Fallout in New York City during 1958

The data indicate that short-lived fission products make a major contribution to radiation dose rates.

George A. Welford and William R. Collins, Jr.

Since 1951 increasing emphasis has been placed on the off-site sampling and measurement of radioactive debris from nuclear detonations. The primary objectives have been the estimation of world-wide gamma-radiation dose and documentation of the deposition of strontium-90 as a major biological hazard. In practice, these two consequences of nuclear testing were calculated from daily total beta-activity levels. More recently, world-wide monthly collections have been made for direct radio-chemical measurements. Of the sampling sites in operation, New York City is of particular interest because of its dense population and its location in the latitude of relatively high deposition of debris.

This article describes the deposition of strontium-90, strontium-89, cesium-137, zirconium-95, cerium-144, yttrium-91, and tungsten-185 measured in New York City during 1958. The chemical state of the debris at the time of deposition is approximated through solubility studies. The effect of rainfall on fallout patterns is discussed, and the age and origin of each monthly fallout collection are estimated through nuclide ratios.

Methods

In January 1958, an auxiliary collection system was established at the Health and Safety Laboratory of the U.S. Atomic Energy Commission for the detailed analysis of fallout debris in New York City. Three collection techniques were used to minimize inter-collector variances, and replicate collections were made to increase reliability. The sampling systems employed were high-walled stainless steel pots and two funnel systems (*1*). The funnel collectors exposed a cylindrical funnel with a conical base to the atmosphere. In one system the material collected was washed directly into a reservoir by rain or by added wash water, while the debris in the other system (an ion-exchange funnel collector) was washed through a paper-pulp filter and a mixed-bed ion exchanger to extract the nuclear debris.

A sequential radiochemical scheme was developed in order to analyze the total sample (3 to 5 grams of ash per month in New York) for the isotopes listed above (*2*). Finally, data collection and calculations were performed by means of an automatic data processing system built around two general digital computer programs. Experimental data were tabulated as the analyses proceeded and were punched into I.B.M. cards to be fed into the computer. The general calculations are fitted to specific analyses by substituting constants pertinent to individual nuclides (*3*).

Fallout Deposition in New York City

Until recently, fallout measurements at sites removed from testing areas were primarily concerned with concentrations of strontium-90 and cesium-137 and were expressed either in terms of cumulative levels or deposition rates. Since many complex biological factors must be considered as strontium and cesium pass through the food chain, it is agreed that deposition of these fission products is not an adequate measure of their hazard to man. When the route is from soil to plants to cattle to milk, entry into the body is a function of the accumulated amount deposited. When surface deposition on vegetation is considered the mode of entry, the rate of deposition is of prime importance.

Recently, estimates of the storage time of nuclear debris in the stratosphere have been reduced, indicating that considerable quantities of shorter-lived nuclides are deposited on a world-wide basis, along with strontium-90 and cesium-137 (*4, 5*). The question to be investigated is whether the concentrations of these nuclides are sufficient to necessitate their evaluation for the assessment of damage to the population.

Monthly activity levels for the fission products strontium-90, cesium-137, cerium-144, zirconium-95, strontium-89, and yttrium-91 are presented in Table 1. These data represent activities found at the time of analysis extrapolated to the end of the sampling month. The levels listed are the averages for a minimum of six collections for each monthly period. Figure 1 illustrates the monthly fallout patterns obtained from the data listed in Table 1. Three peak activity periods are illustrated for each isotope analyzed. The highest deposition occurs in April, with a second high evident in October-November. All isotopes indicate a smaller peak in July, followed generally by a minimum in August and September.

During the first 6 months of 1958, monthly and semimonthly collections were made. The results are reported in Table 2 and show no consistent variation in activity levels between the longer and shorter collection periods. Monthly levels are therefore reported throughout this article.

The half-life values for the nuclides analyzed are as follows (*6*): strontium-90, 28 years; cesium-137, 30 years; strontium-89, 51 days; yttrium-91, 53 days; zirconium-95, 63 days; tungsten-185, 74 days; and cerium-144, 290 days. Because of the long half-lives of strontium-90 and cesium-137 in relation to the 30-day sampling period, their activity levels at the end of the sampling period represent essentially the total amount of activity deposited. However, to obtain total monthly deposition for the individual shorter-lived nuclides, it is necessary to correct for decay during sampling. This is done by assuming a uniform deposition rate during the sampling month. The activity at the end of the month is then corrected mathematically for the effects of both deposition and decay. Since

The authors are staff members of the Health and Safety Laboratory of the U.S. Atomic Energy Commission, New York Operations Office.

fallout is random with respect to time, the net error involved in the year's observations is expected to be small.

Column 2 of Table 3 lists the calculated amounts of the seven nuclides analyzed that were deposited in New York City during 1958. The sum of these activities is 2000 millicuries of beta activity per square mile. Since the total land area of New York City is about 316 square miles, 650 curies were deposited in this area. This corresponds to 200 milligrams of radioactive material. The long-lived fission products, strontium-90 and cesium-137, contributed only 0.9 and 1.0 percent to the total, respectively. Column 3 of Table 3 lists the amount of each nuclide present on 1 January 1959. Totalling these activities we get 690 millicuries per square mile or a 65-percent reduction in beta activity. The contributions of the 12 monthly increments of beta activity to cumulative levels are illustrated in Fig. 2. The effect of radioactive decay on the year's final level (690 mc/mi^2) is obscured by heavy fallout during October and November. The contributions of strontium-90 and cesium-137 to the 1 January level were 2.7 and 3.1 percent, respectively.

Table 1. Monthly isotopic activity levels found in New York City fallout during 1958.

Sampling month	Activity level (mc/mi^2)						
	Sr^{90}	Cs^{137}	Ce^{144}*	Zr^{95}*	Sr^{89}*	Y^{91}*	W^{185}*
January	1.18	1.08	10.4	27.7		19.5	0.0
February	1.40		14.7	28.7		23.6	0.0
March	1.52	3.22	35.9	93.4		29.7	0.0
April	3.68	3.46	52.4	139	23.6	51.5	0.0
May	3.45	2.95	41.2	49.4	16.7	36.1	2.02
June	1.28	0.57	30.2	32.3	14.3	3.28	10.8
July	1.47	1.98	28.6	52.6	43.9	10.7	47.1
August	0.553	1.48	11.7	21.6	17.1	3.52	31.3
September	0.575	1.23	12.5	22.8	14.3	3.50	24.0
October	1.17	1.82	42.2	114	47.0	10.4	22.0
November	1.19	1.74	35.7	126	36.6	13.5	23.4
December	0.790	1.62	23.8	83.4	25.0	9.34	10.0

* Extrapolated to the end of the sampling month.

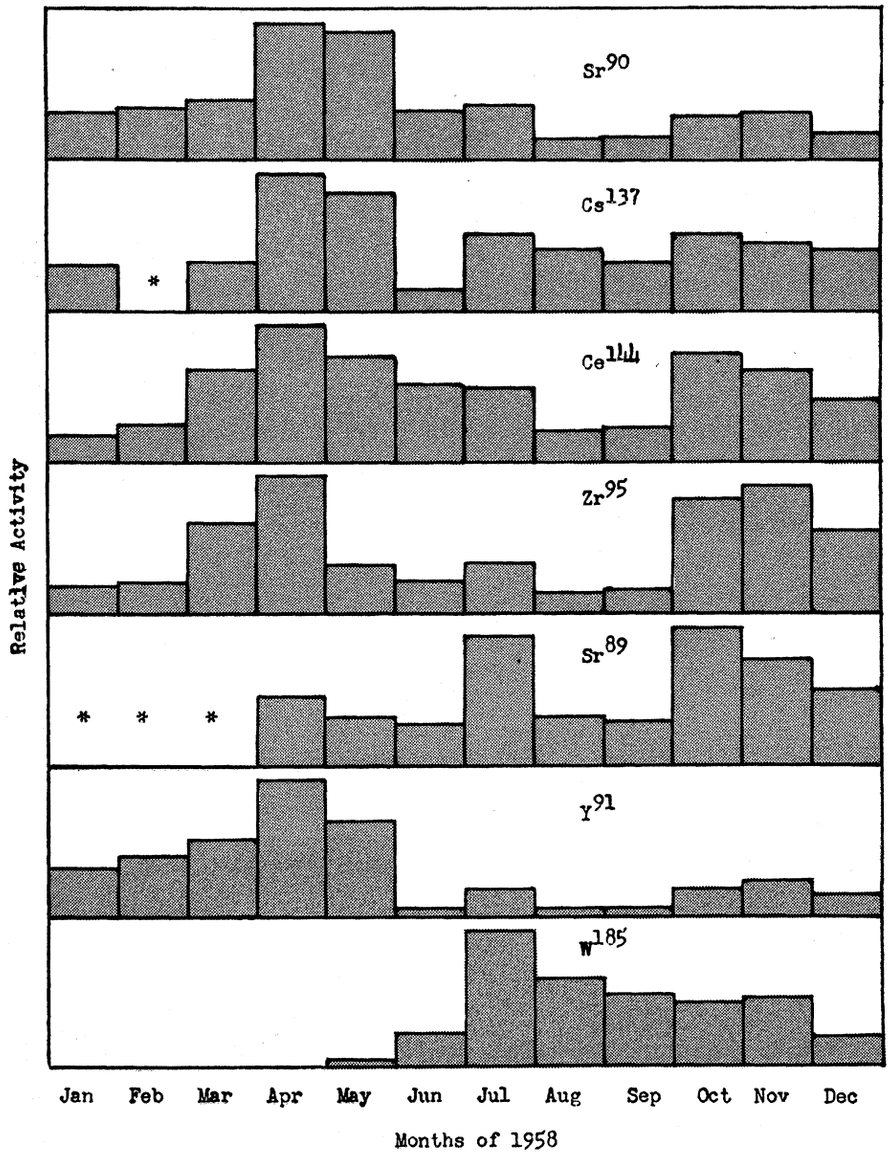

Fig. 1. Monthly fallout patterns in New York City during 1958. Asterisk indicates no data available.

Tungsten Deposition

Tungsten, an element not occurring in fission, was a tracer for the test series conducted by the United States in the Pacific during the spring and summer of 1958. The isotope tungsten-185 was first detected in New York City fallout in the latter part of May 1958. The monthly fallout pattern is illustrated, along with the fission products, in Fig. 1. July was the month of peak deposition of this isotope in the New York City area, and this peak appeared concurrently with the second monthly activity level peak illustrated for the six other nuclides analyzed. From this it appears that the July deposition peak in the New York City area resulted primarily from the Pacific test series, which also contributed to the observed monthly fallout levels for the remainder of 1958.

Solubility of Fallout Nuclides

Strontium-90 and cesium-137 are considered to be biological hazards because of their entry into the food chain and their final uptake by the human body. Martell (*4, 7*) has studied the chemical state of strontium in fallout debris and reports that strontium-90 is formed primarily as the oxide. Through weathering and absorption of moisture and carbon dioxide, the isotope is generally deposited in long-range fallout as the carbonate.

During the first 6 months of 1958, the solubility of nuclides, as deposited on the ground in New York City, was measured with an adaptation of the collection service described by Welford and Harley (*1*). The unit consisted of two polyethylene ion-exchange columns connected in series and attached to a funnel with a leveling device to prevent either column from running dry during the exposure period. As the fallout is deposited in the funnel, the first column, consisting of paper pulp supported on a glass-wool base, filters out the insoluble material. The soluble material passes through and is collected in the second column, which contains a mixed-bed ion exchanger that adsorbs this soluble material quantitatively. The average percentages of the nuclides found in the insoluble portion (paper pulp) and the soluble portion (ion-exchange resins) are listed in Table 4. From these data it is evident that most of the isotopes are deposited in the New York City area in both soluble and insoluble form. Notable exceptions are the strontium isotopes, which occur predominantly in the soluble form. Cesium-137 in fallout debris is 70-percent soluble, and zirconium-95 is the most insoluble of the nuclides deposited in New York City, showing an average of only 29-percent solubility.

Rainfall

It has been established from strontium-90 and cesium-137 measurements that precipitation is a controlling factor in the deposition of fallout from the lower atmosphere (*7*). Since most of the particles present in long-range nuclear debris are not of sufficient mass for significant deposition to occur through gravitation, deposition is caused mainly by condensation of water droplets around the debris or the adherence of debris particles to raindrops already formed. Therefore, the activity content of the air is also a major factor in the activity levels of fallout deposition. For limited areas, weathered by air masses of reasonably consistent activity concentrations, fallout deposition is proportional to rainfall (*8*).

Over the years, this effect has been obscured in the New York area by large seasonal variations in air concentrations and variations caused by debris dispersed from the Nevada testing site. Figure 3 relates monthly depositions of strontium-90 and cesium-137 to monthly rainfall in New York City during 1958. The dependence of fallout on rainfall is shown by the coincidence of peak precipitation and deposition months in April, July, and October. Stewart *et al.* (*9*) observed that heavy rains lower and light rains increase the specific activity of rain water for individual sites. This was illustrated in New York during September and October, when the monthly precipitation rates exceeded the mean rate for the year and the specific activity was low. In November and December, when the monthly rains were below average, specific activity levels were high.

Since 1955, sharply increased fallout deposition during the spring months has been observed in the northern latitudes (*10*). This phenomenon has been attributed to tropospheric dispersion (*11*) and delayed stratospheric deposition (*12*). Martell (*4*) recently cited fall and winter testing of intermediate megaton devices in the northern latitudes as having temporarily lodged fresh debris in the lower levels of the northern stratosphere. For 1958, large amounts of strontium-89, zirconium-95, cerium-144, and yttrium-91, in addition to strontium-90 and cesium-137, were reported in spring depositions in New York City (Table 1). Figure 3 illustrates the influence exerted by rainfall during this period of increased atmospheric activity. Similar depositions are recorded for March and May, when rain levels were average, but high rainfall in April caused an appreciable rise in the deposition rate. Similar correlation between the deposition of shorter-lived material and rainfall is noted in other periods of consistent, but presumably lower, atmospheric concentra-

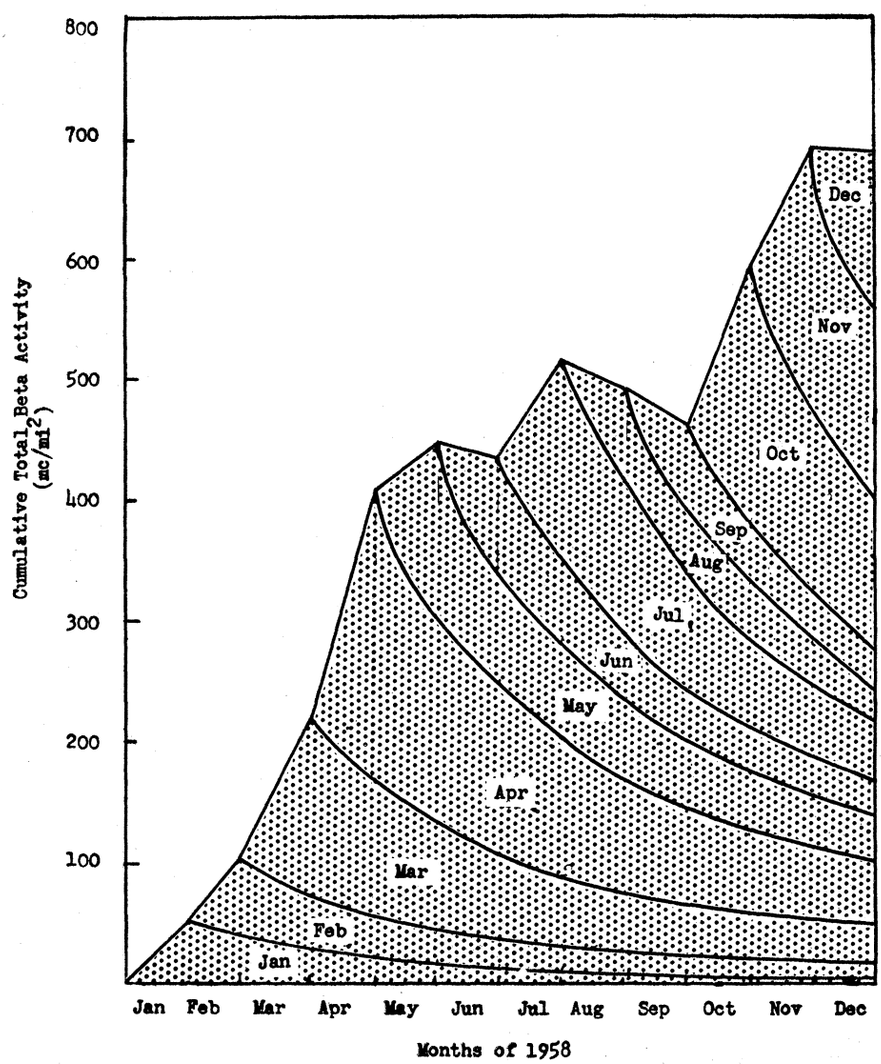

Fig. 2. Contribution of monthly increments to cumulative total beta activity levels in New York during 1958.

Table 2. Comparison of monthly (*a*) and sum of semi-monthly (*b*) activity levels found in New York City fallout during 1958.

Period	Activity level (mc/mi²)					
	Sr^{90}	Cs^{137}	Ce^{144}*	Zr^{95}*	Sr^{89}*	Y^{91}*
			March			
(a)	1.52	3.22	35.9	93.4		29.7
(b)	1.96	3.94	25.2	91.2		47.3
			April			
(a)	3.68	3.46	52.4	139	23.6	51.5
(b)	3.21	4.92	56.4	104	31.3	45.7
			May			
(a)	3.45	2.95	41.2	49.9	16.7	36.1
(b)	3.04	3.49	51.6	88.3	20.8	40.6
			June			
(a)	1.28	0.57	30.2	32.3	14.3	3.28
(b)	1.35	0.89	27.1	67.6	13.7	7.66

* Extrapolated to the end of the sampling month.

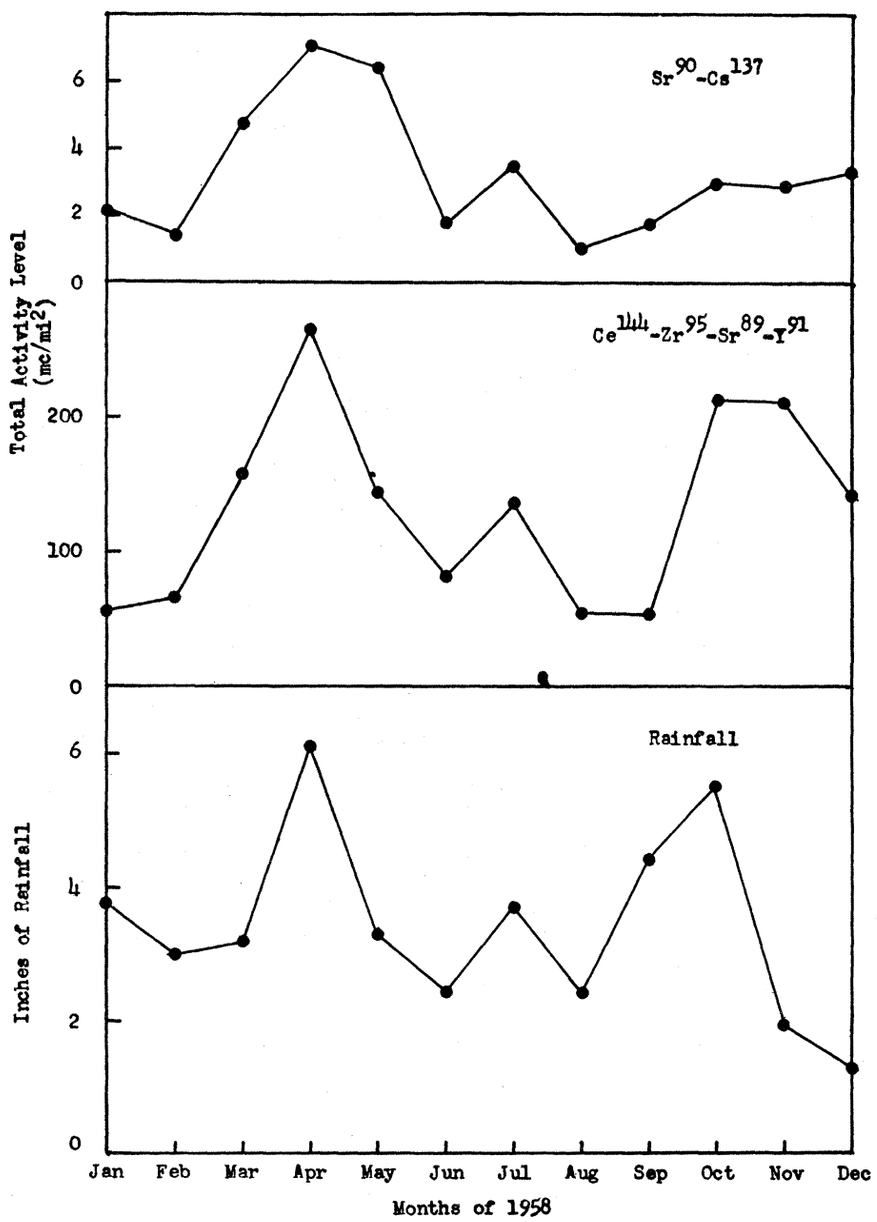

Fig. 3. Comparison of long- and short-lived fallout levels with rainfall in New York during 1958.

tions. July and October (Fig. 1) show simultaneous peaks, and June and August show corresponding minima in the deposition and rainfall curves.

Generalizations on the over-all relationship between fallout and weather are complicated by several sources of error. Sampling, analysis, dry deposition, wind currents, and the eclipsing of short-term phenomena by the extended sampling times may all contribute to the variations appearing in the data. Notable in this respect are the high strontium-90 and cesium-137 activities observed in May (Fig. 3) in conjunction with minimal rainfall and the high short-lived depositions in November, also associated with low precipitation.

Age and Origin of Debris

The approximate age of fallout debris may be established through the relative concentrations of nuclide pairs. Theoretical slow fission yields may be used to approximate the amounts produced by the detonation, and the ratio may be extrapolated by means of the decay constants of two nuclides. Strontium-89 and strontium-90 have been most frequently used in dating long-range debris. Since mixtures of old and fresh debris result in enrichment of strontium-90 with respect to strontium-89, ratios of shorter-lived materials have proved more sensitive.

During 1958, peak depositions of shorter-lived nuclides in New York in April, July, and October–November indicated the arrival of debris from several recent test series. To determine the age of the monthly depositions, Sr^{89}/Sr^{90} and Zr^{95}/Ce^{144} ratios were used. Table 5 lists apparent production dates calculated from these ratios. There is generally good agreement in the two approximations. The debris collected from January through June originated primarily from tests occurring in the fall of 1957. In March and April, debris from the tests conducted in the winter months of 1958 arrived along with older debris. Collections for July through December have already been associated with the U.S. series in the spring of 1958, in which tungsten-185 was produced. The nuclide ratios corroborate this evidence and also show the arrival from October through December of debris from the U.S. and Russian fall tests.

Table 3. Cumulative aspects of New York City fallout during 1958.

Nuclide	Total beta activity deposited during 1958 (mc/mi²)	Calculated total beta activity on 1 Jan. 1959 (mc/mi²)
Sr^{90}	18.2	18.2
Cs^{137}	21.2	21.2
Ce^{144}	349	173
Zr^{95}	925	277
Sr^{89}	294	93.2
Y^{91}	264	33.4
W^{185}	192	71.6

Nuclide ratio approximation is limited by several factors, including variation of actual yields from the theoretical and fractionation of nuclides in the fireball of the detonation. These limitations are compounded, in the dating of long-range fallout, by mixture of debris before deposition. This is especially true of strontium-90, which outlasts strontium-89 in old debris and distorts the dating of mixtures. This effect is observed in April and May samples (Table 5), where the strontium internal ratios yield earlier apparent production dates than the Zr^{95}/Ce^{144} ratios.

Table 4. Solubility of nuclides in fallout.

Nuclide	Percent insoluble (av.)	Percent soluble (av.)
Sr^{90}	0.03	95.6
Sr^{89}	3.56	94.4
Cs^{137}	22.6	70.0
Y^{91}	47.6	52.4
W^{185}	52.3	45.6
Ce^{144}	57.3	42.0
Zr^{95}	67.8	29.1

Table 5. Age of debris estimated from nuclide ratios in New York City fallout during 1958.

Sampling month	Apparent production dates	
	Zr^{95}/Ce^{144}	Sr^{89}/Sr^{90}
Jan.	Nov. 1957	
Feb.	Nov. 1957	
Mar.	Jan. 1958	
Apr.	Feb. 1958	Sept. 1957
May	Nov. 1957	Sept. 1957
June	Dec. 1957	Jan. 1958
July	Mar. 1958	Apr. 1958
Aug.	Apr. 1958	May 1958
Sept.	May 1958	May 1958
Oct.	Aug. 1958	Aug. 1958
Nov.	Oct. 1958	Aug. 1958
Dec.	Nov. 1958	Sept. 1958

Conclusions

Rainfall is clearly a controlling factor in fallout deposition patterns. Proportionality relationships between activity and rainfall, which have been established for areas of consistent atmospheric activity, pertain also to other areas during limited periods when air concentrations are static. The dependence of fallout on rainfall is not notably different for old and fresh debris or for long- and short-lived nuclides.

Strontium-90 and cesium-137, biological hazards in fallout material because of their similarity to calcium and potassium, respectively, are deposited in predominantly soluble form, and this increases the possibility of entry into the food chain.

The deposition pattern of tungsten-185 in New York City during 1958 shows that nuclear tests contribute to off-site fallout within weeks of the detonation. Moreover, this contribution is measurable for at least 8 months, even when testing and sampling occur in widely separated locations.

The approximate age of the debris collected in New York during 1958 is established through fission-product ratios. When an average age of 100 days is assumed, it is estimated that the seven nuclides measured account for about 40 percent of the year's total beta activity. When the calculated amounts of yttrium-90, praseodymium-144, niobium-95, ruthenium-103, cerium-141, barium-140, lanthanum-140, ruthenium-106, rhodium-106, and promethium-147 are added to values for the nuclides measured, it appears that the total deposition of beta activity in New York City during 1958 was 5 curies per square mile, and that the level on 1 January 1959 was 1.7 curies per square mile. Less than 3 percent of either of these values is attributable to the Sr^{90}-Y^{90} and Cs^{137}-Ba^{137} chains.

According to prevailing fallout distribution theory, New York fallout levels are representative of a mid-latitude band covering about 15 percent of the earth's surface. Since the fallout in this area is no more than four times that of other areas, it is obvious that continued testing at a rate similar to, or above, that in 1958 will necessitate the complete documentation of deposition levels for shorter-lived nuclides. Any evaluation of radiation dose to the population from fallout must include these activity levels (*13*).

References and Notes

1. G. A. Welford and J. H. Harley, "A New Method for the Collection of Fallout Material from Nuclear Detonations," *U.S. Atomic Energy Comm. Rept. No. HASL-42* (1958), pt. 4.
2. G. A. Welford, W. R. Collins, D. C. Sutton, R. S. Morse, *U.S. Atomic Energy Comm. Rept. No. HASL-57* (1959).
3. W. R. Collins, *U.S. Atomic Energy Comm. Rept. No. HASL-64* (1959).
4. E. A. Martell, *Science* **129**, 1197 (1959).
5. "Fallout from Nuclear Weapons Tests," *Summary–Analysis of Hearings before the Joint Congressional Committee on Atomic Energy, May 5–8, 1959* (Government Printing Office, Washington, D.C., 1959).
6. D. Strominger, J. Hollander, G. Seaborg, *Revs. Modern Phys.* **30**, 2 (1958).
7. E. A. Martell, *U.S. Atomic Energy Comm. Rept. No. AECU-3297* (195→ W. F. Libby, *Proc. Natl. Acad. Sci. U.S.* **42**, 945 (1956).
8. W. R. Collins and N. A. Hallden, *U.S. Atomic Energy Comm. Rept. No. NYO-4889* (1957); F. Bryant, A. Chamberlain, A. Morgan, G. Spicer, *J. Nuclear Energy* **6**, 22 (1957).
9. N. Stewart, R. Osmond, R. Crooks, E. Risher, "World-Wide Deposition of Long Lived Fission Products from Nuclear Test Explosions," *Atomic Energy Research Establ. G. Brit. Publ. No. AERE HP/R 2354* (1957).
10. "Fallout Monitoring and Documentation," *U.S. Atomic Energy Comm. Rept. No. HASL-42* (1958), pt. 1; "Strontium Program Progress Report," *U.S. Atomic Energy Comm. Rept. No. HASL-65* (1959).
11. W. F. Libby, *Proc. Natl. Acad. Sci. U.S.* **42**, 365 (1956).
12. L. Machta, "Discussion of meteorological factors and fallout distribution," paper presented at the Symposium on Low-Level Irradiation, American Association for the Advancement of Science (December 1957).

13. Without the assistance of our colleagues, Doris C. Sutton and Robert S. Morse, this article would not have been possible. Acknowledgments are also due the other members of the Analytical and Biometrics divisions of the Health and Safety Laboratory who helped in this study.