	2.74	67.17	8.95
Sept. 20-23	1.91	23.81	30.26	43.11	2.83
Sept. 27-30	3.45	14.57	25.66	57.36	2.42
Oct. 4-7	2.49	13.07	26.98	56.81	3.15
Oct. 18-21	2.17	34.83	31.87	33.10	0.20
Oct. 25-28	0.93	25.81	35.21	36.03	2.96
Nov. 1-4	0.82	17.83	49.63	29.29	3.24
Nov. 8-12	0.68	22.41	37.70	37.03	2.86
Nov. 15-18	1.19	17.52	37.05	44.55	0.88
Nov. 19-22	2.39	29.10	35.05	35.57	0.28
Nov. 22-26	1.71	20.99	36.47	41.50	1.05
Dec. 13-16	0.82	23.39	49.81	25.05	1.76
Dec. 20-24	0.48	30.50	40.45	27.22	1.82
Dec. 27-31	0.96	30.18	33.24	35.51	1.08
1964					
Jan. 15-17	1.44	24.85	52.50	22.04	0.62
Jan. 17-21	0.95	37.53	45.93	14.89	1.66
Jan. 24-27	1.79	16.27	38.48	43.03	2.22
Feb. 10-14	1.63	36.73	34.01	28.68	0.58
Feb. 17-20	1.19	27.18	48.87	21.41	2.55
Feb. 24-28	1.33	32.16	34.02	33.73	0.09
Mar. 6-9	1.66	26.43	55.21	16.06	2.30
Mar. 13-16	1.76	25.07	63.42	7.90	3.62
Mar. 20-23	1.43	30.09	50.07	17.99	1.85
Mar. 24-27	1.69	32.44	26.21	38.99	2.36
Apr. 3-7	2.40	24.12	43.41	30.57	1.90
Apr. 7-10	1.49	18.43	38.50	42.26	0.81
Apr. 10-13	2.64	22.30	32.29	44.30	1.10
May 1-4	2.15	25.79	31.14	39.93	3.14
May 8-11	2.87	29.01	20.51	46.25	4.24
May 22-25	2.76	29.86	29.18	38.52	2.44
June 19-22	0.75	33.24	41.56	22.31	2.90
June 26-29	2.38	30.24	30.54	36.53	2.69
July 2-6	1.48	40.87	28.40	29.02	1.71
July 10-16	0.98	39.45	26.60	32.46	1.50
July 23-27	0.16	46.59	51.02	0.09	2.30
July 31-Aug. 3	1.14	33.99	40.39	22.61	3.01
Aug. 7-10	0.86	32.96	41.33	22.61	3.11
Aug. 14-17	0.59	43.04	28.40	25.17	3.39
Aug. 21-24	0.52	37.29	34.59	24.50	3.63
Aug. 28-31	0.26	44.17	41.58	9.68	4.58
Sept. 4-8	0.66	47.10	13.86	36.47	2.58
Sept. 11-14	0.41	51.30	14.99	32.03	1.68
Sept. 18-21	0.39	44.37	42.42	12.44	0.77
Sept. 25-28	0.35	43.99	27.71	26.24	2.06
Oct. 2-5	0.37	38.86	33.74	24.04	3.37
Oct. 16-19	0.22	41.33	39.28	18.44	0.95
Oct. 23-26	0.50	66.64	10.59	21.36	1.40
Oct. 29-30	1.86	61.92	7.23	30.76	0.09
Oct. 30-Nov. 2	0.58	61.00	11.35	26.66	0.99
Nov. 2-4	0.55	62.45	13.77	23.38	0.40
Nov. 4-6	0.45	60.65	9.22	31.28	-1.15
Nov. 6-9	0.40	48.89	26.93	21.27	2.90
Nov. 13-16	0.36	38.19	21.80	36.41	3.60
Nov. 20-23	0.40	42.67	26.98	28.15	2.20
Nov. 27-30	0.40	41.26	33.58	18.54	6.62
Dec. 4-7	0.14	32.28	43.97	10.89	12.86
1965					
Jan. 8-11	0.34	42.79	41.58	9.66	5.97

*Fission product radioactivity averaged over collection period.

of maxima in the spring season, with some decay of Ce [illegible] being noted. The [illegible] also

Table 5
Filter Pack Collections of Natural Radioactivity (RaB+C)

Collection Period	Air Flow		Total Activity	Radioactivity Distribution (%)				Size Distribution (%)			
				IPC	IPC	Type 5G	Type 6				
	(cfm)	(cm/sec)	(counts/min)	(1)	(2)	(3)	(4)	1.1μ	0.6μ	0.3μ	0.15μ
Mar. 15, 1963 0810-0840	18.3	144	136.4±2.8	9.0	4.3	48.2	38.5	4.80	0.48	30.05	64.68
Mar. 27, 1963 0815-0845	18.5	146	270.5±2.8	9.91	5.69	50.57	33.83	3.61	6.80	33.16	56.44
Mar. 18, 1964 0830-0900	19.35	152	151.8±2.1	13.11	6.65	45.78	34.45	5.27	16.42	20.12	58.19

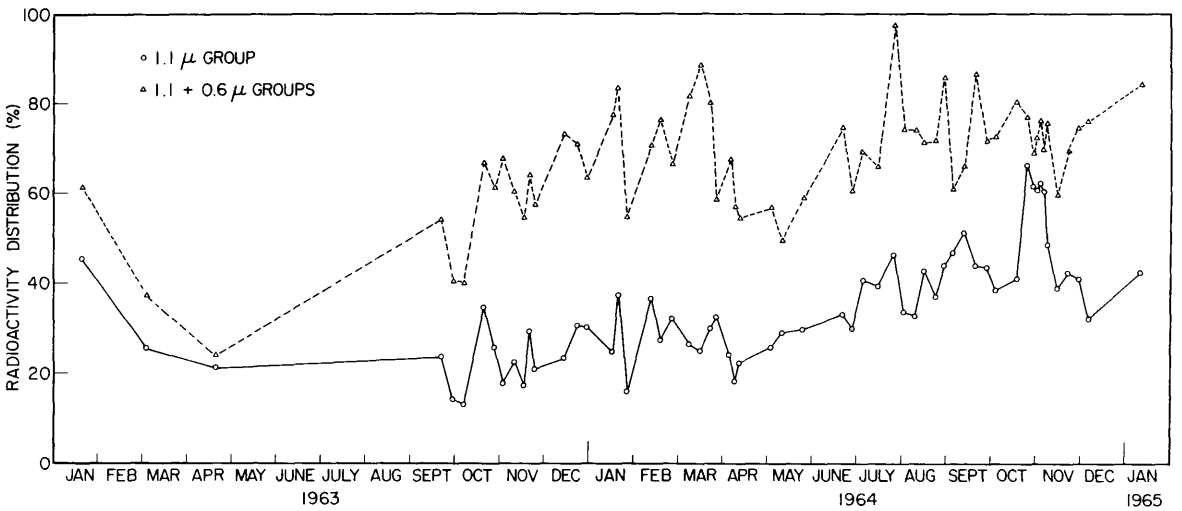

Fig. 4 - Temporal changes in the distribution of fission-product radioactivity with particle size, 1963-1964

Table 6
Changes in the Average Radioactivity Distribution by Size Groups During 1964

Period of Observation	Number of Measurements	Average Radioactivity Contribution of Size Group (%)*			
		1.1μ	0.6μ	0.3μ	0.15μ
1963					
Oct.-Dec.	11	24.2±2.0	37.6±2.1	36.5±2.7	1.7±0.3
1964					
Jan.-Mar.	10	28.9±2.0	44.9±3.6	24.5±3.6	1.8±0.3
Apr.-June	8	26.6±1.7	33.4±2.6	37.6±2.8	2.4±0.4
Jan.-June	18	27.9±1.3	39.8±2.7	30.3±2.7	2.0±0.3
July-Sept.	12	42.1±1.6	32.6±3.3	22.8±3.0	2.5±0.3
Oct.-Dec.†	7	40.5±1.9	32.3±2.9	22.5±3.1	4.6±1.5
July-Dec.†	19	41.5±1.2	32.5±2.3	22.7±2.2	3.3±0.6
Oct. 23-Nov. 6	5	62.5±1.1	10.4±1.1	26.7±2.0	0.35±0.44

*Indicated error is the standard deviation of the mean of the series of measurements.

†Less collections made during period Oct. 23-Nov. 6 during transit of Chinese debris.

These collections were made on a three-filter pack operated at a lower air speed (61 cm/sec linear velocity) than the four-filter packs, so that greater penetration of the larger particulates occurred. The distribution of various size particles among the filters in this pack is described elsewhere (2,3). It would be expected that the top filter (IPC 1478) would contain the bulk of the particles greater than 1.0 μ in diameter, that the second filter (Type 5G) would contain particles primarily in the 0.2 μ to 1.0 μ range, and that the remainder of the activity ($< 0.2\mu$) would be on the final filter. Over quite a wide range of particle sizes (0.3 to 0.8 μ diameter) roughly 75% of the particulate matter should be collected by the middle filter.

The analyses performed on seven collections made during 1963 showed between 65 and 75% of the total activity to be deposited on the middle filter and 8 to 15% to be on the final filter; moreover, there was no strong evidence of isotopic fractionation between the various size groups (2,3). The analyses of the 1964 collections reported here lead to the same conclusions, at least for material settling out from the stratospheric reservoir of old radioactive debris.

The previously noted progressive increase in average particle size with time during 1964 is duplicated in the radiochemical results, with a larger proportion of the various fission products appearing on the top (initial) filter in the pack. The contribution of very small particles containing fission products was less during 1964 than in 1963, but more of the long-lived natural lead (Pb^{210}) appeared in this small-particle grouping.

The February, May, and August collections exhibited quite different distributions of fission-product radioactivity as a function of particle size, but within any given collection the isotopic distribution was remarkably constant. The effects of weather and residence time on the old bomb debris are apparently not too selective, so that fractionation of the radionuclides relative to one another is of little importance. The Pb^{210}, whose average time of residence in the atmosphere is certainly much less than that of fission products from a stratospheric source, is associated with smaller aerosol particles and is strongly fractionated in comparison to the fission-product mixture.

A few changes were noted in the October and November 1964 collections which were made during and immediately following passage over Washington, D.C., of the radioactive cloud from the Chinese bomb test of October 16, 1964. The shorter-lived fission products Y^{91} (58-day half-life) and Ce^{141} (33-day half-life) showed a greater accumulation in the larger sized particles, while the long-lived rare earths (Pm^{147} and Ce^{144}), primarily from tests held prior to 1963, showed a significantly lower concentration in this size range. These fresh fission debris are, of course, from the Chinese bomb test, and the filter pack is in effect permitting a partial resolution of the debris from the old and new sources.

The fresh Sr^{89} appears to be associated with smaller particles than are the rare earths from this test, in agreement with the rather uncertain observation reported earlier (2).

Smaller yield bomb bursts taking place near the earth's surface produce a quantity of radioactive debris associated with particles in excess of 1 μ diameter which may be airborne for several weeks and be transported over great distances by tropospheric air movements. The arrival of these radioactive particles of larger than normal size unfortunately occurred at a time when, for unknown reasons, the stratospheric radioactivity was also associated with the larger aerosols. The differences between the radioactive debris from the two sources were thus not as distinct as might have been expected.

The air concentrations of a few of the isotopes of interest are plotted in Fig. 5 as a function of the time of collection. Both the Sr^{90} and Ce^{144} exhibit the expected patterns of maxima in the spring season, with some decay of Ce^{144} being noted. The Y^{91} also

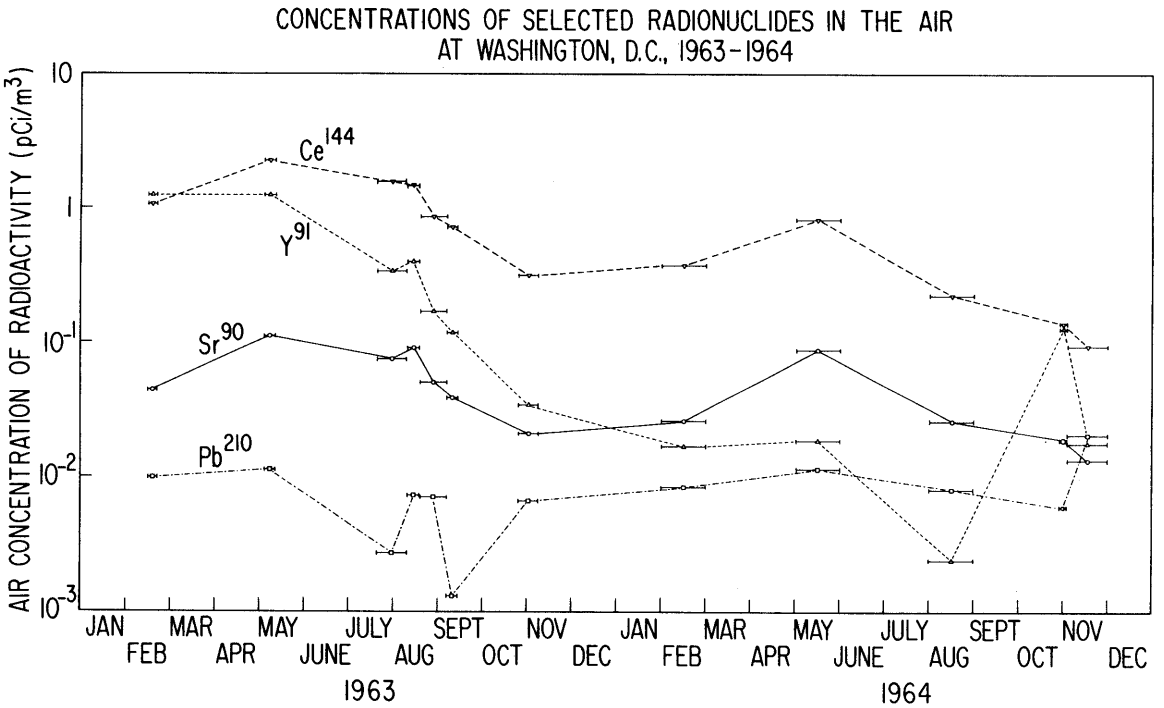

Fig. 5 - Concentrations of selected radionuclides in the air at Washington, D.C., 1963-1964

goes through the same seasonal changes, but the effects are modified by the more rapid decay of this isotope. In contrast, the concentration of the shorter-lived Y^{91} (also Ce^{141} and Sr^{89}) is very strongly influenced by the Chinese debris in the October and November collections, while no appreciable effect on the 285-day Ce^{144} and the 27-year Sr^{90} can be observed.

* Though the limited data obtained do not give a complete picture of the changes in the atmospheric concentration of fission products, they do indicate that the differences between 1964 and 1963 are much less than between 1960 and 1959 (8). The sequential changes in the concentrations of Pb^{210} are different from those of the fission products in conformity with its different mode of formation and vertical distribution.

CONCLUSIONS

The filter-pack method is capable of giving a reasonable resolution of aerosols by size groupings; it should be useful for aerosols of any composition for which sensitive analytical techniques (and a low background of contamination on the filter) are available. It is an even more powerful technique for monitoring changes in the size distribution of aerosols which may occur with time or changing environmental conditions.

In this report the inherent radioactivity of aerosols in the air at ground level was employed as the tracer. It has been found that fission products, particularly fresh bomb debris, are associated with the larger sized particles (in the 0.5 to 1.0 μ range), while the shorter-lived natural decay products of radon are associated with the smallest size range (0.3 μ and smaller). Growth of the small stratospheric aerosols to the near 1 μ size takes place during the several weeks required for their descent from the upper troposphere to ground level. Quite large, real variations in the size distribution of radioactivity from the stratospheric reservoir occur which must be related to the variable meteorological conditions encountered in transit from stratospheric to ground level, as postulated by Junge (9).

Radiochemical analyses indicate a minimum of fractionation of the radioactivity from the stratospheric source but also document the expected different behavior of fresh debris introduced directly into the troposphere by the Chinese atomic test. A third, long-lived radioactive component of the atmosphere, Pb^{210} from decay of radon gas released from the soil, behaves quite differently from the fission products in that a much smaller proportion of it is associated with particles in the larger size ranges.

An investigation of the applicability of this method to the size distribution of sea-salt particles, sulfate-containing particles, and other common atmospheric aerosols through use of chemical analysis, activation analysis, or other specialized techniques is recommended.

ACKNOWLEDGMENT

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REFERENCES

1. Lockhart, L.B., Jr., and Patterson, R.L., Jr., "Filter Pack Technique for Classifying Radioactive Aerosols by Particle Size. Part 1 – Preliminary Report and Evaluation," NRL Report 5970, Aug. 16, 1963.

2. Lockhart, L.B., Jr., Patterson, R.L., Jr., and Saunders, A.W., Jr., "Filter Pack Technique for Classifying Radioactive Aerosols by Particle Size. Part 2 – Isotopic Fractionation with Particle Size," NRL Report 6164, Oct. 28, 1964

3. Lockhart, L.B., Jr., Patterson, R.L., Jr., and Saunders, A.W., Jr., "Distribution of Airborne Radioactivity with Particle Size," in Proceedings of the 2nd AEC Conference on Radioactive Fallout from Nuclear Weapons Tests, Germantown, Maryland, Nov. 3-6, 1964, AEC document CONF-765 (in press)

4. Shleien, B., Glavin, T.P., and Friend, A.G., "Particle Size Fractionation of Airborne Gamma-Emitting Radionuclides by Graded Filters," Science 147:290-292 (1965)

5. Lockhart, L.B., Jr., Patterson, R.L., Jr., and Anderson, W.L., "Characteristics of Air Filter Media Used for Monitoring Airborne Radioactivity," NRL Report 6054, Mar. 20, 1964

6. Mason, R.M., Ryan, P.C., and Hill, S.L., "Filter Pack Technique for Classifying Radioactive Aerosols by Particle Size. Part 4 – Mathematical Considerations," NRL Report 6306, Aug. 9, 1965

7. Lockhart, L.B., Jr., "Atmospheric Radioactivity at Washington, D.C., 1950-1961," NRL Report 5764, Apr. 11, 1962

8. Lockhart, L.B., Jr., Patterson, R.L., Jr., Saunders, A.W., Jr., and Black, R.W., "Summary Report on Fission Product Radioactivity in the Air Along the 80th Meridian (West), 1957-1962," NRL Report 6104, June 15, 1964

9. Junge, C.E., "Air Chemistry and Radioactivity," New York: Academic Press, pp. 250-253, 1963

APPENDIX

EFFECT OF COUNTING ERRORS ON CALCULATED PARTICLE SIZE DISTRIBUTIONS

Several hypothetical cases covering the range of radioactivity distributions among the four filters in the pack (A terms in the system of simultaneous equations in the body of the report) have been considered in order to determine the effect of measurement errors on the assignment of radioactivity to the various size groups (x terms). The range of errors chosen for evaluation (A_1 and A_3, 1%; A_2, 5%; A_4, 10%) correspond roughly in magnitude to the standard deviations in the radioactivity determinations due to counting errors alone; the larger errors associated with the A_2 and A_4 terms are due to the small increment in collected radioactivity on filters 2 (2nd IPC filter) and 4 (Type 6) over the counter background. In one case three error values were chosen for each A term to discover if the effect was linear. As seen in Table A1, this appeared to be the case for the example chosen.

For each case in Table A1, a series of x terms was derived from the corresponding A series by use of a NAREC computational program.* The increase in any individual A term (errors in the positive direction only were considered) was compensated by prorated decreases among the remaining A terms in the series, so that the four A terms (A_1, A_2 A_3, and A_4) always added up to 100%. The percentage error in x_i, the assigned contribution of group i to the total activity, has been plotted against each x_i in the series of graphs shown in Fig. A1. These curves define the magnitude of the error to be expected in the determination of the contribution of radioactive particles of a given size to the total radioactivity for those collections where the total collected radioactivity is about 100 counts/min. In the NRL equipment used this corresponds to a 3-day filtration of air having a fission product content of about 0.5 pCi/m^3. In many cases, of course, the errors were much smaller than those shown, and their effect can be considered to be roughly proportional to the counting error.

From the standard error distributions for the hypothetical cases shown in Fig. A1, calculated errors in x_1 are seen to be roughly twice the error of measurement in A_1 but are dependent also rather significantly on errors in A_2, particularly as x_1 (and A_1) decrease in size. Errors in x_2 on the other hand can best be minimized by improving the accuracy with which A_2 is determined. The error in the assignment to the third size group (x_3) is about three times the error in measurement of A_3 but the feedback of errors in A_2 is significant. In these three cases, significant improvement in determining x_1, x_2, and x_3 can be accomplished by decreasing the counting error of the second filter in the series. For the final grouping, errors in x_4 are governed primarily by the uncertainties inherent in the measurement of low-activity levels over a rather high background; the only practicable solution here (and it would be helpful also in the case of A_2) would be the use of counters having a significantly lower background.

*Mason, Janet P., "Neliac Functions to Solve Systems of Simultaneous Linear Equations," NAREC Bulletin #41A (NELIAC Bulletin #8).

Table A1
Evaluation of the Effect of Measurement Errors on the Determination of the Size Distribution for Some Hypothetical Cases

Assigned Error	Changes in Size Group (x)				Changes in Assignment to Size Group (%)			
	Δx_1	Δx_2	Δx_3	Δx_4	x_1	x_2	x_3	x_4
Case 1:	$A_1 = 26.30$ $A_2 = 11.50$ $A_3 = 58.50$ $A_4 = 3.70$ $x_1 = 13.05$ $x_2 = 27.31$ $x_3 = 56.47$ $x_4 = 3.17$							
$A_1 + 1\%$	+0.35	-0.21	-0.13	-0.01	+2.68	-0.77	-0.23	-0.3
$A_2 + 5\%$	-1.24	+4.48	-3.37	+0.13	-9.50	+16.41	-5.97	+4.1
$A_3 + 1\%$	-0.12	-1.48	+1.78	-0.18	-0.92	-5.42	+3.15	-5.7
$A_4 + 10\%$	-0.07	+0.04	-0.65	+0.67	-0.54	+0.15	-1.15	+21.1
Case 2:	$A_1 = 35.00$ $A_2 = 12.50$ $A_3 = 50.50$ $A_4 = 2.00$ $x_1 = 20.77$ $x_2 = 37.14$ $x_3 = 41.03$ $x_4 = 1.06$							
$A_1 + 1\%$	+0.51	-0.41	-0.09	-0.02	+2.46	-1.10	-0.22	-1.9
$A_2 + 5\%$	-1.42	+4.85	-3.59	+0.16	-6.84	+13.06	-8.75	+15.1
$A_3 + 1\%$	-0.15	-1.20	+1.46	-0.12	-0.72	-3.23	+3.56	-11.3
$A_4 + 10\%$	-0.03	-0.05	-0.27	+0.36	-0.14	-0.13	-0.66	+34.0
Case 3:	$A_1 = 42.00$ $A_2 = 12.00$ $A_3 = 44.50$ $A_4 = 1.50$ $x_1 = 29.30$ $x_2 = 35.71$ $x_3 = 34.43$ $x_4 = 0.56$							
$A_1 + 1\%$	+0.62	-0.54	-0.07	-0.01	+2.12	-1.51	-0.20	-1.8
$A_2 + 5\%$	-1.41	+4.59	-3.33	+0.15	-4.81	+12.85	-9.67	+26.8
$A_3 + 1\%$	-0.19	-0.95	+1.21	-0.08	-0.65	-2.66	+3.51	-14.3
$A_4 + 10\%$	-0.04	-0.03	-0.21	+0.27	-0.14	-0.08	-0.61	+48.2
Case 4:	$A_1 = 46.00$ $A_2 = 11.40$ $A_3 = 41.50$ $A_4 = 1.10$ $x_1 = 34.75$ $x_2 = 32.29$ $x_3 = 33.00$ $x_4 = -0.04$							
$A_1 + 1\%$	+0.68	-0.60	-0.08	+0.01	+1.96	-1.86	-0.24	+25.
$A_2 + 5\%$	-1.35	+4.35	-3.15	+0.15	-3.89	+13.47	-9.54	+375.
$A_3 + 1\%$	-0.21	-0.79	+1.06	-0.07	-0.60	-2.45	+3.21	-175.
$A_4 + 10\%$	-0.04	+0.02	-0.18	+0.20	-0.12	+0.06	-0.55	+500.
Case 5:	$A_1 = 55.50$ $A_2 = 11.80$ $A_3 = 29.70$ $A_4 = 3.00$ $x_1 = 44.16$ $x_2 = 41.61$ $x_3 = 9.74$ $x_4 = 4.49$							
$A_1 + 1\%$	+0.88	-0.98	+0.18	-0.08	+1.99	-2.35	+1. 85	-1.8
$A_2 + 5\%$	-1.47	+4.46	-3.11	+0.12	-3.33	+10.72	-31.91	+2.7
$A_3 + 1\%$	-0.17	-0.51	+0.74	-0.06	-0.38	-1.23	+7.59	-1.3
$A_4 + 10\%$	-0.13	-0.07	-0.33	+0.53	-0.29	-0.17	-3.39	+11.8
Case 6:	$A_1 = 64.00$ $A_2 = 7.20$ $A_3 = 27.60$ $A_4 = 1.20$ $x_1 = 61.89$ $x_2 = 7.13$ $x_3 = 30.63$ $x_4 = 0.35$							
$A_1 + 0.5\%$	+0.43	-0.36	-0.07	-0.01	+0.69	-5.05	-0.23	-2.9
$A_1 + 1\%$	+0.87	-0.71	-0.13	-0.02	+1.41	-9.96	-0.42	-5.7
$A_1 + 2\%$	+1.74	-1.42	-0.27	-0.04	+2.81	-19.92	-0.88	-11.4
$A_2 + 1\%$	-0.19	+0.53	-0.36	+0.02	-0.31	+7.43	-1.18	+5.7
$A_2 + 5\%$	-0.94	+2.72	-1.88	+0.10	-1.52	+38.15	-6.14	+28.6
$A_2 + 10\%$	-1.86	+5.44	-3.76	+0.18	-3.01	+76.30	-12.28	+51.4
$A_3 + 0.5\%$	-0.13	-0.14	+0.28	-0.01	-0.21	-1.96	+0.91	-2.9
$A_3 + 1\%$	-0.22	-0.35	+0.60	-0.03	-0.36	-4.91	+1.96	-8.6
$A_3 + 2\%$	-0.44	-0.69	+1.19	-0.06	-0.71	-9.68	+3.89	-17.1
$A_4 + 5\%$	-0.05	+0.04	-0.10	+0.11	-0.08	+0.56	-0.33	+31.4
$A_4 + 10\%$	-0.07	+0.02	-0.16	+0.22	-0.11	+0.28	-0.52	+62.9
$A_4 + 20\%$	-0.15	+0.03	-0.32	+0.44	-0.24	+0.42	-1.04	+126.

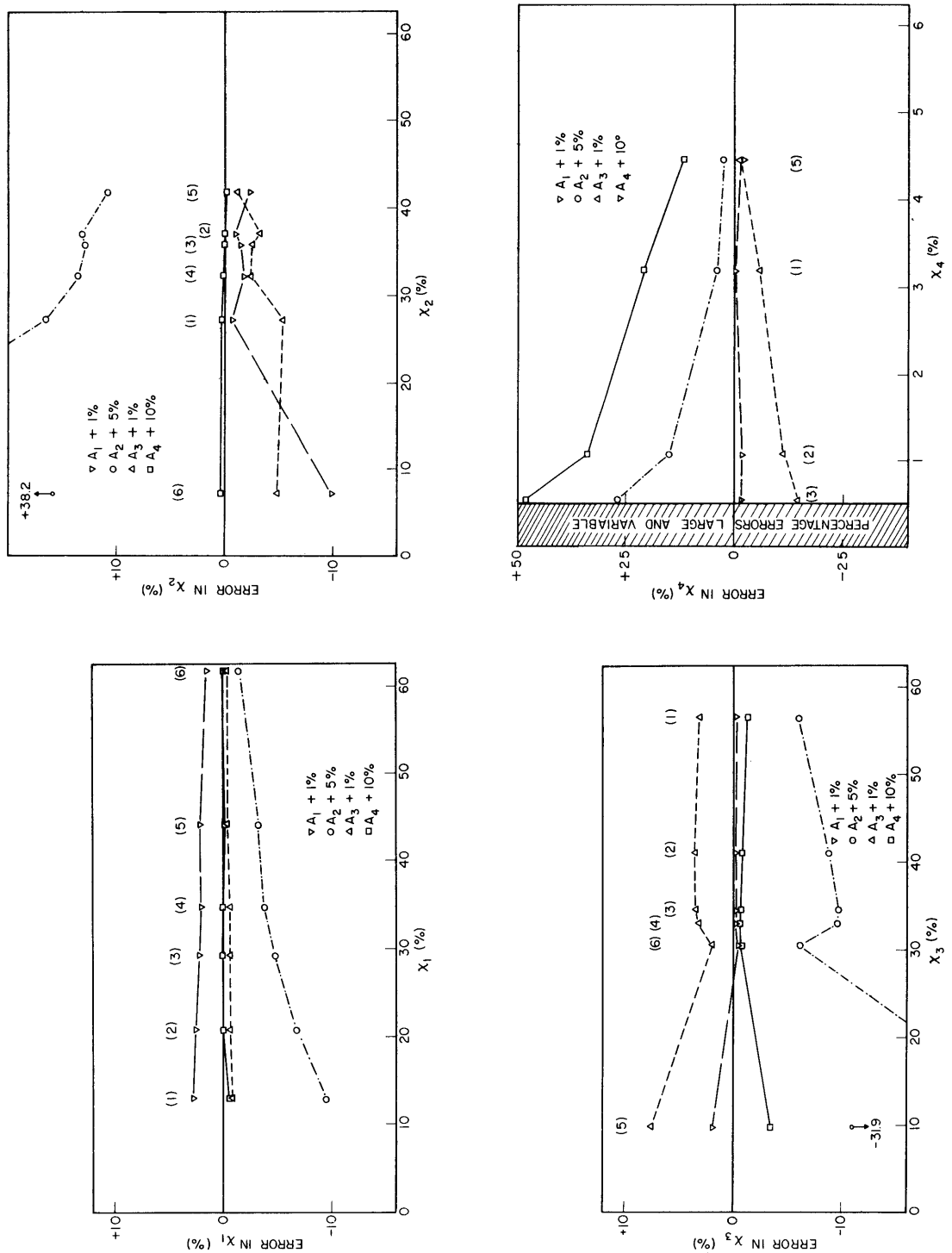

Fig. A1 - Effect of measurement errors on the particle-size group assignments (numbers in parentheses are case numbers from Table A1)

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13. ABSTRACT

Some 60-odd collections of radioactive particulate matter dispersed in the atmosphere have been made during the past two years by use of a filter pack technique utilizing four superimposed filters. The distribution of radioactivity among the filters has been analyzed mathematically by means of four simultaneous linear algebraic equations which relate the particle size of the retained material to the filter characteristics so as to permit the assignment of radioactivity in each collection to four major size groupings: 1.1μ, 0.6μ, 0.3μ, and 0.15μ particle diameter.

The fission-product radioactivity collected at ground level is associated with particles having an "average" diameter between 0.3 and 1.1μ (assuming spherical particles of 1.8 g/cc density). The conclusion is reached that the extremely small radioactive particulates characteristic of nuclear bomb debris which have resided for an extended period in the stratosphere become attached to the larger nonradioactive aerosols typical of troposphere air by the time they arrive at ground level.

A comparison of the relative amounts of various long-lived fission products (Ce^{144}, Pm^{147}, Sr^{90}, Cs^{137}) and of the natural isotope RaD (Pb^{210}) in month-long collections by four-filter packs shows no significant evidence of fractionation effects among the long-lived fission products. This confirms the importance of the attachment process to the size of radioactive fission product conglomerates detected at ground level. Fresh debris from the Chinese nuclear test, however, exhibited a different isotopic distribution with size than did the older bomb debris. Proportionately more of the shorter-lived radioactive fission products collected during transit of debris from the Chinese test appeared in the largest particle size grouping.

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