Deposition of Nuclear Debris and Atmospheric Conditions

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Since any nuclear test explosion had not been conducted during the period from November 1958 to January 1960 and consequently there were no substantial additions of relatively short-lived fission products to the atmosphere by fresh test explosions, it is supposed that the total amount of the nuclear debris from weapon tests in the atmosphere should remain unaltered except for the removal of nuclear debris by the natural deposition to the ground, and that the period was the most useful one for the purpose of studying the rate and mechanism of deposition process of the nuclear debris. As it has been shown in many papers, the rate of deposition of nuclear debris varies considerably at different places and at any particular place at different times of the year. Then in the present paper it is at first studied how the activity concentration of total fission products in rain-water depends upon the atmospheric conditions under which a series of rainfalls occur being accompanied with an extratropical cyclone, for example, the path of a cyclone and the meteorological character of rainfall. Secondly, from the correlation between the day-to-day variation in the fallout activity and the corresponding surface and upper meteorological conditions, some interpretations are given for the mechanism of removal of the nuclear debris from the atmosphere.

Part I. Specific Activity of Nuclear Debris in Rain-water and Weather Conditions near the Earth's Surface.

1. Classification of Radioactive Rain. The meteorological character of rain is generally quite complicated, but to the first approximation rainfalls may be classified at least into the following two types according to the weather conditions, under which each rainfall occurs; the "C-type" and the "W-type". These are shown shematically in Fig. 1. A C-type of rain is a cold front type of rain and is formed in the rear side of a low-pressure. In this case the clouds observed are of the vertically well-developed cumulus or cumulo-nimbus type, showers occur at frequent

intervals and an appreciable extent of mixing of air mass between the lower and higher levels is expected to occur owing to the local turbulent motion, as is described below in detail. The paths of the extratropical cyclones which attend this type of rainfalls in Niigata are indicated in the full line in Fig. 2. Generally this type of rain had a higher value of the specific activity of nuclear debris in precipitation (the radioactivity of total fission products per unit precipitation).

A W-type of rain is of a warm front type. The rain area is wide in advance

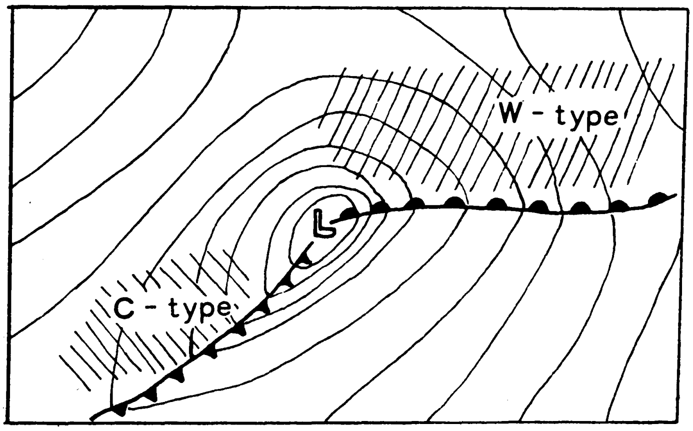


Fig. 1. The C-type and W-type of rainfalls being accompanied with an extratropical cyclone; the cold front is indicated by pointed teeth, the warm front, by rounded teeth and the shaded portion shows the rain area.

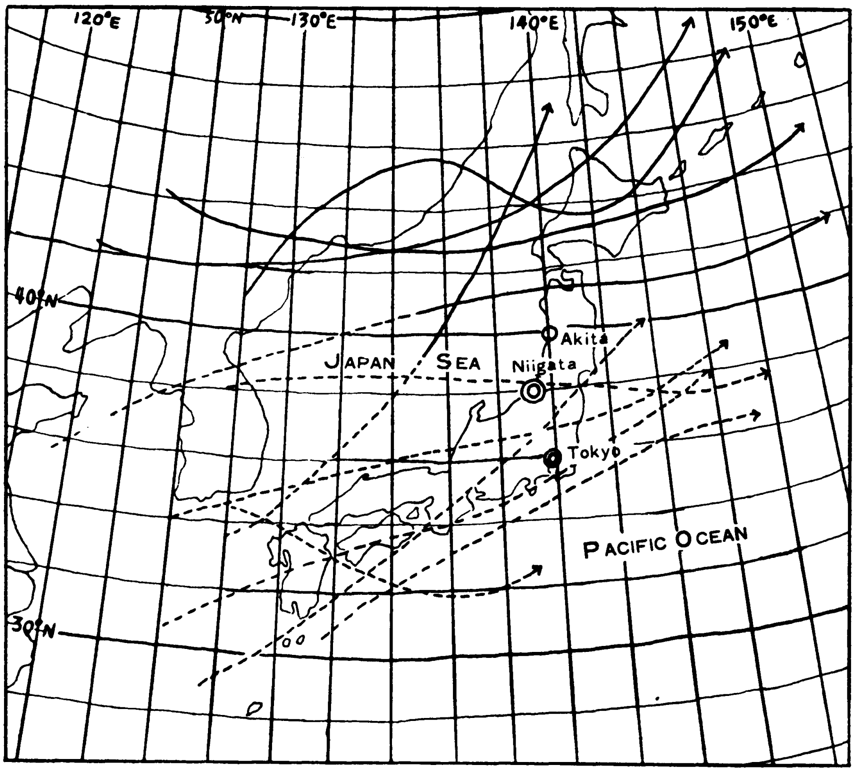


Fig. 2. The paths of extratropical cyclones which referred to radioactive rainfalls during the months March-May, 1959.

of a cyclonic disturbance. This type of rain has the character of steady rain and its rainfall rate is usually more than several times that of a C-type. In this case the clouds observed are of the nimbo-stratus type and the vertical motion of air mass is not violent from the feature of frontal structure of low-pressure. The paths of the cyclones which refer to this type of rainfall in Niigata are shown in the broken line in Fig. 2. Generally this type of rain had a lower value of specific activity of rain.

The rainfalls which occurred from 10 March to 30 June, 1959 are classified into the two types, and each type of monthly total rainfall and the ratio of monthly rainfall of the C-type to that of the W-type are presented in Table 1.

Table 1. Classified monthly rainfall and the ratio of the rainfall of C-type to that of W-type

Period of observation	C* (mm.)	W* (mm.)	C/W	C+W (mm.)
March**	55.4	56.0	1.0	111.4
April	46.0	40.6	1.1	86.6
May	55.5	37.8	1.5	93.3
June	29.4	57.2	0.5	86.6
March-June	186.3	191.6	0.9	377.9

* The figures in the columns denoted by 'C' and 'W' refer to the C- and W- types of rainfalls, respectively.
** Data in March are those observed from 10 to 31.

2. Measurement of Radioactivity of Fallout Sample. During the period from 10 March to 30 June, 1959, fallout samples were collected in a polyethylene-vat from day to day. Each sample was evaporated up and the radioactivity of its residue was measured by an end-window type of G–M tube.

3. Monthly Amount of Fallout Activity. The sum of daily fallout activities in each month is separately presented in Table 2 according to the type described above. It is obvious that to reduce the activity value to a form comparable with a fixed value, allowance is made for the inequality of sampling date; that is to say, all activity values in this paper are reduced to the value on 30 June, 1959. The corrections are applied for decay using the semi-monthly average decay curves which were obtained by the decay measurements of the other fallout samples collected simultaneously at the same site. These decay curves are shown in Fig. 3. These curves show that their half-lives were 110 ± 10 d. during those months.

As is well-known, for long-range fallout of nuclear debris injected originally into the stratosphere the main process of deposition is the wash-out in rain-water after entry into the troposphere, and the amount of fallout activity is approximately proportional to amount of precipitation. However, from the values in Table 2 the amount of fallout activity carried down with a C-type of rainfall was on the average 3.6 times that done with a W-type of one. In comparing each monthly total amount of fallout activity (W+C, in Table 2) with others observed during the months April-June, the value in May was 1.8 and 1.5 times those in April and June, respec-

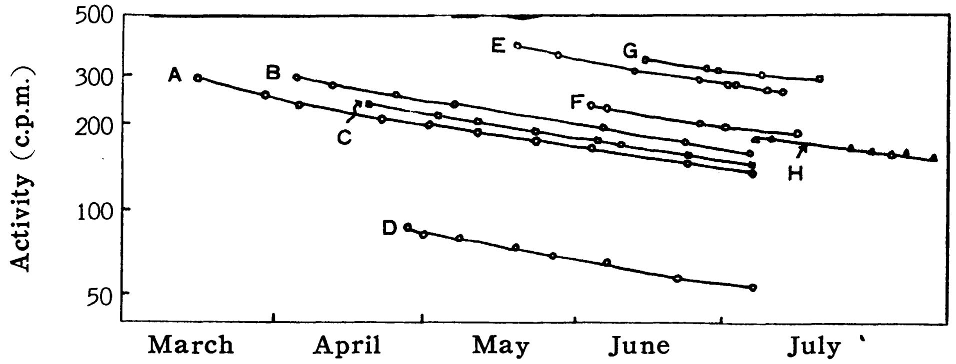


Fig. 3. Semi-monthly average decay curves. A, C, E and G indicate the decay curves of the first halves of Mar., Apr., May and Jun., respectively; B, D, F and H, the second halves of the same months.

tively, though their monthly total rainfalls were all similarly about 90 mm. This means that an increase in the amount of fallout activity is not always proportional to an increase in amount of precipitation. In other words, there appears to be no

Table 2. Classified monthly total fallout activity

Period of observation	C (d.p.m./m².mo.)	W (d.p.m./m².mo.)	C/W	C+W (d.p.m./m².mo.)
March	158,700	34,900	4.6	193,600
April	92,800	26,500	3.5	119,300
May	178,200	23,300	7.7	201,200
June	80,300	56,200	1.4	136,500
March-June	510,000	140,600	3.6	650,600

doubt that the settling speed of nuclear debris in the troposphere depends upon the meteorological conditions near the Earth's surface. Therefore, some interesting results have been derived from a detailed examination of the data of observation on the

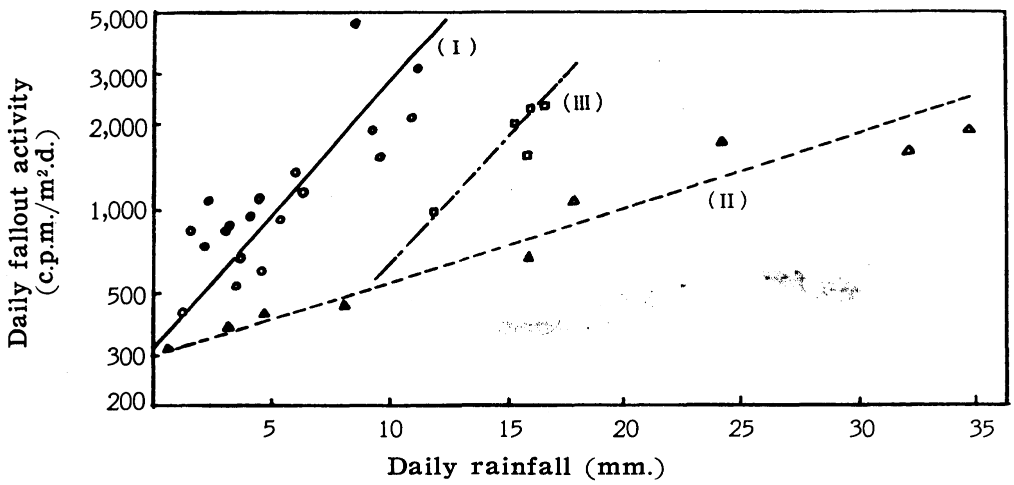


Fig. 4. Relation between daily fallout activity and daily rainfall.

daily rainfalls and daily fallout activities.

In Fig. 4. the activity of daily fallout is plotted on a logarithmic scale against the daily rainfall during March-May, 1959. In this figure it may be seen that the points plotted fall into at least two separate groups, probably three ones, which are indicated by the straight lines I, II and III. Within each group a linear relationship between the daily fallout activity and daily rainfall is approximately followed, but the slopes of the first two lines are quite different. Thus, it appears that the weather conditions under which rain occurred are also to be classified into at least two different types, one of which corresponds to a cold front type (C-type) and the other of which does to a warm front type (W-type), as these types are described in section 1. These two lines intersect the ordinate at a nearly same point and a mean value of the two intercepts seems to be in good agreement with a mean value of dry-fallout activity for that period, that is 300 c.p.m./m^2.d. The third line which refers to the line-III has a nearly same slope as that of the line-I, but intersects the line-II. This may be understood as follows: the rainfalls which refer to the points distributed near the line-III were formed under mixed weather conditions of the two types described above. As will be discussed below, further detailed consideration about the relation between the rainfall and the fallout activity is of help in interpreting the mechanism of deposition process for nuclear debris in the troposphere.

4. Specific Activity of Nuclear Debris in Rain-water. In Table 3 are shown the monthly average values for the specific activity of rain-water and the ratio of the specific activity of a C-type of rain to that of a W-type of rain. These values are evaluated from the values in Tables 1 and 2. In the month of May there was the highest value concerning the C-type of rain and also was found in the same month the effective maximum value* of specific activity of rain-water during the whole observation period; i. e. the value was 15,830 d.p.m./mm. and its rainfall was 8.5 mm. Concerning the values of a W-type of rain, the maximum was found in the month of June, but this may be due to the incompleteness of classification of rain. This shows the limits within which accuracy of such a classification as above is possible or desirable.

5. Time-variation of Specific Activity in a Series of Rainfalls. It has been shown in the preceeding sections that the specific activity of rain observed in the north-eastern sides of a cyclonic center is lower and in the south-western sides, higher. Therefore, it is to be expected that when a series of rainfalls occur being accompanied with an extratropical cyclone which passes from west to east over the

* When the rainfall is less than 1.0 mm., the specific activity of rain shows a higher value, even if the total amount of fallout activitiy is not always large; i. e. 0.3 mm., 32,800 d.p.m./mm. on 22 March, 1959. While the specific activity of rain gives a high value in this case, the contribution of activity to the monthly total amount of fallout activity is insignificant. Therefore, with such exception as this case the maximum value is called the 'effective maximum value'.

north side of an observation point, initially a rainfall of lower specific activity occurs and finally a rainfall of higher one follows. In this case the former is of a W-type and the latter, of a C-type. As is expected, such change in the specific actitviy of rain with time was frequently observed and the data obtained provide further confirmation of the validity of the correlation between the type of rain and the value of specific activity of rain. Some typical examples are presented in Table

Table 3. Classified monthly specific activity of nuclear debris in rain-water

Period of observation	C (d.p.m./mm.)	W (d.p.m./mm.)	C/W	C+W (d.p.m./mm.)
March	2,865	642	4.6	1,697
April	2,020	654	3.1	1,473
May	3,210	608	5.3	2,156
June	2,730	983	3.0	1,360
March-June	2,740	762	3.6	1,675

4. In this table a series of rainfalls which occurred during 12–13 May are the most typical.

When a C-type of rainfall occurs as shown in this case a percentage of the activity contribution to the monthly total amount of fallout activity was sometimes rather large, even though the corresponding rainfall was smaller. In Table 4 in the

Table 4. Time-variation in the specific activity of rain

	Period of observation	Rainfall (mm.)	Specific activity (c.p.m./mm.)	Ratio of spesific activity
[A]	9.00, 12/4– 9.00, 13/4	15.9	41	1 : 6.7
	9.00, 13/4– 9.00, 14/4	3.2	270	
[B]	9.00, 4/5–10.00, 5/5	31.6	41	1 : 3.2
	10.00, 5/5– 9.00, 6/5	4.0	130	
[C]	9.00, 12/5–18.00, 12/5	8.0	129	1 : 2.0 : 4.4
	18.00, 12/5– 9.00, 13/5	15.3	253	
	9.00, 13/5– 9.00, 14/5	8.5	567	

cases of (B) and (C) the specific activity of rainfall which occurred finally was three or four times that of a rainfall which occurred initially, and the rainfalls were about 30 mm. in the both cases, but the average specific activity of (C) is three times that of (B). This difference may be attributed to the upper air conditions over surface lows and therefore, we will discuss the relation between the specific activity of rain and the atmospheric factors in the higher levels in Part II.

6. Effective Maximum Value* and Minimum Value in Specific Activity of Rain. The day-to-day variation in the specific activity of rain is complicated and the complete information on the factors which bring about this variation is not established in the present. However, as a helpful indication, in Table 5 are presented the effective maximum value and the minimum one in each month during

* See Note on pp. 87.

March-June, 1959. In this table it should be noticed that the total amount of fallout activity of the rain which gave an effective maximum value was more than several times larger than that of the rain which gave a minimum value, though the rainfall

Table 5. Daily effective maximum and minimum values of specific activity of rain

Period of observation	Effective maximum value (c.p.m./mm.)	Minimum value (c.p.m./mm.)	Ratio of maximum to minimum
March	268 (11.3)*	48 (32.0)*	1 : 5.3
April	222 (6.0)	41 (15.9)	1 : 5.3
May	567 (8.5)	41 (30.0)	1 : 13.5
June	196 (15.1)	59 (10.0)	1 : 3.3
March-June	567 (8.5)	41 (30.0)	1 : 13.5

* Figures in parentheses show rainfalls (mm.) of the days which gave these extreme values.

of the latter was several times larger than that of the former. In other words, the proportionality of the amount of total fallout activity to the amount of local precipitation was not held in this case. Further it will be seen that the specific activity of a particular rainfall happened to show a very large value, which was more than ten times as a minimum one.

7. Discussion. It is noticed that the average specific activity of a C-type of rain was 3.6 times that of a W-type of rain, during March-June, 1959, as indicated in Table 3. To an explanation for the variation in the specific activity of rain there are some factors which have to be taken into consideration. One of these factors is the air concentration of nuclear debris in the layer where condensation and precipitation occur. The wash-out process in precipitation is considered as a process that the fallout particles are at first adsorbed to such small particles of hygroscopic substances as available for nuclei of cloud particles and that thereafter the dust particles are captured by cloud particles as nuclei or directly by being in collision with raindrops or snowflakes. Consequently, it is generally supposed that the higher the air concentration of nuclear debris is, the larger the specific activity of rain is. Thus the measurement of the specific activity of rain may serve to obtain an indication of average concentration in air of fallout particles in the rain-producing layer, with the exception that the rainfall is extremely large or small. Therefore, that a C-type of rainfall has a higher value of specific activity, suggests that both the following two atmospheric processes take place or at least either of them occurs over an observation point. The first is that there exists a vertical exchange process of air mass on a large scale and as a result of this process, for example, the travelling ranges of cloud particles and raindrops are longer through the air mass contaminated with the nuclear debris. The second is that there exists a process that leads to increase the air concentration of debris in the rian-producing layer and in addition, to maintain the high concentration in air of nuclear debris

during the rainfall interval.

Concerning the first process, this important feature of the exchange process of air mass between the upper and lower layers over the cold frontal surface was made to be sure by the studies of the concentration in rain-water of short-lived daughter products of radon——radium-A, -B and -C. They are naturally present in the lower atmosphere as a result of the leakage of radon from soils and rocks on the Earth's surface. That is, these isotopes were used as tracers to study the mechanism of transport of air mass from lower layer to upper in the troposphere, because it is obvious that their main resident layer lies in the lower levels of the troposphere. Some typical data obtained are presented in Table 6. It is easily seen from Table 6 that a cold front type of rain had also several times larger value of activity concentration than that of a warm front type of rain, the difference of the both rainfall-rates being taken into consideration.

Table 6. Activity concentration of daughter products of radon in rain-water

Type of rainfall	Mean activity concentration (10^{-8}c./1.)	Mean rainfall-rate (mm./hr.)	Date of collection
Warm front type	3.8	3.76	Mar. 26, 1960
Cold front type	19.0	1.96	Mar. 25, 〃

Generally the main radioactivity in rain-water is a combination of two different components, one of which is the raidoactivity of the long-lived fission products coming from the uppermost layer and the other of which is the short-lived (30 min.) daughter products of radon coming from the lower layer. Consequently, on the basis of the fact that these two kinds of radioactive air-born dusts lead to be enriched simultaneously in a cold front type of rainfall, it is acceptable that the high value of specific activity in a cold front type of rainfall reflects a picture of the vertical exchange process of air mass over the cold frontal surface. According to the theory of the cold front the existence of such process as mentioned above can be naturally expected, but concerning the second process here nothing has been said above to this point. It will be discussed in Part II in detail.

Summary

The measurements of fallout activity were made from day to day during the period 10 March—30 June, 1959. From the results obtained we have given some explanations about the rate of fallout at a place far from the site where nuclear explosion tests were conducted. The deposition rate of nuclear debris depends greatly upon the atmospheric conditions near the Earth's surface, for example, the meteorological character of precipitation and the path of an extratropical cyclone.

It is found that the activity concentration of total fission products in a cold front type of rainfall is on the average several times larger than that of a warm front type of rainfall. This fact is also confirmed by the measurements of activity concentration in rain-water of the short-lived daughter products of radon which are present in the lower layer of troposphere.

Part II. Atmospheric Behavior of Nuclear Debris and Upper Air Conditions.

It is described in Part I that the weather conditions near the Earth's surface are important factors to which the day-to-day variation in the specific activity value of rain closely relates. An interesting conclusion has been deduced, that is, the specific activity* of rain is influenced greatly by meteorological character of rainfall. However, as is stated in section 7 in Part I, nothing has been said concerning a mechanism of the downward transport of fission products from the uppermost layer of troposphere to the lower layer where condensation and precipitation occur.

1. Day-to-day Variation in Specific Activity of Rain and Altitude of Constant-pressure Surface at Higher Level. As an indication of changes which are taking place in the upper air, attention is turned to the changes in the 300- and 500-mb. constant-pressure surfaces. For the former, change of meteorological factors at 300-mb. level closely relates to the behavior of an air flow in the vicinity of the tropopause and for the latter, a synoptic chart at 500-mb. level readily comes to hand. In Figs. 1-a, -b and -c the altitudes of 300- and 500-mb. surfaces observed at the Akita Observatory*1 (140°06′ E; 39°43′ N.) are presented as a function of time during March-May, 1959. The reduced amount of total fallout activity and the reduced mean specific activity for a series of rainfalls observed at Niigata (139° E; 38° N) are presented in Table 1. T_i and R_i in this table refer to the i-th valley and peak on the altitude vs. time curves plotted in Figs. 1-a, -b and -c, respectively, in each month. The rain occurred corresponding to these portions of the curves.

The factors that bring about the marked difference** in the specific activity of rain may be more reasonably interpreted by studying the correlation between the specific activity and the day-to-day variation in the altitudes of constant-pressure surfaces at higher levels. Then, for example, we compare the specific activity with each other in the cases of two rainfalls which occurred on 4-5*2 and 12-13*2 May,

* The radioactivity of total fission products per unit precipitation.

** See Table 4 in Part I, pp. 88.

*1. As the Niigata Meteorological Observatory does not perform the upper air observation, the data of the altitudes of 300- and 500- mb. levels are these observed at Akita which is the nearest and meteorologically the most similar place to Niigata.

*2. The pressure distribution of these days at 500- mb. level are presented in Figs. 2 and 3.

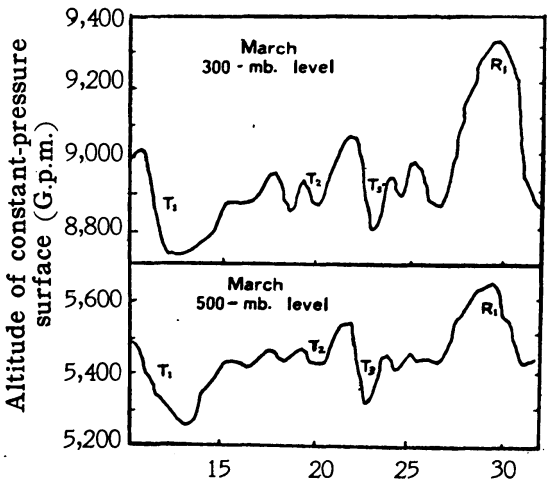


Fig. 1-a. Altitudes of 300- and 500- mb. surfaces vs. time in March, 1959.

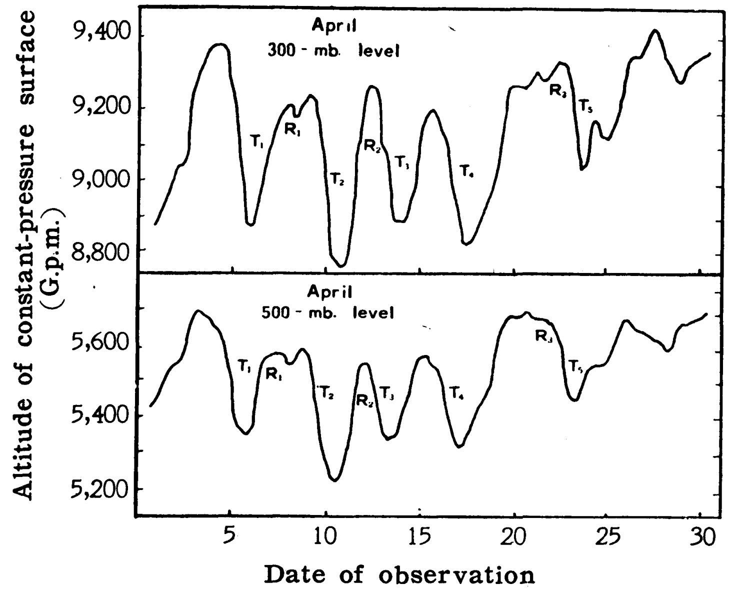


Fig. 1-b. Altitudes of 300- and 500- mb. surfaces vs. time in April, 1959.

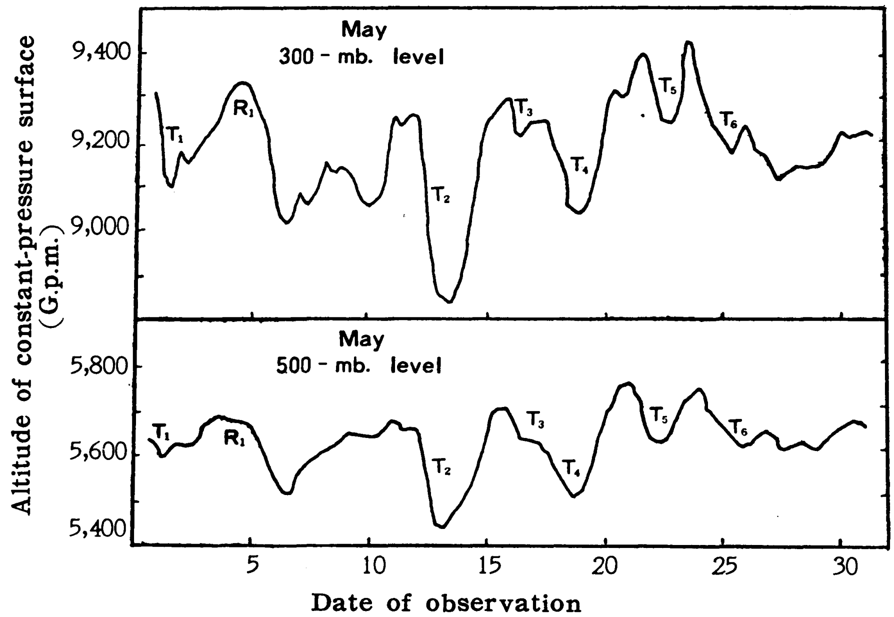


Fig. 1-c. Altitudes of 300- and 500- mb. surfaces vs. time in May, 1959.

1959. Any marked change was not found at the portion (R_1) which corresponded to the former case on the curves plotted in Fig. 1-c, that is, when a value of specific activity of rainfall is lower, no critical change is recognized in the altitudes of constant-pressure surfaces at higher levels. On the other hand, a sudden fall of the altitude was recognized at the portion (T_2) which corresponded to the latter case on the curves in the same figure, that is, a sudden fall of the altitude of isobaric surface at a higher level results in producing a rainfall of higher specific activity. These correlations were further confirmed by studying in detail the data listed in

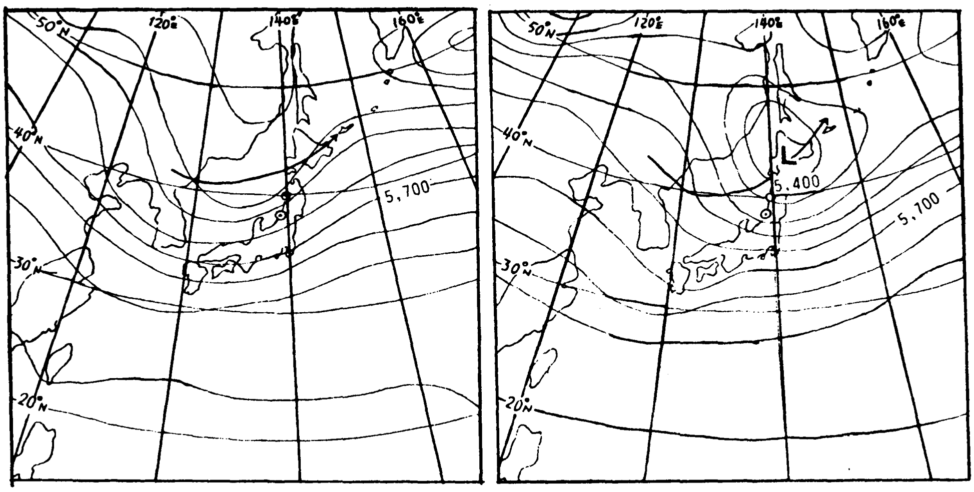
Table 1 in comparison with the curves plotted in Figs. 1-a, -b and -c. This leads to the following assumption: when a remarkable fall of the altitude of constant-pressure surface at a higher level takes place over an observation point, the rainfall observed shows a higher value of specific activity; on the other hand, when a small or no change of the altitude is observed over an observation point, the rainfall observed shows a lower value of specific activity. Since a fall of the altitude of constant-pressure surface at a higher level is, in many cases, caused by the passage of a trough line at a higher level, it will be useful to investigate the shape of contours that pass over an an observation point. However, the rainfall which occurred on 22 March and referred to the portion (T_3), one of minimums of the curve plotted in Fig. 1-a, showed a lower specific activity which was less than one-third, as compared with those of the others (T_1, T_2). It was rather a similar value to that of the rainfall which referred to the peak (R_1) of the curve. This was only an exception for the assumption above mentioned during March-May, 1959.

Table 1. Reduced mean specific activity of total fission products in rain-water during March-May, 1959

Date of collection		Rainfall (mm.)	Total fallout activity (c.p.m./m².)	Specific activity (c.p.m./mm.)
T_1	10–14/3	22.9	7,695	336
T_2	19/3	10.6	2,360	222
T_3	22/3	24.0	1,750	73
R_1	30–31/3	41.9	3,025	72
T_1	4– 5/4	20.0	2,957	148
T_2	9–11/4	12.9	2,800	217
T_3	13–4	3.2	860	270
T_4	16–17/4	7.0	1,520	217
T_5	23/4	3.5	654	154
R_1	8/4	8.1	450	56
R_2	12/4	15.9	654	41
R_3	22/4	11.9	945	79
T_1	1/5	6.6	2,080	316
T_2	12–13/5	31.7	10,470	331
T_3	16/5	2.2	727	330
T_4	18–19/5	5.5	1,452	264
T_5	22/5	6.3	1,160	182
T_6	25/5	4.5	1,090	242
R_1	4– 5/5	35.6	1,820	51

2. Pressure Distribution at 500-mb. Level and Specific Activity of Total Fission Products in Rain-water. At first a general conclusion will be described concerning the correlation between the pressure distribution and the specific activity of total fission products in rain-water. When there is a trough in the upper layer over an observation point and the contour which passes overthere has a large component in the north-south direction, surface lows passing along a V-course in the north side of an observation point, as it often occurs in winter time in Niigata, a series of rainfalls observed on the Earth's surface show generally a higher value

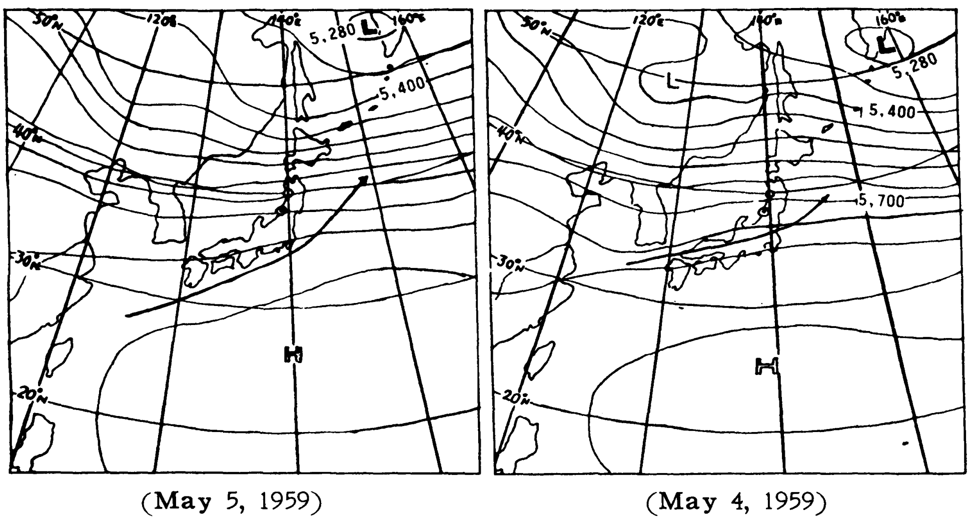
of specific activity and involve inevitably a C-type of rain*3. Thus this type of pressure distribution is called the 'T-type' and typical examples are presented in Fig. 2. The paths of surface lows are shown by heavy line in these figures. On



(May 12, 1959) (May 13, 1959)

Fig. 2. T-type of pressure distribution at 500- mb. level, ⊙, Niigata; ○, Akita.

the other hand, when there is a ridge in the upper layer over an observation point or when the contours which pass overthere lie approximately in the east-west direction, surface lows passing through from west to east just above or in the south side of an observation point, a series of rainfalls show generally a lower value of specific activity and this type of rainfalls observed on the ground-level are frequently of W-type only.*3 This type of pressure distribution is called the 'R-type' and typical examples are presented in Fig. 3. The paths of surface lows, for example, are shown by heavy line in these figures.



(May 5, 1959) (May 4, 1959)

Fig. 3. R-type of pressure distribution at 500- mb. level ⊙, Niigata; ○, Akita.

*3 This journal, Series I, Vol. 3, No. 2, 83 (1962)

It will be easily seen according to the fundamental theory of meteorology that the path of a surface low is a V-course, when a synoptic chart at a higher level shows a T-type of pressure distribution over an observation point. For it is accepted that a center of surface low usually follows the course of upper air current, that is, a cyclonic center moves generally along the contours plotted on a chart at a higher level. Accordingly, the relation between the path of a surface low and the upper air conditions for Niigata is considered as follows; when a chart at 500 mb. level shows a T-type of pressure distribution, the path of a surface low takes a V-course, that is, a surface low which comes from the Siberian Continent and produces a rainfall around Japan, passes across the Japan Sea in the north sides of Niigata. Consequently such a series of rainfalls that are formed in the south-western side of cyclonic center are invariably of C-type*4. On the other hand, when a chart at 500-mb. level shows a R-type of pressure distribution, a surface low takes generally a straight course from west to east and it is to be expected that a surface low which comes from the south-western sea of Japan and produces a rainfall around Japan, passes just above or in the south side of Niigata. Consequenty such a series of rainfalls that are formed in the north or north-eastern side of a cyclonic center are of W-type*4 in many cases.

3. Upper Air Conditions and Deposition Mechanism of Nuclear Debris. From the fact that the specific activity of rain also relates closely to the type of pressure distribution at a higher level, some considerations will be given here concerning a deposition mechanism of nuclear debris. Recently, concerning the behavior of the upper air current Miyake and Kawamura*5,6 have pointed out that the air stream within a trough in the upper layer has generally a southward component being accompanied with a sinking motion, on the basis of the correlation between the day-to-day variation in total ozone and the meteorological conditions at higher levels at Tokyo, Japan.

From this result Miyake *et al.**7 have suggested that the deposition rate of nuclear debris will increase when a trough in the upper layer passes over an observation point and have proposed the 'suction process' as a deposition mechanism of the tropospheric strontium-90; that is to say, tropospheric debris, which re-entered through a break of the tropopause from the stratosphere, comes back to the ground surface being carried with an air current that flows down along the polar

*4. This journal. Series I, Vol. 3, No. 2, 84 (1962).
*5. Miyake, Y., Kawamura, K., "Studies on Atmospheric Ozone at Tokyo," Sci. Proc. Intern. Assoc. Meteorology, Tenth General Assembly, Rome, Sep., 1954, pp. 172
*6. Miyake, Y., Kawamura, K., "Atmospheric Ozone and Its Relation to Meteorological Conditions". ADVANCE IN CHEMISTRY SERIES No. 21, "Atmospheric Ozone" pp. 226. (1959).
*7. Miyake, Y., Saruhashi, K., Katsuragi, T., "The Sr-90 fallout and the Air Motion." Papers in Meteorology and Geophysics Vol. IX Nos. 3-4 (1959) pp. 172.

frontal surface. Thus, on the basis of this suggestion some considerations of the behavior of the debris in the atmosphere will be described here and subsequent sections.

Now, a well-established deposition mechanism of the nuclear debris is the wash-out in precipitation of the troposphere. However, the data of observation given in Part I, particularly concerning the time-variation in the specific activity for a series of rainfalls, can not be explained satisfactorily only by such a simple wash-out process in rain-water. In order to interprete more satisfactorily the results obtained, it is required in addition to the wash-out process to assume a process that successively supplies the rain-producing layer over an observation point with the debris from a region where the debris is not subjected to wash-out in the troposphere, because the debris in the troposphere is generally distributed as aerosol with such a vertical distribution that has an increasing tendency with height. Concerning this process it is acceptable that the southward descending component of the air current within a trough in the upper layer serves as conveyer which transports the debris into the rain-producing layer, when the pressure distribution at a higher level over an observation point is of T-type. Furthermore, this downward supplying-process of debris has the effect of increasing the great compression by piling up of air from above, when the air mass subsides. Therefore, if we permit here a reasonable assumption that the moving mass of air neither receives nor loses the debris during its displacement, the content of debris per unit volume increases in proportion to the magnitude of the downward component of the upper air current. Since the air concentration of debris is originally larger in the upper layer than in the lower, the southward sinking air-component leads to produce a pronounced increase in the air concentration of debris. In consequence, a series of rainfalls formed in case of a T-type of pressure distribution permit to give higher specific activity values.

When a chart at a high level over an observation point gives a R-type of pressure distribution, the specific activity of a rainfall shows a lower value. It is obvious in this case that the effects which cause to increase the specific activity in case of a T-type of pressure distribution almost diminish or result in producing an opposite action. In the other words, the upper air current has generally a northward ascending component when there is a ridge of contour on a chart at a higher level over an observation point, or the vertical component of air flow almost diminishes, the contours lying in the east-west direction on a constant-pressure surface at a higher level. Accordingly the supplying effect of debris from the uppermost layer, where the debris is not subjected to wash-out, is nothing or negligible, that is to say, the debris in the rain-producing layer is not subjected to a fresh supply and decreases by being washed out in rain-water with time. Consequently it is acceptable that a rainfall accompanied with a R-type of pressure distribution does not possess a higher specific activity value owing to such meteorological conditions.

4. Fission Product Debris in the Rain-producing Layer. As is described in the preceeding sections, a mechanism that the fission product debris is enriched in precipitation is considered as follows; at first, it is a process that the debris is simply washed out in rain-water of the rain-producing layer; secondly, a process that the rain-producing layer is subjected to a fresh supply of debris by a downward component of air flow from the uppermost layer of the troposphere. From this point of view, we will describe an alternative interpretation our observational data of fallout activity during the period November, 1958 to September, 1960.

It is generally accepted that the average concentration of total fission product debris in the troposphere has a positive gradient with respect to height and that the wash-out in precipitation is a main deposition process of the debris. Then the mean specific activity for a series of rainfalls, together with the minimum value on the curve*[8] of the altitude of 300-mb. isobaric surface against time are plotted as a function of time during March-June, 1959 on a logarithmic scale in Fig. 4. The

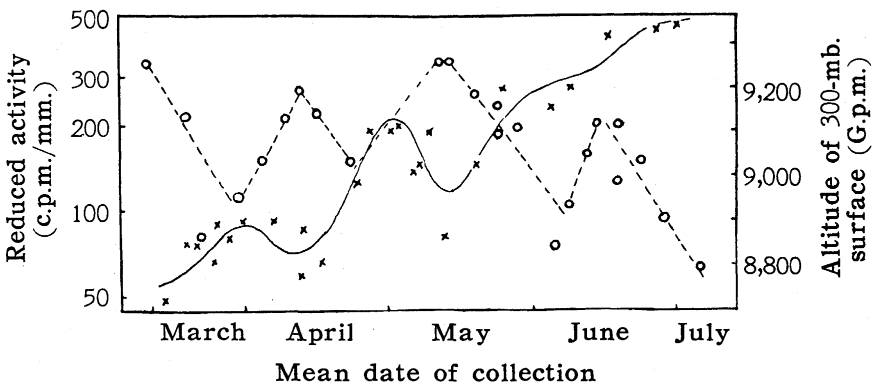


Fig. 4. Mean specific activity of total fission products in a series of rainfalls (---○---, left ordinate) and minimum values in the curves of altitude of 300-mb. isobaric surface (——×——, right ordinate) vs. time during March–June, 1959.

curves plotted in this figure show several interesting features of the tropospheric behavior of nuclear debris, since the curve reflects directly change of the nuclear debris content in the rain-producing layer. It should be noted first of all that our measurements relate to the atmosphere at a single site. However, since a prolonged period for atmospheric mixing elapsed between the last reported test in 1958 and the start of the present observation, it is clear that the results represent conditions in the atmosphere over a comparatively wide region.

At first, peaks were found periodically in the two curves plotted here and both the periods were on the average about one month. Secondly, the values of specific activity indicate roughly exponential rise and fall over the 4-month period of obser-

*[8]. See Figs. 1–a, –b and –c.

vation. From these results obtained it may be possibly considered that the periodic rise is due to the periodic sinking motion of air mass which carries the debris from the uppermost region into the rain-producing region in the troposphere, that is, the existence of an alternative deposition process of the tropospheric debris may be suggested. On the other hand, the exponential fall of specific activity seems to reflect the wash-out of the debris in rain-water from the rain-producing layer. The positive and the negative gradients of the curves provide a measure of the supplying and removal rate of the debris in the same region, respectively. From the negative gradient values an average half-time for removal process of nuclear debris from the rain-producing layer may be roughly estimated to be 12 days for March-June, 1959, provided that the specific activity of total fission product in rain is proportional to the air concentration of debris in this layer. This value is similar to the value (14 d.) that was reported as a half-life of air-born dust for residence in the whole troposphere by Burton and his associates*[9] on the basis of the study of natural long-lived isotopes —— radium-D, -E and -F —— in the atmosphere. However, it should be noted that Burton's value is for the whole troposphere and our value is for the rain-producing layer in the troposphere. The value for the whole troposphere is estmated to be 27-32 d. by us, as will be described below.

5. Nuclear Debris in the Uppermost Region in the Troposphere. The monthly values of the total fallout activity and the specific activity of precipitation during October, 1958—September, 1960 are presented in Table 2. Using the stored samples which were monthly collected for the stated period, the values listed in this table were determined during 4-6 October, 1960, to eliminate the effect of radioactive decay

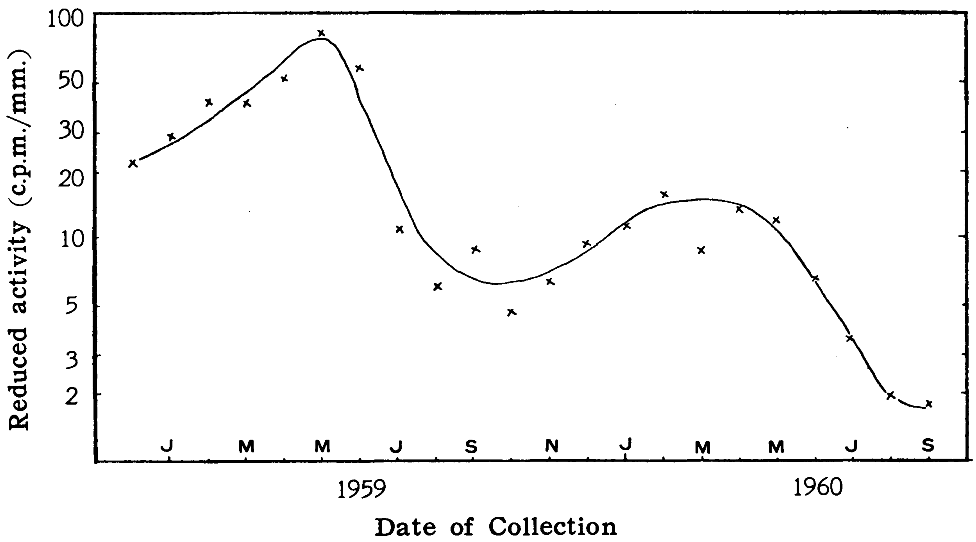


Fig. 5. Monthly specific activity of total fission products in rain-water during December, 1958- September, 1960.

*9. Burton, W. M., Stewart, N. G., Nature 186, 584, (1960).

which occurred after the date of the first sample (Oct. 31, 1958). The monthly value of specific activity of total fission products in rain are plotted against time during the periods January, 1959-September 1960 in Fig. 5 on a logarithmic scale. The curve plotted in this figure seems to reflect important features of the atmospheric processes of the nuclear debris. That is, a spring peak in the specific

Table 2. Reduced monthly total fallout activity and specific activity of rain during October, 1958—September, 1960.

Period of collection	Rainfall (mm.)	Total fallout activity reduced. (c.p.m./m². mo.)	Specific activity reduced. (c.p.m./mm.)
Oct., 1958	206.9	780	3.8
Nov.	214.0	4,900	22.9
Dec.	186.9	7,511	40.3
Jan., 1959	256.3	7,230	28.2
Feb.	129.6	5,160	40.0
Mar.	214.3	8,780	41.2
Apr.	86.3	4,940	52.3
May.	91.7	7,530	82.0
Jun.	89.9	5,140	57.2
Jul.	220.3	2,310	10.5
Aug.	194.0	1,130	5.8
Sep.	126.0	1,102	8.8
Oct.	159.8	730	4.5
Nov.	136.0	860	6.3
Dec.	241.9	2,270	9.4
Jan., 1960	201.2	2,250	11.2
Feb.	104.8	1,750	16.7
Mar.	136.0	1,128	8.3
Apr.	75.5	1,000	13.2
May.	77.8	972	12.5
Jun.	114.5	730	6.4
Jul.	194.1	718	3.6
Aug.	107.8	225	2.1
Sep.	141.6	277	1.9

activity value occurred in May, 1959 and February, 1960, as was reported in many papers. The value of specific activity of rain exhibited a roughly exponential fall from spring to autumn and a rising trend in the subsequent period from autumn to spring over the stated period of observation. The latter rise of specific activity of rain in the left side of a peak value is due to the re-entry of the stratospheric debris into the troposphere. On the other hand, the former fall in the right side of a peak is attributed to the removal of the tropospheric debris. Then from the slopes of these portions of the curve half-times for the removal process of debris in the troposphere are estimated to be 27 d. and 32 d. for 1959 and 1960, respectively. If it is reasonably assumed that a value of specific activity of rain for the total fission products is proportional to an average air concentration of the nuclear debris, as a mean life of residence of the debris in the uppermost troposphere 36- and 47-day values are derived for the stated periods in 1959 and 1960, respectively. A half-time for the removal process of the whole tropospheric debris should be longer than that of the wash-out process of the debris, because a tropospheric deposition process is a combination of two processes described above and because a control step in the deposition process is a process of the periodic entry of nuclear debris into the rain-producing layer from the uppermost tropospheric reservoir adjacent to the relatively persistent stratospheric reservoir. Consequently the whole deposition rate of tropospheric debris leads to be greatly influenced by the mixing process of tropospheric debris between the upper and lower levels. There was a little difference in the deposition rate between 1959 and 1960, but this may be attributed to the fluctuation of meteorological factors which affect the tropospheric circulation and mixing of the debris as described above.

6. Behavior of Stratospheric Nuclear Debris. It will also be seen from Fig. 5 that peaks of the specific activity were found in the months of May and February for 1959 and 1960, respectively. These spring peaks and sharp increases in the activity concentration have been explained satisfactorily by Stewart *et al.**10 on the basis of the Dobson-Brewer's model of atmospheric circulation deduced from measurements of ozone and water vapor and have been further discussed in detail by Martell*11 in consideration of the locations and dates of test explosions conducted.

Table 3. Mean annual specific activity of rain

Period of observation	Reduced total fallout activity (c.p.m./m².)	Amount of precipitation (mm.)	Annual mean specific activity of rain (c.p.m./mm.)	Ratio*
Oct., 1958-Sep., 1959	56,500	2,024	28.2	$\frac{7.8}{28.2}=0.28$
Oct., 1959-Sep., 1960	13,200	1,696	7.8	

* The ratio of the annual mean value of specific fallout activity of 1959 to that of 1960 is indicated.

Now, it may be said that the minimum levels are maintained by a steady leakage from a relatively persistent stratospheric reservoir of fission product debris. From this point of view, the annual mean value of specific activity of total fission products in precipitation is listed in Table 3. From the ratio of these two annual mean values it is proved that the mean annual specific activity in precipitation during October, 1959-September, 1960, is reduced to about 30 per cent of that during October, 1958-September, 1959. This suggests that a half-time for residence of the nuclear debris in the stratosphere may be about 200 d. and consequently a mean life for the residence of debris may be estimated to be 290 d. On the other hand, if it is reasonably assumed that a peak height of each year in the specific activity of rain reflects proportionally the amount of the stratospheric debris which remains without being deposited for each year of observation in the stratospheric reservoir,

*10. Stewart, N. G., Osmond, R. G. D., Crooks, R. N. and Fisher, E. M. R. Atomic Energy Research Establisment, Harwell HP/R, 2354 (1957).
*11. Martell E. A., Science, 129, 1197 (1959).

the ratio of peak heights of adjacent two years may give an estimate of half-time of the nuclear debris for removal process in the stratosphere, that is, about 180–200 d. Accordingly a mean life of residence of the debris in the stratosphere is estimated to be 250–300 d.

Summary

We have described the correlation between the day-to-day variation in the specific activity of precipitation and the weather conditions at higher levels, viz., the day-to-day variation in the altitude of isobaric surface at a higher level over an observation point. From the results obtained some considerations of the behavior of the nuclear debris in the troposphere are described here. That is, in addition to the well-established wash-out process of the debris in rain-water it is proposed here that there is a process that supplies successively the rain-producing layer with the nuclear debris from a region where the debris is not subjected to wash-out in the troposphere. The mean lives for the residence of nuclear debris in the troposphere and stratosphere were estimated to be 40 d. and 300 d., respectively.

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