An Evaluation of Existing Fallout Collection Methods

Analysis of data shows that open vessels and funnels are equally efficient for fallout sampling.

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Since 1951 considerable attention has been focused on the measurement of the deposition of radioactive debris from nuclear detonations. It has been ascertained that about one-third of this material never leaves the immediate vicinity of the detonation and is deposited as close-in fallout (*1*). The remaining by-products of the explosion are dispersed over the earth's surface by transport through the atmosphere.

The greatest single determinant of where and how soon fallout occurs is the original placement of the radioactive particles in the atmosphere. Tropospheric particles are generally limited in latitudinal spread and have a mean atmospheric residence time of about 30 days. Stratospheric fallout has been explained with varying success by several theories (*1, 2*), but it is agreed that particles introduced into the stratosphere deposit more slowly and in patterns which are quite different from those of tropospheric debris. In addition to the size of the detonation, many other factors, such as the site and altitude of burst, determine the placement of the debris in the atmosphere. While the effects of many of these factors may be estimated, the final practical requirement is the measurement of fallout on the ground.

At first glance, native soil would appear to be the best sampling medium. The analysis of soil, however, is complex and subject to large relative errors. Soil sampling is therefore reserved for cumulative or long-term measurements. Many sampling techniques have been tried for the approximation of natural ground deposition conditions. Devices employing planar, conical, and cylindrical collection surfaces have been employed to increase particle retention. Compact and combustible materials such as organic films and ion-exchange resins have been utilized to facilitate sampling, handling, and analysis.

Since individual sampling methods may be affected differently by the physical and meteorological factors controlling fallout, their efficiencies may be quite different. Therefore, an understanding of the characteristics of collectors is necessary before analytical results may be related. Eisenbud and Harley (*3*) have discussed the efficiency of gummed film relative to high-walled pots. Libby (*4*) and Eisenbud (*5*) have evaluated soil samples as a means of measuring deposition rates. Collins and Hallden (*6*) have compared a funnel-reservoir collector with pots and open tubs. Welford and Harley (*7*) have compared a funnel-resin system with pots. Rosinski (*8*) has compared an artificial grass sampler with gummed film and pots.

The over-all relationships among collector types is still vague because of the limited scope of previous observations and the lack of an absolute standard collector. This article presents a broader analysis of existing data and new evidence on the behavior of specific nuclides in fallout material.

Sampling Methods

The open-vessel collectors show great variations in exposure area. The open vessels most commonly used for monthly collections are stainless steel or plastic high-walled pots with an area of about 1 square foot. Shorter-term collections require larger exposure areas, of several square feet, and are usually made with plastic or metal-lined trays provided with runoff reservoirs. These vessels are exposed in the open during the sampling period, then the contents are removed for analysis.

In gummed-film samplers (*3, 9*), the sampling medium is a rubber-base cement spread on a 13- by 13-inch cellulose acetate backing and covered with glassine paper for protection. For sampling, the glassine paper is removed and the film is attached to a cadmium-plated steel frame having an opening of 1 square foot. The frame is held 3 feet above the ground on a stationary metal stand. Each paper is exposed for 24 hours. At the end of the sampling period, fresh films are put in place on spare frames. The exposed films are stripped off their frames and folded, with the gummed sides together, to a size suitable for handling.

There are two types of funnel systems (*6, 7, 10*). One funnel-reservoir system consists of a 1-liter polyethylene bottle equipped with a wide-mouthed cylindrical funnel with a conical base. The assembly is enclosed in a wooden container which is opened manually during rain and closed at other times. After exposure, the bottle may be removed for analysis of the contents.

The funnel ion-exchange collector (Fig. 1) consists of a funnel, an ion-exchange column, and a leveling device constructed from polyethylene. The funnel is welded to a threaded cap which is attached to the top of the ion-exchange column. The bottom of the column is also threaded for a tapered cap with attached leveling tube. The leveling device is a plastic tube extended from the bottom of the column to a Y tube above the filter in the column.

The ion-exchange column is packed with a paper pulp filter (Whatman No. 41 filter paper), anion-exchange resin (the chloride form of Amberlite IRA 400), and cation-exchange resin (the hydrogen form of Dowex 50 X-16). An electric heating device is suspended from the eye hook shown at the top of the wooden collector housing to prevent freezing during cold weather. At the end of the exposure period the entire column may be removed from the funnel and leveling device, capped, and shipped to the laboratory for analysis.

The artificial grass sampler (*8*) consists of two parts: an artificial grass surface and a stainless-steel reservoir. The 3- by 0.5-centimeter "grass" blades are cut from cellulose acetate sheets 0.01 inch thick and are coated with an adhesive—a resin dissolved in a volatile solvent. Antistatic materials are used

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Table 1. Comparison of the efficiencies of gummed film and roof pots as determined by activity levels found in New York during April 1958.

Nuclide	Activity level (mc/mi²)		Relative efficiency of gummed film (%)
	Gummed film*	Roof pot†	
Sr^{89}	1.87	16.6	11
Sr^{90}	0.20	3.33	7
Cs^{137}	0.81	4.37	19
Zr^{95}	97.0	128	76
Ce^{144}	24.0	53.2	46
Y^{91}	25.9	51.5	50
Total β activity	103	191	54

* Collection area, 1.0 ft². † Collection area, 0.82 ft².

to eliminate electrostatic effects. Ion-exchange membranes are used to absorb soluble material in the reservoir. Both the grass and the ion-exchange membrane components of the sample are ashed for analysis.

Evaluation of Data

The final test of any collector is its performance under field conditions. Since there is no absolute collector, this evaluation of performance must be based on an arbitrary standard. The pot has been chosen as the standard collector, since most existing data on fallout levels are based on pot collections.

In 1955, Eisenbud and Harley (*3*) estimated the efficiency of gummed film for collecting mixed fission products to be 63 percent in relation to that of pots, for daily collections. In 1957, Rosinski (*8*) estimated this ratio to be about 22 percent for weekly collections. This difference is reasonable, since gummed film is known to lose its retentiveness with extended exposure. In April 1958, total activity in pooled daily gummed-film samples from New York was 54 percent of total activity in pot samples. Radiochemical analysis of these samples showed that the efficiency of gummed film varied for different nuclides. These data, for averages of replicate collections, are listed in Table 1. When the pot values are taken as 100 percent, the average efficiencies for gummed film for the isotopes listed in the table vary from about 10 percent for the strontium nuclides to 76 percent for zirconium.

Two sources of data are used for comparison of the efficiencies of open vessels of different areas. Strontium-90 data listed by Collins and Hallden (*6*) from tub and pot collections made in 1956 are listed in Table 2. The average relative efficiency of open tubs, calculated from these data, is 124 ± 49 percent. Tables 3 and 4 list reported Sr^{89} and Sr^{90} data from Pittsburgh, Pa., and Westwood, N.J., respectively, in 1958 (*11*). Since short-term tub collections were made, the values shown represent monthly totals. In Pittsburgh, the relative efficiency of open tubs was 100 ± 39 percent for both nuclides, and in Westwood, 89 ± 4.5 and 80 ± 3.6 percent for Sr^{89} and Sr^{90}, respectively.

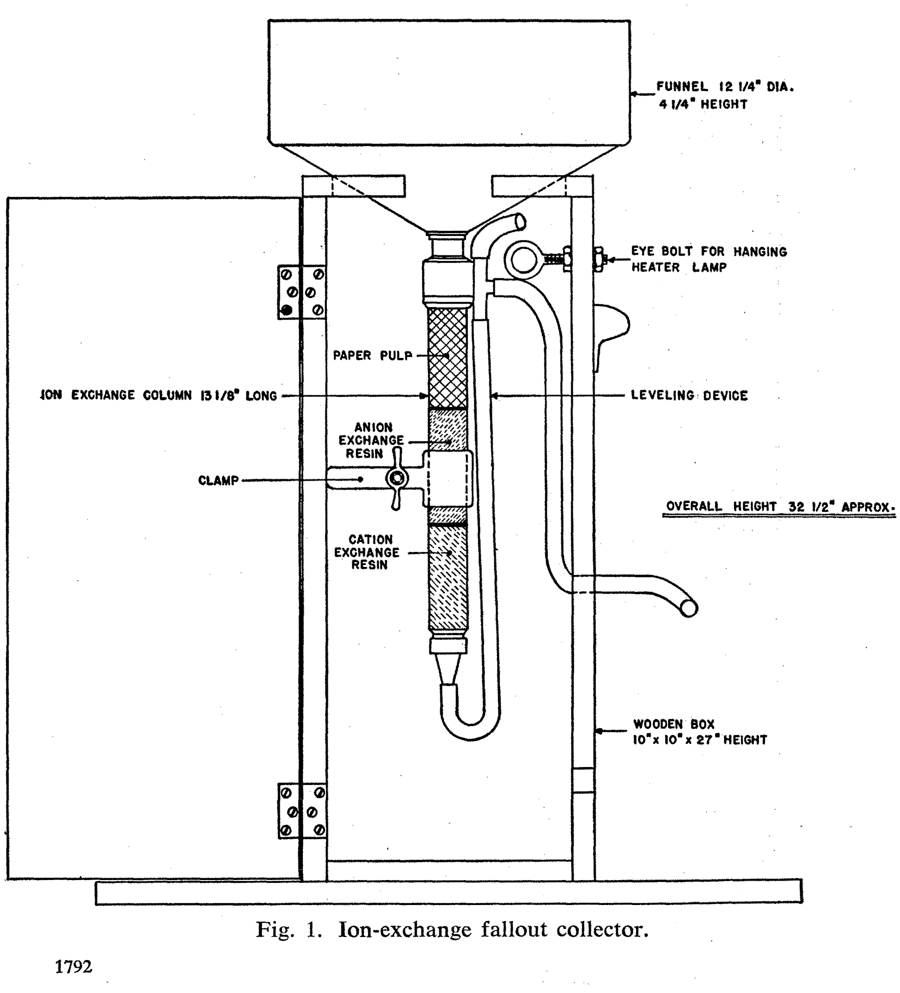

Fig. 1. Ion-exchange fallout collector.

Table 2. Comparison of findings for open-vessel collectors with respect to Sr^{90} activity levels found in the New York area during 1956 (*6*).

Sampling month	Sr^{90} activity level (mc/mi²)	
	Tub*	Pot†
Mar.	1.3	1.9
Apr.	1.0	0.8
May	1.5	1.0
June	1.4	0.8
July	0.6	0.6

* Collection area, 0.82 ft². † Collection area, 2.6 ft².

Funnel systems constitute the third category of collectors. Within this category the major difference in samplers is difference in the depth of the funnel. Two types of funnel, both of 12-inch diameter, were exposed in New York during the fall of 1957, one 6 inches and the other 1 inch in depth. The data obtained are listed in Table 5. Little influence of depth on the efficiency of the collector is evident.

In New York City during 1958, the relative efficiency of funnel collectors with respect to pots was found to be about 88 percent for total beta activity. This was determined from replicate monthly collections made over the 1-year period. Subsequent radiochemical analysis yielded the following efficiencies for individual isotopes: strontium-89, 82 percent; strontium-90, 91 percent; zirconium-95, 94 percent; cesium-144, 79 percent.

The synthetic grass collector described by Rosinski has an efficiency of about 150 percent with respect to pots exposed under the same conditions (*8*). This estimate is based on total beta activity measurements at several sites during 1957.

Finally, the pot as a collector has been evaluated against soil. For a carefully selected sampling site and an accurate strontium-90 analysis, measurement of the soil content of strontium-90 should give the best estimate of accumulated deposition. From data reported for New York for pot analyses

Table 3. Comparison of findings for open-vessel collectors with respect to Sr^{89} and Sr^{90} activity levels found in Pittsburgh during 1958 (*11*).

Sampling month	Sr^{89} activity levels (mc/mi²)		Sr^{90} activity levels (mc/mi²)	
	Tub*	Pot†	Tub*	Pot†
April	20.6	11.2	1.80	0.87
May	21.3	7.4	2.21	0.75
June	16.7	28.7	1.45	2.22
July	67.4	76.2	1.81	2.03
August	29.9	34.1	1.24	1.46
September	13.4	14.0	0.61	0.59
October	11.3	12.7	0.33	0.49
November	20.6	20.3	0.69	0.64
December	15.9	15.8	0.49	0.66

* Collection area, 2.6 ft². † Collection area, 0.82 ft².

Table 4. Comparison of findings for open-vessel collectors with respect to Sr^{89} and Sr^{90} activity levels found in Westwood, N.J., during 1958 (*11*).

Sampling month	Sr^{89} activity levels (mc/mi²)		Sr^{90} activity levels (mc/mi²)	
	Tub*	Pot†	Tub*	Pot†
April	32.4	31.8	1.60	1.73
May	26.0	26.4	2.75	2.81
June	13.2	10.6	1.06	1.05
July	29.0	36.2	1.85	1.11
August	21.0	20.7	0.86	1.10
September	12.5	15.5	0.65	0.70
October	45.9	64.5	1.25	1.64
November	51.8	43.8	0.83	1.38
December	12.8	20.7	0.47	0.79

* Collection area, 2.6 ft². † Collection area, 0.82 ft².

Table 5. Comparison of findings for funnel collectors of different shapes with respect to activity levels found in New York during January 1958.

Nuclide	Activity level (mc/mi²)	
	Funnel depth, 6 in.	Funnel depth, 1 in.
Sr^{90}	1.46	1.09
Cs^{137}	1.02	1.31
Zr^{95}	34.6	34.7
Ce^{144}	13.6	16.8

Table 6. Strontium-90 activity levels found in three replicate pot collections in New York during 1958.

Sampling month	Sr^{90} activity level (mc/mi²)		
	Analyst A	Analyst B	Analyst C
June	1.18	1.76	1.50
July	1.48	1.58	1.61
August	0.52	0.60	0.65
September	0.71	0.65	0.52
October	1.70	1.06	1.41
November	1.18	0.98	1.14
December	0.91	1.77	1.64

made since 1954 and for three annual soil analyses (*11, 12*), the over-all ratio of pot values to soil values was calculated to be 1.11 ± 0.072. By combining this pot-to-soil ratio with the ratio for funnel to pot, the funnel-to-soil ratio was estimated as 0.98.

Although each of these comparisons of sampling techniques was made on collections from a single location, there is still the question of reproducibility of a single type of sampler. Table 6 lists the strontium-90 activity levels in collections from three pots exposed simultaneously for monthly periods from June to December 1958 in New York City. The collections from each pot were analyzed by a different laboratory for strontium-90 content. Variance analysis shows that, at the 95-percent confidence level, there is no significant difference in the results for the three pots.

The results of analysis of pot and funnel collections made in New York City during 1958 and of tub collections made in Westwood (*11*) allow direct comparison of the three most widely used fallout collectors. Table 7 lists average strontium-90 activity levels found for May through December 1958 in the New York City area. Variance analysis of these data show no significant variation between findings for tub, pot, and funnel, at the 95-percent confidence level.

Conclusions

Analysis of the available strontium-90 data shows cumulative activity levels in New York City pot collections to be slightly higher than those in corresponding soils. The discrepancy is small, however, and quite possibly due to uncertainties in the soil sampling or analyses. Allowing for this and also for possible variations for areas which differ from New York City in topological and meteorological characteristics, we have drawn the following conclusions.

1) Gummed film has a considerably lower over-all collection efficiency than pots. Moreover, it is apparently not uniformly efficient for soluble and insoluble elements. This is undoubtedly due to the runoff characteristics of the flat gummed surface during rainfall.

2) The artificial grass collector retains more activity than other samplers. This is due to the three-dimensional aspect of the collection surfaces with respect to the atmosphere, and to the adhesive applied to the grass blades. Apparently the collector simulates conditions of standing vegetation better than conditions of ground deposition.

Table 7. Comparison of findings for tub, pot, and funnel systems with respect to Sr^{90} activity levels in New York and in Westwood, N.J., during 1958.

Sampling month	Sr^{90} activity level (mc/mi²)		
	Tub*	Pot†	Funnel‡
May	2.75	2.81	2.77
June	1.06	1.05	1.18
July	1.85	1.11	1.48
August	0.86	1.10	0.52
September	0.65	0.70	0.71
October	1.25	1.64	1.71
November	0.83	1.38	1.18
December	0.47	0.79	0.91

* Collection area, 2.6 ft². † Collection area, 0.82 ft². ‡ Collection area, 0.85 ft².

3) The shape of the funnel and the size of the pot or tub have little effect on collection efficiency. Although differences were apparent between tubs, funnels, and pots, in the monthly values for nuclides from fallout debris collected in these containers, these variations were not significant, at the 95-percent confidence level, over a year of observation. Therefore, it appears that the three collection systems most commonly employed are of equivalent usefulness within the expected limits of error from analysis and error from nonuniform deposition (*13*).

References and Notes

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