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RELATIONSHIP BETWEEN THE AIR CONCENTRATION OF RADIOACTIVE FISSION PRODUCTS AND FALLOUT

I. H. Blifford, Jr.

Electron Optics Branch
Optics Division

L. B. Lockhart, Jr., and R. A. Baus

High Polymers Branch
Chemistry Division

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NAVAL RESEARCH LABORATORY
Washington, D.C.

ABSTRACT

Comparative data on the daily concentration of fission products in the air and the actual fallout on the ground have been collected. For short times after a test, fallout was very much dependent on rainfall. In many cases, the air concentration was affected relatively little. On the average, the apparent "rate of fallout" was about 4×10^4 feet per day.

Screens made of cloth or metal mounted on a vane and exposed to the wind were efficient collectors of fission products dispersed in the atmosphere. In some cases, 10 to 100 times as much activity was deposited on a vertical screen as on an equal horizontal area. No definite correlation between gummed-paper, screen, and filter collections has been noted.

Direct interception by the small fibers of vegetation, as distinguished from simple fallout, may account for a large fraction of the total fission product activity adsorbed on such ground cover.

Additional measurements were made on the distribution of activity with particle size by the use of filter media of different retention characteristics.

PROBLEM STATUS

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

AUTHORIZATION

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RELATIONSHIP BETWEEN THE AIR CONCENTRATION OF RADIOACTIVE FISSION PRODUCTS AND FALLOUT

Perhaps the most widely used methods for the evaluation of atmospheric and deposited radioactivity are the air-filter and gummed-paper techniques; both methods have the advantage of simplicity. In general, effects associated with meteorological factors, especially rain, make it difficult to compare one method with the other.

The concentration of radioactivity in the ground-level air at Washington, D. C., was measured with the NRL filter equipment (1). The pumping rate was 30 cubic feet per minute, using Army Chemical Corps Type 5 filter paper. Daily values for the fission product concentration per unit volume of air were obtained from the rate of decay of the filter collections. The gummed-paper collections were made with the standard arrangement used by the Weather Bureau and the AEC. The deposited radioactivity per unit area was found from the counting rate of the ignited residue of the gummed papers. Measurements were made from December 1954 through May 1955.

A comparison of daily concentrations of fission products collected by gummed papers and air filters is made in Fig. 1. The rainfall during the period also is shown. As may be seen from the figure, the ground concentration usually rises with rain or snow. This seems to be true for old fission products, that is, those collected between December 1954 and February 22, 1955, as well as for the younger material collected from the TEAPOT tests.

In order to obtain a more accurate comparison, the gummed-paper residues and the daily filter collections for each week were combined and counted as nearly simultaneously as possible. The results are shown in Table 1. The rates of fallout listed in the table were obtained by assuming a uniform distribution of radioactivity with altitude. The expression "rate of fallout" is used throughout this discussion as a convenient means for comparing the fallout activity per unit area per day with the activity per unit volume of air. Examples of the effect of nonhomogeneity in the atmospheric concentration are shown by the high rates of fallout for 4/11-4/18 and 5/2-5/9. The former collection accounts for 65% of the total fallout for the whole period of the measurements. In these cases, activity was apparently rained out of high-level clouds from relatively recent atomic tests. In the period of the 5/16-5/31 collections, however, rain caused a proportionately larger increase in the air concentration than in the fallout.

It will be observed in Table 1 that the rates of fallout vary from 0.7 to 400×10^3 ft/day. The average over the five-month period was 43×10^3 ft/day. The rates of fallout obtained from a similar tabulation of the daily readings are presented in Table 2 for each month from December 1954 through May 1955. The average rate of fall for days without rain was 2.7×10^3 ft/day and, for rainy days, 30×10^3 ft/day.

The extent to which the average rates of fallout can be relied upon to be a measure of the deposition even in the Washington area is by no means clear. Previous measurements (2) on natural radioactive particulates indicated the mean lifetime of these substances in the lower atmosphere to be about 15 days. The present data seem to show a considerably faster rate of fall. Undoubtedly, the distance from the tests at which measurements are made, the frequency of rainfall, particle size, efficiency of rainfall in washing the atmosphere, and height of rainfall are important factors.

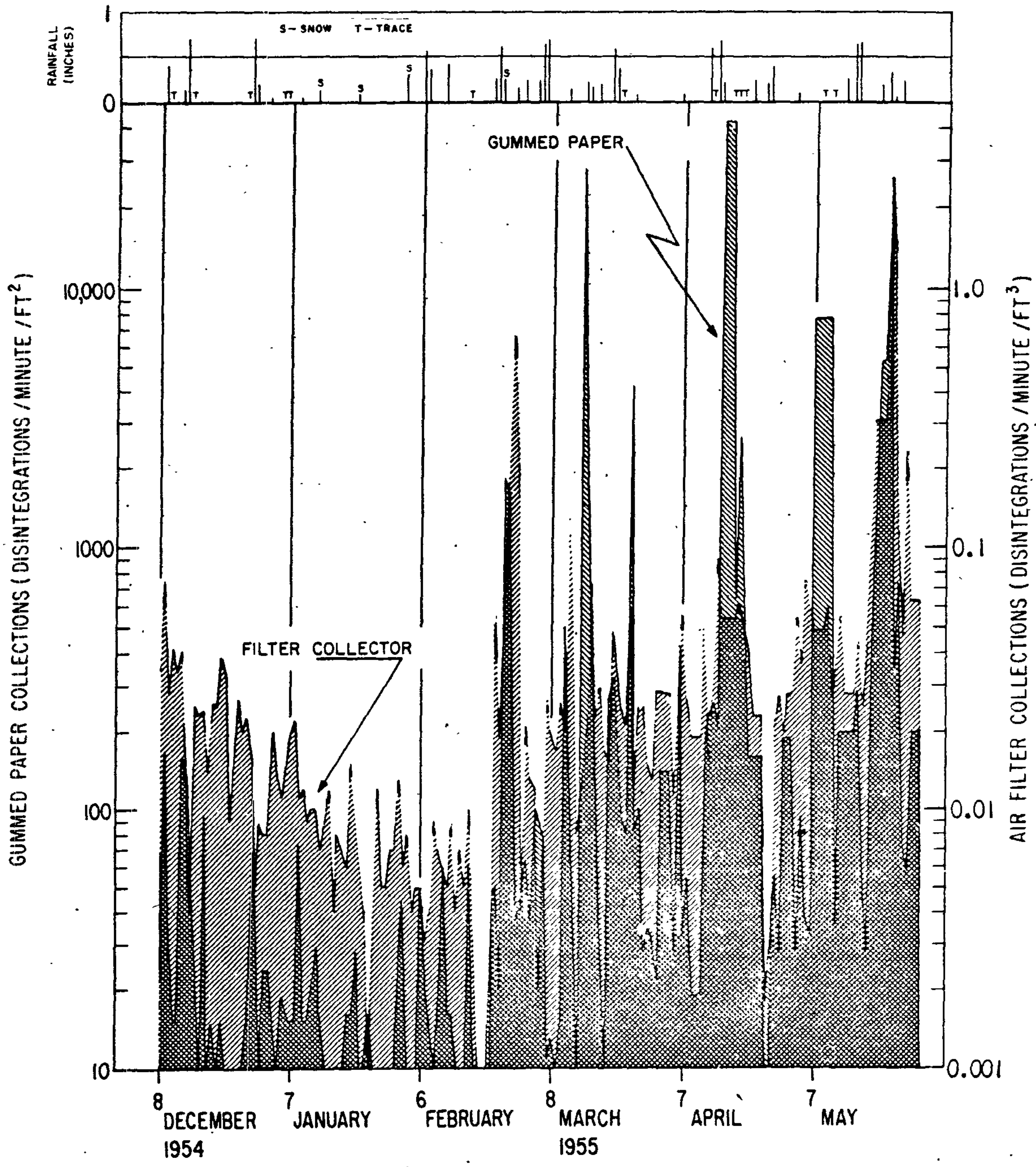

Fig. 1 - Comparison of daily concentrations of fission products collected by gummed paper and air filters

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TABLE 1
Comparison of Collection of Radioactivity by Gummed Paper and Filtration Techniques

Date of Collection	Filter (d/m/ft^3)	Gummed Paper (d/m/ft^2/day)	Rate of Fallout (ft/day)	Rain (in.)
2/21 - 2/28	28×10^{-3}	49	1.8×10^3	1.22
2/28 - 3/7	8.3	29	3.5	1.78
3/7 - 3/14	18	13	0.7	0.13
3/14 - 3/21	19	730	38	1.13
3/21 - 3/28	24	290	12	0.43
3/28 - 4/4	22	24	1.1	0
4/4 - 4/11	27	46	1.7	0.07
4/11 - 4/18	47	19,000	400	1.50
4/18 - 4/25	30	330	11	0.47
4/25 - 5/2	18	100	5.5	0.40
5/2 - 5/9	50	4,400	88	0.01
5/9 - 5/16	31	790	25	1.62
5/16 - 5/23	210	1,400	6.7	0.50
5/23 - 5/31	200	1,600	8.0	1.11
Average			43×10^3	

TABLE 2
Effect of Rain on Apparent Rates of Fallout

Month	No. of Tests	No. of Rains	Rate of Fallout (ft/day)	
			No Rain	Rain
Dec. 1954	0	8	1.0×10^3	1.4×10^3
Jan. 1955	0	7	0.7	1.2
Feb. (before tests)	0	9	1.1	3.7
(after tests)	2	1	9.2	5.2
March	7	12	2.4	50
April	3	15	3.9	133
May	3	18	0.7	12
Average			2.7×10^3	30×10^3

From the available data, it appears that, on the average, and particularly in the absence of well-defined clouds of fission products, the concept of a rate of fallout may be useful in correlating the concentration of activity in the air with the deposition. Obviously, large variations may be encountered in certain instances: i.e., for short distances from the detonation or for short times after a test. The present work indicates the need for more extensive measurements.

In previous experiments, it was found that wires and screens placed at right angles to the wind would collect radioactive atomic bomb debris. In view of the many possibilities which exist for similar collections, both in nature and man-made structures, it was felt that a repetition of some of the earlier work might be valuable. Table 3 gives comparative disintegration rates for weekly collections made by the NRL filter equipment, gummed paper, and an 80-mesh stainless-steel screen. The screen was mounted on a vane so as to face into the wind. The activity was removed from the screens by repeated washing with acetone and nitric acid, after which the washings were evaporated and the residue ignited and counted. Table 3 shows that the total activity collected by the screen was roughly comparable to that of a filter collection of approximately 3×10^6 cubic feet of air and, in some cases, was as much as 10 to 100 times that deposited on an equal horizontal area by fallout. The screen has been found to be of the order of 1% efficient in the absence of rain (on the basis of estimated air flow through it). For periods when there was a marked increase in fallout without a corresponding increase in the air concentration, the screen collections were more nearly equivalent to those of the gummed papers. There are indications that precipitation will wash activity from the screen; this may be the cause of the low screen collection of 4/18-4/25.

TABLE 3
Comparison of One-Week Filter, Gummed-Paper, and Screen Collections at Washington, D. C.

Date of Collection	Filter Collector (d/m)	Gummed Paper (d/m)	Stainless-Steel Screen (d/m)	Rain or Snow (in.)
2/21 - 2/28	8,400	340	3,100	1.22
2/28 - 3/7	2,500	200	2,100	1.78
3/7 - 3/14	4,900	92	3,400	0.13
3/14 - 3/21	5,200	5,100	1,700	1.13
3/21 - 3/28	7,500	2,000	11,000	0.43
3/28 - 4/4	6,700	64	7,700	0
4/4 - 4/11	8,000	320	3,800	0.07
4/11 - 4/18	14,000	130,000	75,000	1.50
4/18 - 4/25	8,900	2,300	1,500	0.47
4/25 - 5/2	2,000	700	1,900	0.40
5/2 - 5/9	9,800	31,000	14,000	0.01
5/9 - 5/16	9,300	5,500	2,600	1.62
5/16 - 5/23	10,000	9,700	4,700	0.50
5/23 - 5/31	110,000	13,000	24,000*	1.11
Total	207,200	200,316	156,000	

*Cloth screen

Ordinary cheesecloth* (about 40 mesh) can be used in place of the metal screens. The cheesecloth can be ignited and counted in the same manner as the gummed paper. Some of the results are shown in Table 4. The cheesecloth seems to be less susceptible to loss of radioactivity through the washing action of rain than does the screen. Flags made of cheesecloth also collect fission activity from the air, but with only about 1/10 the efficiency of the vane-mounted cloth.

Table 5 compares the total activity collected by gummed papers, air filters, and 1-sq ft cloth screens over a two-month period. The measurements included the periods of maximum air and fallout concentrations. The cloth screens collected about 50% more activity than either gummed papers or filters.

*The cheesecloth used in these experiments was found to have an activit of about 100 d/m per sq ft which could be removed by washing, believed to be caused by otassium-40 present in the sizing.

TABLE 4
Comparison of One-Week Screen and Cloth Collections at Washington, D. C.

Date of Collection	80-Mesh Stainless-Steel Screen (d/m)	Cheesecloth Screen (d/m)	Rain (in.)
3/28 - 4/4	7,700	6,300	0
4/4 - 4/11	3,800	6,700	0.07
4/11 - 4/18	75,000	140,000	1.50
4/18 - 4/25	1,500	7,100	0.47
4/25 - 5/2	1,900	3,600	0.40
5/2 - 5/9	14,000	23,000	0.01
5/9 - 5/16	2,600	9,800	1.62
5/16 - 5/23	4,700	59,000	0.50
5/23 - 5/31	---	24,000	1.11

TABLE 5
Comparison of Gummed-Paper and Cloth-Screen Collections at Washington, D. C.
(3/28/55 to 5/31/55)

Total Activity Deposited on Gummed Papers (d/m/ft²)	Total Activity Collected by Cloth Screens (d/m)	Total Activity Collected by Filter Papers (3×10^6 cu ft of air) (d/m)
1.9×10^5	2.8×10^5	1.8×10^6

An attempt was made to relate the mesh size of the screen to the amount of activity collected. Comparative measurements of various one-week collections by electrically grounded screens are shown in Table 6 for four screen sizes. There does not seem to be a significant difference between the collections obtained with any of the screen sizes used in the above experiments. The amount of activity was also not appreciably affected by the screen's being electrically grounded or well-insulated. However, the insulated screen collected considerably more solid matter. The available information indicates that screens collect activity through an impaction process. In the absence of rain, the impaction of particulate matter on small fibers may be the most important means for the removal of fission debris from the air.

Pack filters have been used to effect rough particle size differentiation of fission products dispersed in the air. The filter pack consisted of three separate filter papers: a top paper with a penetration of about 90% at one micron, a middle paper having approximately 50% penetration at this particle diameter, and a bottom paper having essentially 100% efficiency for particles as small as 0.1 micron in diameter.

Table 7 compares data obtained from collections of residual activity before the TEAPOT tests with similar collections during the tests. Both sets of samples were measured at approximately the same time on 30 March, and are not believed to be significantly different. The data are in fair agreement with that obtained during Operation GREENHOUSE.

TABLE 6
Effect of Mesh Size and Electrical Condition on Screen Collections

Screen Mesh Size	Activity (d/m)	Ratio
40	4,500	1.2
80	3,800	
60	1,400	0.9
80	1,500	
200	2,500	1.3
80	1,900	
200	15,000	1.1
80	14,000	
200	2,100	0.81
80	2,600	
200	101,000	1.3
80	75,000	
80 (insulated)	9,400	1.2
80 (grounded)	7,600	
80 (grounded)*	4,700	1.0
80 (grounded)*	4,700	

*Duplicate collections made to check reproducibility

TABLE 7
Comparison of Pack-Filter Collections Made Prior to and During Operation TEAPOT

Date of Collection	Percent of Total Activity		
	Top Filter	Middle Filter	Bottom Filter
Residual Activity Prior to TEAPOT			
1/31 - 2/7/55	41	18	41
2/7 - 2/14	57	24	19
2/14 - 2/21	54	25	21
Average	51	22	27
Activity from TEAPOT			
2/21 - 2/28	53	17	30
2/28 - 3/7	47	21	32
3/7 - 3/14	50	25	25
Average	50	21	29

Table 8 gives the results of additional pack-filter measurements made during Operation TEAPOT. In this case, each sample was measured on the day after collection. Here the average relative activity on the top papers appeared to be appreciably greater than was found from measurements made after the samples had decayed, as shown in Table 7. However, the change in relative activity with decay varied widely in individual samples. After five weeks, the activity of the top paper of the collection of 3/28 to 4/4 increased from 72% to 76% of the total, while that of the top paper of the 4/11 to 4/18 collection decreased from 85% to 66% in 3 weeks. Similar variations in the decay rates of the different fractions had been found previously during Operation GREENHOUSE.

TABLE 8
Pack-Filter Collections Made During Operation TEAPOT

Date of Collection	Percent of Total Activity		
	Top Filter	Middle Filter	Bottom Filter
3/28 - 4/4/55	72	11	17
4/4 - 4/11	60	12	28
4/11 - 4/18	85	7	8
4/18 - 4/25	58	14	28
4/25 - 5/2	57	16	27
5/2 - 5/9	70	11	19
5/9 - 5/16	70	12	18
Average	68	12	20

It seems reasonable to expect that a greater proportion of the activity of old fission products in the air will be in the smaller size ranges, but, from the present data, the evidence for this effect is not conclusive. It appears that the actual number of particles in the small size range is comparatively large for both old and new fission debris, since considerably fewer large particles are required for a given activity. The variability in decay rates may result from a nonhomogeneity which is common to both large and small particles.

CONCLUSIONS

There appears to be little basis for the correlation of individual daily measurements of the air concentration of atomic bomb debris and fallout of this material on the ground. Over extended periods, however, it may be possible to determine experimentally a rate of fallout which, when applied to the figures for the air concentration, will give a reasonable approximation to the deposition. At Washington, D. C., the fallout in a single 3-day period from Operation TEAPOT produced approximately 65% of the total deposit over a two-month period. Similar large variations can be anticipated until the clouds of fission activity become well dispersed, either through time or distance.

In almost every case, greatly increased fallout accompanied rain. However, these instances did not always lead to a corresponding increase in the air concentration. This probably is due to high-level entrainment of activity in the rain droplets. The above phenomena indicate the need for additional experimental studies on the efficiency of rainfall for washing out the fission products.

Direct impaction by small fibers is a significant factor in the deposition of radioactive atomic bomb particulates. Wire and cloth meshes oriented to face into the wind collected, in some instances, as much as 10 to 100 times the activity deposited on the same horizontal area by fallout. The screen and fallout collections were approximately equal during periods of heavy fallout with rain. It seems reasonable to postulate similar impaction phenomena for vegetation or any other loosely compacted filamentary substance. Except for instances when activity is rained out of upper-level strata of fairly limited extent, the available data indicate that impaction accounts for a large part of the total deposition. Variations in screen mesh sizes from 40 to 200 and in the electrical charge had relatively small effects on the amount of activity collected.

Additional work carried out over a longer period of time will be required in order to define clearly the relationship between the various deposition processes. It is possible, however, that the cloth screen may provide a simple means of collection, since it combines some of the advantages of both the air-filter and gummed-paper methods.

Pack-filter measurements for the differentiation of old and new fission debris have not, by themselves, given entirely conclusive results. Apparent differences were found in the particle size distributions similar to those observed during Operation GREENHOUSE. There is some evidence for increased activity in the large size fraction at short times after a test. It is also possible that the observed effects may be due to fundamental problems associated with the method of measurement.

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