Geochemical Study on Strontium-90 Fallout during 1954–1960

By

Takeshi SOTOBAYASHI and Seitarô KOYAMA

(Department of Chemistry)

(Received, December 3, 1961)

Introduction

As a result of fallout from nuclear test explosions, small quantities of fission products now widely have been disseminated. Further, the possiblity of their regulated or accidental discharge must be taken into account in planning peaceful nuclear operations. Therefore, at first a simple and rapid method for routine analysis must be developed for radiostrontium at very low concentration in wide variety of environmental samples, because the isotope strontium-90 represents a major part of the radiation hazard to man caused by nuclear operations by reason of its long half-life (28 years). Secondly, the rate of strontium-90 fallout depends to a large extent on the geographical locality and on the weather conditions. There is evidence that in the northern hemisphere places with high rate of precipitation receive the high fallout. These conditions hold obviously for Japan. The present work was undertaken to establish a rapid method of the determination of radiostrontium in a fallout sample and to obtain more reliable information as possible on the geochemical behavior of strontium-90 in the atmosphere and its rate of deposition in Niigata, Japan.

Part I. Determination of Radiostrontium in the Fallout Sample by Ion Exchange Method.

In the determination of a radionuclide at low levels of activity, various factors must be taken into consideration. First, to obtain a detectable amount of activity a large sample must be processed. Second, the separated isotope should be obtained in high yield and with a good degree of purity. Third, the radiations being measured should be detected with high efficiency. This will depend not only on the instrumentation, but also on the method of a sample preparation.

A conventional procedure[*1] for the determination of radiostrontium in fission products involves precipitation of strontium and barium as the mixed nitrates in a fuming nitric acid solution, followed by removal of barium with precipitation as barium chromate at pH 5 and by separation of yttrium-90 from strontium with precipitation of yttrium as hydroxide, for strontium-90 determination. Furthermore, in the case of a fallout sample large amounts of aluminum and iron must be removed before the treatment of a fuming nitric acid. Each step must be repeated several times to obtain sufficient decontamination for the analysis of a fallout sample at low levels of activity. The radiochemical yield must be measured for the strontium intermediate product. This procedure is tedious and requires trained techniques.

Apparatus and Reagents.

Carrier Solutions. These are not standardized, because recoveries are nearly quantitative and gravimetric yield determinations are not required. Chemically pure reagents are used to prepare the following solutions; 4 mg. of strontium per ml. as nitrate in distilled water and 10 mg. of cesium per ml. as nitrate in distilled water.

Eluting Solutions. Eluent-I. Transfer 60 ml. of methanol and 15 ml. of ammonium hydroxide (1 : 1) to a 100-ml. graduated cylinder and dilute to 100 ml. with distilled water. Dissolve 10 grams of ammonium acetate, analytical reagent grade, in this solution, its volume being made to 100 ml.

Eluent-II. Dissolve 15 grams of ammonium acetate, analytical reagent grade, in distilled water containing 20 per cent (v/v) ammonium hydroxide (1 : 1) and make its volume to 100 ml.

Ion Exchange Columns. Two kinds of columns are used. Column-I and -II are prepared by sealing a 15-cm. length of glass tubing 15 mm. inside diameter and a 5-cm. length of glass tubing 5 mm. inside diameter, respectively. The resin used is Dowex-50 x8 (ammonium form); 100-200 and 200-400 mesh for column-I and -II, respectively.

Preliminary Treatment.

Add 100 ml. of 6 N hydrochloric acid to a fallout sample collected during a month interval and digest the mixture on a water bath for a few hours. Centrifuge and decant the supernatant solution. Repeat this step of acid leaching more than two times and finally wash the solid residue with three 5-ml. portions of hot 2 N hydrochloric acid. Combine these resulting supernatant solutions and washings,

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[*1]. Glendenin, L. E., Paper 3/236, National Nuclear Energy Series IV, 9, "Radiochemical Studies: The Fission Products".

concentrate nearly to dryness, warm on a water bath with 10 ml. of concentrated nitric acid, repeat this step until organic materials are entirely decomposed off and again concentrate nearly to dryness to convert residual salts into chlorides and to accomplish the dehydration of silica. Add 5 ml. of concentrated hydrochloric acid, warm a few minutes on a water bath and dissolve in 200 ml. of distilled water. Filter and wash with hot dilute hydrochloric acid and repeat once more the above operations to complete the removal of silica from the solution. Evaporate the resulting filtrate and washings to dryness and weigh out the solid residue.

Recommended Procedure

Add a few milliliters of concentrated hydrochloric acid to the solid residue obtained by the preliminary treatment. Dissolve the solid salts containing radionuclides leached from a fallout sample in 300 ml. of distilled water, add each 1 ml. of strontium and cesium carrier solutions and add 20 % sodium acetate solution until the color of the solution begins to turn brownish red by the formation of basic acetate of iron. Then, add 5 grams of disodium salt of (ethylenedinitrilo-) tetraacetic acid (EDTA) as powder and add several drops of ammonium hydroxide (1 : 1). Boil for several minutes to accomplish the formation of complex compounds between EDTA and metal ions, especially aluminum. Add 50 ml. of hot solution containing 4 grams of ammonium oxalate at 80° C, make up to pH 4.0 with 2 N hydrochloric acid with pH paper and stand for more than 4 hours. Filter and wash with 0.1 % ammonium oxalate solution adjusted to pH 4.0 with acetate-acetic acid buffer solution. Reserve the filtrate and washings for caesium determination, if required. The collected precipitate consists of chiefly calcium and strontium oxalates. Convert the precipitate into ash in a muffle furnace, dissolve the ash in a minimum amount of hydrochloric acid (1 : 1) and dilute with distilled water. Add 1 ml. of ferric chloride solution containing 5 mg. of iron per ml. and make up to pH 8.0 with carbonate-free ammonium hydroxide. Filter, transfer the filtrate to a reservoir placed in position above the cation-resin column-I and allow it to flow through the resin. Wash the column with 10 ml. of 0.1 N ammonium acetate and test the last few drops of the effluent with pH paper to determine whether the column has been completely converted into the ammonium form. If the conversion is complete, pH will be equal to that of the acetate solution (pH 6). Discard the solution which passed through the column and the washing.

First, elute inert calcium with 80 ml. of the eluent-I at a flow rate of 1 ml. per minute and discard the effluent that contains only calcium. Similarly elute radiostrontium with 100 ml. of the eluent-II and record the time as the beginning of the growth period for yttrium-90. Transfer the effluent to an evaporating glass-dish and evaporate to dryness on a water bath. Cover the dish with a watch glass and set it aside.

After about two weeks dissolve the content of the dish in a minimum amount of dilute hydrochloric acid, wash the dish and transfer the solution to the reservoir of the cation-resin column-II. Allow the solution to pass through the column, wash with 5 ml. of water and run 5 ml. of 0.1 N ammonium acetate for the same purpose as in the case of the first elution of calcium. Discard these resultant solutions which passed through the column.

Elute the strontium with 12 ml. of the eluent-II and record the time. Discard the first 6 ml. of effluent and collect the next 6 ml. of effluent in a small evaporating glass-dish and evaporate nearly to dryness. Transfer the content completely to a counting-dish with a minimum amount of dilute hydrochloric acid, and again evaporate to dryness under an infra-red heat lamp. Mount the dish for beta-counting. Then wash the column with 5 ml. of water and finally elute yttrium-90 with 10 ml. of 6 N hydrochloric acid. Collect the washing and effluent in an evaporating glass-dish, concentrate to about 1 ml. and transfer it to a small porcelain counting-dish. Evaporate to dryness under an infra-red heat lamp. Mount the dish for beta-counting.

Discussion

A problem for the determination of strontium-90 in a fallout sample is the removal of various inert materials from the strontium, in addition to the separation of other radioactive elements from it. The most significant elements of these inert materials are aluminum, iron and calcium. By the present procedure it is simply carried out to remove non-radioactive materials leached from a fallout sample and to obtain the radiostrontium free from ions which interfere with its radiochemical determination.

In Table 1 are shown the mean value of the weight of solid chlorides which are leached with 6 N hydrochloric acid from fallout samples dried at 110°C, and the mean values of aluminum, iron, calcium and magnesium in terms of milliequivalents in fallout samples collected for a month interval.

Table 1.

Solid chloride	6	gram.
Aluminum	20	meq.
Iron	5	//
Calcium	15	//
Magnesium	10	//

Removal of Aluminum and Iron.

Since the solid part of a fallout sample consists of the dust of argillaceous and organic materials, large amounts of aluminum and iron ions are inevitably leached from the sample in the preliminary treatment, as shown in Table 1. In the case of fuming nitric acid method these interfering ions should be previously removed to obtain the pure strontium nitrate with high yield. It is very tedious to separate them by the mixed hydroxide precipitation with ammonium hydroxide, because the bulky hydroxide should be repeatedly precipitated with a minimum loss of strontium into the hydroxide precipitates. It is desirable that there is an alternative and more simple procedure which involves a single precipitation of calcium and strontium while retaining aluminum and iron in solution as soluble complex salts. From this point of view, disodium salt of (ethylenedinitrilo-) tetraacetic acid being used, the removal of these ions was successfully performed, because the stability constants of calcium- and strontium-EDTA complexes are far less than those of aluminum- and iron-EDTA complexes. That is, the presence of EDTA permitted to separate calcium and strontium from aluminum and iron by a single precipitation as oxalate in buffered solution at pH 4.0. A series of preliminary experiments were carried out to ensure this procedure to be applicable. The isotope strontium-89 was used as tracer. It is proved that the coprecipitation of a small amount of strontium on calcium oxalate is nearly quantitative in the solution containing large amounts of aluminum and iron, so far as the number of moles of EDTA present is somewhat larger than the total number of the moles of aluminum and iron present in the solution. The yield of strontium is always more than 97 per cent. Table 2 shows some of these experimental results with various quantities of aluminum and iron. However, the oxalate precipitates are somewhat contaminated with radioactive barium-140 and rare earths, but these contaminants are easily removed in the subsequent steps of cation exchange separation.

Table 2.

No.	Aluminum added (millimole)	Iron added (millimole)	Calcium added (millimole)	Strontium-89 added (c.p.m.)	Strontium-89 found (c.p.m.)	Strontium-89 yield (%)
1	2	0.5	10	4450	4400	99.0
2	5	3	10	4720	4650	98.6
3	7	5	10	3930	3820	97.2

Removal of Calcium.

Calcium, which is widely distributed in nature together with strontium, is one of the inert elements whose separation from strontium have been studied by many investigators. Calcium should be removed in a sample of moderate or high calcium content, when the direct measurement of strontium radiation is required in order to determine the radioactivity ratio of strontium-90 to -89. Recently many eluting reagents have been proposed for ion exchange separation of calcium from strontium, for example, ammonium salts of citric*[2], lactic*[3], acetic*[4] and formic*[5] acids. Among them ammonium acetate and formate are considered to be more suitable for radiochemical analysis at low level of activity, because they are easily eliminated merely by evaporation of the effluent of strontium fraction in preparing a beta-counting sample. Then the separation coefficient of strontium to calcium (Kd^{Sr}/Kd^{Ca}) and the distribution coefficient of calcium (Kd^{Ca}) were re-examined with varying the concentration of these eluting salts and are summarized in Table 3.

Table 3.

Concentration of eluent (Moles)	Ammonium acetate		Ammonium formate	
	Kd^{Ca}	Kd^{Sr}/Kd^{Ca}	Kd^{Ca}	Kd^{Sr}/Kd^{Ca}
1.0	17.5	2.3	17.8	1.9
1.2	12.4	2.2	12.8	1.9
1.4	8.3	2.1	9.7	1.9
1.6	6.3	2.1	7.5	1.9

It is noticed that in the case of acetate eluent the separation coefficient is somewhat larger and that the distribution coefficient of calcium is smaller, as compared with those of formate eluent, especially when the concentration of eluent is higher; that is to say, by means of acetate eluent the separation is performed more effectively and the volume of eluent required for removal of calcium from strontium is reduced. Therefore, acetate is proved to be more preferable to formate as eluting agent. However, for the complete removal of a large amount of calcium (10 millimoles) from a small amount of strontium, this acetate eluent still requires a long column (1.5 cm^2. × 30 cm.) and a large volume of eluent (about 200 ml.) and yet the separation of calcium from strontium is not satisfactorily performed. Then in order to develop a more satisfactory procedure the effect of addition of some organic solvents to these eluting solutions was investigated. Some typical data obtained are assembled in Table 4. The concentration of eluent is 1.6 M. with respect to ammonium acetate or formate.

Inspection of these results reveals first of all that by the addition of alcohol the effectiveness is enhanced for the separation of calcium from strontium and that acetate is more suitable than formate, as expected. This alcoholic solution (6 : 4) of ammonium acetate (1.6 M.) is more excellent as eluent and its separation coefficient is 1.5–1.6 times larger than that of the mere acetate solution. This is caused by the facts that the addition of alcohol reduces the distribution coefficient of calcium

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*[2]. Tompkins, E. R., Khym, J. X., Cohn, W. E., J. Am. Chem. Soc., 69, 2769 (1947).
*[3]. Lerner, M. and Rieman, W., Anal. Chem., 26, 610 (1954).
*[4]. Honda, M., Japan Analyst, 3, 132, (1954).
*[5]. Tsubota, H., Kitano, Y., Bull. Chem. Soc. Japan, 33, 765 (1960)

and has little effect on that of strontium. The reduction of distribution coefficient of calcium is probably due to the formation of a metastable addition compound between calcium acetate and alcohol during a homogeneous ion exchange process, because the precipitation of calcium acetate occurs in some fractions of calcium effluent several hours after elution.

Table 4.

Mixed solvent added		Ammonium acetate (1.6 M.)		Ammonium acetate (1.6 M.)	
		Kd^{Sr}/Kd^{Ca}	Kd^{Ca}	Kd^{Sr}/Kd^{Ca}	Kd^{Ca}
None		2.10	6.3	1.90	7.5
Acetone	(6 : 6)*	2.56	4.9	2.44	4.5
Methanol	(4 : 6)	2.68	4.9	2.44	5.7
	(6 : 4)	3.01	3.9	2.61	4.9
Ethanol	(4 : 6)	2.76	4.9	2.63	6.2
	(5 : 5)	3.01	4.4	2.70	6.0
	(6 : 4)	3.12	4.1	2.74	6.0

* Figures in parentheses show the ratios (v/v) of each organic solvent to water.

As an eluent for calcium the ethanolic solution appears to be somewhat more satisfactory from the above data, but it leads to reduce markedly the flow rate of eluent through a resin column. For this reason methanolic solution was used to dissolve acetate instead of ethanolic one in the present procedure. Fig. 1 shows a typical elution curve of calcium and strontium by methanolic eluent-I and -II described above. From preliminary experiments it was proved that the yield of strontium was more than 99 per cent, when calcium and strontium absorbed on the column were less than 0.4 gram (1 gram as carbonate) and 0.01 gram, respectively. Thus this procedure can be satisfactorily applied to the removal of calcium from strontium for the measurement of strontium activity.

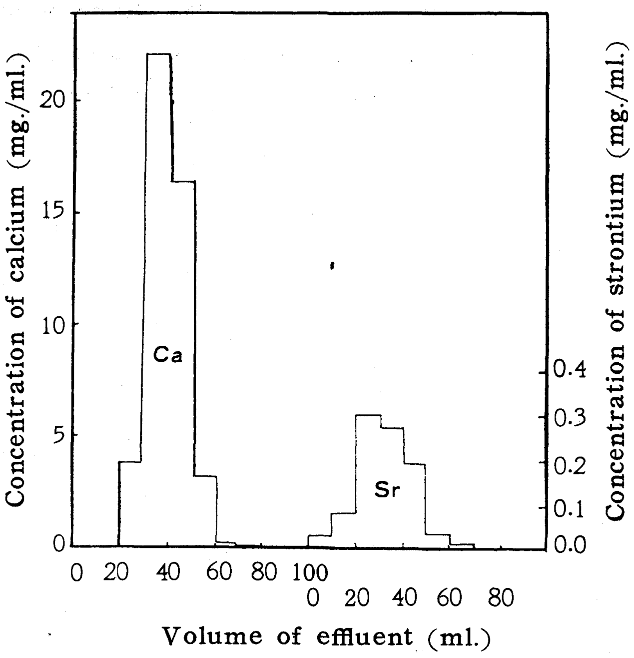


Fig. 1. Elution curve for separation of calcium and strontium on Dowex 50. NH_4^+-form, 100–200 mesh, ϕ 15×150 mm, flow rate 1.0 ml. per minute, eluents "eluent-I and eluent-II".

280.4 mg. of calcium and 6.4 mg. of strontium were absorbed on column. Ten-milliliter fractions were analysed by titration with Mg-EDTA and 2 Na-EDTA using EBT indicator.

Removal of Barium.

Because a small part of barium is

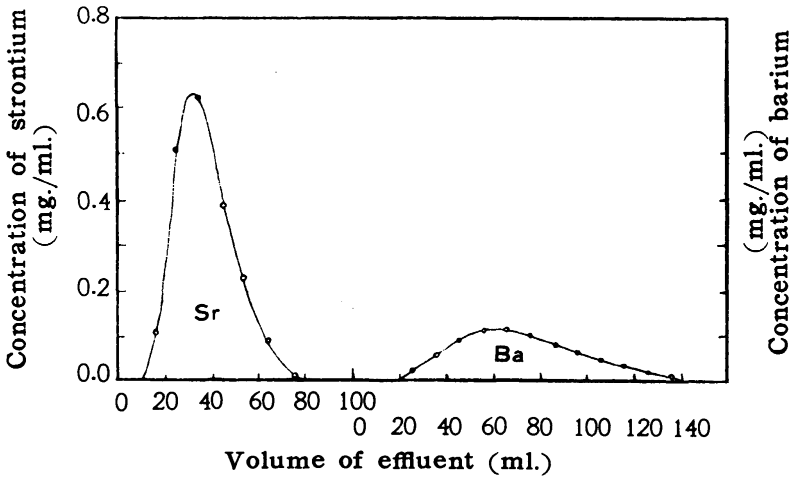
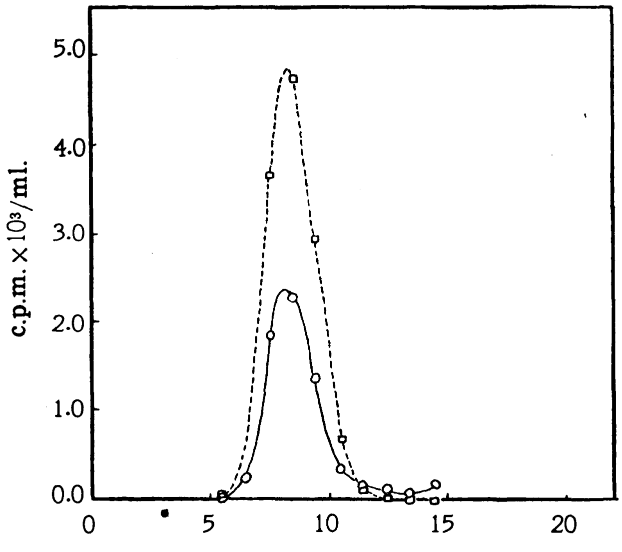


Fig. 2. Elution curve for separation of strontium from barium after calcium elution.

NH_4^+-form, 100–200 mesh, ϕ 15×150 mm., flow rate 1 ml. per minute, eluent for strontium eluent-II, eluent for barium a solution of ammonium chloride (2.5 N) and ammonium acetate (1.2 N) containing 10 % (v/v) ammonium hydroxide.

coprecipitated with the mixed calcium and strontium oxalates as already described, it should be also removed from the strontium fraction. The removal of this ion was successfully performed by cation exchange method, that is to say, barium is not eluted with 100 ml. of eluent-II, by which all the strontium was stripped from the column. If a barium determination is desired, barium can be eluted separately with an ammoniacal solution (1 : 20) of ammonium chloride (2.5 N) and ammonium acetate (1.2 N). Fig. 2 shows a typical elution curve of strontium and barium.



---□---; activity of Sr-90 measured immediately after separation.
——○——; activity of Sr-90 measured two weeks after separation.

Fig. 3. Elution curve for separation of strontium and yttrium. NH_4^+-form, 200–400 mesh, ϕ 5×50 mm., eluent for strontium eluent-II, eluent for yttrium 6 N hydrochloric acid.

Total activity of strontium and yttrium-90 absorbed on column is 30,000 c.p.m. and 1-ml. fractions were measured.

Separation of Strontium and Yttrium

Since an evaporative mounting technique is preferable to that of collecting the precipitate in preparing a sample for beta-counting, the separation of strontium from yttrium was studied by employing a cation exchange column instead of the coprecipitation of yttrium on ferric hydroxide by ammonium hydroxide. This was simply accomplished by using a small cation-resin column-II. Strontium is eluted with eluent-II and yttrium, with 6 N hydrochloric acid. A typical elution curve is shown in Fig. 3.

Summary

The procedure described in this paper involves a simple precipitation of alkaline earths as oxalate in a sample solution containing a large amount of aluminum and iron in the presence of disodium salt of (ethylenedinitrilo-) tetraacetic acid and a rapid separation of radiostrontium from calcium, barium and rare earths by an ion exchange method instead of the conventional fuming nitric acid method. For the determination of strontium-90, the daughter yttrium-90 is separated from the radiostronium by using a small cation-resin column. A counting sample is prepared directly by evaporation of the effluent in a counting-dish. This technique permits a simple and rapid procedure with a radiochemical yield greater than 97 per cent. The high yield eliminates the need for measurements of recoveries of strontium and yttrium.

Part II. Strontium-90 Fallout during 1954-1960 in Niigata and Its Atmospheric Behavior.

We have made measurements of the radiostrontium contents of fallout samples during the period 1954-1960. The results of the present work are summarized here. From the geochemical point of view, we have made some interpretations about the atmospheric deposition process of fission product debris from nuclear test explosions in relation to meteorological principles.

Data of Strontium-90 Fallout

The results measured in Niigata (139° E.; 38° N.) for strontium-90 fallout during the period September, 1954-June, 1960 are listed in Tables 1, 2 and 3. Table 1 shows intermittent data of the activity concentration of strontium-90 in rain-water from September, 1954 to March, 1956. Since nuclear test explosions upto 1955 had not been carried out so many times as they were done from 1956 to 1958, any continuous and systematic analysis of strontium-90 was not performed in our laboratory. The data listed in Table 1 were all calculated by using Halldene-Harley's Table from strontium-89 contents in rain-water samples and the number of days after explosion estimated by decay measurements of gross activities of rain-water samples. Table 2 shows that there exists appreciable difference between the values obtianed by radiochemical determination of yttrium-90, daughter product of strontium-90, and those evaluated by Hunter-Ballou's Table with respect to the percentage of strontium-90 activity to the total fission product activity in the rain-water samples. This is evidence that the influence of each explosion was superimposed one another already in 1956, because the number of nuclear test explosions increased. In Table 2 the strontium-90 values of Sep. 14 and Oct. 2 seem to be very high, as compared with others in the same table. This is due to the weather conditions under which the rainfall occurred, it being taken into consideration that the day-to-day variation in the total fallout activity is considerably affected by the local weather conditions near the Earth's surface. As shown by us[*1], when a rain-

Table 1. Activity concentration Sr-90 in rain-water during 1954–1956

Date of sampling	Activity concentration of strontium-90 ($\mu\mu$c./l.)	Sr-90/Sr-(89+90)* per cent	Sr-(89+90) (d.p.m./l.)
Sep. 24, 1954[a]**	1.8	0.60	680
〃 1954[b]	1.1	0.70	344
Nov. 4, 1954	0.4	0.57	148
Nov. 25, 1955	1.5	0.53	184
Mar. 21, 1956	1.5	0.70	490
〃 24, 〃	0.5	0.74	143
Jun. 23, 〃	0.6	0.66	214
Sep. 18, 〃	0.9	0.57	380

* Figures in this column show percentage of strontium-90 to strontium-(89+90) calculated by Halldene-Harley's Table from number of days after explosion estimated by decay measurement of gross activity.

** Samples of (a) and (b) were collected at different times of the day.

Table 2. Activity contribution of Sr-90 to gross β-activity

Date of Sampling	Sr-90 in rain-water ($\mu\mu$c./l.)	Activity contribution of Sr-90 to gross β-activity (%) by radiochemical analysis	by Hunter-Ballou's Table
Mar. 21, 24 1956	6.2	0.15	0.01 (10)*
Jun. 30	1.5	0.04	0.03 (19)
Aug. 4	1.5	0.32	0.05 (24)
Sep. 14	24.5	2.30	0.04 (22)
Oct. 2	11.0	0.25	0.06 (30)
Nov. 26	3.0	0.30	0.16 (76)
Mar. 14–16**	5.5	0.68	– –

* Figures listed in parentheses show number of days after explosion estimated by decay measurements of gross activities of rain-water samples.

** Date of collection of this sample refers to 1957.

water sample collected in a particular day is measured, its activity concentration happens to indicate an extraordinarily higher value. For example, the ratio of the maximum value to the minimum one for a certain period sometimes reaches more than ten.

Table 3. Sr-90 fallout during April, 1957 -June, 1960

Period of sampling	Amount of precipitation (mm.)	Strontium-90 (mc./km^2.)	(μμc./mm.)	Sr^{90}/Sr^{89} (%)
20/ 4- 7/ 5, 1957	59.2	0.52	8.8	
8/ 5- 8/ 6,	118.2	0.55	4.7	11.6
7/ 6- 3/ 7,	69.1	0.09	1.3	7.6
4/ 7-31/ 7,	189.1	0.06	0.3	
1/ 8-25/ 8,	90.1	0.13	1.5	
26/ 8-20/ 9,	65.8	0.09	1.5	
21/ 9-20/10,	124.2	0.42	3.3	
21/10-22/11,	137.0	0.80	5.9	
23/11-28/12,	213.1	1.19	5.6	
29/12*-3/ 2, 1958	254.4	2.46	9.7	17.2
4/ 2- 6/ 3,	115.6	1.34	11.6	10.7
7/ 3-28/ 3,	55.8	0.63	11.2	11.0
29/ 3-10/ 5,	147.3	1.17	8.0	5.3
11/ 5-23/ 5,	53.5	0.46	8.6	4.4
24/ 5- 2/ 7,	104.3	0.86	7.8	4.0
3/ 7-27/ 7,	571.9	2.14	3.5	6.3
28/ 7-26/ 8,	250.6	1.04	4.2	3.0
27/ 8-30/ 9,	356.7	1.34	3.8	4.0
Oct.	206.9	0.50	2.3	5.1
Nov.	214.0	1.12	4.9	2.6
Dec.	186.9	1.07	5.7	3.3
Jan., 1959	256.3	1.21	5.0	4.0
Feb.	129.6	1.00	7.8	4.8
Mar.	214.2	1.60	7.5	8.2
Apr.	86.2	0.86	10.0	10.5
May	91.7	1.01	11.0	28.4
Jun.	89.8	0.65	7.3	40.
Jul.	220.3	0.17	0.8	58.
Aug.	194.0	0.06	0.3	100.
Sep.	126.0	0.10	0.8	
Oct.	158.9	0.17	1.1	
Nov.	136.6	0.21	1.5	
Dec.	241.9	0.32	1.3	
Jan., 1960	201.2	0.26	1.3	
Feb.	104.8	0.28	2.7	
Mar.	136.0	0.22	1.6	
Apr.	75.5	0.18	2.4	
May	77.8	0.22	2.8	
Jun.	114.5	0.12	1.1	
20/ 4 -28/12, 1957	1065.8	3.85	3.6	
29/12*-31/12, 1958	2518.9	14.06	5.6	
1/ 1 -31/12, 1959	1945.5	7.36	4.0	
1/ 1 -30/ 6, 1960	709.8	1.28	1.8	

The * marked date refers to 1957.

Table 3 shows the amount of precipitation, the rate of strontium-90 fallout (mc./km^2.), the specific activity (μμc./mm.) and the ratio of strontium-90 to strontium-89 (per cent) during the period April, 1957—June, 1960. Radiostrontium

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*[1]. This journal. Series I, Vol. 3, No. 2, 88 (1962)

determination was made following the procedure described in Part I. The values of strontium-89 activity were reduced to values at the middle of each sampling period, the decay correction ($T_{1/2}=51$ d.) being applied.

Discussion

From Table 3 it is obvious that the highest measured value of the rate of strontium-90 fallout in Niigata in the Japan Sea side* of Japan is 14.06 mc./km^2.y. in 1958. This value was more than two times those*2 observed in other places in the Pacific side* of Japan. This is first of all caused by the facts that during 1957-58 a large number of nuclear test explosions had been carried out and that a major part of fission products injected into the atmosphere by the explosions, comes back to the Earth's surface in a relatively short time (less than one year), as shown elsewhere*3 by us. Furthermore, it is an important factor by which the rate of strontium-90 fallout increased in 1958 that Niigata had a larger amount of local precipitation (2518 mm.) in that year, as compared with the annual average value (1782 mm.) for the past thirty years, because the amount of fallout activity is acceptedly considered to be approximately proportional to the amount of local precipitation. However, besides this rough correlation between the rate of strontium-90 fallout and the amount of local precipitation, it should be taken into account that the rate depends to a large extent on the geographical locality of an observation point and in consequence, on the weather conditions overthere. In order to illustrate the locality of the rate of strontium-90 fallout in Table 4 are presented the rate of strontium-90 fallout in 1958 and the amount of cumulative deposit of strontium-90 on the Earth's surface upto 1958 at representative places in the Pacific and the Japan Sea sides. Each place in the both sides lies on the approximately similar latitude.

From Table 4 it is easily seen that the measured values of strontium-90 fallout at places in the Japan Sea side were all several times higher than those in the Pacific side. This is caused not only by the fact that the former places have the larger amount of local precipitation*4, particularly in winter time, but also by the fact that there are a number of showery precipitations which occur more frequently from late autumn to early spring than the other seasons of the year. The annual average value of specfic activity of strontium-90 in the Japan Sea side is larger, as

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*2. For example, the values for Tokyo, Sendai and Osaka were 5.3, 5.4 and 3.5 mc./km^2.y. (1958), respectively. (from Summary of the Observation Results for Nuclear Radiation in Japan and Airborn Radioactive Fallout in the World during IGY. p. 324, 1960, Tokyo, Japan)

*3. This journal. Series I, Vol. 3, No. 2, 91 (1962)

*4 See Fig. 1.

* This journal. Series I, Vol. 3, No. 2, 84 (1962)

Table 4. Geographical locality and rate of strontium-90

Location		Rate of Sr-90 fallout (mc./km^2.y.)	Amount of cumulative deposit of Sr-90* (mc./km^2.)
In the Pacific side	Tokyo	5.3 (1785 ; 3.0)**	
	Kôoriyama		6.
	Maebashi		5.
In the Japan Sea side	Niigata	14.1 (2518 ; 5.6)**	
	Akita		26.
	Takata		28.

* The data listed are taken from "Annual Report of Radiological Measurements", Technical Institute of Agriculture, Japan, pp. 81 (1959).

** The figures in the parenthese show the annual precipitation and the average specific activity of strontium-90 ($\mu\mu$c./mm)..

compared with that in the Pacific side. This may be due to the fact that the activity concentration of showery precipitation is generally higher than that of steady one, as shown elsewhere*[5] by us. This difference of the weather conditions is probably attributed to that of their geographical localities of the both sides of Japan. In other words, the north-west monsoon wind, which is caused by the southward motion of the polar continental air mass, prevails during winter-early sping in Japan. This air mass is rapidly transformed by being warmed from below and by being furnished with water vapor during the passage across the Japan Sea and furthermore, the transformed air mass is hindered and elevated up by a cooled barrier of the Main Island of Japan. As a result of these atmospheric processes a large number of showery precipitations occur in the Japan Sea side, but on the contrary in the Pacific side there is little rainfall in this period.

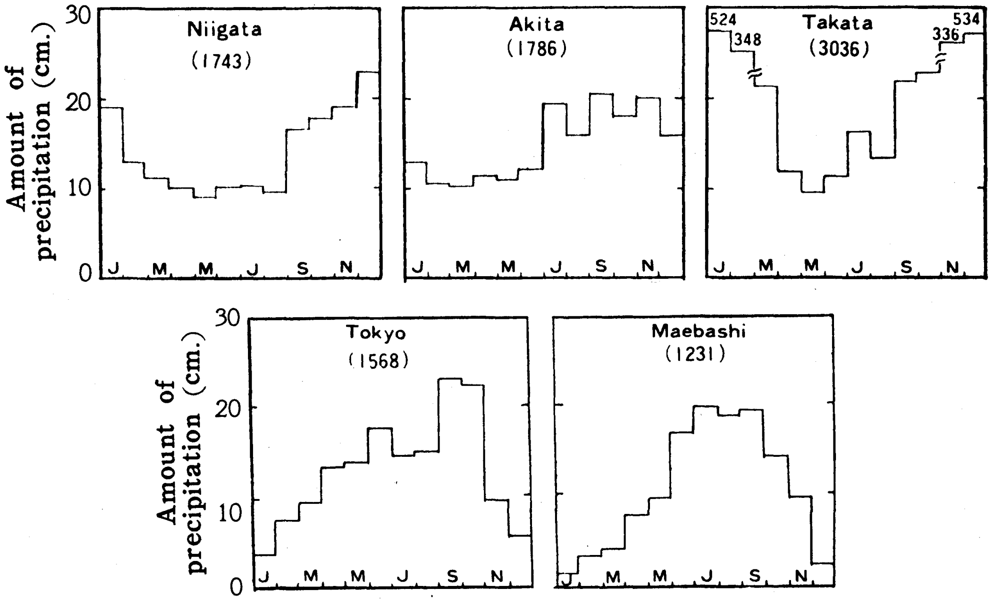

Fig. 1. Average annual precipitation and its type. Values under each place-name are the average annual precipitation for the past 30 years.

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

The monthly average number of days of more than 1.0-mm. precipitation is listed in Table 5 for the past 30 years. The average value of monthly amount of precipitation is shown diagramatically in Fig. 1. These observational data may give quantitative evidence to the considerations mentioned above.

Table 5. Number of days of more than 1.0- mm. precipitation

Location		Number of days of more than 1.0- mm. precipitation
In the Pacific side	Tokyo	102
	Maebashi	100
In the Japan Sea side	Akita	170
	Niigata	176
	Takata	196

Mean Age of Strontium-90

It is of interest to compare the values of activity contribution of the strontium-90 to the total fallout activity with the activity ratio of strontium-90 to strontium-89. Table 6 shows these measured values during 1957-1958. From these data the average values of number of days (mean age) after production of strontium-90 were evaluated by the Hunter-Ballou's and Halldene-Harley's Tables during April, 1957-March, 1958. Each value which was calculated by the both tables is in good agreement. From Table 6 it is obvious that the mean age of strontium-90 was about 220 d. in that period and that the value of the mean age in the latter half of this observational period (240-250 d.; Nov. 3, 1957—Mar. 28, 1958) was large about twice that of the former half (120-140 d.; Apr. 20—Nov. 2, 1957). Generally, the mean age of strontium-90 is affected by the location and date of explosion test and it depends greatly upon the residence time of nuclear debris in the atmosphere.

Table 6. Mean age of strontium-90 during 1957-1958

Period of observation	$\frac{\text{Sr-90 activity}}{\text{Gross activity}}$	$\frac{\text{Sr-90}}{\text{Sr-89}}$	Number of days after explosion
	(per cent)	(per cent)	(day)
20/ 4 - 2/11, 1957	0.26	3.7**	120-140
3/11*-28/ 3, 1958	0.93	12.7	240-250
20/ 4*-28/ 3, 1958	0.52	11.9**	220

*. The * marked date refers to 1957.

**. These ratios seem to be unreliable owing to the lack of data observed, but the values interpolated using the curve plotted in Fig. 2, are in good agreement with those evaluated from the ratio of strontium-90 activity to gross radioactivity.

Since during the former half of the period listed in Table 6 many series of nuclear test explosions were conducted, as indicated in Fig. 2, it seems to be acceptable that the strontium-90 fallout for that period was a combination of two components, one of which was a component that spread laterally and settled down with a relatively high speed in the troposphere and the other of which was a component injected high into the stratosphere. In addition to this fact, concerning the residence time of nuclear debris in the atmosphere it has been pointed out also by us[*6] on the study of time variation in the specific activity of total fission products in precipitation, that is, the value for the troposphere is about 40 d. and for the stratosphere, 300 d. Therefore, it is supposed that the strontium-90 observed for the former half of the year contained an appreciable amount of tropospheric component. In contrast with this case, a major part of the strontium-90 observed for the latter half is considered to come mainly from the stratospheric reservoir, because the mean age is comparable with the mean residence time of nuclear debris in the stratosphere and because the rate of entry of the stratospheric debris into the troposphere has its peak value during the latter half of this observation period, as will be described below.

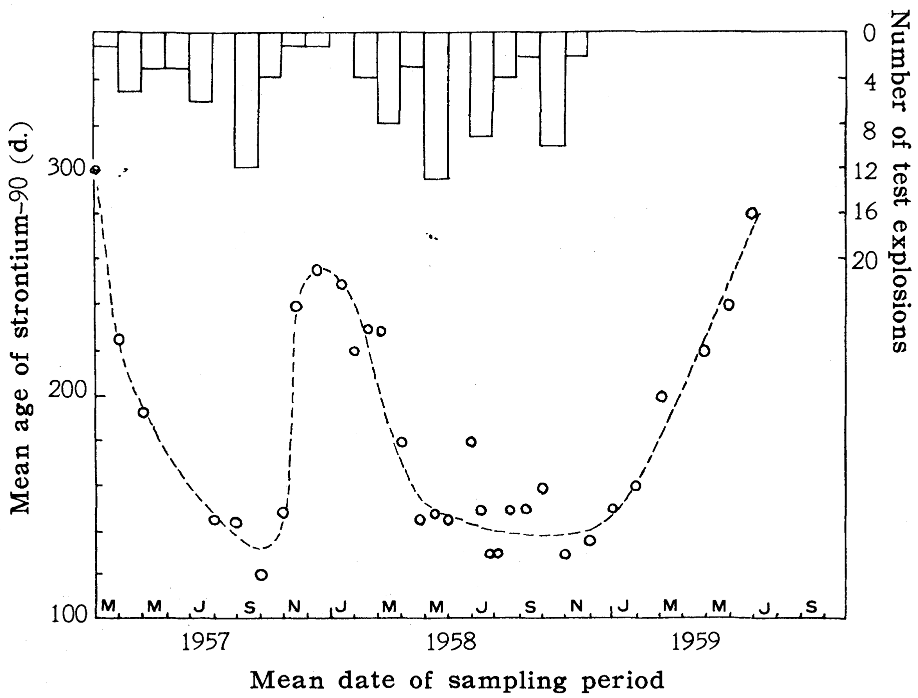

Fig. 2. Mean age of strontium-90 vs. time (left ordinate) and number of test explosions (right ordinate, from top to bottom).

In Fig. 2. are presented as a function of time the mean age of strontium-90 (left ordinate) together with the number of nuclear test explosions (right ordinate, from top to bottom) which were detected by appropriate methods in Japan, or announced by the governments which conducted the explosions during March, 1957—November, 1958. From the curve plotted it will be easily seen that the correlation described above between the values of mean age of strontium-90 fallout and the

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[*6]. This journal. Series I, Vol. 3, No. 2, 91 (1962)

atmospheric origins of strontium-90 fallout also holds for the period June-November, 1958. Therefore, the mean age of strontium-90 would have shown a characteristic seasonal variation similar to that of activity concentration of strontium-90 in rain-water, if nuclear tests were successively conducted. The results obtained concluded from the various lines of evidence as follows: the strontium-90 fallout during summer-autumn in the year involves an appreciable amount of strontium-90 from the pure tropospheric origin, whereas the strontium-90 fallout during winter-spring is derived mainly from a tropospheric reservoir built up during the early months of the year from material injected high into the stratosphere.

Upper Air Conditions and Deposition Rate

In Figs. 3 and 4 the amount of precipitation and the rate of strontium-90 fallout are diagramatically presented, respectivley. It is easily seen that the rate of strontium-90 fallout is approximately proportional to the amount of local precipitation, as stated above. However, the correlation between the amount of local precipitation and the rate of strontium-90 fallout seems not to be linear. This non-lineality may provide an indication on the atmospheric processes of fallout particles. Then for the purpose of studying the correlation between the deposition rate of fallout and the upper air conditions, the specific activity of strontium-90 in precipitation and the mean monthly altitude of 500-mb. constant-pressure surface at Akita* (140° 06′ E; 39° 43′ N.) are given as a function of time in Fig. 5. From this figure at first a seasonal variation in the specific activity was found with a peak in spring and a valley in late summer or autumn. In this connection, Stewart and his associates*[7] have pointed out that the seasonal variation of activity concentration of strontium-90 in rain-water is interpreted by the Dobson-Brewer's model of atmospheric circulation deduced from measurements of ozone and water vapor. Miyake *et al.**[8] have discussed the seasonal variation in the rate of strotium-90 fallout, taking into account the location and the season of injection of nuclear debris as well as the turbulent motion of the air on a larger scale.

Moreover, it will be noticed that a maximum value of the specific activity of strontium-90 in precipitation corresponds to a valley of the curve which is prepared by plotting the monthly mean altitude of 500-mb. surface against time and that a minimum value of the specific activity, to a peak of the curve. Now, that the air

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*[7]. Stewart, N. G., Osmond, R. D. G., Crookes, R. N., Fisher, E. M. R., Ouers, M. J., Atomic Energy Research Establishment, Harwell, HP/R 2354 (1957).

*[8]. Miyake, Y., Saruhashi, K., Katsuragi, Y., Papers in Meteorology and Geophysics, Vol. IX, Nos. 3-4, p. 172 (1959).

* As the Niigata Meteorological Observatory does not perform the upper air observation, the data of upper layer at Akita were used in place of those at Niigata.

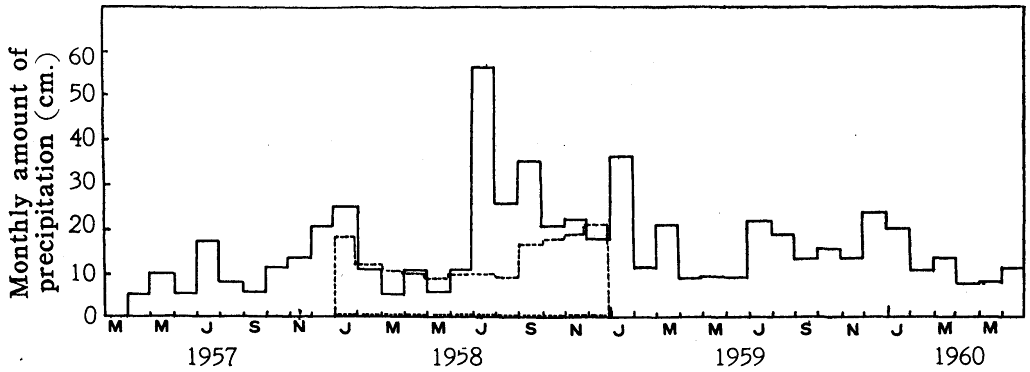

Fig. 3. Monthly amount of precipitation vs. time at Niigata. The dotted lines show monthly average amount of precipitation for the past 30 years.

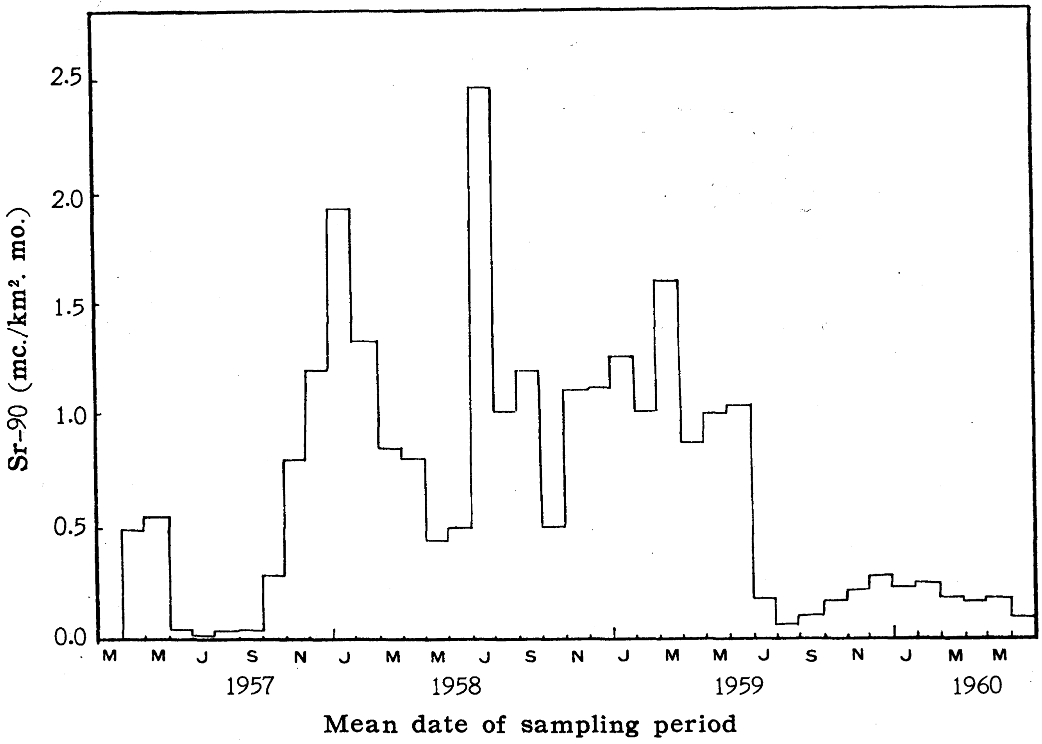

Fig. 4. Monthly amount of strontium-90 fallout at Niigata.

flow over an observation point lying within a trough at a higher level has a descending component in the direction from north to south, is underlined by the measurements of atmospheric ozone*[9]. Furthermore, this component carries down the nuclear debris with the effect of increasing its concentration in air*[10]. Then it is reasonably supposed that this argument holds for an averaged trough* for a month

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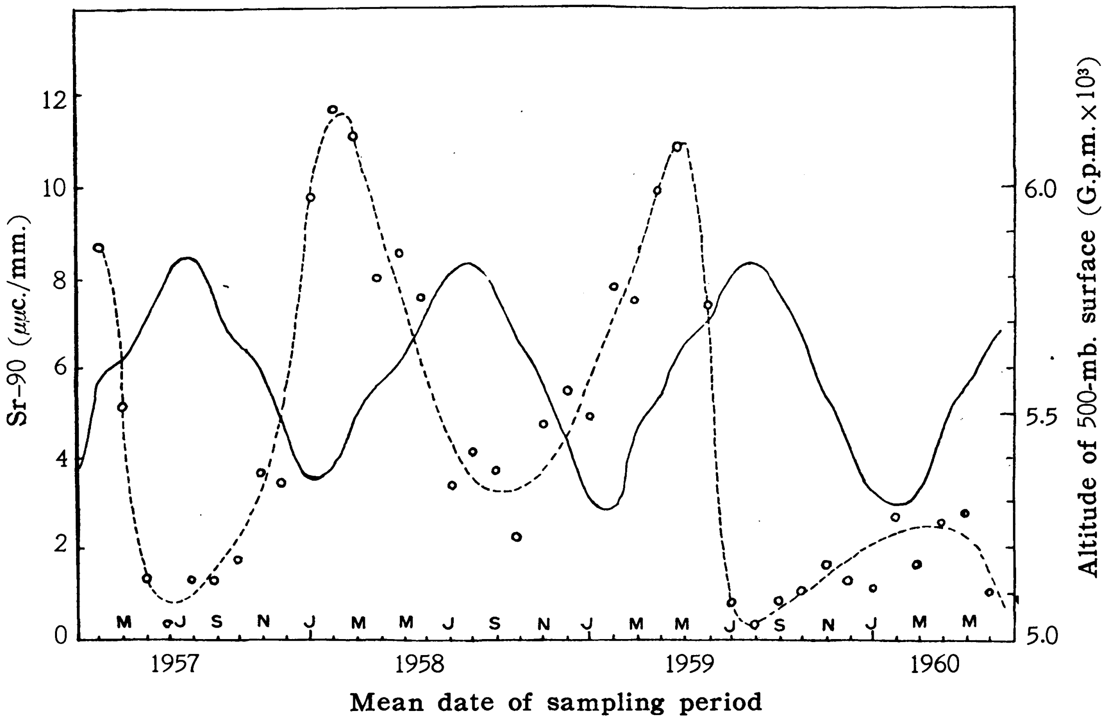
*. An averaged trough means a valley of the contours on the monthly mean chart prepared by plotting monthly mean altitudes of 500-mb. surface.

*[9]. Miyake, Y., Kawamura, K., Studies on Atmospheric Ozone at Tokyo, Sci. Proc. Intern. Assoc. Meteorology, Tenth General Assembly, Rome, September, 1954, pp. 172.

*[10]. This journal. Series I, Vol. 3, No. 2, 96 (1962)



----, specific activity of Sr-90 (left ordinate),
——, monthly mean altitude of 500-mb. surface (right ordinate).

Fig. 5. Seasonal variation in specific activity of strontium-90 in rain-water and monthly mean altitude of 500-mb. surface.

period, as shown in Fig. 6. That is, when this averaged trough at a high level lies over an observation point, the deposition rate of nuclear debris indicates an increasing tendency. Winter and spring correspond to the seasons in which such atmospheric conditions are satisfied. Thus this upper air condition is also considered to be one of the important factors by which the seasonal variation is enhanced. The nuclear debris, which re-entered the uppermost layer of troposphere from the stratosphere in such a manner as Stewart has pointed out, may not come back to the Earth's surface, unless the debris in the uppermost layer, where the debris is not subjected to wash-

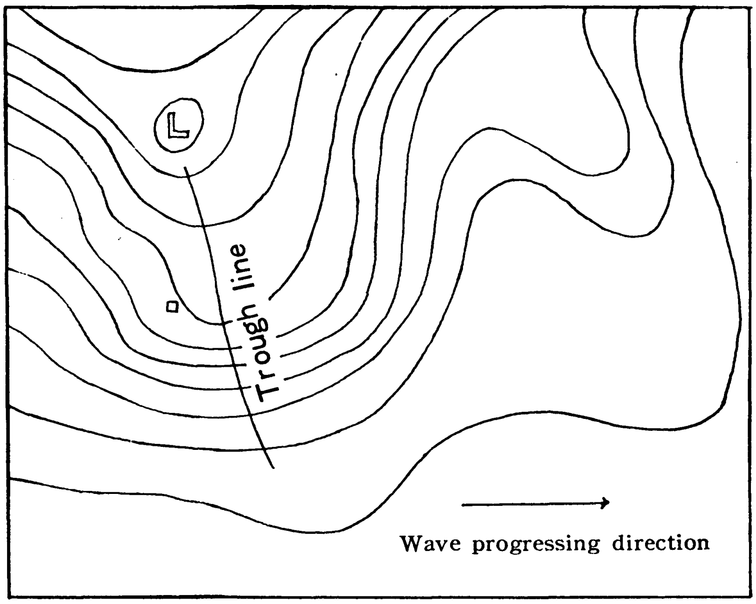


Fig. 6. A model of a trough on isobaric surface at higher level (500-mb) □, a tyipcal site where a rainfall of higher specific activity occurs.

out in rain-water, is carried down into the rain-producing layer with a process described above.

The logarithms of monthly specific activities of strontium-90 in rain-water are plotted as a function of time during the periods May—September of 1957, 1958 and 1959 in Fig. 7. It will be seen from this figure that the points plotted are distributed roughly near a straight line, namely, the specific activity of strontium-90 in rain-water shows approximately an exponential decrease during May-September. Therefore, the apparent rate of removal process becomes greatly reduced and approaches zero in autumn of the year. Concerning the data of 1959 it may be said that in the autumn of 1959 strontium-90 levels were being maintained by material from a stratospheric reservoir, since a prolonged period for atmospheric mixing elapsed after the last reported test in 1958. Then the slope of the line shows that the half-time for removal process of strontium-90 in the troposphere was about a month and this value is in good agreement with that obtained by Anderson *et al.*[*11] for 1959 and that obtained by us[*12] from the measurements of the specific activity of total fission products in rain-water.

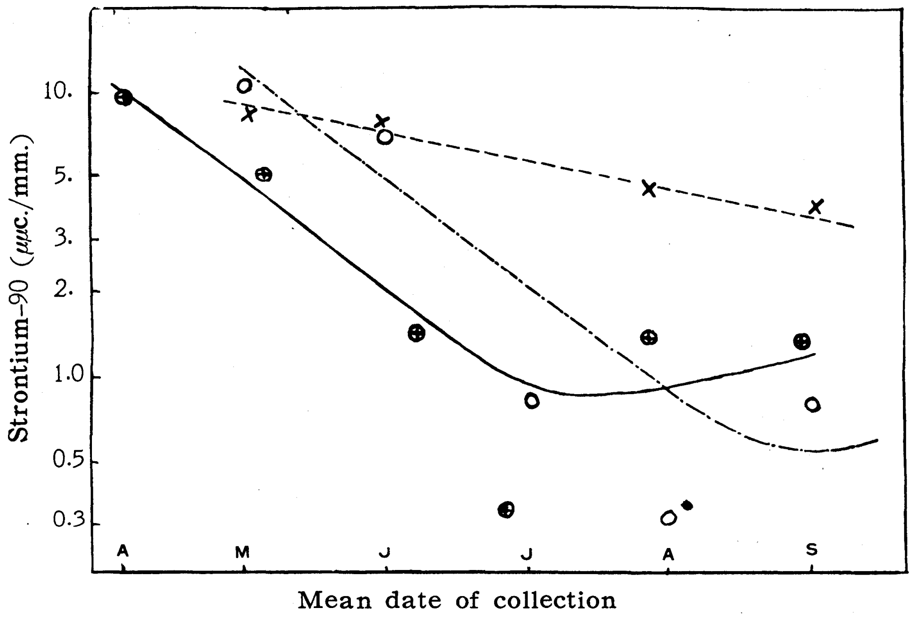


Fig. 7. Monthly specific activity of strontium-90 vs. time, during periods May-September, 1957 (—·—○—·—), 1958 (– – –×– – –), 1959 (——⊕——).

Summary

The strontium-90 contents of fallout samples have been measured during the

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[*11] Anderson, W., Bentley, R. E., Burton, L. K., Crookall, J. O., Greatorex, C. A.. Nature, 186, 925 (1960).

[*12] This journal. Series I, Vol. 3, No. 2, 98 (1962)

period 1954–1960. The results of these observed values are summarized here. From the geochemical point of view, we have discussed these data in relation to the meteorological principles and given some interpretations about the atmospheric deposition process of fission product debris from nuclear test explosions. The half-time for the removal process of strontium-90 in the troposphere was about one month for 1959. We have also found the seasonal variation in the activity concentration of strontium-90 in rain-water, and indicated the geographical locality of strontium-90 fallout in Japan.

We wish to express our sincere thanks to Dr. Y. Miyake of Meteorological Research Institute, for his helpful advice throughout this work.