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# Filter Pack Technique for Classifying Radioactive Aerosols by Particle Size

## Part 2 - Isotopic Fractionation With Particle Size

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## ABSTRACT

An evaluation of the relative distribution of some long-lived airborne fission products among several ranges of particle sizes has been carried out by the radiochemical analysis of individual air filters exposed in a pack. The relative quantities of radioactivity deposited on each of the filters in the packs has been found to vary considerably throughout 1963; however, each of the individual isotopes behaves in much the same manner so that the fractionation of isotopes relative to one another is slight.

Though isotopic fractionation by filters of different characteristics has not been found to be of great significance, there are indications that a more sensitive separating procedure may show more differences between particles in the extreme size ranges. The observed differences have been small and variable, rather than organized, but the variations are too large to be accounted for by experimental error. The long-lived natural radioisotope $Pb^{210}$ has a distribution very similar to that of the fission products, but with some preferential concentration in the smaller size range. The differences are not sufficient to invalidate the use of collections made on less than perfect filters for estimating the relative fission product concentrations in the air or for the approximate age determination of bomb debris.


## 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 2 - ISOTOPIC FRACTIONATION WITH PARTICLE SIZE

### INTRODUCTION

All nuclear explosions generate a wide spectrum of radioactive particles of various sizes and isotopic compositions. The distribution of radioactivity with particle size is extremely complex, depending on the type of detonation, the energy released, and the various materials in the immediate environment which are enveloped by or drawn into the fireball. Surface bursts, or tower bursts where the fireball intersects the earth's surface, generally produce distribution curves skewed in the direction of larger particles. Air bursts, on the other hand, lead to a greater number of the smaller sized particles.

Fractionation of the various isotopes relative to one another can occur during the cooling and condensation phases immediately following a nuclear detonation. During this period it might be expected that the least volatile materials would solidify first and serve as nuclei for later condensation of the more volatile materials or those preceded in the decay chain by volatile precursors. In tower or surface shots where much extraneous material is available for incorporation into the fireball, a considerably greater number and variety of condensation nuclei exist. Moreover, complete melting or volatilization of the incorporated materials does not necessarily occur, so that these nuclei are on the average much larger and are deposited as "fallout" much more rapidly, preferentially carrying with them the most refractory fission products.

In the case of those fission products which have rather long-lived gaseous precursors in the decay chain, the cooling and condensation phases of the fireball may be essentially complete prior to formation of the isotope of interest. Thus such isotopes as $Sr^{89}$, $Sr^{90}$, and $Cs^{137}$, having as precursors in the fission chain 3.2-min $Kr^{89}$, 33-sec $Kr^{90}$, and 3.9-min $Xe^{137}$ respectively, would be expected to accumulate in the outer layers of particles. It is also possible that physical separation of the volatile isotopes from the nonvolatile ones can occur to some extent and that those elements condensing later could be dispersed in a molecular form.

Following a nuclear detonation there is a progressive decrease in the average size of the particles because of the higher settling rate and faster deposition of the large particles. Consequently, radioactive debris which have remained in the stratosphere for any extended period of time must initially have been extremely small; they perhaps eventually become incorporated into the small natural stratospheric aerosols described by Junge and Manson (1). Further growth would be expected to take place through the agglomeration of the radioactive particles with the normally large quantity of ordinary dust particles in the atmosphere, particularly at the lower altitudes. Eventually however, a rather stable size distribution must be reached among the larger particles as the result of a balance between particle growth by coagulation and particle deposition. Their size distribution on arrival at ground level must be related to the period of time they have been undergoing the normal attachment processes to the larger aerosols in the troposphere.

Conditions do exist, therefore, for a variation of the isotopic composition of bomb debris with particle size and for particle size variations with time. As reported in previous work on the distribution of radioactivity with particle size (2), there are observable

quite distinct changes in the particle size distribution with time following a nuclear test series. This report is concerned primarily with the changes occurring in the relative isotopic constitution of several size fractions as a function of time, and whether there exists a relation between the size distribution of radioactive particles and tropospheric and stratospheric residence times, atmospheric dust loadings, and weather.

## EXPERIMENTAL

Collections of fission product radioactivity on packs of 3 or 4 filters each 4 inches in diameter (one or two pieces of IPC 1478 paper superimposed on a Type 5G filter and backed up by a final Type 6 filter) were made routinely through 1963 whenever equipment was not being used for other purposes. These collections were assayed for gross $\beta$ activity several weeks after collection to document the changing radioactivity levels with time and to determine the change in approximate size distribution with time.

Larger collections of radioactive particles, suitable for radiochemical analysis to determine the relative isotopic distribution with particle size, were made on packs of three superimposed filters each 13 × 17 inches (initial filter, IPC 1478; middle filter, Type 5G; final filter, Type 6). The characteristics of these filters are described elsewhere (2,3). Sampling was generally over a one-week period at the rate of about 152 cubic feet per minute ($4.34 \times 10^4$ $m^3$ per week) at an average face velocity of 120 linear feet per minute (61 cm/sec). Little change in flow with time occurred even for collections of 3 week's duration.

These filters with their somewhat selective ranges of fission product sizes were separated from each other and stored in envelopes. Those selected for analysis were wet ashed in the presence of carrier elements and analyzed radiochemically for $Sr^{89}$, $Sr^{90}(Y^{90})$, $Y^{91}$, $Cs^{137}$, $Ce^{141}$, $Ce^{144}(Pr^{144})$, $Pm^{147}$, and $Pb^{210}$ by standardized procedures (4). In a few cases Sb carrier was added and $Sb^{125}$ identified as a component of the fission product mixture. None of the 33-day $Ce^{141}$ was detected in the samples at the time of analysis. In so far as possible, the radioactivity of each sample was determined on several different, but intercalibrated, counting units; moreover, similar isotopes from the different filters of a given pack were counted consecutively on the same counter.

## GROSS FISSION PRODUCT $\beta$ ACTIVITY IN THE AIR

The collection periods of the samples selected for radiochemical analysis and their relation to the gross $\beta$ activity levels existing in the atmosphere during 1963 are shown in Fig. 1. This gross $\beta$ activity data has been obtained from other collections made at NRL for various purposes–filter efficiency determinations, intercalibration of air monitoring procedures, and various filter pack combinations. The monthly average radioactivity concentrations reported by the U.S. Public Health Service Radiation Surveillance Network for Washington, D.C. are also shown. The wide scatter in the values of the short term measurements demonstrates the extreme dependence of the ground level activity concentration on meteorological processes both those bringing upper tropospheric air down to ground level and those cleansing the air of particulate matter.

A strong spring maximum in the concentrations of airborne fission products resulting from the winter-spring subsidence of stratospheric air was observed at Washington, D.C., during 1963 as in previous years, The values reported here cannot be quantitatively related to those of prior years due to changes in the method of collection and analysis beginning in January 1963; however, the activity levels are similar to those observed in 1959 and 1962.

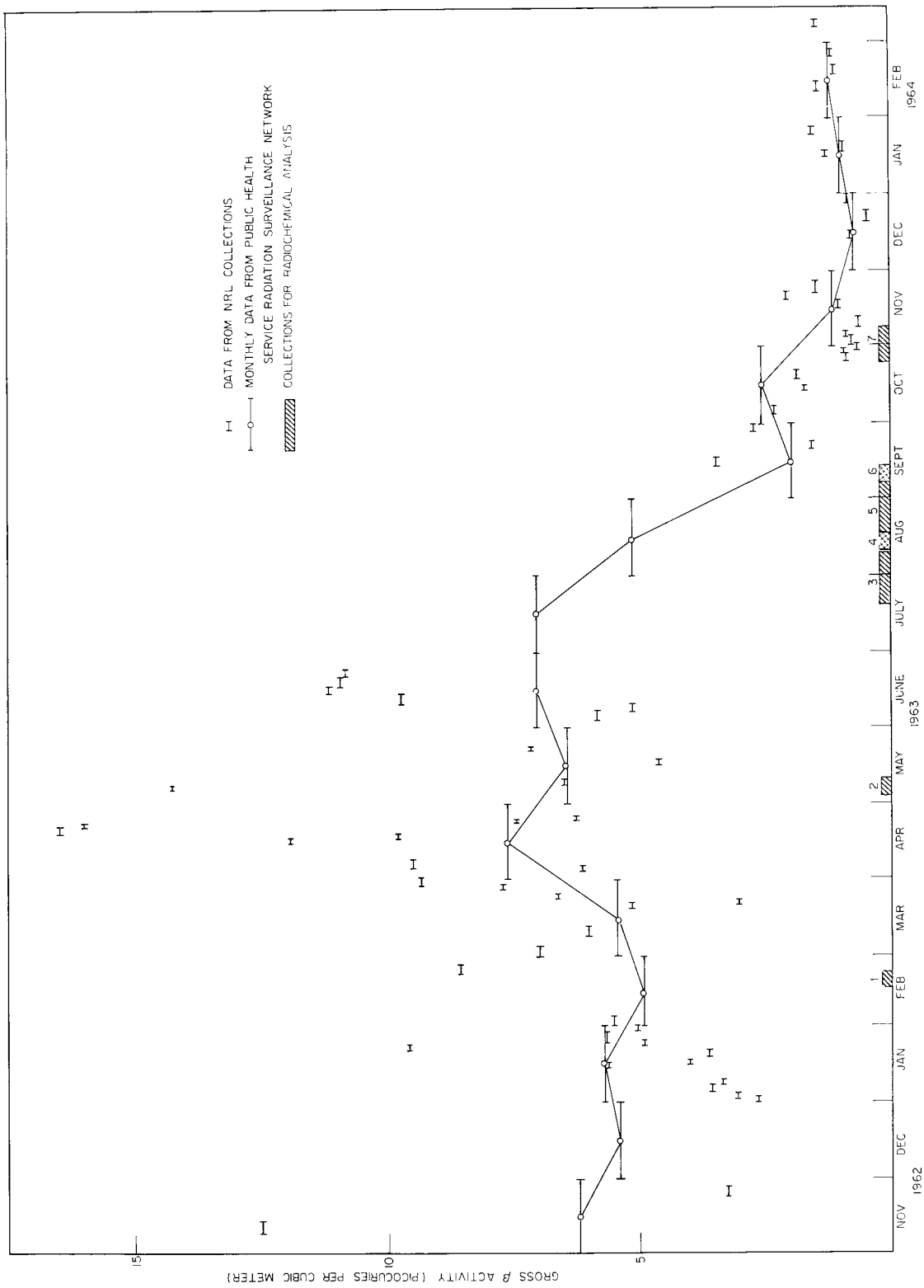


Fig. 1 - Concentration of fission product β activity in the ground level air at Washington, D.C., during 1963

## CHANGES IN PARTICLE SIZE DISTRIBUTION WITH TIME

The fraction of the total radioactive particulate matter retained by the initial (poorest) filter in the pack, shown in Fig. 2 as a function of time, is indicative of the relative amount of radioactive fission products associated with particles having diameters in the $1\mu$ range. If the dispersed material is considered as a homogeneous aerosol consisting of spherical particles with a density of 1.8, the extreme range of retentivity values found between August 1963 and March 1964 (26% to 51% of the incident radioactivity) represents an overall change in the effective particle size from about $0.85\mu$ to $0.55\mu$, based on the measured retentivity of IPC 1478 filter paper toward $H_2SO_4$ aerosols at a face velocity of 280 linear feet per minute (2).

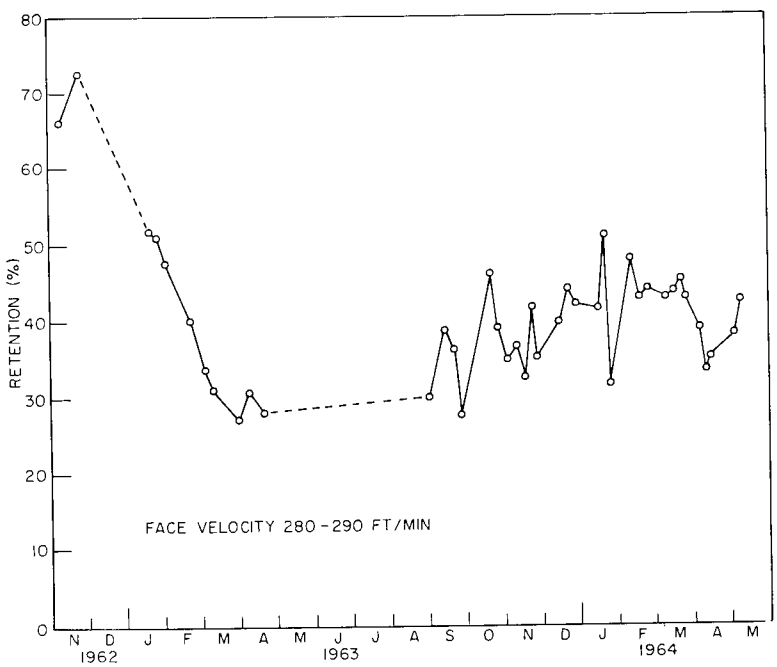


Fig. 2 - Temporal changes in the retention of gross fission product radioactivity by IPC 1478 filters

Other pack collections of gross $\beta$ activity made at a face velocity of 140 feet per minute as well as the large filter collections made at about 120 feet per minute, when compared to the IPC 1478 filter calibrations with $H_2SO_4$ aerosol, also indicate variations in the effective particle size to be in the above range.

The rapid decrease in the larger sized radioactive particles following the ending of nuclear testing in December 1962 appears to be directly related to deposition of the tropospheric component of the radioactive debris. The short term fluctuations observed during the fall and winter of 1963-1964 must, however, be the result of attachment of radioactive aerosols from the stratosphere to normal nonradioactive tropospheric aerosols. This is the season in the Washington area when the output of particles from oil and gas-fired furnaces is high, but variable; the average size of the particles containing radioactivity should thus be a complex function of the quantity of inactive material in the air and the

residence time of the radioactive aerosols in the troposphere. One might thus expect the fraction of activity attached to larger particles to vary directly with the fuel consumption (or "heating degree" days) or inversely with the quantity of activity in the air; no such simple relationships were apparent, however. The discussion of the radiochemical results in the following section indicates equally large variations occurred during the summer months at a time when no filter pack collections for gross $\beta$ measurements were made.

## ISOTOPIC FRACTIONATION WITH PARTICLE SIZE

The results of the radiochemical analyses of filter pack collections made at Washington, D.C., during 1963 are presented in Table 1. The relationship of these samples to the seasonal changes in gross $\beta$ activity in the area and to other measurements of the temporal changes in radioactivity with particle size can be seen by comparison of the collection periods with information shown in Figs. 1 and 2. Short term variations in the radioactivity distribution with particle size predominate over possible seasonal effects, which is probably a direct, but complex, function of local weather conditions. The greatest variation in particle size distribution observed among the seven samples occurred during consecutive collection periods in August 1963.

The particle size ranges represented by the three filters of each pack are roughly as follows: filter 1 (IPC 1478) contains the bulk of the activity associated with the larger particles, that is, those near $1\mu$ in diameter, but also contains some contribution from smaller sized particles; filter 2 contains particles in the range $0.3\mu$ to $1.0\mu$; and filter 3 contains the remainder, primarily in the size range below $0.3\mu$. A series of curves showing the percentage distribution of particles among the three filters as a function of particle size is shown in Fig. 3; this is based partly on calibration data obtained with $H_2SO_4$ aerosol and partly from inferences based on the measured retention of natural radioactive aerosols (3).

Figure 3 indicates that over a wide range of particle sizes roughly 75% of the gross activity would appear on the middle filter of the pack; consequently, isotope ratios measured here should reflect the average of the collection rather than specific fractionation effects. The most likely evidence of isotopic fractionation would show up in activity ratio differences between particles deposited on the initial and the final filters where the largest and smallest particles would be concentrated. The ratio of activity of each isotope relative to $Sr^{90}$ as a reference on the initial filter to that on the final filter ($X/Sr^{90}$) for the IPC 1478 filter vs ($X/Sr^{90}$) for the Type 6 filter, is listed in Table 2 for the seven collections. It should be noted that any inadequacies in counter standardization are cancelled out by this method of presentation, so that the results should be as reliable as counter stability and counting statistics ($\sigma = \pm 1\%$) allow.

Though a comparison of the overall distribution of the various radioisotopes among the filters of a given collection discloses a surprisingly small degree of fractionation with particle size (Table 1), there are some definite trends apparent from Table 2. It is noted that $Sr^{89}$ and $Pb^{210}$ are consistently depleted in the largest size particles relative to the smallest; $Cs^{137}$, $Ce^{144}$, and $Pm^{147}$ are quite variable in distribution; and $Y^{91}$ appears to be fractionated least. One might expect some similarity in behavior between $Sr^{89}$ and $Cs^{137}$ relative to $Sr^{90}$, since these have longer-lived precursors in the decay chain which should cause them to be associated with smaller sized particles. The $Ce^{144}$, $Pm^{147}$, and $Y^{91}$, with short-lived precursors, should be associated with larger particles. No such consistency in behavior was found, however. The $Sr^{89}$ results are the least accurate of those reported, however, due to the low activity of $Sr^{89}$ at the time of measurement and the necessity for obtaining its value by difference from two larger quantities, the total $Sr^{89,90}$ activity and the $Sr^{90}$ activity obtained form $Y^{90}$ determinations. The concentration of $Pb^{210}$ in the smaller particles is expected on the basis of its shorter average lifetime in the troposphere, and its continual reintroduction through decay of radon, primarily in the lowest levels of the atmosphere.

Table 1
Results of Radiochemical Analyses of Filter Pack Collections Made at NRL

| Collection Date (1963) | Sample Volume ($m^3$) | Filter | Order | Activity Distribution on Filters (%) | | | | | | | Selected Activity Ratios | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | $Sr^{89}$ | $Sr^{90}$ | $Y^{91}$ | $Cs^{137}$ | $Ce^{144}$ | $Pm^{147}$ | $Pb^{210}$ | $Sr^{89}/Sr^{90}$ | $Sr^{89}/Y^{91}$ | $Ce^{144}/Sr^{90}$ | $Cs^{137}/Sr^{90}$ |
| Feb. 15-21 | $3.72 \cdot 10^4$ | IPC 1478 | 1 | 16.3 | 18.7 | 21.3 | 14.3 | 29.4 | 22.0 | 10.9 | 22.1 | 0.70 | 37.7 | 1.10 |
| | | Type 5G | 2 | 75.0 | 74.2 | 71.9 | 77.3 | 64.6 | 71.2 | 74.4 | 25.7 | 0.95 | 20.9 | 1.50 |
| | | Type 6 | 3 | 8.7 | 7.1 | 6.8 | 8.4 | 6.0 | 6.8 | 14.7 | 31.2 | 1.16 | 20.5 | 1.71 |
| | | | | | | | | | | | 25.4 | 0.91 | 24.0 | 1.44 |
| | Total Activity (dis/min)* | | | $9.28 \cdot 10^4$ | $3.65 \cdot 10^3$ | $1.02 \cdot 10^5$ | $5.25 \cdot 10^3$ | $8.78 \cdot 10^4$ | $1.49 \cdot 10^4$ | $8.17 \cdot 10^2$ | | | | |
| May 3-10 | $4.34 \cdot 10^4$ | IPC 1478 | 1 | 13.1 | 13.3 | 13.9 | 10.3 | 17.3 | 17.0 | 14.8 | 7.8 | 0.68 | 25.8 | 1.11 |
| | | Type 5G | 2 | 74.2 | 75.2 | 74.1 | 80.7 | 70.5 | 72.2 | 67.2 | 7.9 | 0.73 | 18.7 | 1.55 |
| | | Type 6 | 3 | 12.7 | 11.5 | 12.0 | 9.0 | 12.2 | 10.8 | 18.0 | 8.9 | 0.77 | 21.3 | 1.14 |
| | | | | | | | | | | | 8.0 | 0.73 | 20.0 | 1.45 |
| | Total Activity (dis/min)* | | | $8.63 \cdot 10^4$ | $1.08 \cdot 10^4$ | $1.19 \cdot 10^5$ | $1.56 \cdot 10^4$ | $2.16 \cdot 10^5$ | $3.27 \cdot 10^4$ | $1.10 \cdot 10^3$ | | | | |
| July 19-Aug. 9 | $1.30 \cdot 10^5$ | IPC 1478 | 1 | 23.2 | 23.9 | 23.6 | 20.7 | 22.0 | 22.0 | 24.7 | 3.4 | 0.74 | 19.6 | 1.25 |
| | | Type 5G | 2 | 64.1 | 63.7 | 64.8 | 69.2 | 66.8 | 67.7 | 62.2 | 3.5 | 0.75 | 22.3 | 1.57 |
| | | Type 6 | 3 | 12.7 | 12.4 | 11.6 | 10.1 | 11.2 | 10.3 | 13.1 | 3.6 | 0.83 | 19.2 | 1.18 |
| | | | | | | | | | | | 3.5 | 0.75 | 21.3 | 1.45 |
| | Total Activity (dis/min)* | | | $7.42 \cdot 10^4$ | $2.13 \cdot 10^4$ | $9.85 \cdot 10^4$ | $3.08 \cdot 10^4$ | $4.52 \cdot 10^5$ | $1.09 \cdot 10^5$ | $7.93 \cdot 10^2$ | | | | |
| Aug. 10-17 | $4.34 \cdot 10^4$ | IPC 1478 | 1 | 10.2 | 14.0 | 12.4 | 15.1 | 16.5 | 11.6 | 10.9 | 2.4 | 0.61 | 19.3 | 1.31 |
| | | Type 5G | 2 | 73.5 | 72.4 | 75.3 | 77.2 | 69.7 | 75.8 | 69.0 | 3.4 | 0.72 | 15.8 | 1.30 |
| | | Type 6 | 3 | 16.3 | 13.6 | 12.3 | 7.7 | 13.8 | 12.6 | 20.1 | 4.1 | 0.98 | 16.8 | 0.69 |
| | | | | | | | | | | | 3.4 | 0.74 | 16.4 | 1.22 |
| | Total Activity (dis/min)* | | | $2.90 \cdot 10^4$ | $8.62 \cdot 10^3$ | $3.92 \cdot 10^4$ | $1.05 \cdot 10^4$ | $1.42 \cdot 10^5$ | $3.26 \cdot 10^4$ | $7.07 \cdot 10^2$ | | | | |
| Aug. 17-Sept. 6 | $1.24 \cdot 10^5$ | IPC 1478 | 1 | 21.6 | 21.6 | 21.9 | 34.0 | 23.8 | 23.6 | 18.7 | 2.2 | 0.64 | 18.4 | 1.22 |
| | | Type 5G | 2 | 68.2 | 69.6 | 68.6 | 58.4 | 59.4 | 64.7 | 65.9 | 2.1 | 0.65 | 14.3 | 0.65 |
| | | Type 6 | 3 | 10.2 | 8.8 | 9.5 | 7.6 | 16.8 | 11.7 | 15.4 | 2.5 | 0.70 | 31.9 | 0.67 |
| | | | | | | | | | | | 2.2 | 0.65 | 16.7 | 0.77 |
| | Total Activity (dis/min)* | | | $3.07 \cdot 10^4$ | $1.41 \cdot 10^4$ | $4.71 \cdot 10^4$ | $1.09 \cdot 10^4$ | $2.36 \cdot 10^5$ | $4.95 \cdot 10^4$ | $1.97 \cdot 10^3$ | | | | |
| Sept. 6-13 | $4.34 \cdot 10^4$ | IPC 1478 | 1 | 14.3 | 17.5 | 15.3 | 16.7 | 16.3 | 15.2 | 14.2 | 2.0 | 0.75 | 17.2 | 1.02 |
| | | Type 5G | 2 | 76.5 | 73.4 | 75.4 | 72.5 | 74.9 | 75.7 | 67.7 | 2.6 | 0.81 | 18.8 | 1.06 |
| | | Type 6 | 3 | 9.2 | 9.1 | 9.3 | 10.8 | 8.8 | 9.1 | 18.1 | 2.5 | 0.79 | 17.6 | 1.26 |
| | | | | | | | | | | | 2.5 | 0.80 | 18.4 | 1.07 |
| | Total Activity (dis/min)* | | | $9.23 \cdot 10^3$ | $3.76 \cdot 10^3$ | $1.16 \cdot 10^4$ | $4.03 \cdot 10^3$ | $6.92 \cdot 10^4$ | $1.68 \cdot 10^4$ | $1.27 \cdot 10^2$ | | | | |
| Oct. 24-Nov. 8 | $9.30 \cdot 10^4$ | IPC 1478 | 1 | 14.0 | 24.1 | 23.4 | 22.0 | 23.7 | 23.0 | 16.5 | 0.8 | 0.52 | 15.4 | 1.49 |
| | | Type 5G | 2 | 77.1 | 68.2 | 70.0 | 70.7 | 68.8 | 69.2 | 61.1 | 1.6 | 0.95 | 15.8 | 1.69 |
| | | Type 6 | 3 | 8.9 | 7.7 | 6.6 | 7.3 | 7.5 | 7.8 | 22.4 | 1.7 | 1.16 | 15.2 | 1.55 |
| | | | | | | | | | | | 1.5 | 0.86 | 15.7 | 1.63 |
| | Total Activity (dis/min)* | | | $6.16 \cdot 10^3$ | $4.23 \cdot 10^3$ | $7.14 \cdot 10^3$ | $6.90 \cdot 10^3$ | $6.62 \cdot 10^4$ | $2.00 \cdot 10^4$ | $1.37 \cdot 10^3$ | | | | |

*Activity corrected for decay to midpoint of collection period.

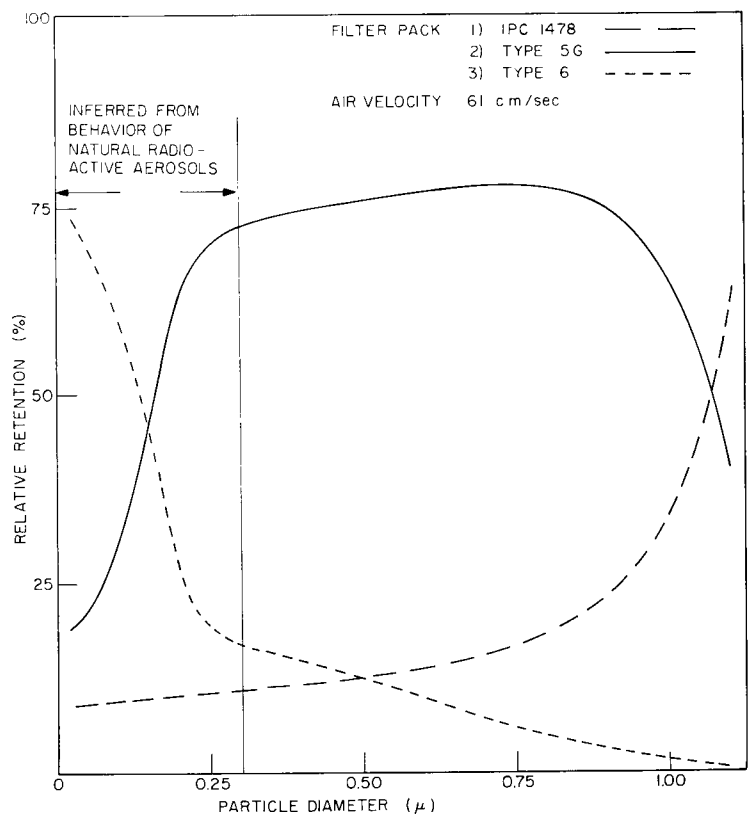


Fig. 3 - Effect of size on the distribution of particles among filters in a pack

A comparison of the percentage retention of radioactive materials by the initial and final filters (Table 1) with the filter calibration data (Fig. 3) indicates, qualitatively, that the effective particle size distribution is rather narrow, falling between about 0.5$\mu$ and 0.9$\mu$ diameter. This would lead to a rather incomplete separation of particle size groupings by the filter pack employed with a consequent lessening of sensitivity to fractionation effects. On the positive side, however, it would indicate that absolute filters are not essential for air monitoring purposes at ground level, even if the collections are to be used for isotope ratio determinations. Filters operated under conditions giving a retentivity of 90% or better for 0.3$\mu$ DOP (dioctyl phthalate) aerosols should retain nearly 100% of incident fission debris with an unmeasurable degree of fractionation. Many of the filter media being employed at the present time fall in this category (3).

The $Cs^{137}/Sr^{90}$ ratios show peculiarities observed before for collections made during 1961 and 1962, namely, an apparent depletion of $Cs^{137}$ in some collections (5,6). This depletion is particularly notable in two collections made in August and September 1963 which give indications of abnormally low concentrations of $Cs^{137}$ in the smaller particles. There is apparently no relationship between the residence time of the radioactive particles in the troposphere, indicated by the percentage of larger sized particles, and the degree of fractionation of $Cs^{137}$ relative to $Sr^{90}$. The Local Climatological Data for the National Airport, Washington, D.C., issued monthly by the Weather Bureau, U.S. Department of Commerce, disclosed no unusual weather during this period except for heavy rain on August 19-20; perhaps a more detailed examination into the past history of the air masses would uncover some reason for the observed fractionation.

Table 2
Comparison of Activity Ratios Relative to $Sr^{90}$ for the Largest and Smallest Particle Size Fractions

| Collection | Ratio of Activity Ratios to $Sr^{90}$ for Initial vs Final Filter | | | | | |
|---|---|---|---|---|---|---|
| | $Sr^{89}$ | $Y^{91}$ | $Cs^{137}$ | $Ce^{144}$ | $Pm^{147}$ | $Pb^{210}$ |
| Feb. 15-21 | 0.71 | 1.19 | 0.65 | 1.85 | 1.23 | 0.28 |
| May 3-10 | 0.89 | 1.01 | 0.99 | 1.23 | 1.36 | 0.71 |
| July 19-Aug. 9 | 0.95 | 1.05 | 1.06 | 1.02 | 1.11 | 0.97 |
| Aug. 10-17 | 0.61 | 0.99 | 1.89 | 1.17 | 0.89 | 0.53 |
| Aug. 17-Sept. 6 | 0.86 | 0.94 | 1.83 | 0.58 | 0.82 | 0.49 |
| Sept. 6-13 | 0.81 | 0.85 | 0.80 | 0.96 | 0.87 | 0.41 |
| Oct. 24-Nov. 8 | 0.50 | 1.13 | 0.96 | 1.01 | 0.94 | 0.23 |
| Average | 0.76 | 1.02 | 1.17 | 1.12 | 1.03 | 0.52 |
| Standard deviation of the mean | ±0.06 | ±0.04 | ±0.19 | ±0.15 | ±0.08 | ±0.10 |
| Standard deviation of a single ratio | ±0.16 | ±0.11 | ±0.50 | ±0.39 | ±0.21 | ±0.25 |

In past work where low $Cs^{137}/Sr^{90}$ activity ratios were observed (primarily in collections made at Miami, Florida), the possible loss of $Cs^{137}$ during sample ashing at 650°C was considered. In the analyses reported here, however, wet ashing was employed with the same result. Meanwhile, other experiments have demonstrated to our satisfaction that no $Cs^{137}$ activity is lost from asbestos ash at 1000°C or at 650°C after addition to the ash of 1 percent by weight of sodium chloride to permit formation of the more volatile CsCl.

## OTHER ACTIVITIES

Antimony activity was separated from the middle filter (Type 5G) of several filter collections made in 1963 to determine if any nonfission product $Sb^{124}$ activity could be isolated. Though quite large amounts of the fission product $Sb^{125}$ (2.7 yr half-life) were formed, neither $\beta$ spectral analysis through absorption measurements at NRL nor $\gamma$ analysis at the AEC Health and Safety Laboratory (7) indicated the presence of any $Sb^{124}$ (60.0 day half-life) at the time of measurement.

The $Sb^{125}/Sr^{90}$ activity ratios on the Type 5G (middle) filters were 1.59, 1.55, and 1.52 for the collections of Aug. 10-17, Sept. 6-13, and Oct. 24-Nov. 8, 1963, respectively. This essentially constant ratio is consistent with the formation of $Sb^{125}$ by the fission process; however, the ratio is about 50% higher than expected, possibly due to inadequacies of the indirect method of calibrating the $\beta$ counters toward the complex spectrum of $\beta$ particles and conversion electrons emitted by this isotope.

The yttrium fraction, which contains all rare earth activities except cerium and is normally counted to determine $Y^{91}$ and $Pm^{147}$ activities, would be expected to also carry any $Y^{88}$ that might have been produced in a bomb burst by neutron activation (for diagnostic purposes). Such a fraction from the large filter pack collection of Aug. 17-Sept. 6 was

examined at HASL for this isotope (7). Indications were that there were traces of $Y^{88}$ present to the extent of less than one part per thousand relative to $Y^{91}$. Subsequently, a series of $Y^{91}$ separations covering collections made at Washington during 1962 and 1963 have been supplied to HASL for further study; no results of these analyses have yet been reported.

## DISCUSSION

There is no question that the isotopic content of fission debris varies with particle size immediately following a nuclear detonation; this fractionation can be detected in collections made soon thereafter, particularly in the tropospheric component of the debris. The question is, however, whether this initially varying radioactivity distribution among particles of different size will later become essentially uniform as the particle size distribution becomes more heavily weighted with small particles following deposition of the larger ones.

The effective particle size range of fission product-containing aerosols collected at ground level during 1963 appears to be rather narrow with most of the radioactivity associated with particles falling in the range $0.5\mu$-$1.0\mu$ diameter. Since during most of 1963 the radioactive fission products collected at ground level must have resided for an extended period of time in the stratosphere, their initial size must have been considerably smaller. Growth to the larger size range must have been accomplished either by self-condensation or by agglomeration with the ubiquitous nonradioactive aerosols present in quantity throughout the atmosphere.

Since such growth has obviously occurred, the original dimensions and chemical properties of the fission debris will be subordinated to the properties of the larger particles to which they become attached. Hence, separation of the conglomerate particles into size fractions would not in general be expected to result in isotopic fractionation. The data presented earlier suggests that a limited amount of fractionation does occur however.

There is presumably some intermixing of stratospheric and tropospheric air going on more or less continuously with the maximum rate occurring in the winter-spring season. Varying amounts of unattached particles, or particles having a smaller size distribution where the isotopic content would exert more influence on the properties of the aerosol, could thus exist in the upper troposphere for a short time at any season in a position where some fractionation could occur. Since thunderstorms penetrate this area, they might preferentially collect particles of a particular physical size or chemical nature (hydrophilic compounds) with resulting isotopic separation. A comparison of the relative isotope ratios in air before and after a rainstorm with the isotope ratios in the rainwater is suggested as a more sensitive means of determining the existence of a fractionation effect. Attempts are being made to secure samples under these conditions.

## CONCLUSIONS

Radioactive bomb debris which have resided several months in the stratosphere or upper troposphere become associated with nonradioactive aerosol particles by the time they arrive at ground level. The resulting particles have an effective or "average" size between $0.5\mu$ and $1.0\mu$ diameter (assuming spherical particles of density 1.8) and a rather restricted particle size distribution. Short term variations in the effective particle size are apparently independent of the activity concentrations. No connection is evident between the effective size or size range of radioactive particulate matter collected by filtration at ground level and the spring maximum in stratospheric subsidence of radioactive debris.

Isotopic fractionation by filters of different characteristics has not been found to be of great significance; however, there are indications that a more selective means of separating particles may show more differences between the isotopic constituents of particles of extreme size ranges. As a practical matter, isotopic differences with particle size would be expected to be slight since the radioactive particles collected at ground level are agglomerates consisting primarily of nonradioactive normal atmospheric aerosols. Fractionation should be greater under those atmospheric conditions which decrease the contact time between subsiding stratospheric debris and normal dust particles in the troposphere. Such a period might exist during thunderstorms when air from the upper troposphere or lower stratosphere is rapidly conveyed to ground level. Unfortunately, such conditions may not persist long enough for useful filter pack collections to be made. An alternative, however, would be to compare the various isotope ratios in the rainwater itself with those existing in the atmosphere before and after a thunderstorm.

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An evaluation of the relative distribution of some long-lived airborne fission products among several ranges of particle sizes has been carried out by the radiochemical analysis of individual air filters exposed in a pack. The relative quantities of radioactivity deposited on each of the filters in the packs has been found to vary considerably throughout 1963; however, each of the individual isotopes behaves in much the same manner so that the fractionation of isotopes relative to one another is slight.

Though isotopic fractionation by filters of different characteristics has not been found to be of great significance, there are indications that a more sensitive separating procedure may show more differences between particles in the extreme size ranges. The observed differences have been small and variable, rather than organized, but the variations are too large to be accounted for by experimental error. The long-lived natural radioisotope $Pb^{210}$ has a distribution very similar to that of the fission products, but with some preferential concentration in the smaller size range. The differences are not sufficient to invalidate the use of collections made on less than perfect filters for estimating the relative fission product concentrations in the air or for the approximate age determination of bomb debris.

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