25 November 1960, Volume 132, Number 3439 SCIENCE

Atmospheric Transport of Artificial Radioactivity

Isotope-ratio and tungsten-185 data show large behavior differences for various stratospheric sources.

E. A. Martell and P. J. Drevinsky

The atmospheric aspects of world-wide fallout from nuclear-weapon tests have been much studied and discussed (*1–6*), and many details of the physical behavior of fallout are now reasonably well known. Nevertheless, there remains a general lack of understanding of large-scale atmospheric transport and mixing processes. Widely different views have been offered to explain the marked spring increase in fallout rate which has been observed at some Northern Hemisphere locations every year over the period 1955 to 1959. The explanation for the excessive accumulation of fallout in north temperate latitudes also is confused. The confusion is due partly to the unsatisfactory nature or quality of much of the fallout data, but more particularly to the use of invalid assumptions with respect to the origin or atmospheric behavior of the fallout debris.

Radioactive fallout usually is classed as local, tropospheric, or stratospheric. Local fallout is radioactive debris which is deposited within a few hundred miles of the test site and is comprised principally of large particles from surface or subsurface nuclear explosions. Tropospheric fallout is nuclear-test debris which is confined to the lower atmosphere below the tropopause throughout its mixing history. Stratospheric fallout is that component of world-wide fallout derived from radioactive clouds which initially rose to heights above the tropopause. For surface or near-surface explosions, tropopause penetration occurs when the total energy yield exceeds about 200 kilotons for the equatorial tropopause and about 100 kilotons for the polar tropopause. In world-wide fallout, it is well known that most long-lived radioisotopes are of stratospheric origin. It usually has been assumed that most short-lived radioisotopes in fallout over areas remote from nuclear-test sites are of tropospheric origin.

Stewart, Crooks, and Fisher (*7*) estimated the mean atmospheric residence time of tropospheric fallout to be about 1 month. After the high-yield U.S. Castle tests in early 1954, Libby (*1*) first pointed out the long holdup of strontium-90 in the stratosphere and estimated the mean stratospheric storage time to be 5 to 10 years. At that time the spring increases and north temperate latitude peak in Sr^{90} fallout were considered to be due to tropospheric fallout from Nevada and Soviet tests. Stewart *et al.* (*3*) attempted to distinguish the tropospheric and stratospheric components of Sr^{90} fallout by use of Sr^{89} and Sr^{90} concentration data for rainfall at Milford Haven, Wales. Taking 35 days as the mean tropospheric storage time and assuming that all the Sr^{89} was tropospheric in origin, they showed that most of the Sr^{90} fallout after 1954 was of stratospheric origin. They attributed the spring increases in fallout rate to a more rapid downward mixing of stratospheric air during the spring, and they related the north temperate latitude peak in Sr^{90} fallout to a selective zone of downward mixing of stratospheric air at middle latitudes. Machta and List (*8*) offer a similar interpretation of the seasonal and latitudinal pattern of fallout. Kuroda (*9, 10*) has derived a general equation for distinguishing the stratospheric and tropospheric contributions of fallout from isotope-ratio data. The incorrect assumption of a rapidly and uniformly mixed stratospheric reservoir is inherent in his analysis scheme.

The principal shortcoming common to these attempts to interpret the atmospheric behavior of fallout is the failure to distinguish the influence of latitude, altitude, and time of injection on the storage time and fallout pattern for stratospheric sources. In most instances, arbitrary assumptions with respect to the tropospheric fallout contribution also have given rise to confusion. On the basis of evidence provided by fission-product concentration ratios in precipitation, Martell (*4*) pointed out that the short-lived radioisotopes in world-wide fallout were principally of stratospheric origin and that stratospheric storage times decrease markedly with latitude and increase with altitude.

In the present article we use 12.8-day barium-140, 53-day strontium-89, 28-year strontium-90, and 74-day tungsten-185 radioactivity concentrations in individual precipitation samples as a basis for interpreting some of the major features of the transport and deposition patterns for stratospheric fallout. The measurement of the concentration in rainfall of two or more fission-product radioisotopes makes it possible to relate the fallout which is being deposited on the earth's surface to known atmospheric sources of different age, strength, and location in space. It is well established (*1–4*) that the scavenging action

The authors are members of the Geophysics Research Directorate, Air Force Research Division, Bedford, Mass.

of precipitation is the principal mechanism for the removal of radioactive particles from the atmosphere. Thus, the concentration of a given radioisotope in rains and the rainfall records provide a reasonably accurate measure of the accumulation of the radioisotope in soils and surface waters.

The usefulness of data on fission-product ratios as a means of estimating the relative contribution of various nuclear-test sources to total fallout has been questioned on the basis of possibly serious physical and chemical fractionation effects and of uncertainties in fission-product production-yield ratios. For the high-yield nuclear explosions which have been dominant as sources of short-lived as well as long-lived radioisotopes in world-wide fallout, the relatively late condensation times minimize fractionation effects due to delayed formation of radioisotopes with short-lived krypton and xenon precursors. The chemical similarity of barium and strontium limits chemical fractionation effects on Ba^{140}/Sr^{90} ratio data; Sr^{89}/Sr^{90} ratio data obviously are unaffected by chemical factors. For the major sources of fission products in world-wide fallout, the principal production reactions are undoubtedly 14-Mev neutron fission of uranium-238 and fast-neutron fission of plutonium-239 and uranium-235. On the basis of fission yields reported by Katcoff (*11*), calculated production activity ratios of 170 for Sr^{89}/Sr^{90} and 1200 for Ba^{140}/Sr^{90} are unlikely to be in error by more than 20 percent and 40 percent, respectively. Uncertainties due to the use of these production-ratio values, or due to the assumption that there are no fractionation effects, do not seriously affect the usefulness of isotope-ratio data for distinguishing the major differences in atmospheric behavior of nuclear-cloud sources of different altitude and latitude for well-spaced nuclear tests.

A useful tracer for studying the behavior of fallout debris from equatorial tests is provided by the tungsten-185 radioisotope. This radioactivity was produced in a number of surface nuclear explosions in the U.S. Hardtack nuclear-test series at 11° north latitude during the period May through July 1958. Libby (*12*) has estimated the total Hardtack production of W^{185} as 250 megacuries at a reference date of 1 August 1958. Since most of the W^{185} was produced in several surface shots of sufficient yield to thrust the activity into the lower stratosphere, the atmospheric mixing and world-wide fallout patterns for the W^{185} should be characteristic of sources at lower levels of the equatorial stratosphere. Data on W^{185} concentration for rainfall samples from both hemispheres are presented and discussed below.

Experimental Procedures

Individual rains were collected during 1958 and 1959 in a Lucite funnel fitted to a polyethylene bottle at selected sites in both hemispheres. Concentrations of selected radioisotopes are reported for Burlington, Vermont (44°N, 73°W); Bedford, Massachusetts (42°N, 71°W); and Pôrto Alegre, Brazil 30°S, 51°W). The particulate matter of each sample was filtered off and rinsed with dilute hydrochloric acid, with additional water and dilute ammonium hydroxide rinses when W^{185} was to be determined. The liquid fraction of each sample was combined with the rinse solutions, concentrated by evaporation, and analyzed for Sr^{89}, Sr^{90}, Ba^{140}, and W^{185} by standard radiochemical techniques (*13–15*). Analysis of solid fractions of samples was discontinued after early measurements indicated that these contained a negligible fraction of the total activity of each radioisotope being measured.

To samples analyzed for Ba^{140} and Sr^{90}, fuming nitric acid was added to precipitate the combined nitrates. Several ferric hydroxide scavengings were followed by a separation of barium from strontium by the precipitation of barium chromate. Barium was purified by repeated chloride precipitations and ferric hydroxide scavengings. The final sample was barium chloride monohydrate. Strontium was purified by barium chromate and ferric hydroxide scavengings and precipitated as the carbonate, which was milked for 64.6-hour yttrium-90 after Sr^{90}-Y^{90} equilibrium was attained. Yttrium phosphate precipitations were followed by an oxalate precipitation. The final sample was yttrium oxide.

When samples were analyzed sequentially for W^{185}, Sr^{89}, and Sr^{90}, tungsten was separated from strontium by the precipitation of tungstic oxide with hydrochloric acid. Tungsten was purified by sodium carbonate fusion and reprecipitated as the oxide from the aqueous leach solution. The final sample was tungsten 8-hydroxyquinolate. Strontium was precipitated as the carbonate and purified by repeated nitrate precipitations and ferric hydroxide scavengings. Barium chromate and ferric hydroxide scavengings were followed by the precipitation of strontium carbonate, which was counted for total strontium. When secular equilibrium was attained, strontium carbonate was dissolved in dilute hydrochloric acid and Y^{90} was milked by precipitations of yttrium hydroxide and yttrium oxalate. The final sample was yttrium oxide.

All beta measurements were carried out by means of thin-wall, cylindrical, flow-type Geiger counters, of 1.4-centimeter diameter and 3.7-centimeter active length. Details of construction and operation of counters of this type have been given elsewhere (*14–16*). Use of massive-steel and anticoincidence shielding provided background count rates of about 0.2 count per minute. Radioisotope disintegration rates were determined by means of the thick-solid sample technique of Suttle and Libby (*17*). Observed activities were corrected for counter background, sample self-absorption, external absorption, counter geometry, chemical yield, and sample volume to obtain specific activities in disintegrations per minute per liter.

Barium-140 to Strontium-90 Ratios

The Ba^{140}/Sr^{90} activity ratios for individual rainfall samples collected at Bedford, Massachusetts, in 1958 and early 1959 are presented in Fig. 1. A limited number of similar data obtained by Kuroda (*9*) at Lemont, Illinois, and by Suess (*18*) at La Jolla, California, during early 1958 are included for comparison. The Ba^{140} activity data were corrected for decay to the mean time of precipitation. Estimated errors in isotope ratios for Bedford are shown in Fig. 1; these are based on counting errors and decay-curve resolution errors in determining Ba^{140} and Sr^{90} specific activities. The data which show errors exceeding 10 percent represent either very small samples from light rains or samples of very low Ba^{140} activity.

The curve for Ba^{140}/Sr^{90} activity-ratio data in Fig. 1 displays several instructive features. Unlike data for concentration in rainfall of individual radioisotopes, which are very sensitive to rainfall intensity and scatter widely from rain to rain, the isotope-ratio values change smoothly with time. The 12.8-day half-life of the Ba^{140} activity makes the ratio a sensitive indicator of each new test source. The source of

the Ba^{140} component for any data point can be approximately dated by extrapolating back along a Ba^{140} decay-slope line to the intercept date corresponding to the production ratio. Less than a one-week uncertainty in dating is introduced by using a production ratio of 1200. Because of the presence of some Sr^{90} from older stratospheric sources, the actual production date for the Ba^{140} component will be more recent than the intercept date. It is clear that the Ba^{140} activity in rain in north temperate latitudes during March to May 1958 came from the February–March 1958 Soviet tests; that during July to September 1958, from the May–July 1958 U.S. Hardtack tests; and that during October 1958 to March 1959, from the October 1958 Soviet tests. The corresponding Sr^{90} contribution of each test series can be only crudely estimated from the production ratio. However, Sr^{89}/Sr^{90} activity-ratio data for well-spaced tests would provide a more reliable estimation of the Sr^{90} contribution of various tests because the production ratio is more reliable and the Sr^{89} half-life of 53 days is comparable to the durations of nuclear-test series.

It usually has been assumed that the short-lived fission-product activity observed in rainfall and surface air is tropospheric contamination from small weapon tests and is washed out of the troposphere by rains with a half-time of three or four weeks. The data on Ba^{140} and Sr^{90} concentration support the opposite conclusion, that Ba^{140} and other short-lived radioisotopes in fallout are primarily of stratospheric origin. During the periods from mid-April to the end of May 1958, July through September 1958, and January through March 1959, the Ba^{140}/Sr^{90} activity ratio (Fig. 1) decreased according to the decay rate of Ba^{140}. Within each of these periods the average concentration of Sr^{90} activity in rainfall and air either increased with time or remained approximately constant. The Ba^{140} activity, corrected for decay, varied in the same manner. It is clear that the observed Sr^{90} fallout is principally of stratospheric origin. Thus, for these periods it also must be concluded that even the short-lived Ba^{140} activity is of stratospheric origin. A stratospheric origin for both Ba^{140} and Sr^{90}, even during early April 1958 when the decrease in the Ba^{140}/Sr^{90} ratio was more rapid than the decay of Ba^{140}, is plausible on the basis of differences in initial arrival times for debris from stratospheric sources of different altitude or latitude. The rapid decrease in the Ba^{140}/Sr^{90} ratio at mid-June 1958 is explained on a different basis below. The stratospheric origin of the Ba^{140} activity is consistent with both the magnitude of the Ba^{140}/Sr^{90} activity ratios and their gradual and systematic change with time (Fig. 1). For random mixtures of tropospheric and stratospheric sources, one would expect large fluctuations in the Ba^{140}/Sr^{90} ratios and occasional values indicating an apparent age of less than one month. It appears necessary to conclude that most of the short-lived and essentially all of the long-lived radioisotopes in world-wide fallout are of stratospheric origin.

The data on the Ba^{140}/Sr^{90} activity-ratio (Fig. 1) for the period July through September 1958 appeared to indicate an alternation of contribution

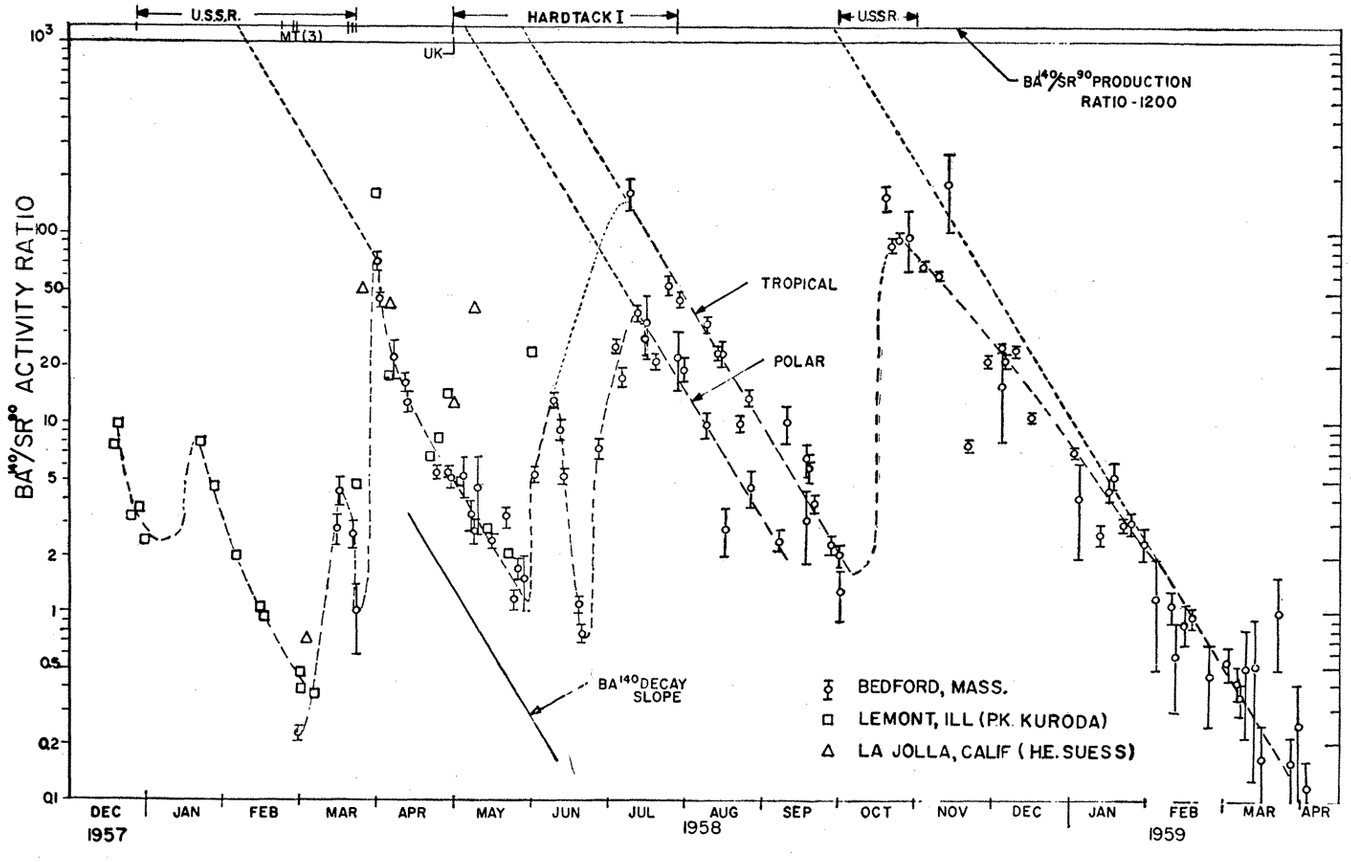

Fig. 1. Ba^{140}/Sr^{90} activity ratios for individual rains collected at Bedford, Massachusetts. The Ba^{140} activities are corrected for decay to the mean time of precipitation. Ratios for a limited number of rains collected at Lemont, Ill. (*9*), and La Jolla, Calif. (*18*), are also included. Test sources for Ba^{140} are indicated. Ratios for July to September 1958 are grouped into two classes, according to the association of a given rain with an air mass of tropical or polar origin.

from two regions of the stratosphere, one containing debris from an early portion of the U.S. Hardtack nuclear-test series and the other containing fresher debris produced several weeks later. This possibility was checked by plotting, at 6-hour intervals, the mean trajectories at several levels (5000, 10,000, and 14,000 feet) for air associated with each rain during this period. Air-mass trajectories were followed for periods of as long as two weeks preceding a given rain in order to establish the origin of the air masses associated with that rain. Without exception, the trajectory data indicated a polar air source for the older debris and a tropical air source for the fresher debris. The persistence of the distinction for nearly three months demonstrates that early Hardtack debris must have mixed to arctic latitudes and that later Hardtack debris was restricted to lower latitudes. It also indicates that mixing of particulate debris across the polar front is slow as compared to the washout rate.

Analysis of the trajectories of air masses associated with individual rains for other periods is helpful in clarifying additional features of the isotope-ratio data. Thus, the June 1958 ratio data (Fig. 1) can be explained as (i) an initial rise due to the first appearance of Hardtack test activity in the tropical troposphere; (ii) a marked decrease in the Ba^{140}/Sr^{90} ratio during mid-June, due to a transition to rains associated with polar air; and (iii) a subsequent rise due to the arrival of Hardtack test activity in the polar troposphere.

Strontium-89 to Strontium-90 Ratios

The Sr^{89} activity in rainfall during periods of well-spaced nuclear tests provides the best basis for estimating the Sr^{90} contribution of the most recent series of tests to the total measured Sr^{90} activity. The accuracy of such estimates is limited by uncertainties in production dates and fission production ratios, errors in radiochemical analysis, and possible fractionation effects. With measurements of good quality for periods of well-spaced, high-yield nuclear tests, however, it is unlikely that the combined uncertainties will exceed a factor of 2. It is shown below that an uncertainty of this magnitude does not seriously affect the usefulness of data on Sr^{89} and Sr^{90} activity as indicators of the influence of latitude and altitude on the behavior of the stratospheric source.

Stewart (*3*) has provided data on Sr^{89} and Sr^{90} activity for rainfall collected at Milford Haven, Wales, over the period from April 1954 to April 1957. High-yield tests conducted during this period, together with the estimated strength of the stratospheric source in megatons of fission, are listed in Table 1. For the well-spaced Castle, fall 1955 Soviet, and Redwing tests, the Milford Haven data make it possible to distinguish behavior differences for stratospheric Sr^{90} sources of different location in space. When 170 is taken as the Sr^{89}/Sr^{90} production ratio and the midpoint of the test series is taken as the production date, the Sr^{89}/Sr^{90} activity ratios of Milford Haven rains for February to July 1956 identify about one-half of the Sr^{90} activity with the Soviet tests of the fall of 1955. Similarly, essentially all of the Sr^{90} for September to December 1956 is associated with Redwing test production. For the 18-month period following the Castle tests, the assumption that all Sr^{90} in Milford Haven rains was due to these tests can lead to only a small overestimation of the Castle contribution.

Figure 2 shows the Sr^{90} concentration of Milford Haven rains per megaton of stratospheric contamination following the Castle, Redwing, and 1955 Soviet tests. Each data point represents the Sr^{90} rainfall concentration divided by the stratospheric contamination in megatons (from Table 1) for the given test series. After each of the Redwing and 1955 Soviet tests, the data on Sr^{90} concentration were taken only for rains with a Sr^{89}/Sr^{90} ratio indicating a 50- to 100-percent Sr^{90} contribution from that test series. The results are very striking, showing an intensity of stratospheric Sr^{90} fallout soon after the 1955 Soviet tests which is 10 times that for Redwing and nearly 60 times that for Castle.

The large differences in north temperate latitude fallout intensities estimated for these tests can be attributed to several factors: (i) possible overestimation of the stratospheric component for equatorial tests; (ii) the degree of mixing of equatorial test debris into the Southern Hemisphere; (iii) differences in stratospheric storage times; and (iv) more selective deposition of Soviet test debris in north temperate latitudes. W. H. Langham (*6*) suggests that stratospheric injection of debris from surface-water shots may be only 30 percent. Use of this value would reduce the estimates of stratospheric contribution for Redwing and Castle given in Table 1 by factors of

Table 1. Estimated strength of stratospheric source for high-yield nuclear tests.* PPG, Pacific proving ground (includes Eniwetok and Bikini atolls).

Test series	Period	Location	Source strength (10^6 tons of fission)†
U.S. (Ivy)	Nov. 1952	PPG (11°N, 166°E)	1.4
U.S. (Castle)	Mar.–May 1954	PPG (11°N, 166°E)	20
U.S.S.R.	Aug.–Nov. 1955	(~ 52°N)	1.8
U.S. (Redwing)	May–Jul. 1956	PPG (11°N, 166°E)	6.7
U.S.S.R.	Aug.–Nov. 1956	(~ 52°N)	2.7
U.S.S.R.	Jan.–Apr. 1957	(~ 52°N)	2.7
U.K.	May–June 1957	Christmas Island (2°N, 157°W), Malden Atoll (4°S, 155°W)	
U.S.S.R.	Aug.–Dec. 1957	(~ 52°N)	5.3
U.K.	Nov. 1957	Christmas Island (2°N, 157°W)	
U.S.S.R.	Feb.–Mar. 1958	(~ 52°N), Novaya Zemlya‡ (75°N, 55°E)	3.3
U.S. (Hardtack I)	May–Jul. 1958	PPG (11°N, 166°E)	4.0
U.S.S.R.	Oct. 1958	Novaya Zemlya‡ (75°N, 55°E)	12.5–15
Total			~ 62

* U.K. tests of 3 high-yield devices, April and September 1958, not tabulated. See K. Telegadas (*6*). †The stratospheric component, based on stratospheric inventory estimates of Libby (*12*), assumes stratospheric injection of 20 percent for surface land shots, 80 percent for surface water shots, and 100 percent for air shots. ‡(*23*).

1.8 and 2.1, respectively. Assuming confinement of Soviet test debris to the Northern Hemisphere and equal partitioning of equatorial test debris between hemispheres, one could account for a factor of 2 of the difference in the values between Soviet and equatorial tests. Since most equatorial tests were conducted at 11° north latitude, however, a somewhat greater contribution to the Northern Hemisphere may be expected for them. Storage time and latitude selectivity factors, therefore, must account for most of the difference in the fallout intensities between Soviet and equatorial tests (Fig. 2). The factor of 6 difference between the values for Castle and for Redwing appears to be accounted for best on the basis of longer stratospheric storage times associated with greater cloud heights for the higher-yield Castle test shots.

The results are not inconsistent with mean stratospheric storage times of a few months or more for Soviet test debris in the polar stratosphere, 1 to 3 years for debris in the lower equatorial stratosphere, and 5 to 10 years for the debris at higher levels near the equator. In view of the marked influence of altitude and latitude on the storage time and the transport and distribution pattern of stratospheric fallout, the concept of a mean global stratospheric residence time is not applicable to the interpretation of stratospheric fallout.

Tungsten-185 Concentration

A substantial quantity of the W^{185} radioisotope was produced in the U.S. Hardtack tests during the period May through July 1958. Most of the W^{185} radioactivity was produced in several surface explosions of sufficient energy to thrust the debris into the lower stratosphere. A total production of 250 megacuries of W^{185}, corrected for decay to 1 August 1958, has been reported (*12*). An estimated 40 percent of the total was retained in the stratosphere, with most of the remainder deposited as local fallout. The stratospheric W^{185} component has afforded a unique opportunity to follow the fate of debris originating from the lower stratosphere near the equator for a single test series.

The concentration of W^{185} in individual rains collected at Burlington, Vermont, and Bedford, Massachusetts, is presented in Fig. 3. The W^{185} data are corrected for decay to 15 June 1958, an arbitrary date about midway through the production period. The estimated errors in W^{185} activity determination are shown, with large errors only for rain samples of low total W^{185} activity.

The W^{185} data exhibit a rapid rise over the May–July 1958 production period. The stratospheric origin of Hardtack Ba^{140} in New England rains during July to September 1958 (Fig. 1 and the related discussion) testifies to the concurrent stratospheric origin of the W^{185} activity. Except for a drop in December 1958 and an appreciable rise in April to June 1959, the mean W^{185} concentration, corrected for decay, remained approximately constant over the year that followed production. The scatter in the concentration data is due in part to the inverse relationship between rainfall intensity and isotope concentration, with the higher concentrations corresponding to the lighter rains (for examples, see *3* and *19*).

For several short periods, the data

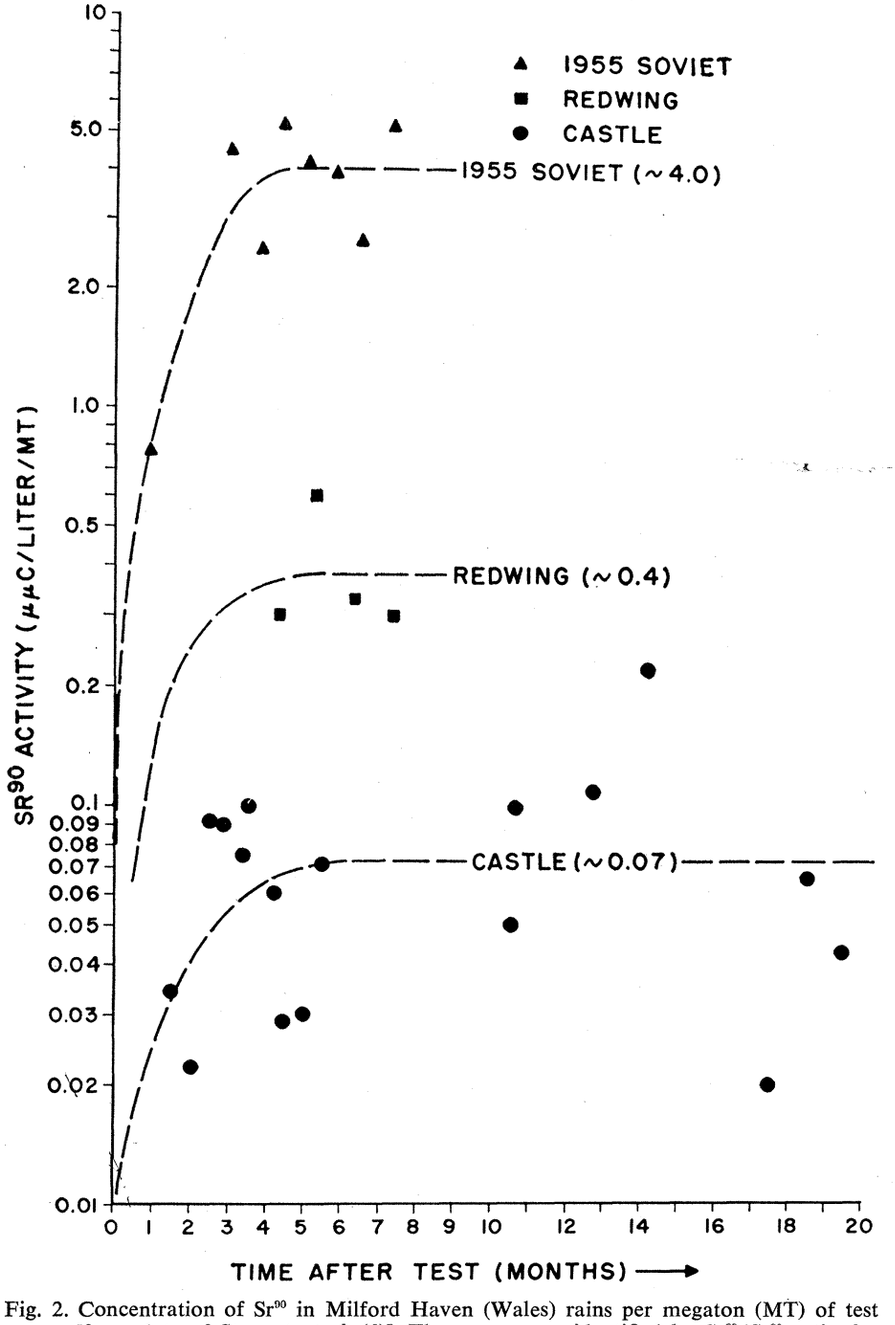

Fig. 2. Concentration of Sr^{90} in Milford Haven (Wales) rains per megaton (MT) of test source [from data of Stewart *et al.* (*3*)]. The source was identified by Sr^{89}/Sr^{90} ratio for well-spaced tests as follows: U.S. (Castle), March–May 1954 (11°N, 166°E); U.S.S.R., August–November 1955 (~ 52°N); and U.S. (Redwing), May–July 1956 (11°N, 166°E). The estimated strength of the stratospheric source, in megatons of fission, for each of these high-yield test series is given in Table 1.

Table 2. Rainfall data for Pôrto Alegre, Brazil.

Collection period	Rainfall (in.)	Specific activity on mean collection date (disintegration/min. per lit.)			Activity ratios corrected to 15 June 1958	
		Sr^{89}	Sr^{90}	W^{185}	Sr^{89}/Sr^{90}	W^{185}/Sr^{90}
1958:						
4 Jul.–1 Aug.	1.5	279 ± 19	4.63 ± 0.35	224 ± 19	91 ± 10	65 ± 8
1 Aug.–9 Aug.	4.4	242 ± 13	2.40 ± 0.34	191 ± 7	201 ± 30	131 ± 19
27 Aug.–5 Sept.	2.5	535 ± 240	5.30 ± 0.50	234 ± 8	274 ± 126	90 ± 9
6 Sept.–15 Sept.	4.6	94.4 ± 6.6	1.84 ± 0.20	80 ± 2	156 ± 22	97 ± 11
15 Sept.–30 Sept.	0.8	240 ± 21	2.45 ± 0.38	86 ± 14	350 ± 63	88 ± 20
25 Oct.–19 Nov.	1.4	162 ± 14	4.84 ± 0.72	105 ± 9	223 ± 39	84 ± 15
20 Nov.–2 Dec.	2.3	45.0 ± 3.5	2.73 ± 0.30	60 ± 6	138 ± 19	100 ± 15
2 Dec.–18 Dec.	4.0	33.5 ± 1.9	2.35 ± 0.19	41.9 ± 2.0	143 ± 14	94 ± 9
1959:						
22 Jan.–28 Jan.	1.7	14.9 ± 2.6	1.67 ± 0.11	19 ± 2	159 ± 30	87 ± 11
28 Jan.–30 Jan.	2.5	9.5 ± 2.2	0.96 ± 0.09	9.6 ± 1.9	187 ± 47	80 ± 17
30 Jan.–7 Feb.	1.7	19.0 ± 3.9	1.34 ± 0.16	25.5 ± 1.3	283 ± 67	159 ± 22
14 Feb.–25 Feb.	2.6	6.6 ± 1.9	0.72 ± 0.12	8.5 ± 0.8	236 ± 79	118 ± 23
26 Feb.–7 Mar.	0.7	19.2 ± 5.3	2.15 ± 0.22	12.0 ± 1.3	260 ± 77	62 ± 10
Average					208 ± 48	97 ± 17

of Fig. 3 show a gradual and almost regular transition in concentration for successive individual rainfalls. In an attempt to explain this observation, trajectories were plotted for the air masses associated with each rain. The method was identical to that described for the Ba^{140}/Sr^{90} activity-ratio data. The rains were classed as polar or tropical, in accordance with the indicated origin of the associated air mass. The mean monthly W^{185} concentrations of rains in each category are plotted in Fig. 4. The number of rains and the fraction of the monthly precipitation represented are noted beside each data point. Rains associated with air masses of doubtful origin were omitted.

The data on W^{185} concentration for tropical rains (Fig. 4) show only a moderate variation with time after initial build-up, suggesting an approximately uniform rate of downward mixing from the stratosphere into the tropical troposphere. The observed changes in concentration can be attributed, at least in part, to differences in rainfall intensity. Thus, lower concentrations are observed for heavy rains like those of September 1958 and July 1959 and higher concentrations for light rains like those of April and May 1959. The general decrease with time after initial build-up represents a gradual depletion of the stratospheric reservoir of W^{185}.

For rains of polar origin, however, the W^{185} concentration continues to build up until the spring of 1959 and then falls off more sharply in the summer of 1959. The slow build-up reflects the rate of mixing into the polar stratosphere as well as time variations in the rate of downward mixing at polar latitudes. The low W^{185} concentration of polar precipitation in December 1958 may be explained on the basis of reduced downward mixing due to a drop in height of the polar tropopause at this time. The very high concentration of W^{185} in polar rains in January, April, and June, 1959, may be explained partly by differences in rainfall intensity but may also be indicative of intensified downward mixing or subsidence of polar air. The large changes in relative W^{185} concentration in polar and tropical rains reinforce the conclusion drawn from the Ba^{140}/Sr^{90} data, that downward mixing is taking place from two regions of the stratosphere, separated in latitude. The remarkable reduction in W^{185} concentration in polar rainfall after June 1959 may be due either to virtually complete removal

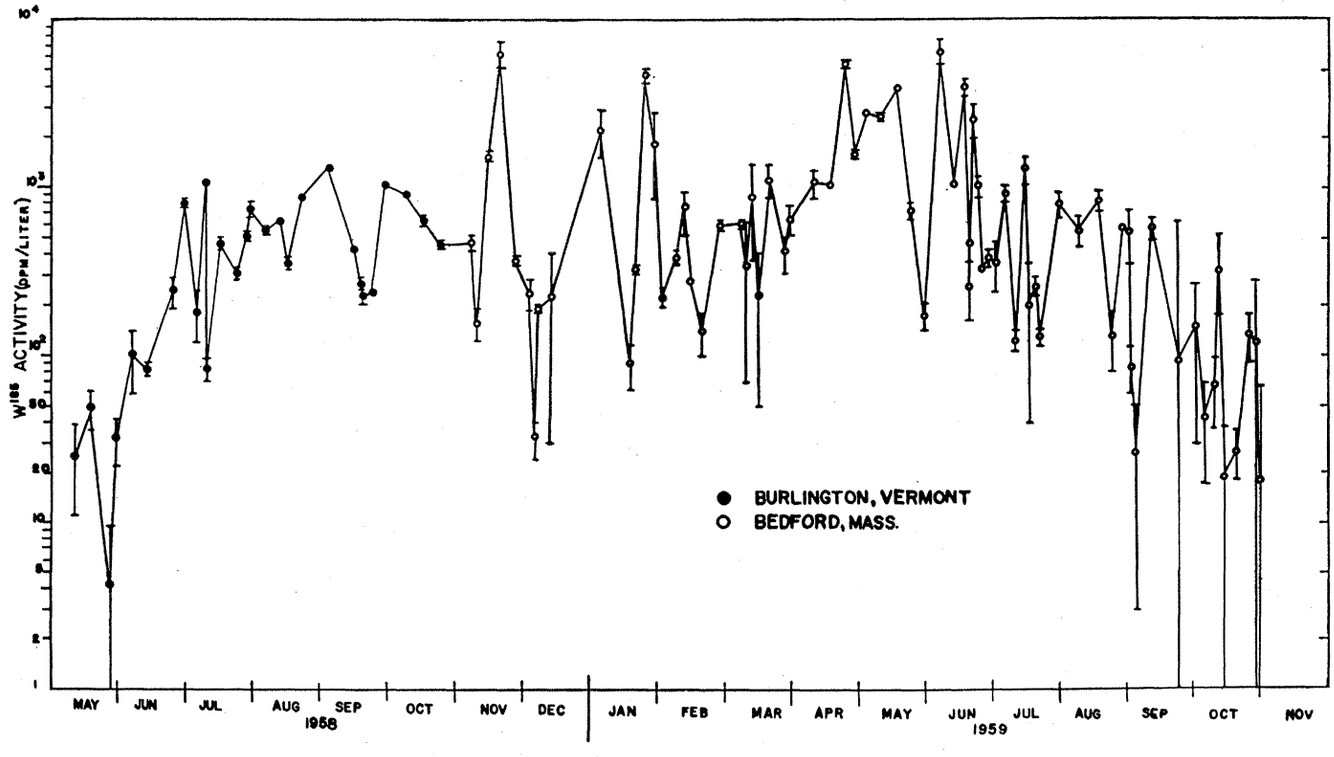

Fig. 3. Concentration of W^{185} in individual New England rains. Data are corrected for decay to 15 June 1958, assumed as the mean production date for the U.S. Hardtack tests.

of W^{185} activity from the polar stratosphere or to inhibition of downward mixing at this time.

It should be noted that the differences in W^{185} concentration observed for polar and tropical rains are not a direct measure of relative concentration in air. For a given concentration of particulate debris in air, higher corresponding concentrations in rainfall are to be expected in polar latitudes than in tropical latitudes, because polar rain clouds have a lower liquid-water content.

Origin of Recent Strontium-90 Fallout

Using W^{185} as a measure of Sr^{90} from the May to July 1958 Hardtack tests and Sr^{89}/Sr^{90} activity-ratio data for the period that followed the October 1958 Soviet tests to distinguish Sr^{90} fallout from that source, one should be able to distinguish the approximate contributions of Sr^{90} fallout from these two sources and from combined older sources. Data for radioactivity concentration in rainfall collections at Pôrto Alegre, Brazil, for an extended period following the Hardtack tests are presented in Table 2. The Sr^{89}/Sr^{90} data clearly indicate that most of the Sr^{90} fallout over the period of observation came from the 1958 U.S. and U.K. equatorial tests. The few large deviations in the Sr^{89}/Sr^{90} ratio from the average for the period must be due principally to the influence of tests conducted earlier or later than the assumed reference production date of 15 June 1958. The Sr^{90} data for closely spaced rains show the inverse relation between concentration of radioactivity and amount of rainfall. The gradual reduction in mean Sr^{90} concentration with time over the period of observation is attributed to the depletion of recent test debris from the lower layers of the stratosphere. In view of the evidence for an increase in stratospheric storage time with altitude for equatorial tests, the decrease in Sr^{90} concentration in Pôrto Alegre rains (Table 2) is not a reliable measure of the rate of depletion of the stratospheric reservoir.

The W^{185}/Sr^{90} data (Table 2) show a similar trend for Sr^{90} and Hardtack W^{185}. The observed W^{185}/Sr^{90} ratio of 97 ± 17 for the Southern Hemisphere, however, is substantially lower than would be expected from the reported Hardtack production data alone. Assuming that 40 percent of the reported total amount of W^{185} produced in the Hardtack tests and 0.4 megacurie of Sr^{90} from these tests was retained in the stratosphere, one obtains a value of 380 for the W^{185}/Sr^{90} ratio for the 15 June 1958 reference date. It is unlikely that uncertainties in production estimates account for much of the difference. Variations in W^{185}/Sr^{90} production ratios and in cloud heights for the individual Hardtack nuclear tests gave rise to initial differences in spatial distribution and may have resulted in uneven partitioning between hemispheres. The observed low W^{185}/Sr^{90} ratio for the Southern Hemisphere (Table 2) is attributed to the combined influence of these factors and to the Sr^{90} contribution of the 1958 U.K. equatorial tests.

The data on mean monthly Sr^{90} concentration in Bedford, Massachusetts, rainfall for 1958 and 1959 are presented in Fig. 5. These data are based on the Sr^{90} concentration of individual rains, weighted by precipitation amount. The two dashed curves connect points representing the estimated Sr^{90} contribution for the U.S. Hardtack tests and for the October 1958 Soviet tests, respectively. The Hardtack test contribution is estimated by assuming a W^{185}/Sr^{90} ratio of 380 for W^{185} activity corrected for decay to 15 June 1958. The October 1958 Soviet test component is based on the data on Sr^{89} activity. The total Sr^{89} activity was reduced by the estimated Hardtack Sr^{89} activity, based on the W^{185}-concentration data and the assumption that the W^{185}/Sr^{89} ratio was 380/170 in the Northern Hemisphere stratosphere at a reference date of 15 June 1958. This correction amounted to less than 10 percent except during June and August 1959, months for which corrections were about 10 and 20 percent, respectively. The Sr^{90} activity associated with the October 1958 Soviet tests was estimated from the corrected data on Sr^{89} activity, a Sr^{89}/Sr^{90} ratio of 170 at a

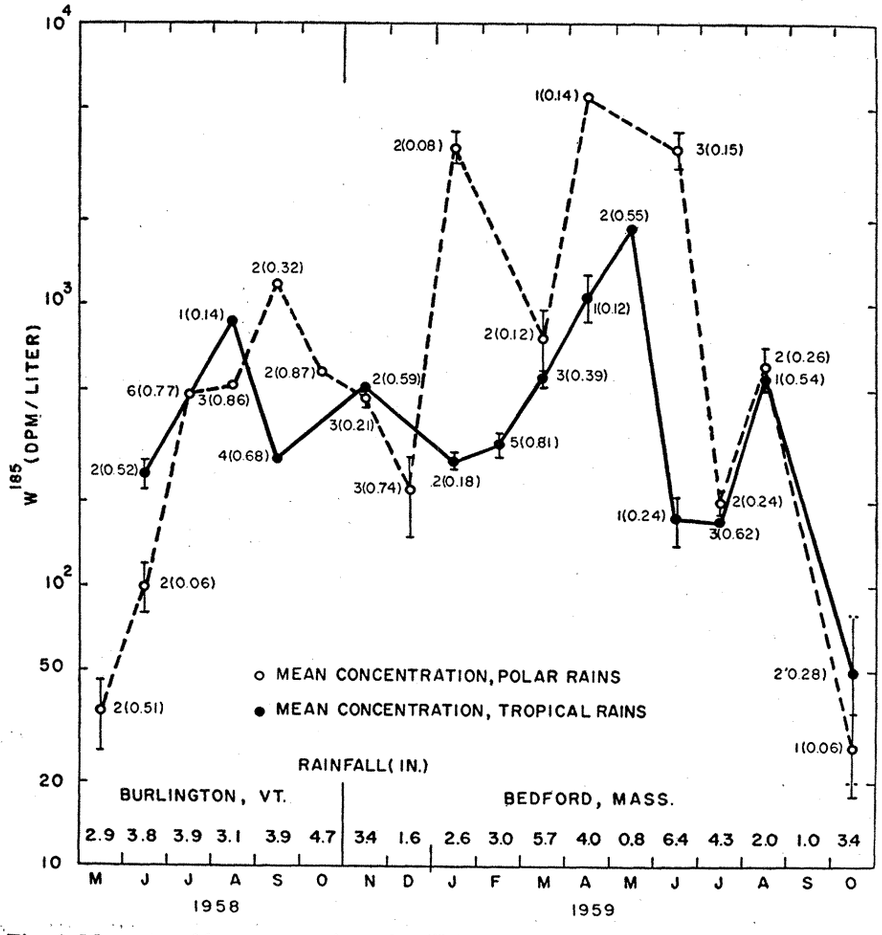

Fig. 4. Mean monthly concentration of W^{185} in New England rains classed by origin of air mass, from data in Fig. 3. Data are corrected for decay to 15 June 1958. Only individual rains for which air-mass trajectories indicated an unequivocal arctic or low-latitude source were used.

reference production date of 15 October 1958 being assumed.

It is clear that the heavy peak in Sr^{90} fallout over the first half of 1959 is due principally to the October 1958 Soviet tests. Immediately after these tests, the residual Sr^{89} activity from the Hardtack tests, as estimated from production amounts (Table 1) plus data on mixing between hemispheres and decay, accounted for less than one-thirtieth of the total Sr^{89} activity in the Northern Hemisphere stratosphere. For the first half of 1959, uncertainties due to errors in analysis in estimating the Sr^{90} component from the Soviet tests of October 1958 should not exceed 20 percent. A significant reduction in the assumed W^{185}/Sr^{90} ratio of 380 for Hardtack debris would account for more than the observed Sr^{90} concentrations. Similarly, it can be deduced from the Ba^{140}/Sr^{90} data (Fig. 1) and from Sr^{89}/Sr^{90} data that the heavy fallout during the first half of 1958 was due mainly to the Soviet tests of February–March 1958.

There is uncertainty about the Sr^{90} component from the Hardtack tests (Fig. 5) because of possible variations in the W^{185}/Sr^{90} ratio from the estimated value of 380. However, the curve accurately represents the relative mean monthly concentration of W^{185} in New England rains as determined from the individual-rainfall data of Fig. 3. The data show a marked increase in W^{185} concentration in Bedford rainfall nearly a full year after production. It is indicated (Fig. 4) that most of the increase is due to the much higher concentration in rains associated with arctic air. For New England, both the mean concentration in rainfall and the total deposition of W^{185} corrected for decay were nearly 2.5 times as high during the first six months of 1959 as they were during the last six months of 1958. By comparison, the concentration and deposition of debris from the Soviet tests of October 1958, on the basis of the analysis given in Fig. 5, was about 10 times as high in the first half of 1959 as in the latter half.

The dramatic removal in early 1959 of debris from these Soviet tests is attributed to intensified downward mixing or subsidence of air from the polar stratosphere in late winter. The spring peak for the decay-corrected concentration of W^{185} in rains associated with arctic air can be explained similarly. The very low concentration of both W^{185} and measured Sr^{90} in December 1958, a month in which all New England rains were associated with polar air, indicates a quite low rate of downward mixing at high latitudes at that time. The more rapid late-winter rise in rate of downward mixing of debris from the October 1958 tests as compared with the rate for W^{185} from the Hardtack tests may be due to differences in spatial distribution in the polar stratosphere.

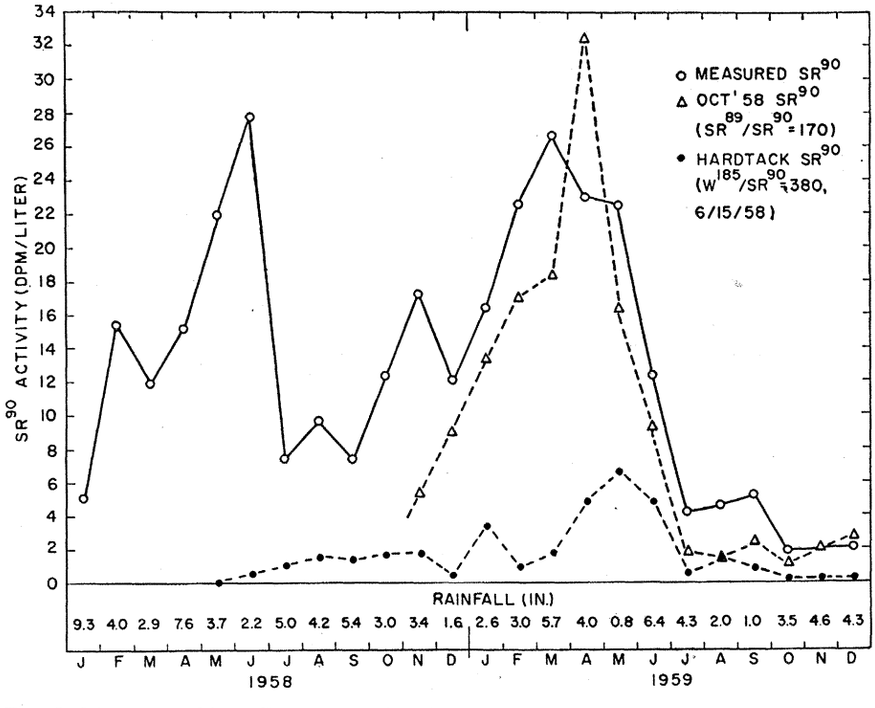

Fig. 5. Mean monthly Sr^{90} concentration in New England rains and contributions of the U.S. Hardtack (May–July 1958) and Soviet (October 1958) nuclear test series to Sr^{90} fallout. The Hardtack contribution of Sr^{90} is estimated by assuming $W^{185}/Sr^{90} = 380$ for W^{185} corrected for decay to 15 June 1958. The estimate of the Soviet October 1958 contribution is based on Sr^{89} data.

Summary

The foregoing discussion of fission-product ratios and concentrations of W^{185}, in which we have attempted to relate known sources of stratospheric debris to deposition in rains, has indicated a number of the complexities involved in the interpretation of atmospheric transport and deposition phenomena. The close spacing and variety of nuclear tests up to October 1958 and the fact that individual-radioisotope data of acceptable quality and type are limited have made it almost impossible to study the atmospheric behavior of individual nuclear-cloud sources. Even the W^{185} activity in equatorial latitudes was produced in a number of Hardtack tests of different date, yield, cloud height, and W^{185}/Sr^{90} ratio. Single, well-spaced nuclear explosions would offer a much-improved opportunity to study the physical aspects of fallout. Even in this case, however, the interpretation would be complicated by changes in the spatial distribution of the source with time under the influence of variable meteorological factors, and thus the results would have limited applicability at other seasons or in other years.

In spite of these complications and limitations, the isotope-ratio and W^{185} results have clarified a number of important points relative to the atmospheric transport of particulate debris from nuclear tests. The stratospheric origin of substantially all the short-lived, as well as the long-lived, radioisotopes in world-wide fallout appears to be well established for locations and periods for which reliable Ba^{140}/Sr^{90} and Sr^{89}/Sr^{90} activity-ratio data have been reported. The relative intensity of fallout per unit of strength of stratospheric source for different regions of the stratosphere shows a marked influence of latitude and altitude on fallout rates and fallout pattern. Debris injected into the lower regions of the polar stratosphere is substantially removed by subsidence or intensified mixing during the late winter and early spring. In this case, the storage time is a matter of months and depends on the time inter-

val between injection and removal. Storage times for debris in the equatorial stratosphere, however, are a matter of years and increase appreciably with altitude. Thus, the concepts of a well-mixed stratosphere and a mean stratospheric storage time appear largely inapplicable to the interpretation of stratospheric fallout.

Both the data on Ba^{140}/Sr^{90} ratio (Fig. 1) and the data on W^{185} concentration (Fig. 4) for rains classified in accordance with associated air-mass trajectory provide evidence for downward transport of debris from two regions of the stratosphere. The data on radioactivity indicate that downward mixing at high latitudes takes place to a limited extent throughout the year and is accompanied by a marked late-winter subsidence of the type described by Moser (*20*) and Dobson (*21*). Poleward movement of stratospheric air which must accompany this subsidence may account for the rapid appearance of early Hardtack debris in polar air (Fig. 1) and for the high W^{185} activity associated with polar air in the spring of 1959. The transfer of stratospheric debris into the temperate and tropical troposphere presumably takes place at middle latitudes near the subtropical jet. Downward mixing in this region continues throughout the year. The apparent seasonal variations in concentration of fallout in tropospheric air and rains at temperate and tropical latitudes to a large extent must be due to shifts in origin of the stratospheric air source and to changes in its concentration of radioactivity.

The sustained differences in isotope ratio and the large temporal changes in the relative concentrations of a given radioisotope in polar and in tropical rains indicate that tropospheric mixing of particulate debris across the polar front is inhibited. Thus, selective precipitation scavenging of radioactive fallout in frontal storms may be an important factor in explaining the selective deposition of fallout in north temperate latitudes. Seasonal distribution of rainfall, rainfall amount, average rainfall intensity, and the origin of the scavenged air are factors which markedly influence fallout accumulation. Areas receiving substantial rainfall in the spring, with a high proportion of light rains associated with arctic air, undoubtedly have had relatively high levels of fallout.

Thus, the marked differences in transport, distribution, and consequences for equatorial and high-latitude nuclear tests are becoming clearer. The rapid late-winter and early-spring removal of debris injected into the polar stratosphere leads to greater selectivity in the surface distribution. The corresponding short storage time makes the short-lived radioisotopes and their biological consequences more significant. The radioactive products of air bursts at high latitudes are distributed quantitatively in the form of widespread fallout within the hemisphere. Thus, the type, timing, and location of past Soviet tests have tended to maximize fallout in the heavily populated North Temperate Zone. The high proportion of surface explosions in the U.S. equatorial tests has resulted in deposition of the major fraction of the debris as local fallout. The resulting reduction in debris available for worldwide fallout, the more widespread distribution over both hemispheres, the longer residence in the stratosphere, and the relatively low seasonal and latitudinal selectivity of deposition have made fallout from equatorial tests substantially less significant per test unit than fallout from tests in other latitudes (*22*).

References and Notes

W. F. Libby, *Proc. Natl. Acad. Sci. U.S.* **42**, 365 (1956).

——, *ibid.* **43**, 758 (1957).

3. N. G. Stewart, R. G. D. Osmond, R. N. Crooks, E. M. Fisher, "The 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).
4. E. A. Martell, *Science* **129**, 1197 (1959).
5. "The nature of radioactive fallout and its effects on man," *Hearings before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy, May 27–29, June 3–7, 1957* (U.S. Government Printing Office, Washington, D.C., 1957), pts. 1 and 2.
6. "Fallout from nuclear weapons tests," *Hearings before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy, May 5–8, 1959* (U.S. Government Printing Office, Washington, D.C., 1959), vols. 1–3.
7. N. G. Stewart, R. N. Crooks, E. M. Fisher, "The radiological dose to persons in the U.K. due to debris from nuclear test explosions," *Atomic Energy Research Establ. (G. Brit.) Publ. No. AERE HP/R 1701* (1955).
8. L. Machta and R. J. List, "Meteorological interpretation of strontium-90 fallout," *U.S. Atomic Energy Comm. Health and Safety Lab. Publ. No. HASL-42* (1958), p. 327.
9. P. K. Kuroda, "On the stratospheric strontium-90 fallout," *Argonne Natl. Lab. Rept. No. ANL-5920* (1958).
10. L. Fry and P. K. Kuroda, *Science* **129**, 1742 (1959).
11. S. Katcoff, *Nucleonics* **16**, No. 4, 78 (1958).

W. F. Libby, *Proc. Natl. Acad. Sci. U.S.* **45**, 959 (1959).

13. L. E. Glendenin, "Determination of strontium and barium activities in fission," in "Radiochemical Studies: The Fission Products," *Natl. Nuclear Energy Ser., Div. IV* **9**, 1460 (1951); W. R. Collins, Jr., and G. A. Welford, "The radiochemical determination of tungsten," *U.S. Atomic Energy Comm. Health and Safety Lab. Publ. No. 58-3* (1958).
14. E. A. Martell, "The Chicago sunshine method: Absolute assay of strontium-90 in biological materials, soils, waters, and air filters," *U.S. Atomic Energy Comm. Rept. No. AECU-3262* (1956).
15. P. J. Drevinsky, C. E. Junge, I. H. Blifford, Jr., M. I. Kalkstein, E. A. Martell, "Natural aerosols and nuclear debris studies," *Geophysics Research Directorate Notes No. 8* (1958).
16. T. Sugihara, R. Wolfgang, W. F. Libby, *Rev. Sci. Instr.* **24**, 511 (1953).
17. A. D. Suttle and W. F. Libby, *Anal. Chem.* **27**, 921 (1955).
18. H. E. Suess, Interim Progr. Rept., project 40, Univ. of California (1958).
19. J. F. Bleichrodt, J. Blok, R. H. Dekker, C. J. H. Lock, *Tellus* **11**, 404 (1959).
20. H. Moser, *Ber. deut. Wetterdienstes No. 11* (1949), p. 28.
21. G. M. B. Dobson, *Proc. Roy. Soc. (London)* **236A**, 187 (1956).

22. This work was supported in part by the U.S. Atomic Energy Commission under contract AT (49-7)-1431. We are grateful to W. S. Hering and P. B. Storebo for helpful discussions of the meteorological aspects. The assistance of Miss N. A. Dimond and J. Pecci, who carried out many of the radiochemical analyses, is gratefully acknowledged. Some of the samples were assayed by Controls for Radiation, Inc., Cambridge, Mass., under contract AF 19(604)-5742. We are indebted to Dr. Jose Luiz Paranhos de Araujo, Sr., head of I.R.M. Coussirat Araujo, Pôrto Alegre, Brazil, who provided the Pôrto Alegre precipitation samples.

23. M. Baath, *Tek. Tidskr.* **89**, 765 (1959).