気象研究所研究報告 第26巻 第1号 1-8頁 昭和50年3月
Papers in Meteorology and Geophysics Vol. 26, No. 1, pp. 1-8. March 1975

Plutonium Fallout in Tokyo

by

Yasuo Miyake

Geochemistry Research Association, Tokyo

Yukio Katsuragi and Yukio Sugimura

Meteorological Research Institute, Tokyo

(Received December 20, 1974)

Abstract

The monthly fall rates of plutonium 239 and 238 in Tokyo during the period from Jan. 1967 to the end of 1973 are given. The cumulative amounts of plutonium fallout in Tokyo from the beginning of nuclear explosion to the end of 1973 are estimated to be 1.2 mCi/km^2 and 55 μCi/km^2 respectively for 239 and 238. Owing to the accidental release of 238 due to a satellite failure, ratio of 238 to 239 increased abruptly in 1967 from 1.8% in 1966 to 16% in 1967. The maximum value of the ratio, 31%, was observed in 1970, and then the value decreased gradually to 6.3% in 1973. The ratio of 239 to Sr-90 during the period of 1968 to 1973, in which the Chinese bomb tests prevailed, was about 1%, which was lower than the former value of 1.6% in the tests conducted by the USA and the USSR. The total amounts of deposition in the northern hemisphere of 239 and 238 were calculated to be about 200 kCi and 9 kCi respectively. In 9 kCi of 238, 2 kCi was of satellite origin.

1. Introduction

It is well known that small amounts of plutonium isotopes with mass numbers 238, 239 and 240 are contained in the radioactive fallout derived from nuclear bomb explosions. Among the above isotopes the amount of 240 is much smaller than that of 239 which has almost the same alpha energy spectra as those of 240. Therefore, in this paper, the plutonium isotopes in the fallout are represented by two isotopes, i.e. 238 and 239.

In our laboratory, detailed records have been kept since 1958 on the monthly fall rate of the plutonium isotopes in Tokyo. Concerning the monthly depositions of 239 and 238 in Tokyo during the period from March 1958 to April 1969, the reports were presented already (MIYAKE et al., 1968, 1970). According to the previous reports, the cumulative amount of 239 and 238 during the period mentioned above were 0.97 mCi/km^2 and 43 μCi/km^2 respectively. The average activity ratio of 239 to Sr-90 was 1.6%. On the other hand, average activity ratio of 238 to 239 was 3.9% until the end of 1966, which was a little higher than those observed by deBORTOLI et al. (1969) and HARDY et al. (1972), i.e. respectively 2% and 2.4%. The 238 to 239 ratio began to increase

suddenly in 1967 in Tokyo (see also TELEGADAS et al., 1969), and a maximum of 31% was reached in 1970 as shown later in Table 2. This strange relative increase in 238 was due to a mistake by the USA in orbiting the SNAP-9A satellite in the southern hemisphere over the Indian Ocean in the spring of 1964, which was loaded with 17 kCi of 238 as a power source. With respect to the residence time in the stratosphere of 239, it was estimated to be about 1.7 years, which was a little longer than that of Sr-90 (MIYAKE et al., 1968).

In this paper, the results of further observations of the plutonium fallout carried out in Tokyo during the period from January 1967 to the end of 1973 are reported. Some discussions are presented concerning the inventory of the plutonium isotopes both of bomb- and satellite-origins.

2. Samples and the method of determination

The samples of meteoritic precipitations together with falling dusts were collected monthly. A water sample of about 10 to 200 liters was dried up and the residue was digested with 6 M HCl a few times, and the solution was subjected to analysis. The method of analysis was almost the same as described in the previous paper (MIYAKE et al., 1968). Plutonium in the solution was coprecipitated with a small amount of ferric hydroxide, and was isolated from a larger amount of other substances by means of an anion exchange resin in the nitrate form. The plutonium isotopes were electroplated on a disk cathode made of stainless steel (30 mm dia.) for 90 min. in the ethyl alcohol-HCl media at a voltage of 15 volts and a current of 300 mA (MIYAKE and SUGIMURA, 1968). The chemical yield of separation was 60 ±10%. Measurement of alpha-ray intensities of 239 and 238 was done with an alpha-ray spectrometer which consists of a solid state detector coupled with a multichannel pulse height analyzer.

3. Results and discussions

In Table 1 and Fig. 1, the monthly fall rates of 239 and 238, and the activity ratio of 238 to 239 in Tokyo during the period from January 1967 to the end of 1973 are shown. In Table 1, the amounts of precipitation are also given. In Table 2 and Fig. 2, the annual amounts of fall of 239 and 238 are given. In Table 2, the estimated value of the plutonium fallout from the beginning of nuclear explosions through February 1958 is also given.

The calculation was done by assuming the activity ratios of 239 to Sr-90 and 238 to 239 respectively to be 1.6% and 3.9%, on the basis of the observed value of Sr-90 deposition until February 1958 (MIYAKE et al., 1963).

As shown in Table 2, the cumulative amounts of plutonium fallout in Tokyo to the end of 1973 are 1.2 mCi/km^2 and 55 μCi/km^2 respectively for 239 and 238. As mentioned above, owing to the accidental release of 238 due to a satellite failure, the ratio of 238 to 239 increased abruptly in 1967—from 1.8% in 1966 to 16% in 1967. The maximum value of the ratio of 31% was observed in 1970 and then the value decreased gradually to 6.3% in 1973.

Since the cessation of surface tests by the US and the USSR in 1963, the annual rate of radioactive fallout in Tokyo decreased rapidly with a half-life of about one year. However, it is to be noted that the fall rate in Tokyo of Sr-90 etc. has not

Table 1. Monthly deposition of plutonium 239 and 238 and the ratio of 238 to 239 in Tokyo.

Date		239 μCi/km^2	238 μCi/km^2	238/239 (%)	Precipitation mm
1967	Jan.	1.4±0.1	0.10±0.01	7.7	33
	Feb.	2.7±0.3	0.14±0.01	5.4	55
	Mar.	1.9±0.2	0.43±0.04	22	69
	Apr.	1.6±0.2	0.26±0.03	16	111
	May	3.6±0.4	1.41±0.1	40	52
	June	1.0±0.1	0.27±0.03	27	147
	July	1.9±0.2	0.21±0.02	11	130
	Aug.	0.9±0.1	0.03±0.003	3.5	140
	Sept.	1.6±0.2	0.18±0.02	12	211
	Oct.	2.3±0.2	0.02±0.002	0.8	158
	Nov.	1.4±0.1	0.20±0.02	15	64
	Dec.	0.7±0.1	0.09±0.01	13	28
Sum.		21	3.3	av.* 15.7	1,208
1968	Jan.	1.2±0.1	0.08±0.01	7.0	9
	Feb.	0.3±0.03	0.03±0.003	9.6	72
	Mar.	2.6±0.3	0.20±0.03	7.4	77
	Apr.	4.2±0.4	0.11±0.01	2.6	131
	May	2.2±0.2	0.06±0.01	2.7	174
	June	4.2±0.4	0.17±0.02	4.1	203
	July	3.6±0.4	0.22±0.02	6.1	177
	Aug.	1.5±0.2	0.30±0.03	20	268
	Sept.	1.9±0.2	0.46±0.05	24	111
	Oct.	2.1±0.2	0.59±0.06	27	160
	Nov.	0.5±0.05	0.13±0.01	25	39
	Dec.	0.8±0.1	0.19±0.02	24	215
Sum.		25	2.5	av.* 10	1,644
1969	Jan.	1.2±0.1	0.15±0.02	13	74
	Feb.	1.0±0.1	0.12±0.01	12	114
	Mar.	1.0±0.1	0.24±0.02	24	140
	Apr.	1.0±0.1	0.15±0.02	15	72
	May	2.3±0.2	0.65±0.07	29	104
	June	0.4±0.04	0.25±0.03	58	185
	July	0.5±0.05	0.23±0.02	46	188
	Aug.	0.6±0.06	0.15±0.02	27	120
	Sept.	1.8±0.2	0.30±0.03	17	181
	Oct.	1.4±0.1	0.33±0.03	24	148
	Nov.	0.4±0.04	0.26±0.03	66	110
	Dec.	0.2±0.02	0.18±0.02	89	27
Sum.		12	3.0	av.* 25	1,471
1970	Jan.	0.2±0.02	0.32±0.03	178	74
	Feb.	0.4±0.04	0.28±0.03	77	40
	Mar.	0.4±0.04	0.34±0.03	90	51

Table 1. (continued)

Date		239 μCi/km^2	238 μCi/km^2	238/239 (%)	Precipitation mm
1970	Apr.	0.9 ±0.1	0.43 ±0.04	46	95
	May	1.0 ±0.1	0.10 ±0.01	9.7	150
	June	1.4 ±0.1	0.20 ±0.02	15	209
	July	0.5 ±0.05	0.03 ±0.003	7.7	56
	Aug.	0.4 ±0.04	0.04 ±0.004	9.8	62
	Sept.	0.3 ±0.03	0.04 ±0.004	12	84
	Oct.	0.4 ±0.04	0.07 ±0.01	17	100
	Nov.	0.2 ±0.02	0.02 ±0.002	13	127
	Dec.	0.03±0.003	0.01 ±0.001	19	28
Sum.		6	1.9	av.* 31.7	1,082
1971	Jan.	0.01±0.001	0.001±0.0005	9.1	61
	Feb.	0.3 ±0.03	0.02 ±0.002	4.7	46
	Mar.	0.8 ±0.08	0.05 ±0.005	6.6	79
	Apr.	1.0 ±0.1	0.12 ±0.01	12	111
	May	1.1 ±0.1	0.08 ±0.01	6.9	95
	June	3.5 ±0.4	0.09 ±0.01	2.7	98
	July	3.9 ±0.4	0.16 ±0.02	4.1	119
	Aug.	0.8 ±0.08	0.03 ±0.003	4.5	185
	Sept.	0.7 ±0.07	0.05 ±0.005	6.7	276
	Oct.	0.4 ±0.04	0.03 ±0.003	7.5	255
	Nov.	0.2 ±0.02	0.01 ±0.002	6.0	26
	Dec.	0.1 ±0.02	0.01 ±0.002	7.0	39
Sum.		13	0.7	av.* 53.1	1,395
1972	Jan.	0.2 ±0.02	0.01 ±0.002	3.5	129
	Feb.	0.3 ±0.03	0.01 ±0.001	4.4	158
	Mar.	0.4 ±0.04	0.02 ±0.002	4.1	40
	Apr.	1.3 ±0.13	0.10 ±0.01	7.6	175
	May	0.1 ±0.02	0.07 ±0.01	51	100
	June	1.1 ±0.1	0.07 ±0.01	6.3	138
	July	0.3 ±0.03	0.07 ±0.01	25	359
	Aug.	0.1 ±0.02	0.06 ±0.01	46	100
	Sept.	0.1 ±0.02	0.02 ±0.002	18	322
	Oct.	0.1 ±0.02	0.05 ±0.005	47	39
	Nov.	0.9 ±0.1	0.02 ±0.002	2.2	21
	Dec.	0.1 ±0.02	0.02 ±0.002	17	105
Sum.		5	0.5	av.* 10	1,701
1973	Jan.	0.04±0.004	0.01 ±0.002	21	135
	Feb.	0.09±0.009	0.02 ±0.002	25	51
	Mar.—Apr.	0.20±0.02	0.02 ±0.002	8.4	145
	May—Aug.	1.6 ±0.6	0.02 ±0.02	1.4	442
	Sept.—Dec.	0.6 ±0.06	0.02 ±0.02	2.6	432
Sum.		2.6	0.1	av.* 3.9	1,207

* The average ratio of 238 to 239 was calculated by the annual sums.

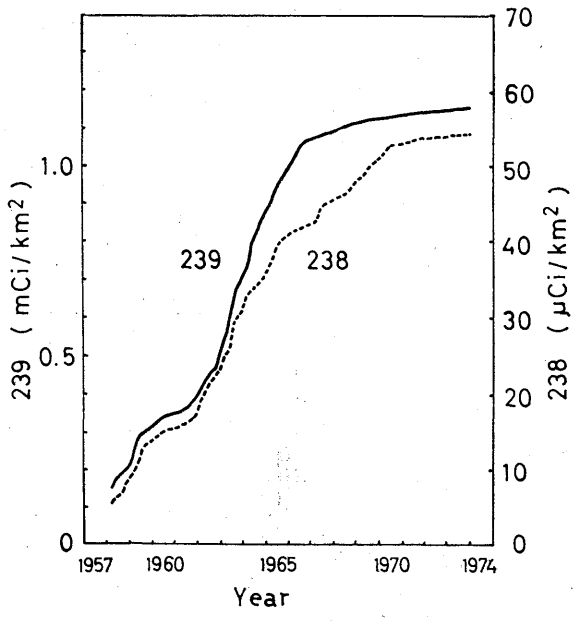

Fig. 1. The cumulative amount of plutonium 239 and 238 deposition in Tokyo.

Table 2. The annual deposition of plutonium 239 and 238 and 238/239 activity ratio.

Year	239 μCi/km²	238 μCi/km²	238/239 (%)	Precipitation mm
through Feb. 1958	147	5.7	3.8	
1958 (Mar.—Dec.)	56	2.9	5.1	1,654
1959	97	5.1	5.3	1,612
1960	43	1.7	3.9	1,175
1961	37	1.6	4.4	1,232
1962	110	6.5	5.9	1,152
1963	200	7.3	3.6	1,656
1964	185	5.3	2.9	1,132
1965	121	4.9	4.1	1,761
1966	73	1.3	1.8	1,796
1967	21	3.3	16.1	1,208
1968	25	2.5	10.0	1,644
1969	12	3.0	25.7	1,471
1970	6	1.9	31.3	1,082
1971	13	0.7	5.1	1,395
1972	5	0.5	10.1	1,701
1973	3	0.1	6.3	1,207
Year	1,162	54.3	av.* 4.7	22,881 (Mar. 1958—1973)

* The average ratio of 238 to 239 was calculated by the total sums of 239 and 238.

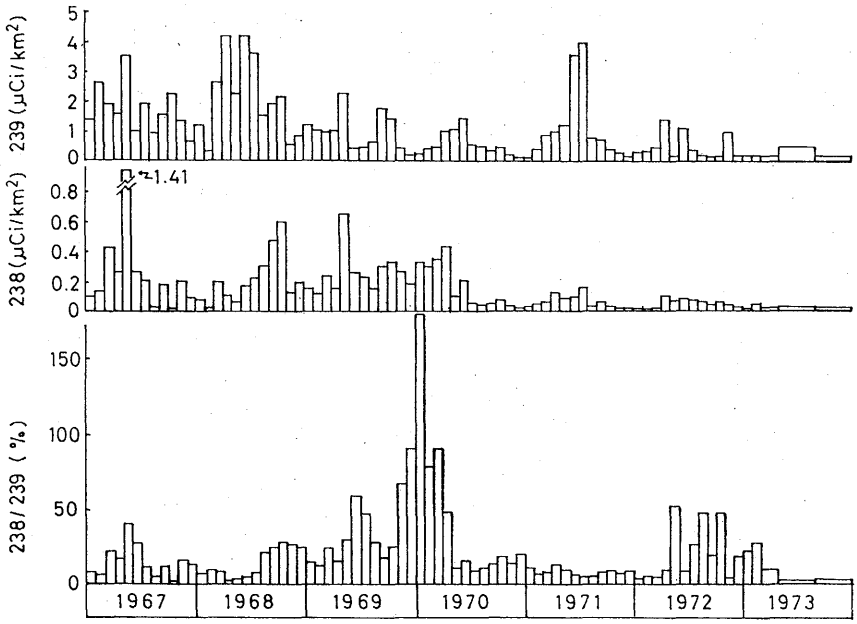

Fig. 2. Monthly deposition of plutonium 239 and 238 and 238 to 239 ratio in Tokyo.

shown any appreciable decrease since 1966 (MIYAKE and KATSURAGI, 1974). This situation is caused by the series of test explosions carried out by the People's Republic of China and France since 1964.

According to MIYAKE and KATSURAGI (1974), most of the radioactive fallout in the northern hemisphere since 1968 comes from the Chinese tests. For example, the portion occupied by the debris of Chinese bombs in the fallout increased from 62% in 1968 to 98% in 1973 in Tokyo. The ratio of 239 to Sr-90 during the period of 1968 to 1973 in which the PRC tests prevailed was about 1%, which was considerably lower than the former value of 1.6% in the tests conducted by the USA and the USSR (Table 3).

Since the year 1967, when the effect of 238 of satellite origin was clearly observed in Tokyo, the fall amounts of 239 and 238 were respectively 85 μCi/km^2 and 12 μCi/km^2. By using the isotopic ratio of 238 to 239 of 3.9% of bomb produced plutonium, the estimation on the amount of 238 of the satellite origin was done. The result showed this to be 9 μCi/km^2.

In the previous report (MIYAKE and KATSURAGI, 1970, 1974) was given the numerical value of 6.3, which was the simple ratio of Sr-90 fallout in Tokyo (mCi/km^2) to the total amount of deposition of Sr-90 on the northern hemisphere (MCi). By applying the same ratio to the plutonium fallout, the total amounts of deposition in the northern hemisphere of 239 and 238 were calculated to be about 200 kCi and 9 kCi respectively. Of the 9 kCi of 238, 2 kCi was of the satellite origin. The results of the above estimations agreed approximately well with those obtained in the soil observations carried out by HARDY et al., (1972).

Table 3. The activity ratio of annual deposition of 239 to Sr-90.

Year	239/Sr-90 (%)	Year	239/Sr-90 (%)
through the end of 1958	1.6	1966	4.1
1959	1.2	1967	2.6
1960	1.8	1968	2.0
1961	1.8	1969	1.0
1962	1.4	1970	0.4
1963	1.0	1971	1.2
1964	2.2	1972	0.9
1965	2.8	1973	1.4
			av.* 1.6

* Calculated by the total sums of Sr-90 and 239.

References

deBORTOLI, M. C. and P. GAGLIONE, 1969: SNAP plutonium-238 fallout at Ispra, Italy. Health Phys., **16**, 197–204.

HARDY, E. P., P. W. KREY and H. L. VOLCHOK, 1972: Global inventory and distribution of Pu-238 from SNAP-9A. USAEC. HASL-250, 1–32.

MIYAKE, Y., K. SARUHASHI, Y. KATSURAGI, T. KANAZAWA and S. TSUNOGAI, 1963: Deposition of Sr-90 and C_s-137 in Tokyo through the end of July 1963. Pap. Met. Geophys., **14**, 58–65.

MIYAKE, Y., Y. KATSURAGI and Y. SUGIMURA, 1968: Deposition of plutonium in Tokyo through the end of 1966. Pap. Met. Geophys., **19**, 267–276.

MIYAKE, Y. and Y. SUGIMURA, 1968: Plutonium content in the western North Pacific waters. Pap. Met. Geophys., **19**, 481–485.

MIYAKE, Y., Y. KATSURAGI and Y. SUGIMURA, 1970: A study on plutonium fallout. J. Geophys. Res., **75**, 2329–2330.

MIYAKE, Y. and Y. KATSURAGI, 1974: Origin of ^{90}Sr fallout in recent years. Tenki, a journal of Meteorological Society of Japan (in Japanese). **21**, 639–644.

TELEGADAS, K., 1969: The seasonal stratospheric distribution of plutonium-238 and strontium-90 March through November 1967. USAEC. HASL-204, I-2-16.

東京におけるプルトニウムの降下

三宅泰雄，葛城幸雄，杉村行勇

1967年1月から1973年末までの東京におけるプルトニウム-239と238の月間降下量を測定した。核実験開始以来，1973年末までの東京におけるプルトニウムの積算降下量は，239が1.2 mCi/km^2，238が55μCi/km^{2}になる。原子力電池搭載の人工衛星打上げ失敗によるプルトニウム-238の大気圏放出によって，238/239比は1966年の1.8%から1967年の16%に急増し，1970年に最大値31%に達した。この比は徐々に減少し1973年には6.3%になった。

239/Sr-90比についてみると，中国核実験に由来する放射性降下物が，全降下物中の98%をしめる1968年から1973年の期間において，約1%を示し，米国およびソ連の核実験に由来する放射性降下物の比1.6%よりも低い。東京におけるプルトニウム降下量から，北半球における全降下量を計算すると，239は200 KCi，238は9 KCiとなる。238の9 KCiのうち，2 KCiは人工衛星起源である。