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INTERPRETATION OF WESTWOOD, NEW JERSEY RAINFALL DATA

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INTERPRETATION OF WESTWOOD, NEW JERSEY RAINFALL DATA

I. INTRODUCTION

During the period covered by this report attention has been given to the compilation of the existing rainfall data, methods of data handling, definition of the most important areas of investigation and preliminary consideration of several specific questions.

For the past several years the collection and fission product analysis of rainfall samples from many stations has been proceeding. As the data becomes more numerous and complex the interpretative aspect becomes more important and time consuming. It is the purpose of this project to assist in the evaluation of the fission product concentration in rain as a function of time, location and meteorological setting. The Atomic Energy Commission has been the primary force behind this program having gradually established an extensive domestic and foreign rainfall collection network. At the time of the last data summary by the New York Operations Office of the A.E.C.[1], a total of 27 continental United States and 39 other monthly fallout sampling stations were operative. In addition three of the sites, Pittsburgh, Pennsylvania; Richmond, California; and Westwood, New Jersey collect and analyze individual rains. This relatively large amount of data will contribute to the solution of many of the important questions concerning fallout. Some of these are:

1. The reliability of samples taken for the purpose of evaluating world-wide fallout.
2. The adequacy of coverage by the existing sampling stations.
3. The deposition mechanism(s), and in particular the manner in which precipitation brings the debris to the earth.

4. The relations between fallout and meteorológical conditions, perhaps leading to an understanding and evaluation of dry fallout versus rainóut.

5. The validity and mechanism of the apparent seasonal variation of fallout.

6. Gross rate of transfer from the stratosphere to the ground.

7. Mechanism and location of the stratosphere troposphere passage.

8. Correlation between measurements of fission products in precipitation and other (e.g. soil and air) measurements.

9. The origin and age of debris on the basis of isotopic ratios.

10. The specific activity of the rain and its relation to total precipitation.

The research under this contract will be directed toward answering as many of these questions as is possible within the time and funds provided.

In the first quarterly report four subjects have been pursued:

(a) The Mechanism of Fallout Deposition,

(b) Seasonal Variation in Fallout.

(c) The Stratospheric Residence Time of the HARDTACK Produced Tungsten-185, and

(d) Accuracy of the Pot Sampler.

Based on the work accomplished to date and the insight provided by the problems and data encountered, it is becoming increasingly clear that an expanded program in rainfall-fallout evaluation is justified and would yield great return.

II. THE WESTWOOD, N. J. RAINFALL COLLECTION STATION

Isotopes, Inc. at Westwood, New Jersey has been collecting and analyzing rainfall since August 1957, under contract with the A. E. C. At the present time Westwood has a comprehensive program of both monthly and individual rain collections for fallout analysis in addition to rain-gaging equipment and occasional fractional precipitation collections. Figure 1 shows the entire array of routine rain collection devices on the roof at Isotopes, Inc. Shown are two steep-walled stainless steel monthly pots (0.842 square foot collection area), the monthly plastic funnel ion exchange collector (0.815 square foot collection area) developed by New York Operations Office, three large area plastic individual rain collectors (2.58 square foot collection area), a standard copper and an aluminum automatic recording (weighing type) U. S. Weather Bureau rain gage. Figure 2 shows the three types of fallout collectors in more detail.

In addition to the routine collection program samples of fractions of individual rains are collected in order to study the mechanism of rain scavenging. Earlier collections were made with nearly rectangular (12" x 13 1/2") plastic pans. At the present time, a new set of four circular, steep-walled plastic pails are being used. These have screw-on lids and after scouring are covered and placed on the roof. During a rain one pail at a time is exposed for a predetermined interval.

Measurements of the local concentration of radioactivity in ground level air at Westwood, New Jersey has also been initiated using a Staplex type TFIA high volume air sampler. The purpose of this analysis is to attempt to cast some light on the depletion of tropospheric radioactivity by rain and to study the relation of the specific activity of ground level air to that in rain.

All of the data routinely developed at Isotopes, Inc. is reported monthly to the Health and Safety Laboratory, New York Operations Office, of the A. E. C. and distributed by that office in monthly Original Data Reports and Quarterly Strontium Program Summary Reports. The latest Summary Report is HASL-65 dated May 29, 1959 and was followed by an August 31, 1959 Data Report.

Figure 3 illustrates the monthly fallout of strontium-90 in Westwood, New Jersey since inception of the station, as measured by the stainless steel

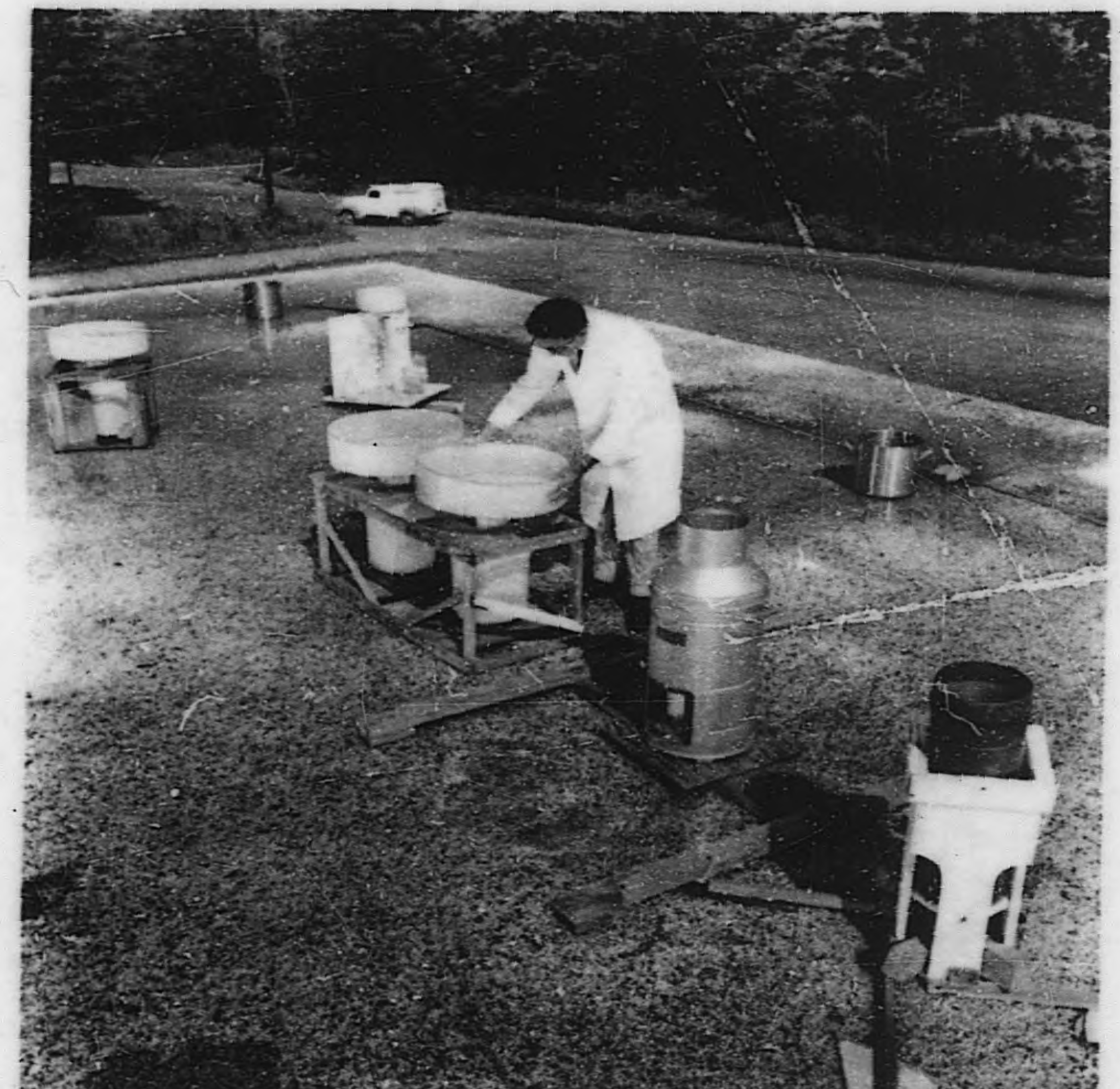


Figure 1. Array of Rain Collection Devices on the Roof at Isotopes, Inc.

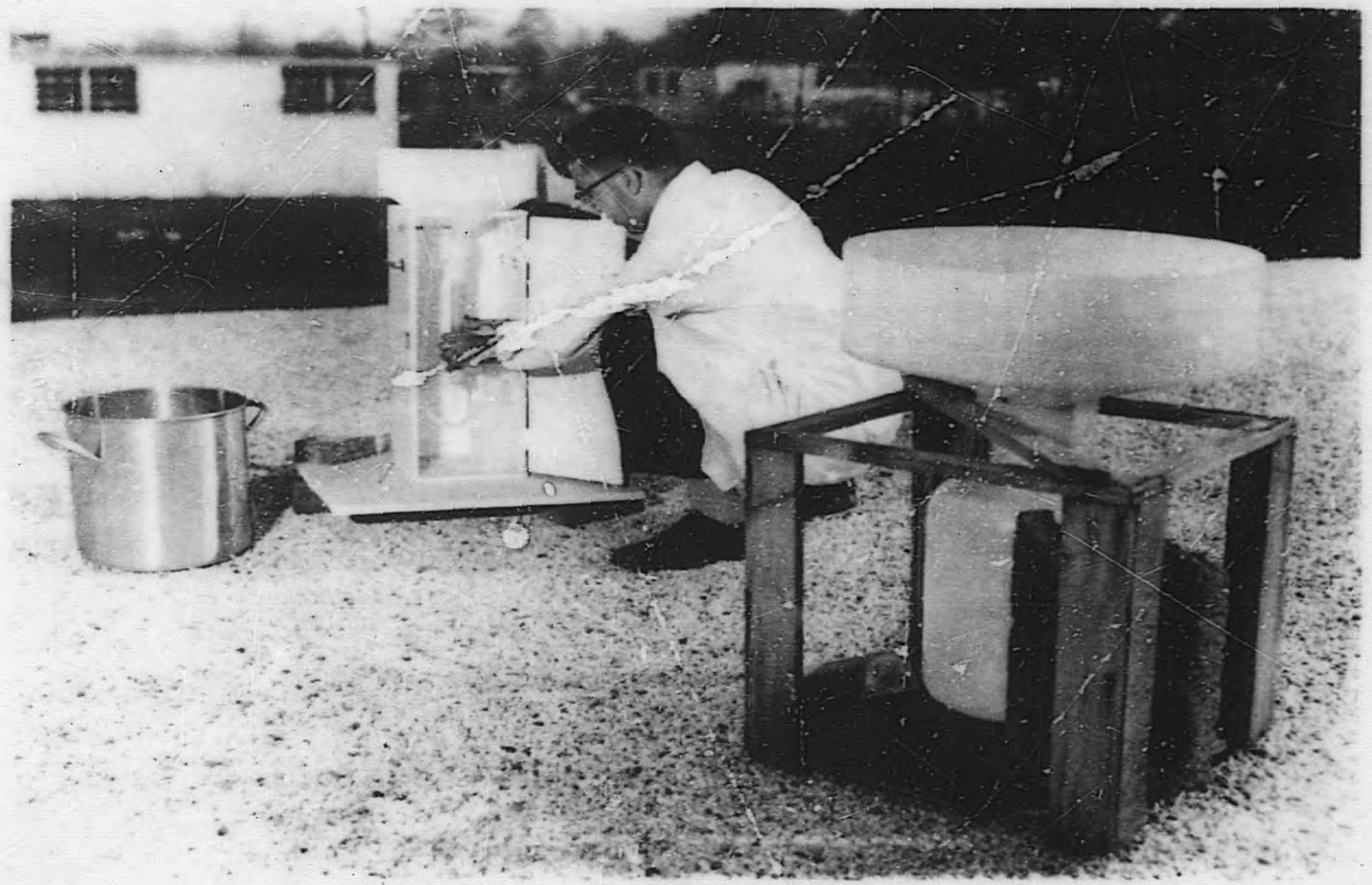


Figure 2. Detailed Picture of Three Types of Fallout Collectors

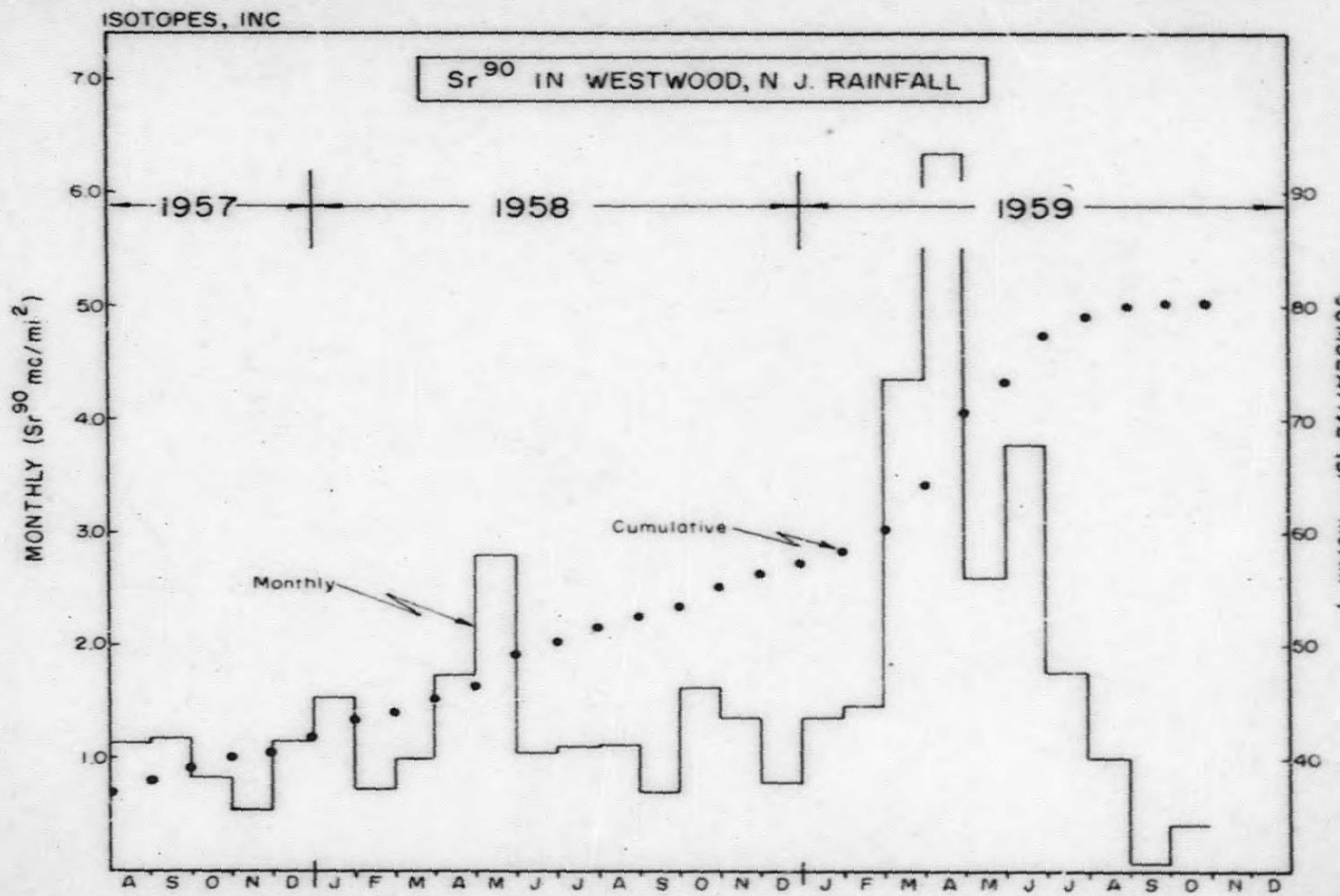

ISOTOPES, INC
Sr^{90} IN WESTWOOD, N J. RAINFALL
1957
1958
1959
MONTHLY (Sr^{90} mc/mi^2)
CUMULATIVE (Sr^{90} mc/mi^2)
Monthly
Cumulative
7.0
6.0
5.0
4.0
3.0
2.0
1.0
90
80
70
60
50
40
A S O N D J F M A M J J A S O N D J F M A M J J A S O N D

FIGURE 3

pot collector. For the cumulative data curve it was assumed that on July 31, 1957, the integrated fallout in Westwood, New Jersey was equal to that in New York City. Thus the initial value of 37 millicuries per square mile was obtained from HASL data[1].

III. INTERPRETATION

A. The Mechanism of Fallout Deposition

It is generally accepted that the principal agent for the deposition of delayed radioactive fallout on the earth is precipitation. However, the mechanism of radioactive "rainout" is not well understood. The following possible mechanisms have been suggested:

(1) incorporation of radioactive particles in cloud drops as condensation nuclei during cloud formation ("condensation");

(2) mechanical capture of radioactive particles by cloud drops resulting from random collisions within the cloud ("coagulation");

(3) capture of radioactive particles by falling raindrops and snowflakes ("cleansing").

Greenfield[2] has shown that the ionization of atomic debris is not sufficient to give these particles priority as condensation nuclei. This fact, however, does not eliminate radioactive particles from the list of possible ordinary nuclei, even though they are probably not highly effective. It is, therefore, not unreasonable to assume, as Holland[3] does, that radioactivity may be incorporated in precipitation during the condensation stage. On the basis of this assumption Holland has hypothesized that the local depletion of tropospheric strontium-90 by local rain showers should exceed that resulting from large scale precipitation, for in the latter case the radioactivity is replenished with the moisture influx.

The efficiencies of the latter two processes have been calculated by Greenfield[4] who has shown that the "cleansing" process is very inefficient for particles with diameters smaller than about one micron. On the other hand the "coagulation" process does account for the efficient removal of sub-micron particles, according to Greenfield. These particles are then placed in a more advantageous condition, in cloud droplets, for removal by precipitation.

Holland's hypothesis would follow from the coagulation as well as the condensation mechanism. However, if cleansing is the principle rainout mechanism, the local depletion of tropospheric radioactivity may be independent of the character of the precipitation, unless local showers and large scale steady rain have different "cleansing" efficiencies.

For the purpose of elucidating the mechanism of fallout by precipitation scavenging, a program of collecting successive fractions of individual rains was initiated at Westwood in July 1959. The rain fractions have been analyzed only for total beta activity thus far.

The first four sets of rain collections were obtained with two soft plastic dishpans which were exposed alternately on the roof of the laboratory. The pans have nearly rectangular cross sections, about 12 by 13 1/2 inches, and are about 6 inches deep. At the onset of rain a pan is scoured and placed on the roof at the laboratory. After a certain time interval (which varied from 10 minutes to 16 hours depending on the time of day and on the rain intensity) the pan is replaced by a fresh collector. The two pans are then alternated for the duration of the rain. The volume of rain collected each time is measured, and the pan is scoured to collect surface deposits before processing the water for counting. The beta counting procedure is the same as that used for the total rain collection.

The fractional rain collection program is expected to cast some light on the mechanism of radioactive rainout, and possibly also on the precipitation process itself. Among the questions to which the experiment should provide answers are these:

(1) How does the specific radioactivity of rain water change during the course of a rainstorm?

(2) What fraction of the total activity is contributed by each of the rain fractions?

(3) How does the variation in rainout during the storm depend on the intensity and duration of the rain?

(4) How does the variation in rainout depend on the character of the precipitation and on the general meteorological situation?

The answers to these questions should help to solve the more fundamental problem of how precipitation removes radioactivity from the air. Some preliminary data relevant to these problems are given in Table 1.

Fractional rain samples have been collected on July 14, 20, 23 and 31, 1959, thus far. The data are presented in Table 1.

The highest specific activity was associated with the lightest, and most protracted rain (No. 150) of the four. On July 14-15, 1959 a stationary front oriented northeast-southwest lay almost over Westwood, New Jersey. Light, steady rain began about 0400 EST/14 and continued, with variable intensity, for more than 30 hours. The total accumulation of rain was only 0.22 inches, but the average specific activity, 0.73 dpm/ml, was 2 to 6 times as large as that of any of the other rains.

Rain number 153 on July 20, 1959 consisted almost entirely of two brief periods of intense shower activity, one about 1600 EST produced by a pre-frontal squall line, the other about 1940 EST when the cold front passed the laboratory. Almost half the total rainfall of 0.67 inches fell in each shower, only about 0.1 inch having accumulated in intermittent rain before and between the two showers. The mean specific activity in this case, 0.12 dpm/ml, was the lowest of the four.

The third rain, number 154 on July 23, 1959, fell in a violent tropical air mass thunderstorm, and was not associated with any frontal activity. The

Table 1. Total Beta Activity in Fractional Rain Samples. The total rainfall measured in the raingage for each case is expressed in inches. Sample rain volumes collected are given in millileters (ml). Beta activity is expressed both in dpm per sample and in dmp/ml. The "rain numbers" refer to the series of rains in the regular collection program. Fractional rain samples are designated by letter. Collection times are in Eastern Standard Time (EST).

Rain No.	Date	Sample: Time(EST)	Rain Vol. (ml)	Total Beta (dpm) Sample	Per ml
150	July 14-15, 1959	a: 0300/14-0800	48	51	1.06
		b: 0800-1100	176	76	0.43
		c: 1100-1310	74	58	0.78
		d: 1310-1430	30	30	1.00
		e: 1430-1620	25	32	1.28
		f: 1620-0800/15	80	95	1.19
		g: 0800-1000	160	93	0.58
		h: 1000-1330	16	9	0.56
		Total	609	444	0.73
		Total Rain Depth:	0.22 inches		
153	July 20, 1959	a: 1500-1547	152	33	0.22
		b: 1547-1615	405	31	0.08
		c: 1615-1900	160	31	0.19
		d: 1900-2000	450	30	0.07
		e: 2000-2030	460	71	0.15
		Total	1627	196	0.12
		Total Rain Depth:	0.67 inches		
154	July 23-24, 1959	a: 1400-1425	470	192	0.41
		b: 1425-1440	900	308	0.34
		c: 1440-1455	1540	621	0.40
		d: 1455-1525	1770	519	0.29
		e: 1525-1545	550	97	0.18
		f: 1545-1630	330	96	0.29
		g: 1630-0800/24	100	44	0.44
		Total	5660	1877	0.33
		Total Rain Depth:	2.08 inches		
157	July 31, 1959	a: 1345-1355	790	343	0.43
		b: 1355-1415	415	48	0.12
		c: 1415-1435	235	44	0.19
		d: 1435-1450	1230	191	0.16
		e: 1450-1500	1085	129	0.12
		f: 1500-1515	615	105	0.17
		g: 1515-1530	575	89	0.15
		h: 1530-1545	285	72	0.25
		i: 1700-1930	120	20	0.17
		Total	5350	1041	0.19
		Total Rain Depth:	2.14 inches		

storm began at 1400 EST, and in a series of heavy showers, produced more than 2 inches of rain in less than 2 1/2 hours. This storm produced the highest total activity of the four (1877 dpm), and the second highest specific activity (0.33 dpm/ml).

Rain number 157 on July 31, 1959 was again a series of air mass thunderstorms in advance of a slowly moving cold front. Most of the accumulation of 2.14 inches of rain fell in less than 2 hours. Despite the similarity of rains 154 and 157, the former deposited almost twice as much radioactivity as the latter.

The variation of total beta activity in rain water during the course of each rain is shown in Figure 4. The abscissa in the figure is the cumulative rainfall in percent of the total rain, while the ordinate represents the cumulative beta activity in percent of the total beta activity present in the entire rainfall. The 45-degree line is drawn to represent the hypothesis that equal fractions of rain deposit equal fractions of radioactivity.

No attempt will be made to draw any definitive conclusions from the limited data available. However, Figure 4 does suggest that, for all rains grouped together, equal fractions of rain deposit approximately equal fractions of radioactivity. On the other hand the graph also suggests that short, heavy rains (e.g., thunderstorms) tend to deposit a greater proportion of their total radioactivity in the early part of the storm, while the light, protracted rains appear to deposit a relatively smaller proportion of the radioactivity in the early part of the rain. This highly tentative result indicates that either "cleaning" is unimportant, in which case the hypothesis of greater tropospheric depletion by local showers than by large scale precipitation is supported, or the "cleansing" efficiency of heavy rain is greater than that of light rain. Although he has not computed the cleansing efficiencies in the sub-micron range,

VARIATION OF TOTAL BETA ACTIVITY IN RAINFALL DURING THE COURSE OF VARIOUS RAINS AT WESTWOOD, N. J.

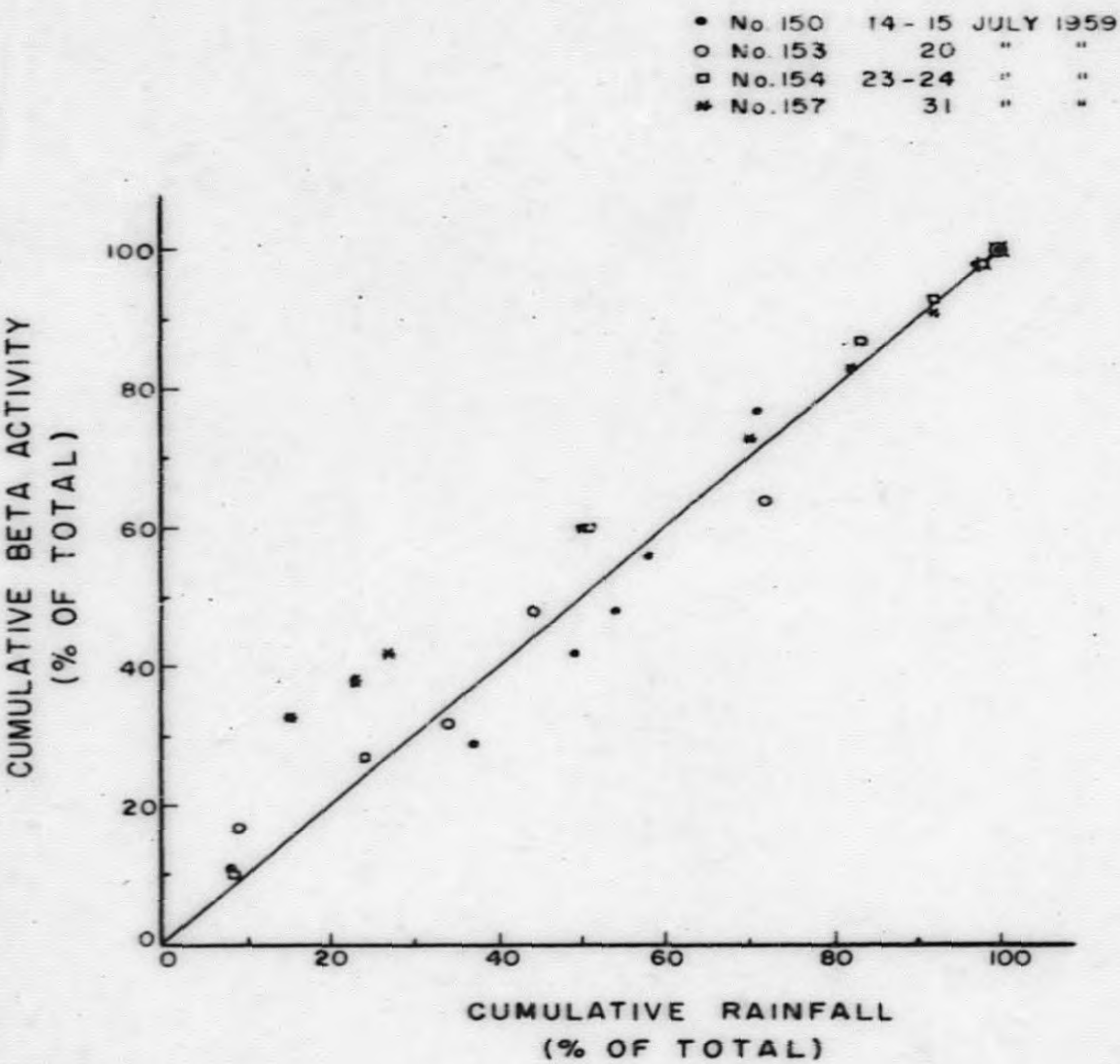


FIG. 4

Greenfield's[4] calculations down to particle diameters of 2 microns do show much greater efficiencies for the heavy rains. It should also be noted that despite the fact that the cleansing efficiencies are low in the sub-micron range, the absolute effect of cleansing may not be negligible.

Another aspect of the data is shown in Figure 5, where the relative specific beta activity is plotted against cumulative rainfall for each sample of each rain. (Cumulative rainfall serves here as a kind of uniform, relative time scale for each storm.) The relative specific activity, expressed as percent, is the ratio of the specific activity of the sample to the mean specific activity of the entire rain. In each case the specific activity of the second sample was lower than that of the first, while that of the third sample exceeded the second. The specific activity was lower at the end of sampling than at the beginning in three out of four rains. The decrease of activity following the first sample suggests a cleansing effect, while the subsequent increase indicates possibly that later replenishment of radioactivity resulting from moisture influx at the cloud level or from low level convergence. These data do not as yet indicate any consistent difference between light and heavy rains. Indeed it remains to be seen whether or not even the apparently consistent initial drop in activity proves to be statistically significant.

B. Seasonal Variation in Fallout

The possibility of a seasonal variation of fallout has been discussed extensively in the literature by Stewart[6], Machta[7] and recently reviewed by Martell[8]. It is generally acknowledged that seasonal variations have been observed at least in the Northern Hemisphere, with maximum fallout occurring in the spring and minimum in the fall. The cause of the seasonal variation, however, has been a matter of controversy. Meteorologists have tended to accept the view (see e.g. Machta and List[9]) that the cause of the seasonal variation

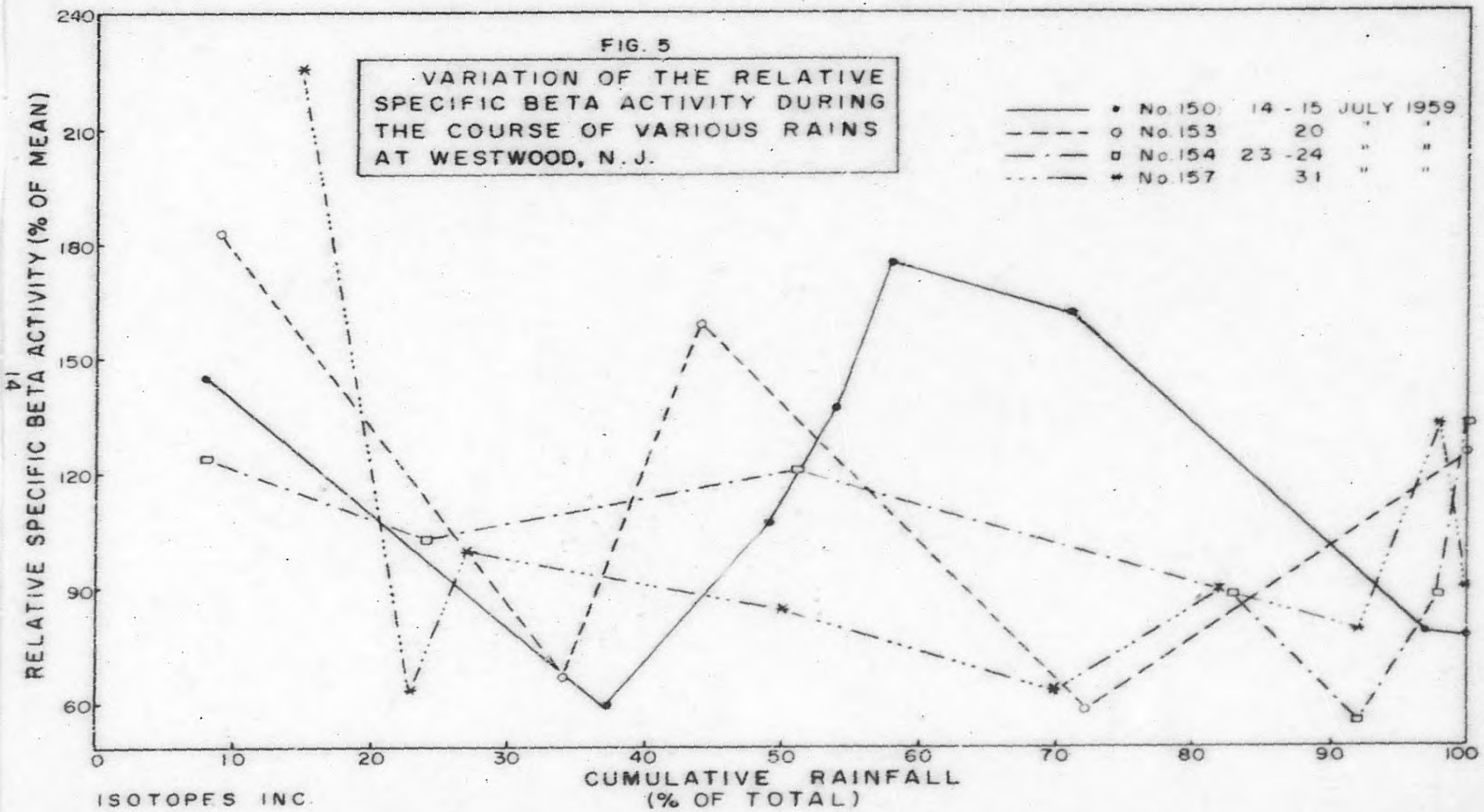


FIG. 5

VARIATION OF THE RELATIVE SPECIFIC BETA ACTIVITY DURING THE COURSE OF VARIOUS RAINS AT WESTWOOD, N.J.

is meteorological, being associated with a maximum transfer of radioactive debris from the stratosphere to the troposphere in late winter and early spring. Martell[8] has argued that the apparent seasonal variation may be attributed to the schedule of nuclear testing, i.e. Russian testing at high latitudes in the fall followed by spring deposition.

Among the meteorologists there are two prevalent viewpoints regarding the transfer mechanism. Stewart[6] and Machta[7] follow generally a model suggested by Brewer, which predicts a general entry of polar stratospheric air directly into the troposphere with peak values in winter and spring. Spar in a recent report[10] argues that a seasonal variation in the turbulent transfer through the tropopause break, associated with the seasonal variation in jet stream intensity, is responsible for the seasonal variation in fallout.

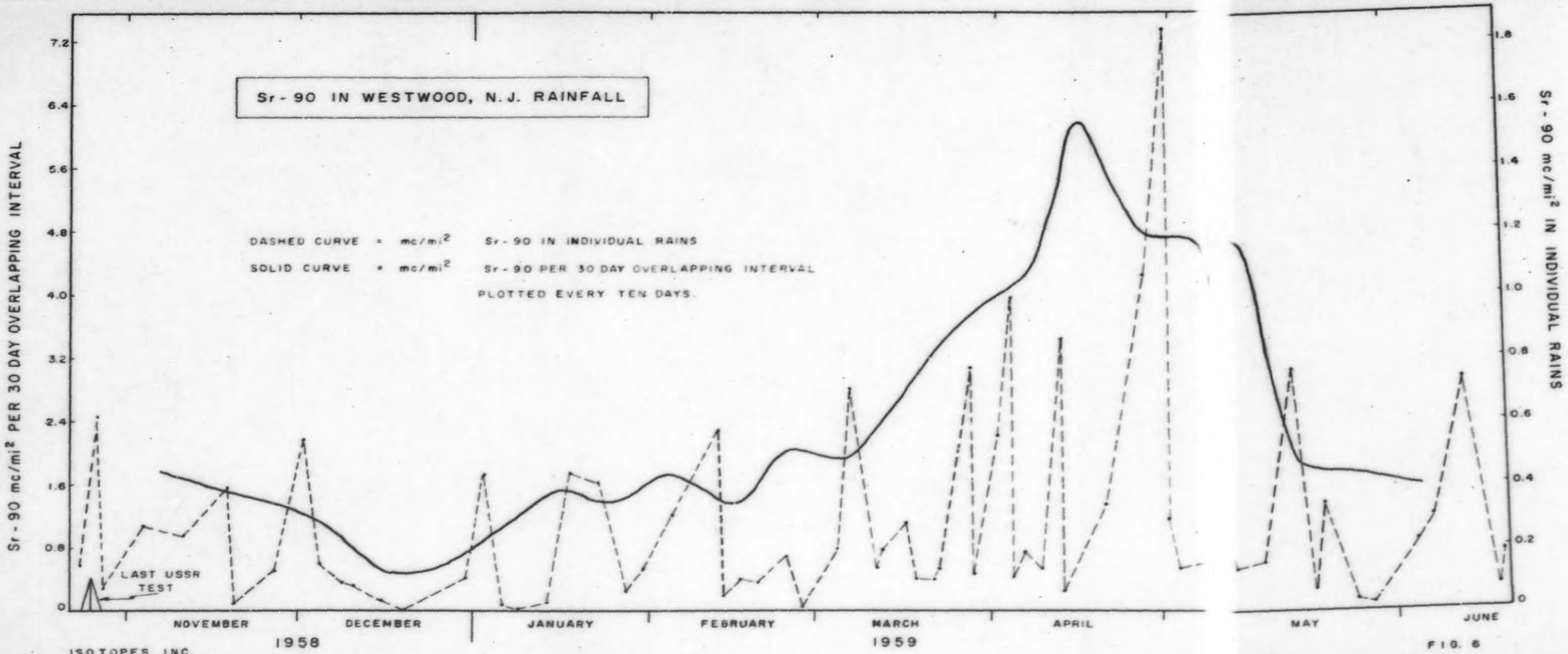
The moratorium on testing of nuclear weapons that began in November 1958 provides an opportunity for a critical evaluation of the various theories of delayed fallout. The last significant test was conducted by the U. S. S. R. on October 25, 1958 at their high latitude test site. As no large weapons were detonated after this date, and the tropospheric residence time is known to be quite short, there is little likelihood that the spring 1959 fallout contained a significant tropospheric component. Thus the strontium-90 fallout data for this period provides unambiguous information about the stratospheric fallout.

In May and June 1958, during the HARDTACK test series in the Pacific (latitude 11°N), tungsten-185 was produced. This unique tracer may be used to estimate the transport of stratospheric radioactive debris from an equatorial source. It may also cast light on the mechanism of stratospheric fallout, and in particular on the seasonal variation. For if tungsten-185 fallout exhibits a maximum in spring, Martell's hypothesis must be rejected (at least as the sole explanation), and the meteorological theory is strongly supported.

The seasonal variation of strontium-90 fallout at Westwood is illustrated in Figure 6 for the period beginning October 23, 1958. The total activity of each rain is plotted on the graph, and a smooth curve has been drawn to represent the total fallout in overlapping 30-day intervals. The latter was constructed by adding the strontium-90 activity in all rains occurring within overlapping 30-day periods, where the 30-day interval is advanced through the time series in 10-day steps. Figure 7 is a similar graph showing the seasonal variation of specific strontium-90 activity in overlapping 30-day intervals, and the seasonal variation in the same intervals of specific tungsten-185 activity in Westwood rains. The latter values have been corrected for decay back to October 1, 1958. The specific activity values in this figure were computed by dividing the total activity over the summation period by the total rain over that period.

Figure 6 shows the expected large variations in strontium-90 fallout from one rain to the next, associated with variable rainfall amounts, but with an increasingly high frequency of "hot" rains beginning in March. The smoothed fallout curve based on the running summation shows an abrupt increase in fallout beginning early in March and reaching a peak in mid-April, after which the activity declined. (The last entry on the graph is for June 19.)

The fact that the spring increase is not due just to higher rainfall is clearly shown in Figure 7 where the specific strontium-90 activity, expressed in mc/mi^2/inch of rain, is seen to exhibit almost the same seasonal variation as the total strontium-90 fallout. The slow, irregular rise in activity from November through January may be partly due to residual tropospheric debris and partly the result of a slowly increasing stratospheric "drip-out", but the abrupt rise in early March can only mean that the transfer of radioactive debris from the stratosphere was suddenly accelerated, probably starting in February, and strongly suggest a meteorological cause for the spring maximum. This



ISOTOPES INC.

FIG. 6

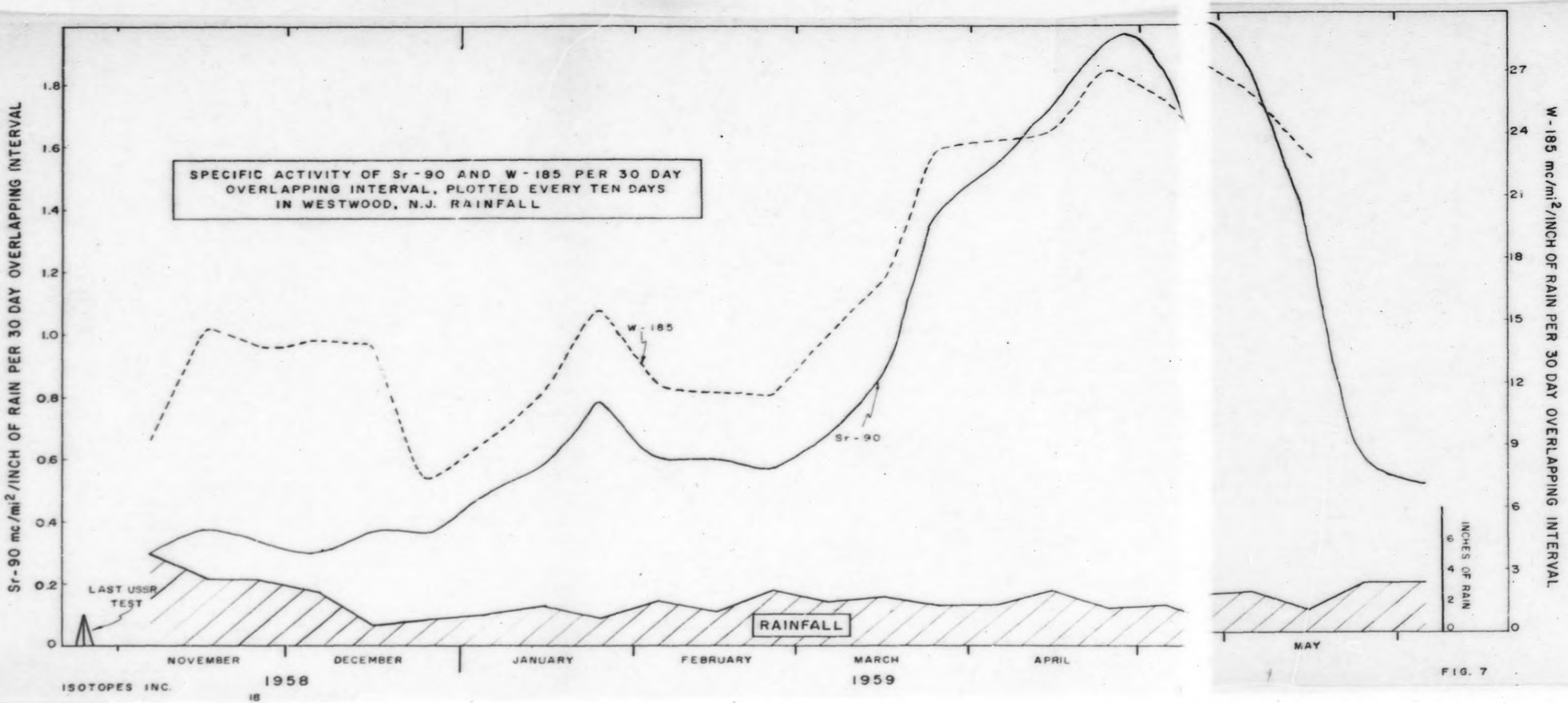

SPECIFIC ACTIVITY OF Sr-90 AND W-185 PER 30 DAY OVERLAPPING INTERVAL, PLOTTED EVERY TEN DAYS IN WESTWOOD, N.J. RAINFALL
Sr-90 mc/mi²/INCH OF RAIN PER 30 DAY OVERLAPPING INTERVAL
W-185 mc/mi²/INCH OF RAIN PER 30 DAY OVERLAPPING INTERVAL
W-185
Sr-90
LAST USSR TEST
RAINFALL
INCHES OF RAIN
NOVEMBER
DECEMBER
JANUARY
FEBRUARY
MARCH
APRIL
MAY
1958
1959

FIG. 7

conclusion is borne out by the curve of specific tungsten-185 activity on the same behavior as that for strontium-90, at least after December 1958. The tungsten curve provides clear evidence that the spring rise in fallout is not due merely to the Soviet testing schedule, but is rather a meteorological phenomenon, and is independent of the geographical origin of the debris.

The data from Pittsburgh, Pennsylvania, which are received through the monthly HASL reports, were analyzed in the same manner to determine if the seasonal variation there is consistent with that shown by the Westwood data. The tungsten-185 values were corrected for decay back to October 1, 1958 from mid-point of each of the overlapping 30-day intervals, rather than from each rain date. The results are shown in Figure 8.

Both the tungsten and strontium curves for Pittsburgh show the abrupt rise in specific activity at the beginning of March that was found at Westwood. The last observation available from Pittsburgh at the time of writing was for April 13, and it is not yet known if the specific activity reached a peak in late April, as it did at Westwood. The high tungsten activity in December, like the two winter maxima at Westwood, occurred during a period of very low precipitation, so that the specific activity values are questionable. The total monthly fallout of strontium-90 in March and April at Pittsburgh was more than four times as great as in December, and the total tungsten-185 fallout (corrected for decay) was twice as great.

The precipitation data for Richmond, California were also studied, but did not lend themselves to a similar analysis due to the extreme variability of the rainfall. However, the monthly fallout collections (pot data) from Richmond and from Houston, Texas, which were available through April 1959, have been examined with regard to the seasonal effect. The data for these stations are shown in Table 2.

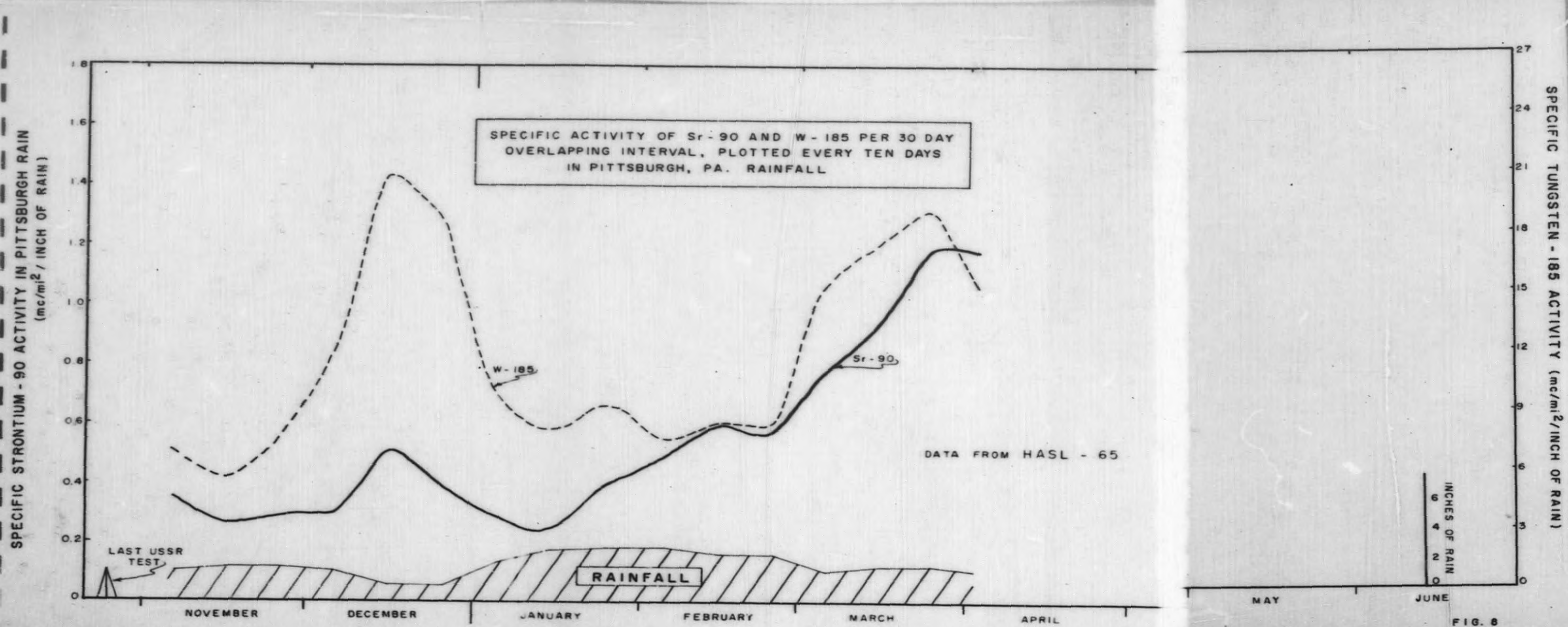

SPECIFIC ACTIVITY OF Sr - 90 AND W - 185 PER 30 DAY OVERLAPPING INTERVAL, PLOTTED EVERY TEN DAYS IN PITTSBURGH, PA. RAINFALL
SPECIFIC STRONTIUM - 90 ACTIVITY IN PITTSBURGH RAIN (mc/mi^2 / INCH OF RAIN)
SPECIFIC TUNGSTEN - 185 ACTIVITY (mc/mi^2/INCH OF RAIN)
W - 185
Sr - 90
DATA FROM HASL - 65
LAST USSR TEST
RAINFALL
INCHES OF RAIN
NOVEMBER
DECEMBER
JANUARY
FEBRUARY
MARCH
APRIL
MAY
JUNE
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
3
6
9
12
15
18
21
24
27
2
4
6

FIG. 8

Table 2. Total and Specific Strontium-90 and Tungsten-185 Activity from Monthly Pot Collections at Richmond, California and Houston, Texas. The tungsten data are corrected for decay back to October 1, 1958

A. Richmond, California

Month	Precip. (in.)	Sr-90 (mc/mi^2)	Sr-90 ($mc/mi^2/in.$)	Corrected to Oct. 1, 1958 W-185 (mc/mi^2)	Corrected to Oct. 1, 1958 W-185 ($mc/mi^2/in.$)
Dec. 1958	1.77	0.249	0.14	3.99	2.25
Jan. 1959	4.42	1.20	0.27	28.47	6.44
Feb. 1959	6.12	1.70	0.28	20.54	3.36
Mar. 1959	0.81	0.49	0.60	10.62	13.11
Apr. 1959	0.44	0.31	0.70	6.13	13.93

B. Houston, Texas

Month	Precip. (in.)	Sr-90 (mc/mi^2)	Sr-90 ($mc/mi^2/in.$)	Corrected to Oct. 1, 1958 W-185 (mc/mi^2)	Corrected to Oct. 1, 1958 W-185 ($mc/mi^2/in.$)
Oct. 1958	5.90	0.64	0.11	40.0	6.8
Nov. 1958	1.65	0.25	0.15	13.6	8.2
Dec. 1958	0.54	0.47	0.87	21.6	4.0
Jan. 1959	5.58	1.21	0.22	31.2	5.6
Feb. 1959	6.11	1.85	0.30	39.3	6.4
Mar. 1959	0.84	1.43	1.70	48.6	57.8
Apr. 1959	6.92	4.09	0.59	73.7	10.6

Both the tungsten and strontium data for Richmond and Houston show an abrupt rise in specific activity in March. Unfortunately the very low March rainfall at both stations casts doubt on the reliability of this result.

C. Calculation of the Equatorial Stratospheric Residence Time from Tungsten-185 Rainfall Data

The tungsten-185 produced in Operation HARDTACK has been monitored constantly in the rainfall at Westwood, New Jersey since its first appearance in May, 1958. From the data thus far developed, and a number of assumptions concerning total tungsten-185 yield, total tungsten-185 stabilized in the stratosphere and world-wide distribution of stratospheric fallout, a calculation defining the stratospheric half residence time of the tungsten-185 may be made. This computation is similar to the one made by Libby in a recent paper[11], in which he used the Pittsburgh, Pennsylvania data of November and December, 1958.

Total Tungsten-185 Yield

Although the actual yield of tungsten-185 produced in Operation HARDTACK is still classified at this time Libby[11] estimated 250 megacuries as of August 1, 1958. For the purpose of this calculation, all of the data will be normalized to June 1, 1958, thus it is assumed that effective June 1, 1958 the total yield was 404 megacuries of tungsten-185 produced.

Total Tungsten-185 Stabilized in the Stratosphere

A study of the actual altitude distribution of various clouds from surface water detonations in the high yield range, suggests that the best assumption of the amount which actually entered the stratosphere was between 20% and 40% of the total tungsten-185 yield from the HARDTACK tests. Based on the total yield of 404 mc described above, it is, therefore, assumed that no more than 162 mc of tungsten-185 stabilized in the stratosphere.

World-Wide Distribution of Stratospheric Fallout

In this calculation it is necessary to estimate the U. S. produced stratospheric fallout per square mile on the earth's surface relative to that in Westwood, New Jersey. In a recent Isotopes, Inc. report[10], it was calculated that from the inception of nuclear testing through June 30, 1958, a total of 2.1×10^9 millicuries of strontium-90 has fallen on the entire surface of the earth. This gives an average of 10.5 mc per square mile.

On the basis of data presented at the Congressional Hearings on Fallout before the joint Committee on Atomic Energy on May 5, 1959, revealing the dates and yield data of nuclear detonations, it would appear that about 10% of the total fallout through June 1958, could be attributed to U. S. S. R. tests. Thus a reasonable world-wide average from U. S. tests might be 9.4 mc of strontium-90 per square mile.

Using a combination of New York City and Westwood, New Jersey rainfall data[1], a total of about 50 mc per square mile of strontium-90 is calculated to have fallen in this area, through June 30, 1958. Since this number is based on data from the pot sampler, it is probably high by about 5 mc per square mile, due to resuspension of soil and dust (as described in a later section). Thus a more realistic figure for true fallout would be 45 mc per square mile. As a result of extensive soil measurements in the New York City and Philadelphia, Pa. areas in 1957 and 1958 by HASL[1] and Lamont Geological Observatory[12] an average value for the cumulative fallout as of June 30, 1958 may be calculated as 37 mc per square mile. If it is assumed that these soil analyses, which were carried out by the HCl extraction procedure, are low by 10%, the cumulative fallout value is raised to 41 mc per square mile, in fair agreement with the corrected pot data. On the basis of the foregoing a fair assumption for the total strontium-90 fallout in the Westwood area is taken as 43 mc per square mile. From this value it is now necessary to subtract all tropospheric fallout and also debris which originated in U. S. S. R. tests. It is difficult to estimate this correction without resorting to classified data. However, it would appear that based on the ratio of high yield detonations and the altitude of the bursts approximately 20% of the total on the ground on June 30, 1958 is of Russian origin. Assuming an additional 5% is due to purely tropospheric fallout, the calculation of the stratospheric residence time for equatorial shots is made using 75% of the total Westwood, N. J. fallout (.75 x 43 = 32.3 mc per mile of strontium-90).

Using the values derived above for stratospheric fallout from equatorial shots, the ratio of Westwood to the world average (32.3/9.4) gives a correction factor of 3.44 to apply to the observed tungsten-185 fallout on the earth's surface.

The tungsten-185 fallout for a full year (June 1, 1958 through May 31, 1959) measured at Isotopes, Inc. is shown in Table 3, on a monthly basis. These measurements are the cumulative values based on individual rain collections, and are believed not to be subject to the errors noted for the pot sampler.

Of this total (1230.5 mc/mi^2) it was assumed that all of the fallout in the months of June through September was tropospheric, the remainder being stratospheric. This assumption appears reasonable on the basis of the widely accepted 30-day mean washout rate of the troposphere. This correction results in 1168 mc/mi^2 of tungsten-185 being assigned to stratospheric fallout in Westwood, New Jersey through May 31, 1959. Dividing this number by the correction factor 3.44 obtained earlier for strontium-90 fallout gives a value for the average tungsten-185 stratospheric fallout of 339.6 mc/mi^2 and a total (2×10^8 mi^2 on the earth's surface) fallout of 68 MC of tungsten-185 on the entire earth. Subtracting this from the 162 MC of tungsten-185 injected into the stratosphere by the HARDTACK tests, it is seen that on June 1, 1959 only 94 mc

Table 3. Tungsten-185 Fallout in Westwood, New Jersey Rainfall (All values are normalized to June 1, 1958)

Month	W^{185} mc/mi^2
June 1958	6.0
July 1958	25.4
August 1958	48.4
September 1958	62.5
October 1958	111.9
November 1958	117.5
December 1958	171.8
January 1959	112.0
February 1959	91.0
March 1959	176.5
April 1959	278.5
May 1959	108.8

Total for the 12 months = 1230.5 mc/mi^2.

was still in the stratosphere. These values indicate a stratospheric half residence time for the HARDTACK produced tungsten-185 of about 16 months. For clarity, the calcclation is illustrated stepwise on Table 4.

The 16 month half residence time calculated here is very likely a maximum value, based on the following considerations:

(a) There is evidence that less than 40% of the original yield of tungsten-185 stabilized in the stratosphere. Any lesser fraction would shorten the T 1/2 calculated.

(b) It is likely that of the total strontium-90 deposited at Westwood, New Jersey less than the assumed 75% is of equatorial stratospheric origin. Again a lesser amount would shorten the residence time.

(c) The assumption that none of the tungsten-185 in Westwood fallout for June through September of 1958 was stratospheric is probably extreme, and likewise maximizes the T 1/2 calculation.

D. The Validity of the Pot Sampler

For some time the data developed in the various rainfall collection projects have indicated a puzzling inconsistency between the values obtained from the steep-walled stainless steel pots, and those found in either soil or individual rain analyses. In the New York City area, where fallout in rain has been monitored consistently since early 1954 and periodic soil samples have been carefully analyzed, it can be seen[1], that in all of the cases tested, the soil was found to be lower in strontium-90 than the cumulative rain. The most recent soil data for the New York Area sampled in October 1958 give the most puzzling results. The data are summarized in Table 5. These comparisons seem to indicate a fairly systematic difference which could be partially attributable to incomplete recovery of strontium-90 from the soil.

Taking another line of evidence, further puzzling results are shown by comparison of the pot data and the individual rain collection data for the past 18 months in Westwood, New Jersey. Table 6 summarizes the data. In two

Table 4. Calculation of Stratospheric Half Residence Time for W^{185}
(All W^{185} Values are Corrected to June 1, 1958)

1.	Total Sr^{90} fallout in Westwood, N. J. through June 30, 1958	43 mc/mi^2
2.	Assumed percent of Westwood, N. J. Sr^{90} fallout, U. S. stratospheric origin	75%
3.	U. S. stratospheric Sr^{90} fallout in Westwood, through June 30, 1958	32.3 mc/mi^2
4.	World average Sr^{90} U. S. stratospheric fallout	9.4 mc/mi^2
5.	Westwood/world fallout correction factor (f)	3.44
6.	Westwood, N. J. W^{185} stratospheric fallout (June 1, 1958-May 31, 1959)	1168 mc/mi^2
7.	Westwood W^{185} fallout/f = average world stratospheric W^{185} fallout	339.6 mc/mi^2
8.	Total world stratospheric W^{185} fallout (June 1, 1958-May 31, 1959)	68 MC
9.	Total W^{185} stabilized in the stratosphere after detonation	162 MC
10.	W^{185} remaining in the stratosphere after 1 year	94 MC
11.	Stratospheric half residence time for HARDTACK produced W^{185}	16 months

Table 5. Comparison of Soil and Cumulative Pot Data for the New York Area
Data Taken from HASL 65

	Strontium-90, mc/mi^2	
	Soil	Pot
1955	12.4	15.3
1956	24.0	26.0
1957	33.9	37.8
1958	36.6 (N.Y.C.) 44.0 (Phila.)	50.0

Table 6. Summary of Pot and Individual Rain Data

Collection Period	Sr^{90} (mc/mi^2)		Inches of Rain
	Individual Rains	Pot	
March 1958	0.75	1.00	3.88
April 1958	1.61	1.73	7.04
May 1958	2.72	2.80	4.36
June 1958	1.07	1.05	2.72
July 1958	0.76	1.10	3.19
August 1958	0.88	1.11	2.85
September 1958	0.52	0.70	3.86
October 1958	1.12	1.64	5.26
November and December 1958	2.07	2.17	6.53
January and February 1959	2.85	2.85	4.85
March 1959	2.76	4.34	3.76
April 1959	6.17	6.35	3.23
May 1959	1.85	2.63	1.08
June 1959	3.21	3.78	3.27
Total	28.35	33.22	

cases the table combines the values for two months in order to eliminate confusing numbers due to rainfall over weekends, which also occurred at the end of the months. Although some of the comparisons are within the probable experimental errors (generally not greater than 5%), the observation that all of the pot data in Table 6 are either equal to or greater than the individual rain values suggests that some systematic phenomenon is responsible.

The most apparent difference between the two modes of rain sampling is that in most months, after the first rain has fallen, some water remains in the bottom of the pot throughout the remainder of the month. Thus, for a sizeable fraction of the collection period, the pot presents a wet collection surface. The individual rain collector, on the other hand, while much larger in over-all area, presents a dry surface at all times except during actual precipitation. This comparison suggests that any resuspended soil or dust would be held in the water in the bottom of the pot and build up in concentration over the sampling interval, to a much greater degree than would occur in the individual rain device. It has not been possible to relate the effect unambiguously to either total rainfall per month or number of rains, although there is a suggestion of a partial correlation, i.e. March 1959 had the lowest rainfall of any month in the period shown in Table 6.

While no quantitative estimate of the magnitude of the inaccuracy of the pot sampler can be stated, the two lines of evidence i.e. soil and individual rain collections versus pots, suggest that use of the monthly pot data may have caused the total fallout for the New York and Westwood, N. J. areas to be overestimated by about 10%.

IV. CONCLUSIONS AND RECOMMENDATIONS

Four specific areas of interest related to fallout of nuclear debris associated with rainfall have been examined in the course of the year's first three months of this project. The conclusions based on this effort may be summarized as follows:

1. Analysis of strontium-90 and tungsten-185 debris in rainfall clearly indicates a spring high in fallout, related to neither the amount of rainfall nor the U. S. S. R. test schedule. The cause of the well-known spring increase must be meteorological in nature, i.e. an acceleration of the stratosphere-troposphere transfer rate, independent of the initial source of the debris.

2. The stratospheric half residence time of the HARDTACK produced tungsten-185 has been calculated by use of Westwood, New Jersey rainfall data and assumptions as to stratospheric yield and world-wide fallout distribution, resulting in an upper limit value of 16 months. This value is in good agreement with estimates of the stratospheric half residence time made in the High Altitude Sampling Program[10], for U. S. equatorial high yield debris.

3. Observation of soil, pot, and individual rain collection data suggests that the widely used pot sampler probably gives values higher than true fallout by upwards of 10%, due to contamination by resuspended soil and dust.

4. Data of fractions of rain tentatively suggest that "cleansing" is more efficient for heavy then for light rains. The variation in specific activity of samples noted in all of the four cases tested suggests an initial cleansing of the air followed by replenishment of radioactivity.

As a result of the work reported above several specific recommendations may be made pertinent to increasing the productivity of the rainfall program:

1. To finally understand the mechanism of rainfall scavenging of nuclear debris, a sustained program of fraction rain collection and analysis should be initiated. In addition, at the site of this project a system for sampling large volumes of air should be developed, to enable monitoring of the ground level air before, during and after rains.

2. Continued investigation of the relationship of strontium-90 in soil, pots, individual rain collectors and the ion-exchange collector should be carried out to clarify the inconsistencies in cumulative fallout data. To do this comprehensive soil sampling programs should be initiated at the stations where all of the rain samplers are operative.

3. To enable continued observation of the HARDTACK tracer, lower levels of tungsten-185 detection should be developed and/or

analysis for tungsten-181 in all of the rain samples.

4. Routine measurement of tritium in rainfall might shed light on the various theories of stratospheric mixing and residence times in addition to other useful parameters of geochemical interest, as suggested by Libby[13].

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