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Atmospheric Aspects of Strontium-90 Fallout

Fallout evidence indicates short stratospheric holdup time for middle-latitude atomic tests.

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The production of fission-product strontium-90 in nuclear weapon tests, its world-wide distribution in fallout, and its possible biological consequences are matters which have received increasing public attention over the past several years. The particular concern with Sr^{90} stems from its high fission yield, its long half-life of 28 years, and its chemical similarity to calcium, which it follows from the soil, through vegetation and dairy products, to its eventual incorporation into human bone.

The physical and biological distribution of Sr^{90} activity was initially investigated on a world-wide basis in studies carried out by the University of Chicago Sunshine Project (*1*). The results of this work and related studies have been extensively discussed by W. F. Libby (*2–5*). Methods of analysis and summaries of the experimental results have been presented elsewhere (*6–8*). For a comprehensive treatment of the general aspects of radioactive fallout, the reader is referred to two excellent summaries (*9–10*).

One of the important areas of uncertainty in our knowledge of Sr^{90} fallout is the atmospheric history of Sr^{90} debris and the consequent world-wide distribution of this debris. Experimental evidence (*2, 8*) has indicated that, aside from local fallout from surface bursts, debris from small nuclear weapons is circulated in the lower atmosphere, is confined to the hemisphere in which it was produced, and exhibits a mean atmospheric residence time of about 1 month. By contrast, the debris from weapons of megaton yield is largely injected into the stratosphere, exhibits a long stratospheric residence time, and may have world-wide distribution. The rate and extent of stratospheric mixing and the rate and mechanism of transport of stratospheric debris through the tropopause into the lower atmosphere are not completely understood. Details of the mixing and scavenging of the Sr^{90} debris in the troposphere have not been fully explained, although it has been well established (*3, 8, 11*) that precipitation plays the dominant role in the deposition of Sr^{90} debris.

In view of our limited knowledge of large-scale atmospheric circulation, and in view of the inherent experimental difficulties in the synoptic evaluation of either the atmospheric distribution or the surface distribution of Sr^{90} debris on a world-wide basis, the present stratospheric burden of Sr^{90}, the deposition rate and the consequent ground distribution of Sr^{90} can be only crudely estimated. It is the object of this article to review some of the experimental evidence relating to the nature and atmospheric history of Sr^{90} debris and to discuss a number of experimental uncertainties and special considerations which affect the interpretation of world-wide fallout.

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Production of Strontium-90

The half-life of Sr^{90} has been well established as about 28 years (*12, 13*). The corresponding fission yield of Sr^{90} from slow-neutron fission of U^{235} ranges from 5.4 to 5.8 percent (*13, 14*); the uncertainty with respect to the absolute value is 10 to 20 percent. When 1.0×10^{12} calories is taken as the energy equivalent of 1 kiloton of TNT and 5.6 percent, as the U^{235} fission yield of Sr^{90}, the production of Sr^{90} is 1.14 grams per kiloton, corresponding to 146 curies per kiloton. Yields of Sr^{90} will be substantially lower for fissionable materials of mass greater than 235 and will be further modified by the neutron energy spectra of nuclear weapons, with reduced yields for higher neutron energies. The amount of Sr^{90} produced by nuclear weapons is usually taken as 100 curies per kiloton of fission energy release. It is unlikely that the uncertainty in this value exceeds 30 percent.

The Sr^{90} activity is produced in the mass-90 fission chain as follows:

$$Kr^{90} \xrightarrow{33\ sec} Rb^{90} \xrightarrow{2.7\ min} Sr^{90} \xrightarrow{28\ yr} Y^{90} \xrightarrow{64.6\ hr} Zr^{90}(stable)$$

Estimates of the most probable charge distribution for U^{235} thermal-neutron fission (*13*) indicate high independent yields only for Kr^{90} and its very short-lived precursor, Br^{90}, with independent yields of Rb^{90}, Sr^{90}, and Y^{90} contributing only about 15, 0.1, and 10^{-4} percent of the total chain yield, respectively. The distribution of independent yields for the mass-90 chain for nuclear weapons will vary somewhat with mass of fissioning nuclide and with neutron energy.

Adams, Farlow, and Schell (*15*), in a detailed discussion of bomb debris con-

densation and of the physical and chemical characteristics of local fallout particles, give evidence for the occurrence of spherical radioactive particles as small as a few tenths of a micron in diameter for surface shots over coral and of similar particles as small as 0.01 micron in diameter for surface shots over sea water. They also give evidence for the isotopic enrichment of Ba^{140} and Sr^{89} on fallout particles which condense after a relatively long time interval has elapsed.

For weapons of low yield, independent yield distribution and condensation rate clearly indicate that Sr^{90} and other daughter products of volatile and gaseous fission products condense later than most of the vaporized materials in the fireball. For high-yield shots the times of debris condensation and Sr^{90} formation are more nearly comparable, and less fractionation of gaseous precursor products is to be expected. However, in this case condensation takes place at stratospheric altitudes following extensive cloud dilution, and this leads to the formation of particles less than 1 micron in diameter. The long stratospheric storage time that is observed for stratospheric debris is perhaps the most convincing evidence that the particles are of submicron size. It is indicated that Sr^{90} produced in weapon tests is to a large extent concentrated in very fine particles which can remain airborne for sufficiently long periods to make extensive distribution over the world possible.

The delayed condensation of Sr^{90} suggests that, except possibly in the case of high-yield surface bursts over silicate soils, the chemical form of the Sr^{90} activity is strontium oxide. During its subsequent circulation in air, the strontium oxide would be readily converted to the hydroxide by the action of atmospheric water vapor. Exposure to atmospheric carbon dioxide would result in slow, partial conversion of the hydroxide to carbonate. Experimental verification of the chemical nature of Sr^{90} debris would be complicated by the attachment of the very small Sr^{90} particles to larger particles of bomb debris and to natural particulate and liquid aerosols in the atmosphere. It has been observed (*8*) that the Sr^{90} activity deposited in rains is substantially all in water-soluble form and thus is generally available for uptake in the biosphere.

Libby (*5*) has provided estimates of the cumulative amount of Sr^{90} introduced into the stratosphere and troposphere for all United States, United Kingdom, and Soviet nuclear tests up to December 1957. Total stratospheric injection is estimated (*5*) as 3.6 megacuries of Sr^{90}, about two-thirds of this amount resulting from United States tests in the Pacific and one-third from Soviet tests. In addition, nearly 0.6 megacurie is estimated to be the total distributed as tropospheric debris. These estimates were made on the assumption of a local fallout of 80 percent for surface land shots and 20 percent for surface water shots. The remaining 20 and 80 percent, respectively, was assigned to the stratosphere for shots with total energy release of 1 megaton or more and to the troposphere for smaller shots.

Such estimates are subject to the several uncertainties with respect to Sr^{90} fission yield discussed above, and uncertainties in estimation of the total fission energy release for the various nuclear devices tested and in the determination of the fraction of Sr^{90} production which falls out locally. Obtaining essential information about Soviet nuclear tests and determining the local fallout fraction for high-yield surface shots in the Pacific test areas entail obvious difficulties. Estimates of the total amount of Sr^{90} which has been distributed as world-wide contamination may, therefore, be in error by as much as a factor of 2.

Fallout Models

Several alternative interpretations of Sr^{90} fallout observations have been proposed, by Libby (*2, 3, 5*), Stewart *et al.* (*11, 16*), and Machta (*10, 17*), respectively. The principal features of the experimental data which require explanation are the strong peak in cumulative Sr^{90} deposition in the middle latitudes of the Northern Hemisphere and the sharp increase in Sr^{90} deposition rate which has been observed in the spring in each of the past several years.

According to Libby's model (*2, 3, 5*), Sr^{90} debris which is injected into the stratosphere is uniformly mixed over both hemispheres in the first year or two following injection and exhibits an average storage time of 10 ± 5 years. Libby proposes that stratospheric debris will be distributed nearly uniformly over the world except for differences due to variations in rainfall. Tropospheric debris from submegaton weapons, distributed in a narrow latitude band for each test site, is thought to account for both the high mid-latitude deposition and the observed spring peaks in Sr^{90} fallout.

Stewart *et al.* (*11*) conclude from measurements of the ratio of Sr^{89} to Sr^{90} in rains that since 1954 nearly all Sr^{90} deposited at places remote from test sites has been derived from delayed fallout of stratospheric debris. They also indicate that the seasonal variation in concentration of Sr^{90} in rain is in step with a similar variation in the concentration of the lower stratosphere. These authors suggest that the results are consistent with the circulation model proposed by Brewer (*18*) and Dobson (*19*).

Brewer explains the observed distribution of water vapor in the lower stratosphere as being the result of air entering the stratosphere near the equator, moving northward, and sinking into the troposphere in temperate and polar regions. Dobson explains the spring rise of ozone concentration in the lower atmosphere at high latitudes as being due to the sinking of a cold air mass which forms above the winter pole during the late winter. Stewart and his associates attribute the high peak of Sr^{90} deposition in the middle latitudes of the Northern Hemisphere to a Brewer mode of circulation with selective downward mixing of stratospheric debris at middle latitudes. The spring peak in the Sr^{90} deposition rate is attributed to Sr^{90}-rich air at higher stratospheric levels, carried down in the same manner as ozone.

Machta (*10, 17*) has discussed some of the meteorological factors involved and has raised a number of objections to both proposed models. He favors a shorter stratospheric residence time of about 5 years and, like Stewart, attributes the high level of deposition in middle latitudes of the Northern Hemisphere and the spring increase in deposition rate to stratospheric fallout, considering selectivity with respect to latitude and season of deposition to be due to meteorological factors.

An alternative interpretation of the Sr^{90} fallout observations is presented below, following discussion of some of the principal features and limitations of the experimental evidence.

Concentrations of Strontium-90 in Air

A number of large-volume surface air samples from several Northern Hemisphere locations were collected by the Naval Research Laboratory and analyzed for Sr^{90} at the University of Chicago (*8*). Measurements were made of collections from Washington, D.C.; Kodiak, Alaska; Port Lyautey, French Morocco; and Yokosuka, Japan. The results are summarized in Fig. 1. Each point of Fig. 1 represents the volume-weighted average of only several days' collection dur-

ing any one month at each location and thus may not provide a very reliable measure of average monthly air concentration. For locations other than Washington, D.C., the data may be in error by as much as 50 percent, due principally to uncertainty with respect to volume. The relative concentration values at each location are reasonably accurate.

These data exhibit several interesting features. There is a roughly exponential rise in concentration of Sr^{90} over the 4-year period of observation and a general equivalence in range of Sr^{90} concentration at all four locations. The 1952 data for Kodiak, Alaska, show contribution from the spring 1952 Nevada tests, followed by complete absence of Sr^{90}—a typical finding for contamination that is purely tropospheric. The data for the period 1953 through 1955 for all four locations demonstrate the build-up of delayed fallout from the stratosphere following the November 1952 Ivy test and the spring 1954 Castle tests, with some superimposed contribution of tropospheric debris from intermittent small-weapon tests. The higher concentrations observed for Washington and Port Lyautey during the spring of 1955 may be due to the spring 1955 Nevada tests or to other production or circulation factors.

Data for total fission-product beta activity for surface air at Washington, D.C., furnished by I. H. Blifford, Jr. (*20, 21*), are presented in Fig. 2, together with data on Sr^{90} concentration for the same location. The plotted fission-product activity data have not been corrected for decay. Mixed fission products decay exponentially with time, with approximately an order of magnitude decrease in activity corresponding to a sevenfold increase in age of the fission products. Thus, the observed fission-product activity levels are quite sensitive to considerations of production date and rate of arrival. The peak fission-product concentrations shown in Fig. 2 provide, with this complication, a history of tropospheric contamination from small-weapon tests. Peak heights for a given test area reflect tropospheric injection amounts, modified substantially by tropospheric circulation and mixing factors. The lower peak heights for Russian tests as compared with Nevada tests are due to greater decay and dilution due to mixing, consistent with later arrival times. The very low concentrations observed following high-yield United States tests at the Pacific Proving Grounds reflect the limited extent of early fallout contamination from these tests.

When the steep slope of the fission-product activity curve following small-weapon tests is corrected for decay, the curve provides a measure of the dilution and removal rate for tropospheric contamination. Using similar data for tropospheric air samples and assuming virtually complete lateral and vertical diffusion several weeks after production date, Stewart, Crooks, and Fisher (*16*) have estimated a mean tropospheric residence time of about 1 month. Appreciable continued lateral diffusion would correspond to longer residence times. Considerations of particle size and meteorological factors which affect diffusion and washout suggest the tropospheric residence time may be quite variable.

The data for fission-product activity given in Fig. 2 indicate essentially complete removal of tropospheric contamination between tests before November 1952. Following the November 1952 Ivy test and the spring 1954 Castle test, the minimum mixed fission-product concentrations observed can be attributed to the slow downward mixing of fission products from the stratosphere. Curve *A* (Fig. 2) is approximately fitted to the data and represents the slow mixing of Operation Castle stratospheric contamination into surface air at north temperate latitudes. Curve *B* is similarly constructed, the same buildup with time that followed Ivy and Castle being assumed and the Ivy contribution being based on the January and February 1954 fission-product activity data. Comparison of curves *A* and *B* indicates the ratio of the post-Castle and post-Ivy stratospheric inventories to be about 5 or 6 to 1. When correction for decay is made for curve *A*, the transport of Castle debris to surface levels at latitude 39° north is shown

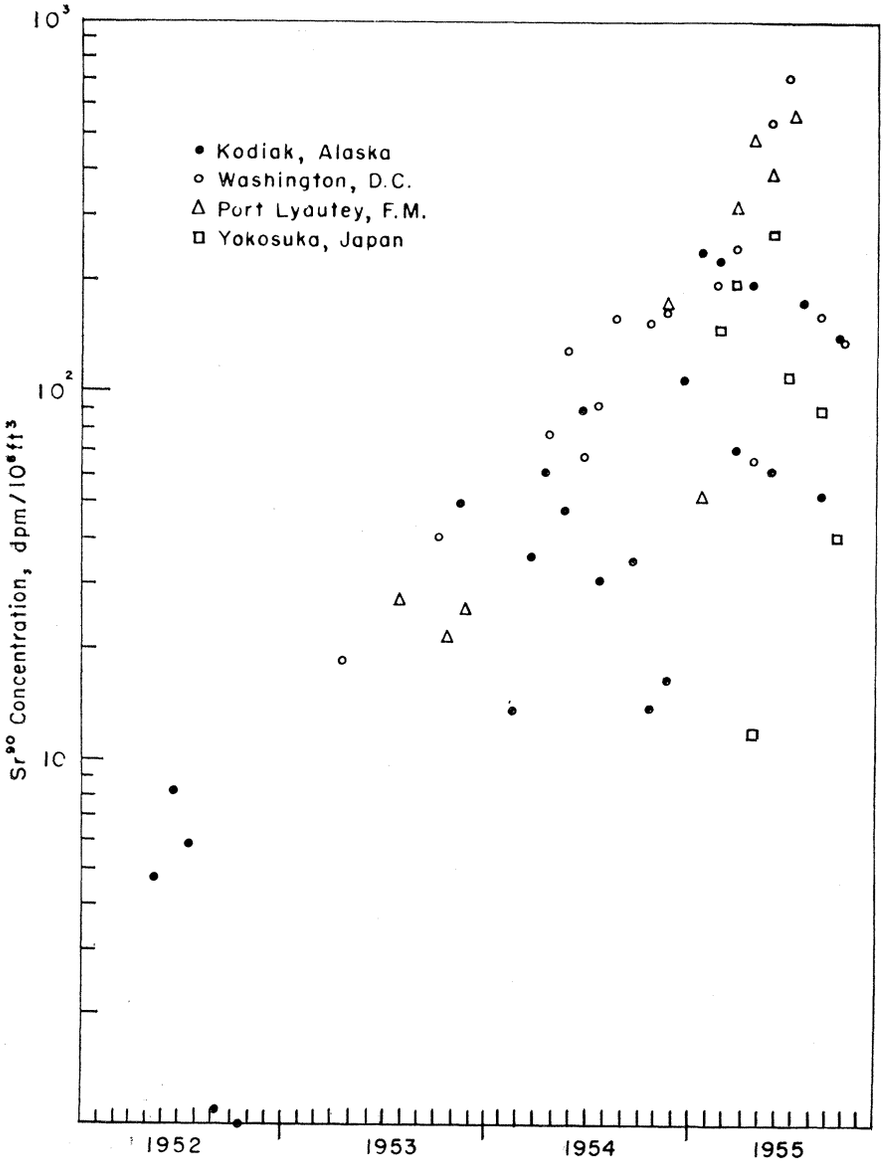

Fig. 1. Concentration of Sr^{90} in surface air.

to be negligible in the first three months and, subsequently, to increase continuously over a period of about one year.

The data for concentration of Sr^{90} in air for Washington, D.C. (Fig. 2, curve *C*) are higher by factors of 2 to 3 than Sr^{90} values computed from the resolved stratospheric component of fission product activity data (Fig. 2, curve *A*). The difference equals or exceeds the combined uncertainties in sampling and analysis. This result lends credence to the separation of the fission product data (Fig. 2) into tropospheric and stratospheric components, since the discrepancy can be reduced only by an upward revision of the stratospheric contribution (curves *A* and *B*).

The resolution of stratospheric and tropospheric components of the fission-product beta-activity data (Fig. 2) makes it possible to estimate the contribution of tropospheric contamination to total Sr^{90} fallout during this early period. If average tropospheric contamination arrival times at Washington, D.C., are assumed to be 1 week for Nevada tests and 2 weeks for Soviet tests, and if 30 days is taken as the half-time of removal due to further dilution and deposition, it is estimated that tropospheric fallout accounts for less than 50 percent of the total Sr^{90} fallout in 1953 and for less than 10 percent of that in 1954 and the first 10 months of 1955.

Blifford *et al.* (*20*) have presented records of the average monthly fission-product beta activity of the surface air, for the period 1950 to 1955, for five locations in the Northern Hemisphere ranging from Subic Bay, Philippine Islands (14°45′N), to Kodiak, Alaska (57°30′N). These records show responses to Nevada and Soviet tests at all locations, indicating that tropospheric debris is very widely distributed over the hemisphere. Records for Subic Bay and for Pearl Harbor, Hawaii, show peak responses following the Ivy and Castle tests which do not exceed those observed in Washington, D.C., following Nevada tests. If the yield of the Pacific tests is assumed to be greater by about two orders of magnitude, it is evident that initial tropospheric contamination from multimegaton weapon tests, excluding local fallout, amounts to only about 1 percent of the total fission production.

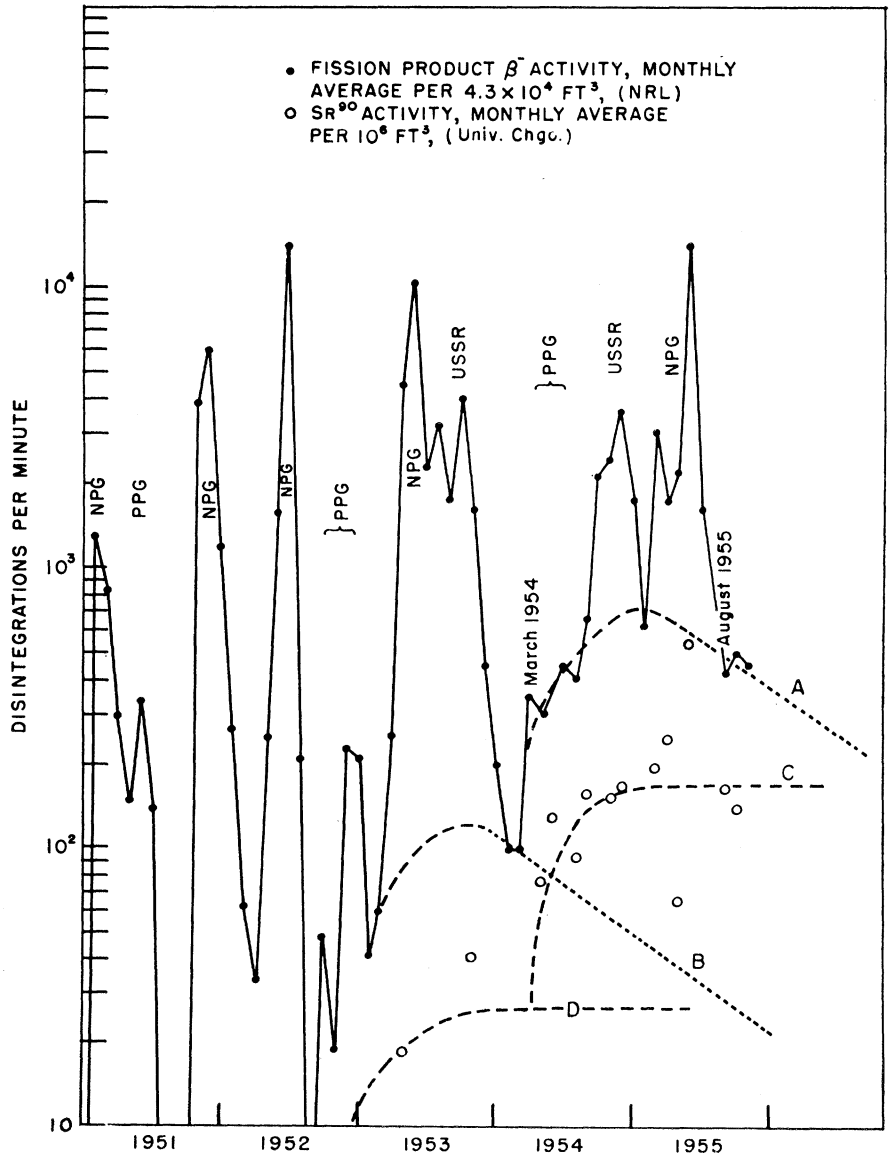

Fig. 2. Fission-product-β^- activity and Sr^{90} in surface air, Washington, D.C., from NPG, Nevada Proving Grounds; PPG, Pacific Proving Grounds; USSR, Soviet tests. Curves *A* and *B* represent the approximate stratospheric component of fission-product activity following the Castle and Ivy nuclear tests, respectively. Curves *C* and *D* represent the Sr^{90} concentration for corresponding periods.

Precipitation Data

The cumulative deposition of Sr^{90} in rains and snows at Chicago (*6–8*), Pittsburgh (*8, 22*), New York City (*22*), and Milford Haven (Wales) (*11*) is summarized in Fig. 3. The Chicago and Milford Haven deposition curves have been normalized in conformity with the observed cumulative burden of Sr^{90} of soils in the two areas. The Pittsburgh and New York City data are fitted approximately to the 1954 Chicago levels. Results for two New York City soils (*22*) are included for comparison. Altogether, these rainfall data provide a nearly complete history of the deposition of Sr^{90} in the middle latitudes of the Northern Hemisphere. Other areas at these latitudes which have similar fallout histories show considerable variation in amount of Sr^{90} accumulated, due principally to differences in annual rainfall. Other factors that affect Sr^{90} deposition include seasonal distribution of rainfall, type of rainfall, pattern of regional air circulation, and proximity to nuclear test sites.

The rainfall data (Fig. 3) indicate that only a small—perhaps a negligible —fraction of present levels of Sr^{90} fallout was deposited before the advent of large-scale nuclear tests. A substantial increase in annual Sr^{90} fallout occurred in each successive year up to the end of 1957. An important feature of the rainfall data is the sharp increase in Sr^{90} deposition rate in the spring—an increase which accounts for a large proportion of the total fallout for each year.

This spring increase has been attributed by Libby (*2, 5*) to tropospheric contamination from submegaton weapon tests; by Stewart *et al.* (*11*) and Machta (*17*), to more rapid downward mixing of stratospheric air at middle and higher latitudes in the spring. I submit the alternative view that the spring increases are due largely to Soviet tests of intermediate- and high-yield weapons which have injected debris into lower and intermediate levels of the stratosphere—debris which exhibits relatively short stratospheric residence times. I attribute the substantial spring rise that has occurred in each of the past 3 years principally to the immediately preceding fall and winter series of Soviet tests. Substantial evidence to support this interpretation is available and is discussed in subsequent sections of this article.

The wide range in the Sr^{90} burden observed for soils in northern hemisphere locations of equivalent annual rainfall and latitude for some time masked the dominance of rainfall as a factor in Sr^{90} deposition. The low Sr^{90} content of soils (*3, 8*) in low-rainfall areas was the first evidence that rainfall is the controlling factor. Brawley, California, with less than 2 inches of annual rainfall, held less than 0.6 millicurie of Sr^{90} per square mile in the topsoil in January 1956. This is less than one-tenth the levels concurrently observed in high-rainfall areas of similar latitude. At Antofagasta, Chile, where it almost never rains, the January 1956 Sr^{90} level in soil was 0.02 millicurie per square mile—about 0.01 of the concurrent level in high-rainfall areas of the Southern Hemisphere. Such evidence convincingly demonstrates that the gravitational settlement of dry particles is a negligible factor in the world-wide fallout of Sr^{90}.

Results for the Mediterranean area soils (Fig. 4) provide striking evidence of the dependence on rainfall of the Sr^{90} level in soils. The remarkable rainfall correlation for this set of soils was first pointed out by L. T. Alexander (*23*). These Mediterranean lands all receive the same seasonal distribution of rainfall, with a marked winter maximum and extremely dry summers. In general, the rainfall over the whole area is derived from air masses of similar mixing history. Similar correlation of total Sr^{90} deposition with total rainfall has been observed in other limited geographical areas (*24, 25*). It is indicated that within a given limited geographical region the average Sr^{90} concentration in rains will be approximately constant. It has also been shown (*8, 11*) that for individual rains at a given location, the specific concentration of Sr^{90} decreases with increasing amounts of rain. Thus, within areas showing total Sr^{90} deposition proportional to total rainfall, a location which received twice as much Sr^{90} fallout as another must have experienced about twice as many rains of the same general type and intensity.

The data given in Fig. 4 for the Southern Hemisphere show a roughly similar relationship between Sr^{90} deposition and total rainfall. It is quite evident from data (*2, 7, 8*) for South American soils and Antarctic snows that delayed stratospheric fallout has been almost exclusively the source of Sr^{90} fallout in the Southern Hemisphere. Two soils from Belo Horizonte, Brazil, held 0.11 and 0.17 millicurie per square mile, respectively, in March 1954. Nearby areas of

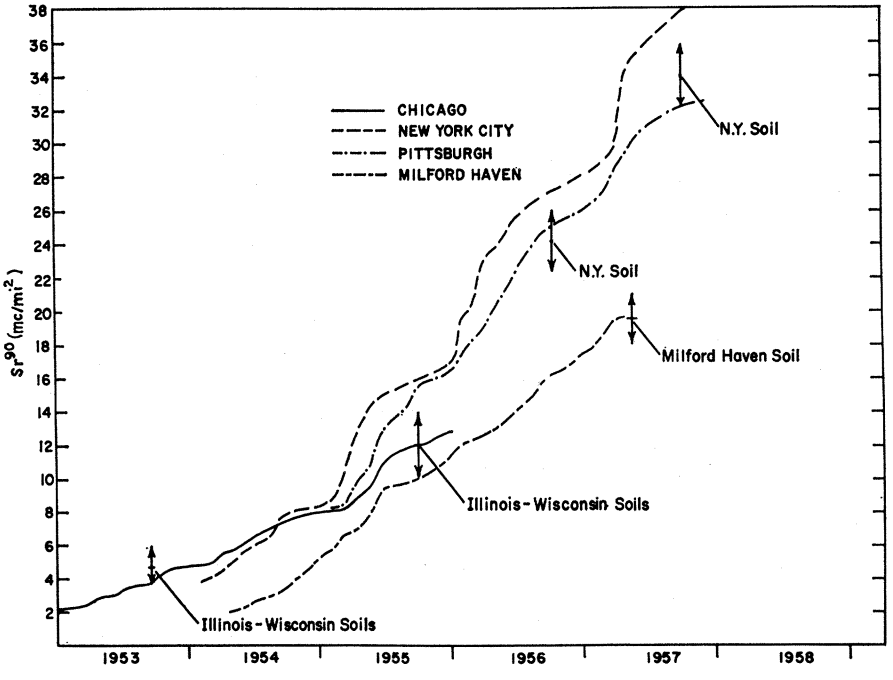

Fig. 3. Cumulative Sr^{90} deposition in rain.

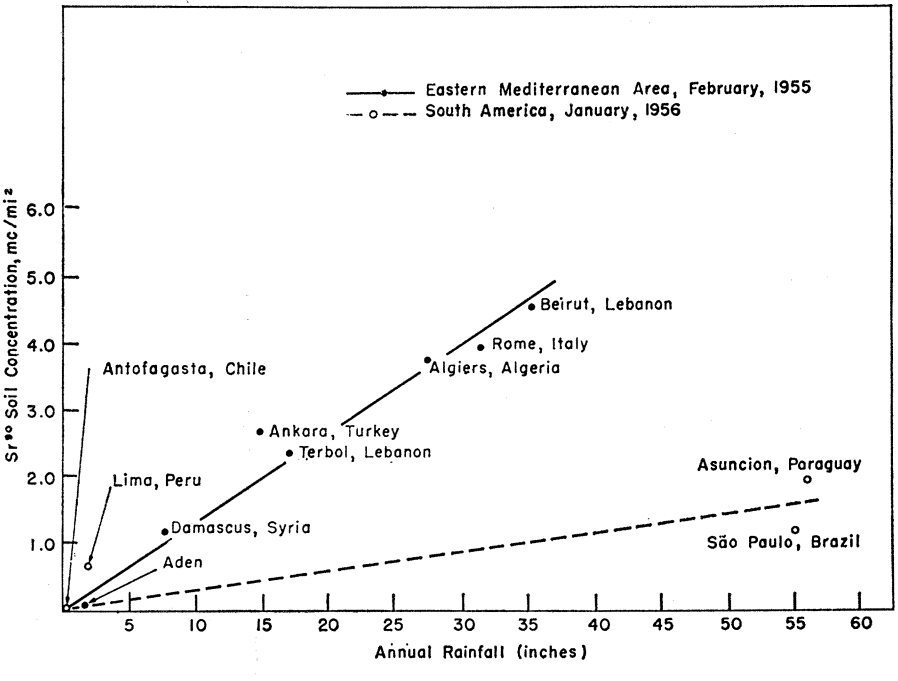

Fig. 4. Concentration of Sr^{90} in soil versus annual rainfall.

similar high rainfall at São Paulo, Brazil, and Asunción, Paraguay (Fig. 4), show 1.2 and 2.0 millicuries, respectively, per square mile in January 1956, indicating an intervening annual Sr^{90} fallout rate of nearly 1 millicurie per square mile. A somewhat lower precipitation rate has been estimated (*2*) from snow-core data from Antarctica (*7*) for the same period. Recent results (*5, 22*) for rain collections in the Southern Hemisphere indicate about 1 to 2 millicuries per square mile per year as the 1957 rate of Sr^{90} fallout.

The results (*8, 26*) of Sr^{90} assay of snow cores and surface snows from polar areas are presented in Tables 1–3. Greenland snow-core samples were obtained from Site II of the Snow, Ice, and Permafrost Research Establishment. This site provides reliable annual precipitation layers. The dating of layers has been checked by measurement of density and oxygen isotope-ratio profiles, both of which show seasonal variation. Additional snow cores and fresh precipitation samples from both polar areas are currently being collected and analyzed for Sr^{90} (*26*).

The Greenland core (Table 3) shows negligible Sr^{90} deposition up to the summer of 1953, indicating that essentially no tropospheric fallout occurred in this latitude and that at least 9 months' mixing time was required for arrival of stratospheric debris from the November 1952 Ivy test. If about 9 months is assumed to be the time required for stratospheric debris from shots at the equator to arrive in Antarctica, the Admiral Byrd Bay core (Table 2) can be approximately dated. Post-Ivy and post-Castle precipitation concentrations are indicated to be about 0.85 and 3.8 disintegrations of Sr^{90} per minute per liter for Greenland and about 0.6 (Table 2, 0.5 to 1.5 years) and 3.4 (Table 1) disintegrations of Sr^{90} per minute per liter in Antarctica, respectively. Direct comparison of the data for Greenland and Antarctica cannot be made with confidence because of the difficulty of dating the cores from Antarctica and because local and seasonal factors may affect both the depth distribution and the concentration of Sr^{90} in snows. However, the two sets of data show comparable Sr^{90} concentration, and the ratio of post-Castle to post-Ivy Sr^{90} levels is similar for the two areas. The ratios are also comparable to the ratio observed for average surface air concentration of stratospheric Sr^{90} for the same periods at Washington, D.C. (Fig. 2).

F. Begemann (*27*) has reported average levels of tritium, from cosmic rays, of 12.6 ± 1.0 and 16.8 ± 1.5 tritium atoms, respectively, per 10^{18} hydrogen atoms for Site II, Greenland, and for Antarctica. If the average amounts of precipitation in Antarctica and in the Arctic are assumed to be inversely proportional to the levels of tritium from cosmic rays, the Arctic would thus appear to have a 33 percent higher annual precipitation than the Antarctic. This leads to the conclusion that deposition of debris from the Ivy and Castle Pacific tests is about 1.5 to 2 times higher in the Arctic than in Antarctica. The values for the Arctic may include some contribution from Soviet tests. The difference may also be due to the fact that the equatorial tests were conducted at 11° north latitude and that hemispheric distribution was therefore unbalanced. Recent collections from both polar areas, including dated cores from Antarctica, should make more accurate comparison possible.

Age of Debris

Measurements of the ratio of Sr^{90} activity to total fission product activity and to the activity of Sr^{89} and other isotopes have been carried out (*11, 25*) in order to distinguish the relative contribution of tropospheric and stratospheric debris to total Sr^{90} deposition and thus to try to account for the sharp increase in the rate of Sr^{90} deposition in the spring (Fig. 3). Results of measurements of Sr^{89} and Sr^{90} in rains at Milford Haven reported by Stewart *et al.* (*11*) are reproduced in Fig. 5. Assuming a mean age of 35 days for tropospheric debris from small weapon tests and further assuming that all Sr^{89} is derived from such tests, Stewart (*11*) estimates that, at most, 5 and 12 percent, respectively, of the total Sr^{90} fallout in the spring of 1955 and 1956 was tropospheric debris. He then concludes that practically all Sr^{90}

Table 1. Deposition of Sr^{90} in surface snow from Antarctica, collected in January–February 1955.

Location	Depth (in.)	Sr^{90} content (disintegration/min/lit.)	Sr^{90} fallout increment* (mc/mi^2)
Little America III (78°S, 170°W)	0 to 8	3.2 ± 0.3	0.31
Little America III (½ mile to the east)	0 to 8	3.1 ± 0.7	0.30
Atka Bay (70°S, 8°W)	0 to 8	5.3 ± 0.5	0.51
Admiral Byrd Bay (69°S, 1°W)	0 to 12	2.0 ± 0.2	0.29
Average		3.4 ± 0.5	

* Density, 0.4.

Table 2. Deposition of Sr^{90} in snow core from Admiral Byrd Bay (69°S, 1°W), collected 19 February 1955.

Estimated age (yr)	Depth (in.)	Sr^{90} content (disintegration/min/lit.)	Sr^{90} fallout rate* ($mc/mi^2/yr$)
0 to 0.25	0 to 12	2.0 ± 0.2	1.2
0.25 to 0.50	12 to 24	1.7 ± 0.2	1.0
0.50 to 0.75	24 to 36	0.48 ± 0.04	0.28
0.75 to 1.0	36 to 48	0.90 ± 0.06	0.52
1.0 to 1.25	48 to 60	≤ 0.48	≤ 0.3
1.25 to 1.5	60 to 72	0.29 ± 0.03	0.16

* Average density, 0.42.

Table 3. Deposition of Sr^{90} in snow core from the Snow, Ice, and Permafrost Research Establishment's site II, in Greenland (77°30′N, 50°30′W), collected 13 September 1956.

Period of sample	Depth (in.)	Density	Sr^{90} content (disintegration/min/lit.)	Sr^{90} fallout rate ($mc/mi^2/yr$)
Summer 1955 to summer 1956	0 to 49	0.36	3.8 ± 0.2	2.0
Summer 1954 to summer 1955	49 to 98	0.40	1.34 ± 0.16	0.79
Summer 1953 to summer 1954	98 to 126	0.41	0.85 ± 0.08	0.29
Summer 1952 to summer 1953	126 to 169	0.41	0.05 ± 0.12	0.03
Summer 1951 to summer 1952	169 to 206	0.42	0.01 ± 0.08	0.00

fallout since 1954 has been derived from old stratospheric debris and that the spring peaks are due to more rapid downward mixing of stratospheric air during the spring.

The assumptions made in Stewart's analysis warrant further consideration. The mean residence time for tropospheric debris may vary appreciably with latitude, season, and weather pattern. However, the range of possibilities is unlikely to alter the conclusion that tropospheric debris from small weapon tests accounts for only a small fraction of the total Sr^{90} fallout. The assumption that the increase in Sr^{89} activity (Fig. 5) is due to tropospheric contamination from small weapon tests appears reasonable at first sight, and is in accord with the widely accepted view that stratospheric debris exhibits a mean residence of 5 to 10 years. However, each increase in rate of Sr^{90} deposition (Fig. 5) appears to be accompanied by a relatively greater increase in Sr^{89} deposition, indicating contribution from recent tests. If these substantial increases in both Sr^{89} and Sr^{90} are related, an explanation other than that of small weapon tests or seasonal mixing effects is needed. A reasonable possibility is that the Sr^{89} is largely of stratospheric origin and that each peak of deposition involves debris injected into the stratosphere only a few months earlier. It should be noted that Soviet tests of high-yield devices have been conducted in the fall or winter of each of the past three years, preceding the three periods of most intense Sr^{90} fallout at north temperate latitudes.

Results of measurements of Sr^{89} and Sr^{90} for rains in the United States, carried out by the Atomic Energy Commission's Health and Safety Laboratory and reported by Collins and Hallden (*25*), are presented in Table 4. The calculated production dates (Table 4, column 4), computed from the average monthly concentration data for March, April, May, and June, clearly suggest the fall 1955 Soviet tests as a prominent source. It would be quite a coincidence if the apparent age of a mixture of fresh tropospheric and old stratospheric contamination should remain so nearly constant for four months. The Sr^{89}/Sr^{90} ratios for rains in Milford Haven (Fig. 5) show the same trend during this period, with values about one-half those observed for rains in the United States (Table 4). The difference in ratios may be due to errors in analysis or to real differences in the relative contribution of older stratospheric debris to the Sr^{90} fallout in these two regions. In view of the methods of analysis (*25*, *28*) and the magnitude of the Sr^{89}/Sr^{90} ratios (Fig. 5 and Table 4), it is thought that analytical errors should not introduce more than a 50 percent error in ratios, amounting to about a 1-month variation in apparent age.

Table 4. Age of debris determined from Sr^{89}/Sr^{90} ratio in rains in the United States, March–July 1956.

Sampling month	Av. Sr^{90} deposition (mc/mi²/mo)	Av. Sr^{89}/Sr^{90} ratio	Calculated production date
March	1.2	22	Sept. 1955
April	1.1	9.7	Aug. 1955
May	1.6	9.3	Sept. 1955
June	1.0	7.0	Oct. 1955
July	1.5	29	Feb. 1956

The foregoing suggestion that the winter-spring 1956 Sr^{89} concentration in rains is largely stratospheric in origin implies that the tropospheric contribution of Sr^{90} from small weapon tests is even lower than Stewart's estimates. Blifford *et al.* (*20*) show appreciable mixing of debris from Nevada and Soviet tests over the whole range of latitudes from 15° to 57°N. A cloud height of about 77,000 feet for a 1-megaton explosion has been reported (*29*). Tropopause heights range from about 55,000 feet at the equator to 30,000 feet at high latitudes. It is indicated that tropospheric contamination results only from surface or low-altitude nuclear detonations of less than about 100 to 200 kilotons in yield; if this is true, downward revision of estimates of tropospheric contamination is necessary. The reduced inventory and widespread distribution of tropospheric debris are consistent with the supposition that tropospheric contamination constitutes only a small percentage of total Sr^{90} fallout.

Results of recent experiments (*26*) support the view that stratospheric contamination from high-yield Soviet tests appears in high concentration at lower stratospheric levels and exhibits relatively short stratospheric residence time. Air-filter samples collected from lower stratospheric levels in the Northern Hemisphere during April 1958 showed very high concentrations of short-lived fission products at middle latitudes and only old fission products at low latitudes. The Ba^{140}/Sr^{90} ratios for a series of 15 individual rains in Bedford, Massachusetts (*26*), in the period from 2 to 8 weeks following the last announced Soviet test in March 1958, decreased regularly with the 12.8 day half-life of Ba^{140}. Since the Sr^{90} deposition rate was slowly increasing during this period, the only reasonable physical interpretation appears to be that even the short-lived Ba^{140} activity is of stratospheric origin. Rapid downward mixing of stratospheric debris from Soviet tests is indicated.

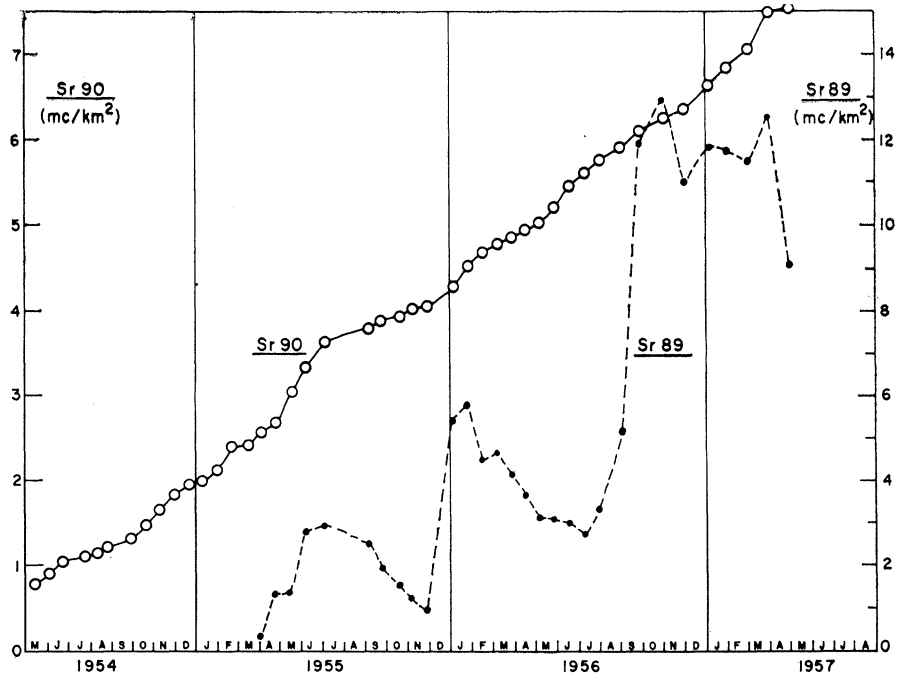

Fig. 5. Cumulative deposition of Sr^{89} and Sr^{90} at Milford Haven, Wales. [According to N. G. Stewart (*11*)]

Surface Distribution

The approximate world-wide distribution of Sr^{90} over land areas in mid-1956, based on Health and Safety Laboratory soil measurements (*22*), is presented in Fig. 6. This representation somewhat overemphasizes United States levels in order to show recent annual changes in the levels at north temperate latitudes where adequate soil data were available. These soils were sampled to a depth of 6 inches, and the Sr^{90} was quantitatively extracted with hydrochloric acid before assay. A few soils from areas of abnormally low or high precipitation were omitted in order that the distribution might be reasonably representative. The Arctic and Antarctic levels of 1.8 and 1.2 millicuries per square mile, respectively, for mid-1956 are based on the data in Tables 1–3 and assume mean annual precipitation of 10 and 8 inches of water equivalent, respectively, in the Arctic and in the Antarctic.

The Southern Hemisphere distribution includes representative soil data from South America, South Africa, Australia, and New Zealand. Variations in soil levels appear to correlate roughly with total annual rainfall, the principal cause of variation in observed Sr^{90} burden (Fig. 4). Other variations are at least partly due to errors in sampling and analysis and to differences in sampling times. Except for the lower deposition in Antarctica, there is no striking variation in latitudinal distribution that can be related to features of the general atmospheric circulation. Since atomic tests in the Southern Hemisphere have been very limited in number and yield, fallout in the Southern Hemisphere is essentially all derived from stratospheric debris. The 1956 levels for the Southern Hemisphere may be accounted for by United States equatorial tests alone.

In the Northern Hemisphere the striking feature is the heavy fallout in the middle latitudes. Comparable high levels are observed in northern United States, southern Canada, and northern Europe. Within a limited geographical area, the cumulative level in soil is approximately proportional to total rainfall. Large variations for areas of similar latitude and annual rainfall occur as a result of seasonal distribution of rainfall in relation to the spring peak in deposition rate. There is also a striking variation in deposition with latitude in relation to the zone of entry for stratospheric debris from Soviet tests. Proximity to small weapon test sites, variations in type of rainfall, and circulation features have some additional effect on distribution.

Discussion and Conclusions

It is inferred from considerations of the age of debris and other evidence discussed above that the spring peak in Sr^{90} fallout (Fig. 3) may involve a substantial contribution from stratospheric debris of short residence time injected during the immediately preceding Soviet test series. This possibility is strongly reinforced by consideration of alternative interpretations. Estimates based on Sr^{89}/Sr^{90} ratios and other evidence discussed above indicate that the Sr^{90} contribution from tropospheric debris following small weapon tests is minor. Selective downward mixing of old, and thus well-mixed, stratospheric debris during the late winter and spring (a theory in accord with Dobson's interpretation of the ozone data) may also be questioned as an explanation for the spring peaks in Sr^{90} deposition rate. If seasonal variation in downward mixing of stratospheric Sr^{90} debris were an important factor independent of considerations of production, the effect should be evident in precipitation data for the Southern Hemisphere. Stewart's results (*11*) for Ohakea, New Zealand (40°12′S; 175°23′E), show minor seasonal variation in Sr^{90} content of rains as compared to variations observed at Middle latitudes of the Northern Hemisphere. Unless there are large differences in degree of effect for the two hemispheres, the striking spring peaks in Sr^{90} fallout in north temperate latitudes must be attributed largely to stratospheric debris from preceding Soviet tests. It should also be mentioned that the spring peak in ozone concentration (*30*) is very broad and of low amplitude whereas the Sr^{90} fallout peaks at north temperate latitudes are of greater amplitude and are not confined to the spring season (Figs. 3 and 5).

Since tropospheric injections of Sr^{90} contamination may be reasonably assumed to contribute only a small fraction of total Sr^{90} fallout, the surface distribution (Fig. 6) must be explained in terms of the production schedule, the injection latitudes, and the mixing history of intermediate and large-scale detonations. The 1956 distribution in the Southern Hemisphere is without question due largely to the November 1952 Ivy and the spring 1954 Castle tests, with stratospheric injection at latitude 11° north. The distribution in the Southern Hemisphere exhibits no appreciable variation with latitude. This implies that stratospheric debris from equatorial tests is not selectively mixed downward at middle latitudes to an appreciable degree. If distribution of equatorial test debris is assumed to be similar in the two hemispheres, the north temperate latitude peak in distribution must to a very large extent be the consequence of Soviet tests.

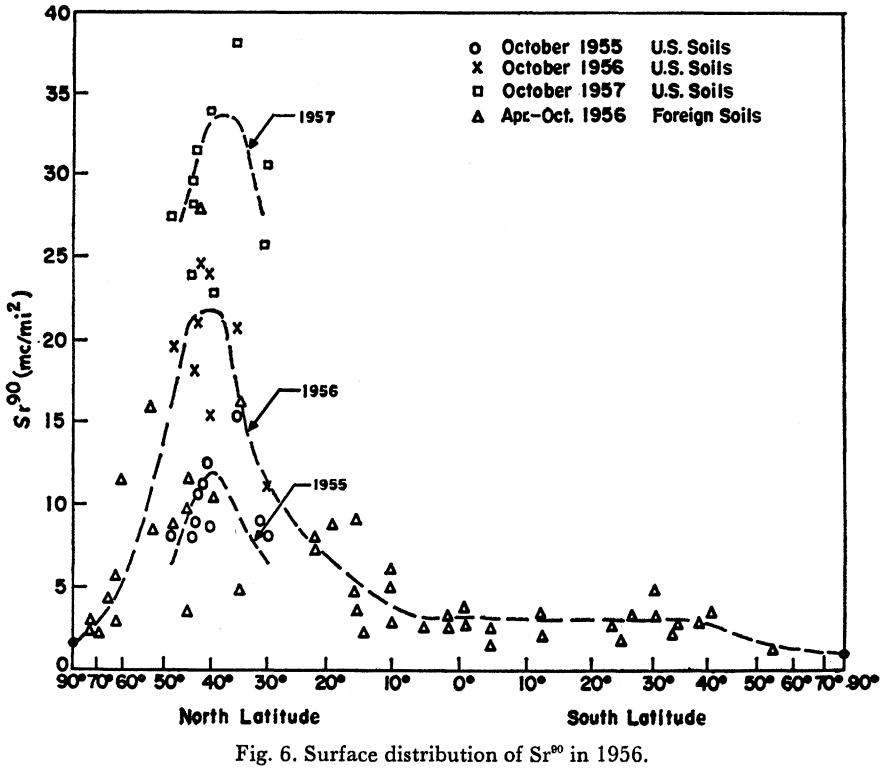

Fig. 6. Surface distribution of Sr^{90} in 1956.

Comparison of the post-Ivy and post-Castle deposition levels in polar snows makes possible an approximate interpretation of the 1956 surface distribution (Fig. 6) in terms of United States and Soviet test contributions. The deposition rate in the Arctic for equatorial tests is estimated to be 1.5 times that for Antarctica. If 1.5 times the Southern Hemisphere is taken as the Northern Hemisphere distribution for equatorial tests and if 5 percent of the Northern Hemisphere levels is assumed to be tropospheric fallout, the balance is attributable to Soviet tests. On the basis of this analysis, Soviet tests are the source of roughly three-fourths of the Sr^{90} fallout in north temperate latitudes.

When the possible histories of circulation and mixing of clouds of stratospheric debris injected at equatorial and middle latitudes are considered, nothing seriously inconsistent with the foregoing analysis is found. The Brewer (*18*) and Dobson (*19*) views of atmospheric circulation indicate poleward flow in the stratosphere and downward mixing at middle and higher latitudes, with more rapid downward mixing in the spring. The influence of the jet stream may further increase the downward mixing of stratospheric air at middle latitudes. If these views of circulation are assumed to be correct, it is indicated that the timing and latitude of Soviet tests fortuitously lead to short storage times and to a quite selective zone of downward mixing and deposition. A markedly different mixing history is to be expected for stratospheric debris injected at equatorial latitudes. The debris may be expected to diffuse rapidly throughout a broad zone of the equatorial stratosphere, under the influence of the strong easterlies in this region. Debris in the equatorial stratosphere will subsequently mix into higher latitudes of both hemispheres; perhaps somewhat more debris will move into the Northern than into the Southern Hemisphere from United States tests at 11° north latitude.

A long stratospheric residence time for debris in the equatorial stratosphere would mean a slow rate of exchange from this region into higher latitudes. The rate of poleward mixing in the stratosphere may vary appreciably with altitude and season. H. Wexler (*31*) has pointed out that the average meridional geostrophic wind speeds, (at 10° to 70°N) at 19 kilometers in altitude, are at a minimum in August and at a maximum in March and April; this suggests more rapid northward migration of debris for spring tests. The observed initial arrival times for equatorial test debris at mid-latitude ground levels [about 3 months (*21*)] and later at high latitudes [9 months (Table 3)] are qualitatively consistent with observations of the rate of northward spread of the Krakatoa volcanic dust cloud (*31*) and of the vertical mixing of ozone (*30*). In his discussion of the Krakatoa cloud spread, Wexler (*31*) has pointed out that the average meridional wind speeds, at an altitude of 19 kilometers, are a maximum at low latitudes, decrease to a minimum at middle latitudes, and increase somewhat at higher latitudes. The ozone data for middle latitudes show that the peak in surface air concentration postdates the period of maximum total ozone content by about 2 months, indicating the approximate time required for vertical mixing from the 20-kilometer ozone layer to ground levels. Thus, the 3 months required for Castle debris to arrive at middle latitudes must correspond to about 1 month required for northward mixing and 2 months required for vertical mixing. The subsequent slow increase in Sr^{90} concentration in middle latitudes and the late arrival of Sr^{90} fallout in polar areas reflect a slow exchange out of the equatorial stratosphere northward through the region of low meridional winds. Clouds of nuclear debris from Soviet tests at 52° north latitude, injected in the region and usually in the season of lowest meridional wind speeds, probably undergo little diffusion before mixing downward, perhaps quantitatively, into the lower atmosphere.

It appears that debris from equatorial detonations of high-yield devices is reasonably well mixed in the equatorial stratosphere and that subsequent circulation does not effect appreciable latitudinal or seasonal variation in Sr^{90} fallout. Middle-latitude tests of intermediate and high-yield weapons must account to a large extent for the observed seasonal and latitudinal variation in Sr^{90} deposition in the Northern Hemisphere. The differences in mixing history and ultimate distribution for stratospheric debris injected at low and middle latitudes are important consequences of stratospheric meteorology. Seasonal effects undoubtedly influence the circulation and deposition of fallout but may be assigned a secondary role in the explanation of major variations in fallout rate.

On the basis of the foregoing consideration of the Sr^{90} fallout evidence, the following summary remarks and tentative conclusions may be stated.

Uncertainties about the amounts of Sr^{90} that have been injected into the stratosphere, coupled with lack of adequate information about the origin and distribution of the Sr^{90} already deposited over the earth's surface, make it difficult to assess stratospheric holdup times and limit the accuracy of fallout distribution predictions.

Precipitation washout is the predominant mechanism of Sr^{90} fallout. Gravitational fall of dry particulate material appears to contribute a negligible fraction of the total Sr^{90} fallout.

Within a limited geographical zone of uniform circulation and of characteristic rainfall type, the cumulative S^{90} fallout will be approximately proportional to total rainfall. In the Northern Hemisphere there is a marked variation in Sr^{90} fallout, dependent on the location of the area with respect to the zone of entry for stratospheric debris from Soviet tests. The seasonal distribution of rains further modifies the Sr^{90} distribution. In the Southern Hemisphere the annual rainfall appears to be the major factor in Sr^{90} distribution.

Stratospheric debris injected by high-yield atomic tests at equatorial latitudes appears to undergo diffusion throughout a broad region of the equatorial stratosphere and subsequently mixes poleward, roughly equivalent amounts being contributed to each hemisphere. A relatively long stratospheric storage time of perhaps 5 to 10 years and an approximately uniform hemispheric distribution, reduced toward the pole, are indicated in this case.

Clouds of stratospheric debris from middle-latitude nuclear detonations apparently reach lower stratospheric levels after only limited diffusion and release substantially all of their contamination into a zone of restricted latitude during the first 6 months or so. The spring peaks in Sr^{90} deposition rate and the band of heavy fallout at north temperate latitudes are attributable principally to high-yield Soviet tests.

The direct injection of contamination into the troposphere is apparently limited to detonations not exceeding 100 to 200 kilotons. Strontium-90 activity from this source is quite widely dispersed and has amounted to only a few percent of total fallout to date.

The total Sr^{90} fallout at north temperate latitudes from high-yield tests at these latitudes is higher by a substantial factor than that from equivalent equatorial tests. Perhaps even more important, the short stratospheric holdup time for middle-latitude tests allows for substantial deposition before decay of the shorter-lived fission products which con-

tribute to external gamma radiation and its possible genetic effects. It is clear that estimates of future contamination from high-yield detonations at north temperate latitudes must be revised upward from estimates based on 5- to 10-year periods of stratospheric storage (32).

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Recent Saprolite

Thick saprolite has formed in northern New Jersey since the last Pleistocene glacial stage.

James P. Minard

Thick saprolite is present in glaciated terrane of northern New Jersey under field conditions suggesting formation within a comparatively short time interval. The saprolite has developed on a low spur lying along the east side of a glaciated valley, normal to the direction of former ice movement. It is quite probable that the saprolite has weathered from consolidated rock since the latest (Wisconsin) glacial period.

A gneissic saprolite, the maximum age of which is possibly determinable, is present in northern New Jersey, 5 miles north of the southern static limit of Wisconsin glaciation. The saprolite is exposed in a pit along the east side of route 206, east of Cranberry Lake, Sussex County. The saprolite is along the west side and near the base of a north-trending bedrock ridge; it forms a benchlike spur, the upper surface of which is 50 to 60 feet above the swampy terrain west of the highway.

The rock from which the saprolite formed is shown on the geologic map as Pochuck gneiss of Precambrian age (Fig. 1). The rock is an amphibolite gneiss containing much hornblende, considerable oligoclase and diopside, and some quartz, magnetite, and microperthite.

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Outcrop Description

A thickness of nearly 15 feet of saprolite is exposed in a pit approximately 200 feet wide and 300 feet long. The saprolite is easily excavated and crumbles readily, under hand pressure, into a sticky, clayey, gritty mass. At least 12 feet of saprolite underlies the pit bottom, giving a minimal thickness of 25 feet. Because of the gentle slope and absence of bedrock downhill from the pit, the total thickness of saprolite may be as much as 40 feet. Overlying the saprolite in the pit is several feet of well-weathered glacial till. Except for a shallow A horizon of the soil profile, the till and the upper few inches of the saprolite apparently comprise the 2- to 3-foot-thick B horizon of the soil profile, and the rest of the saprolite forms the deep C horizon. By comparison, saprolite formed from unglaciated gneiss 3 miles south of Chester (see Fig. 2) is nearly 60 feet thick and has a substantial A horizon and a thick (5 to 6 feet) B horizon.

Remarkably continuous light and dark gneissic layering can be seen in the exposure (see Figs. 3–5). These layers are one-eighth to 1 inch thick and are many feet long. The layering dips approximately 15° south, except in the west