気象研究所研究報告 第19巻 第2号 267—276頁 昭和43年7月
Papers in Meteorology and Geophysics Vol. 19, No. 2, pp. 267—276, July 1968

551.510.721

Deposition of Plutonium in Tokyo through the End of 1966

by

Yasuo Miyake, Yukio Katsuragi and Yukio Sugimura

Meteorological Research Institute, Tokyo

(Received May 17, 1968)

Abstract

Integrated amounts of ^{239}Pu during the period from March 1958 to the end of 1966 and ^{238}Pu (from 1959 to 1966) in Tokyo are respectively 0.92 mCi/km^2 and 34 μCi/km^2. ^{238}Pu to ^{239}Pu ratio from 1959 to 1966 is 3.9% which is nearly the same as that in fallout prior to the satellite failure. The mean ratio of $^{239}Pu/^{90}Sr$ deposition is 1.6%. Assuming 1.6% activity ratio of $^{239}Pu/^{90}Sr$, the total release of ^{239}Pu and ^{238}Pu associated with the nuclear test is estimated to be about 0.22 MCi and $<$10 kCi respectively in this period.

The mean residence time of ^{239}Pu is 1.7 years in both periods from 1959 to 1961 and from 1963 to 1966, which is slightly longer than that of ^{90}Sr (1.4 yr., 1963 to 1966).

Introduction

The fallout plutonium was derived by neutron capture of ^{238}U or by escape of fissile materials from nuclear devices.

Among plutonium isotopes, ^{239}Pu (a half-life 24,360 yr.) is most abundant (SENTYURIN, 1967). A small amount of a lighter isotope of plutonium, ^{238}Pu (a half-life 89.6 yr.) is also produced in bombs as a daughter product of ^{238}Np or ^{242}Cm. The presence of a few percent of bomb producee ^{238}Pu to ^{239}Pu was reported (SALTER, 1964). In April of 1964, an isotope-powered satellite failed to orbit and apparently reentered the atmosphere. The power unit contained 17 kCi of ^{238}Pu (HARLEY, 1964, LIST *et al*, 1966).

The plutonium fallout is much less than that of ^{90}Sr and ^{137}Cs, but because of the long-life of ^{239}Pu and the emission of α-ray, the evaluation of plutonium deposition is important.

The present paper reports on the result of study of the amount of deposition of plutonium, the activity ratios of $^{238}Pu/^{239}Pu$ and $^{239}Pu/^{90}Sr$ in fallout in Tokyo during the period from March 1958 to the end of 1966.

Samples and method of analysis

Monthly samples of rain water, snow melt and falling dust in Tokyo which were collected since March 1958 through the end of 1966 were used. A sample was

dried up and a residue was digested with 6 M HCl. After several digestions, solutions were combined and dried up followed by the digestion with concentrated nitric acid and hydrogen peroxide to decompose organic matter.

Plutonium in the extract was coprecipitated with ferric hydroxide by adding ammonium hydroxide and the precipitate was separated by centrifugation. The hydroxide containing plutonium was dissolved in 100 ml of 8 M HNO_3 and the solution was heated with potassium bromate to make plutonium tetravalent.

Separation of plutonium isotopes was carried out by means of anion exchange resin (Dowex 1×2, NO_3 form, from 50 to 100 mesh in a column of 7 mm dia. and 8 cm long) at a flow rate of 1 ml per minute. Washing of the resin was done successively with 8 M HNO_3 and 12 M HCl. Plutonium isotopes which were adsorbed on the resin were eluted with 0.5 M HCl.

The effluent containing plutonium was evaporated under an infra-red lamp, and the residue was dissolved in 1 ml of 8 M HNO_3. The solution was placed in an electroplating unit and pH of the solution was adjusted adding dropwise ammonium hydroxide solution with methyl red as an indicator.

Plutonium isotopes were electroplated on a cathode of stainless steel disk (30 mm dia. and 0.2 mm thick) for one hour at a voltage of 4.5 volts and a current of 1.0 amp. Then the disk was removed and heated with flame.

Measurement of α-ray intensities of ^{239}Pu (5.15 Mev) and ^{238}Pu (5.49 Mev) was done with an α-ray spectrometer which consists of a Frish screen grid ion chamber or a solid state detector coupled with a multichannel pulse height analyzer. The results of duplicate analyses by using ^{239}Pu or ^{238}Pu tracer showed that the chemical yield was $56 \pm 8\%$.

A typical α-spectrum of plutonium isotopes in a fallout is given in Fig. 1.

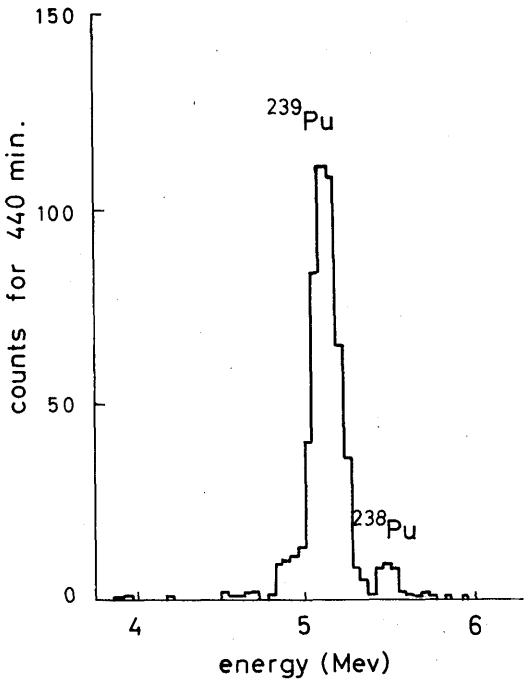

Fig. 1. An example of the α-ray spectrum of ^{239}Pu and ^{238}Pu in fallout.

Results and discussion

(1) *Deposition of ^{239}Pu and ^{238}Pu*

Table 1 and Fig. 2 show the monthly rate of deposition of ^{239}Pu fallout and the activity ratio of ^{238}Pu to ^{239}Pu in Tokyo since March 1958 to the end of 1966. The integrated annual amount of deposition of ^{239}Pu and ^{238}Pu are shown in Table 2 and Fig. 3. It is shown that deposition of plutonium in Tokyo is remarkable in the period from 1962 to 1965.

Table 1. Monthly deposition of ^{239}Pu and ratio of $^{238}Pu/^{239}Pu$.

site: Meteorological Res. Inst., Tokyo.

		^{239}Pu $\mu Ci/km^2$	$^{238}Pu/^{239}Pu$ %	Precipitation mm
1958	Mar.	10.0		63.4
	Apr.	6.4		26.1
	May—June	9.5		140.3
	July	8.2		184.4
	Aug.	0.5		80.6
	Sept.	3.9		704.0
	Oct.	11.0		275.0
	Nov.	3.6		91.5
	Dec.	2.8		89.0
	Sum	55.9		1654.3
1959	Jan.—Mar.	17	4.94	257.0
	Apr.—June	46	3.30	497.8
	July—Sept.	25	8.90	404.9
	Oct.—Dec.	9	—	452.4
	Sum	97	Av. 5.3	1612.1
1960	Jan.—Mar.	14	3.25	126.2
	Apr.—June	20	3.81	342.6
	July—Sept.	8	4.85	381.0
	Oct.—Dec.	1.3	7.50	326.1
	Sum	43.3	Av. 4.0	1175.9
1961	Jan.	1.5	2.70	31.7
	Feb.	1.9	10.2	38.6
	Mar.	5.7	1.71	102.5
	Apr.	3.8		166.9
	May—June	7.0	3.70	348.3
	July—Aug.	1.5	3.20	47.8
	Sept.—Oct.	9.7	5.90	400.0
	Nov.—Dec.	5.8	—	96.5
	Sum	36.9	Av. 4.4	1232.3
1962	Jan.—Mar.	26	10.5	103.7
	Apr.	17	4.23	112.6
	May—June	31	1.60	388.4

		^{239}Pu $\mu Ci/km^2$		$^{238}Pu/^{239}Pu$ %	Precipitation mm
1962	July	4.3		11.1	155.0
	Aug.	2.5		9.52	102.9
	Sept.	2.3		5.95	4.1
	Oct.	8.5		8.02	82.4
	Nov.—Dec.	18.1		5.31	203.8
	Sum	109.7	Av.	5.9	1152.9
1963	Jan.—Mar	45		3.37	106.0
	Apr.	21		1.53	79.0
	May	32		3.60	118.7
	June	12		10.3	239.8
	July	42		3.00	125.6
	Aug.	25		3.50	416.9
	Sept.	13		2.10	156.5
	Oct.—Dec.	10		6.30	414.2
	Sum	200	Av.	3.6	1656.7
1964	Jan.—Feb.	32		6.79	173.5
	Mar.	11		4.03	107.0
	Apr.—May	61		0.30	171.3
	June	11		4.98	120.6
	July	11		1.78	46.0
	Aug.—Sept.	14		3.33	275.0
	Oct.	26		1.00	131.7
	Nov.	13		3.02	56.3
	Dec.	6		11.1	54.4
	Sum	185	Av.	2.9	1135.8
1965	Jan.	5.9		4.10	45.1
	Feb.	6.3		—	17.0
	Mar.	5.7		11.1	61.0
	Apr.	20.0		2.20	81.3
	May	18.0		6.80	431.7
	June	14.0		4.21	217.6
	July	2.0		7.78	105.7
	Aug.	6.9		6.57	300.5
	Sept.	13.6		2.85	247.5
	Oct.	5.2		1.57	73.3
	Nov.	13.9		2.27	124.7
	Dec.	9.1		1.21	55.7
	Sum	120.6	Av.	4.1	1761.1
1966	Jan.	3.8		2.33	46.5
	Feb.	10.3		1.37	140.5
	Mar.	9.1		1.80	110.7
	Apr.	5.7		2.15	138.0

		^{239}Pu μCi/km²	^{238}Pu/^{239}Pu %	Precipitation mm
1966	May	15.8	0.95	210.9
	June	9.0	1.40	551.7
	July	9.4	1.96	166.1
	Aug.	3.5	3.66	100.0
	Sept.	1.7	5.57	195.2
	Oct.	1.6	2.00	98.3
	Nov.	2.0	1.27	28.2
	Dec.	1.4	4.44	10.2
	Sum	73.3	Av. 1.8	1796.3

Average is calculated on the basis of the total fallout of ^{239}Pu and ^{238}Pu in the given period.

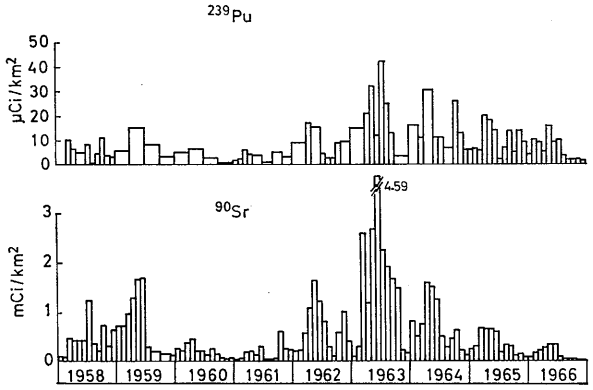

Fig. 2. The monthly deposition of ^{239}Pu and ^{90}Sr in Tokyo.

Table 2. Annual deposition of ^{239}Pu, ^{238}Pu and ratio of ^{238}Pu/^{239}Pu.

		^{239}Pu μCi/km²	^{238}Pu μCi/km²	^{238}Pu/^{239}Pu %
1958	Mar.—Dec.	56	—	—
1959	Jan.—Dec.	97	5.1	5.3
1960	〃	43	1.7	4.0
1961	〃	37	1.6	4.4
1962	〃	110	6.5	5.9
1963	〃	200	7.3	3.6
1964	〃	185	5.3	2.9
1965	〃	121	4.9	4.1
1966	〃	73	1.3	1.8
Sum		922	34 Av.	3.9

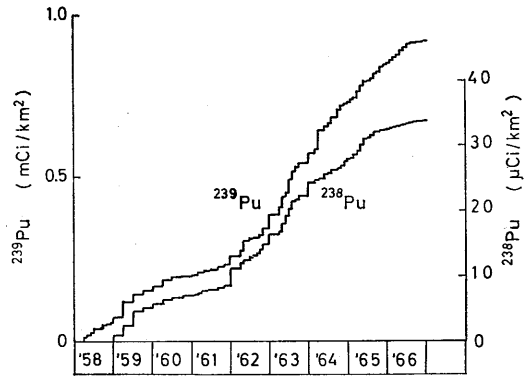

Fig. 3. The cumulative amount of ^{239}Pu and ^{238}Pu deposition in Tokyo.

The total cumulative amount through the end of 1966 is calculated to be 0.92 mCi/km^2 for ^{239}Pu and 34 μCi/km^2 for ^{238}Pu. The spring peak was clearly observed every year since 1959. The activity ratio of ^{238}Pu/^{239}Pu varied from 0.3% to 11.1% with a mean value of 3.9%.

The total atmospheric inventory in early 1964 was estimated to be 1×10^5 and 1 to 7×10^3 curies respectively for ^{239}Pu and ^{238}Pu (HARLEY, 1964). Therefore, the isotopic ratio of plutonium obtained in Tokyo is not much different from the background value prior to the satellite failure. This suggests that in Tokyo ^{238}Pu of the satellite origin would not have given an appreciable effect yet to ^{238}Pu deposition by the end of 1966.

(2) *^{238}Pu and ^{90}Sr ratio*

Table 3. The ratio of ^{239}Pu/^{90}Sr in monthly deposition.

		^{239}Pu/^{90}Sr %			^{239}Pu/^{90}Sr %
1958	Mar.	2.27	1959	Jan.—Mar.	0.72
	Apr.	1.52		Apr.—June	1.00
	May—June	1.16		July—Sept.	3.57
	July	0.68		Oct.—Dec.	2.14
	Aug.	0.14			
	Sept.	2.05			
	Oct.	1.53			
	Nov.	1.20			
	Dec.	0.44			
	Av.	1.1		Av.	1.2
1960	Jan.—Mar.	1.66	1961	Jan.	5.00
	Apr.—June	2.38		Feb.	4.75
	July—Sept.	1.57		Mar.	3.35
	Oct.—Dec.	0.72		Arp.	1.90
				May—June	1.59
				July—Aug.	2.14

		$^{239}Pu/^{90}Sr$ %			$^{239}Pu/^{90}Sr$ %
1960			1961	Sept.—Oct.	1.49
				Nov.—Dec.	1.21
	Av.	1.8		Av.	1.8
1962	Jan.—Mar.	2.63	1963	Jan.—Mar.	1.50
	Apr.	1.57		Apr.	1.81
	May—June	1.09		May	1.20
	July	0.54		June	0.26
	Aug.	0.86		July	1.88
	Sept.	2.30		Aug.	1.32
	Oct.	1.47		Sept.	0.78
	Nov.—Dec.	1.28		Oct.—Dec.	0.54
	Av.	1.4		Av.	1.1
1964	Jan.—Feb.	2.46	1965	Jan.	2.46
	Mar.	1.45		Mar.	2.17
	Apr.	1.98		Mar.	0.84
	June	0.89		Apr.	3.08
	July	2.08		May	2.81
	Aug.—Sept.	1.92		June	2.37
	Oct.	4.13		July	1.11
	Nov.	6.50		Aug.	2.03
	Dec.	4.62		Sept.	4.12
				Oct.	4.73
				Nov.	9.93
				Dec.	11.38
	Av.	2.2		Av.	2.8
1966	Jan.	4.75			
	Feb.	5.72			
	Mar.	3.96			
	Apr.	2.11			
	May	4.51			
	June	2.50			
	July	9.40			
	Aug.	5.83			
	Sept.	2.83			
	Oct.	4.00			
	Nov.	6.67			
	Dec	7.00			
	Av.	4.1			

Averages are calculated on the basis of the total fallout of ^{239}Pu and ^{90}Sr.

As shown in Table 3, the ratio of the annual deposition of $^{239}Pu/^{90}Sr$ in Tokyo ranged from 1.1 to 4.1%. The integrated amount of deposition of ^{90}Sr and ^{239}Pu in Tokyo from March 1958 to the end of 1966 are 59 mCi/km^2 and 0.92 mCi/km^2 respec-

tively. Therefore, the mean ratio of ^{239}Pu to ^{90}Sr deposition is 1.6%.

A systematic study of ^{239}Pu and ^{90}Sr in the stratosphere was carried out by United States High Altitude Sampling Program from August 1957 to June 1960 (HASP, 1961). The result showed that the $^{239}Pu/^{90}Sr$ ratio in the stratospheric air in the latitudinal zone between 30°N and 90°N ranged from 1.4% to 2.2% with a mean value of 1.7% in the period from April 1958 to June 1960.

As shown in Table 4, the ratio of $^{239}Pu/^{90}Sr$ in the fallout in Tokyo ranged from 1.1% to 1.8% during the same period with a mean ratio of 1.3% which is in fairly good agreement with the HASP result.

Table 4. The ratio of annual deposition of ^{239}Pu and ^{90}Sr.
site: Tokyo

		$^{239}Pu/^{90}Sr$ %
1958	Mar.—Dec.	1.1
1959	Jan.—Dec.	1.2
1960	〃	1.8
1961	〃	1.8
1962	〃	1.4
1963	〃	1.1
1964	〃	2.2
1965	〃	2.8
1966	〃	4.1
	Av.	1.6

The comparison of $^{239}Pu/^{90}Sr$ ratio in fallout in Tokyo and in seven cities in USA was done in Table 5 (HASL 144 (1964), 193 (1968)). As seen in Table 5, there is no significant difference in the ratio among different stations during the period except in 1966.

Table 5. $^{239}Pu/^{90}Sr$ ratio in precipitation at different stations (%).

	1960	1961	1962	1963	1966
USA					
Richmond		2.9	1.6	1.8	
Louisville	2.0	2.8			
Westwood	0.9	2.7	1.2	0.6	
Pittsburgh	0.3	3.2	1.5	0.9	
Houston	3.7	2.6	3.0	1.4	
New York					1.2
Seattle					2.1
Japan					
Tokyo	1.8	1.8	1.4	1.1	4.1

(3) *Mean residence time of ^{239}Pu in the stratosphere*

By the method proposed by the present authors (1963) the mean residence time of ^{239}Pu in the stratosphere is estimated for the period from 1959 to 1961 and 1963

to 1966 respectively. As shown in Figs. 4a and 4b, the estimated residence time is 1.7 years in both periods.

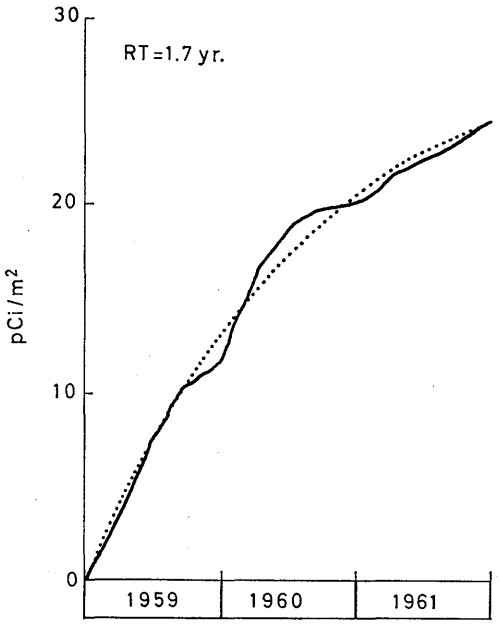

Fig. 4a. The residence time of the stratospheric ^{239}Pu during the period from 1959 to 1961.
The solid line represents the cumulative amount of the estimated airborne ^{239}Pu in the rain bearing air column calculated on the basis of observation.
The dotted line represents the calculation for 1.7 yr. residence time.

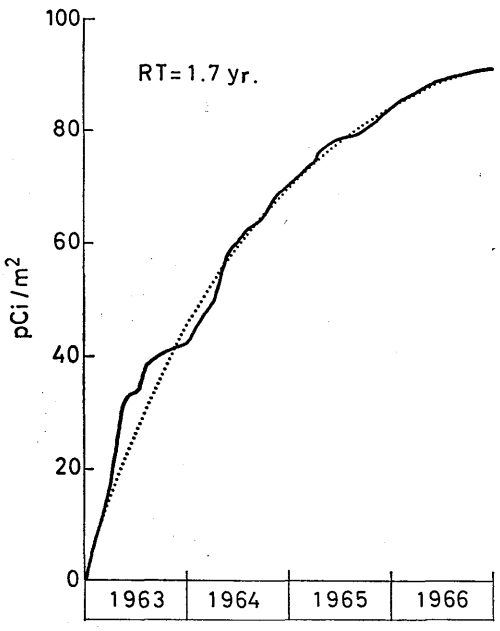

Fig. 4b. The residence time of the stratospheric ^{239}Pu during the period from 1963 to 1966.
The solid line represents the cumulative amount of the estimated airborne ^{239}Pu in the rain bearing air column calculated on the basis of observation.
The dotted line represents the calculation for 1.7 yr. residence time.

The present authors (1967) gave the residence time of about 1.4 years for the stratospheric ^{90}Sr for the period of 1963 to 1966. It is to be noted that the mean residence time of ^{239}Pu seems to be a little longer than that of ^{90}Sr.

(4) *Estimation of ^{239}Pu and ^{238}Pu production through nuclear tests*

According to the USAEC (1963) the estimated total explosion was 422 MT during the years from 1957 to 1962, in which the fission energy was regarded to be about 141 MT or 14 MCi of ^{90}Sr. Assuming the activity ratio of 1.6% for $^{239}Pu/^{90}Sr$ in fallout, the total release of ^{239}Pu is estimated to be about 0.22 MCi which is equivalent to 3.6 tons of ^{239}Pu. As the activity ratio of ^{238}Pu to ^{239}Pu is 4%, the production of the ^{238}Pu by nuclear tests will be 10 kCi. Because, some of ^{238}Pu originated from the satellite failure may have been mixed in the nuclear-test debris, the amount of 10 kCi would be the maximum value for the production of this radio-nuclide in the bombs.

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1966年末までの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

（気象研究所）

東京における ^{239}Pu（期間：1958年3月～1966年末）と ^{238}Pu（期間：1959年～1966年）の積算降下量は各々0.92mCi/km^2 と 34μCi/km^2 である。1959年から1966年末までの ^{238}Pu と ^{239}Pu の降下量の比は3.9%である。^{239}Pu と ^{90}Sr の降下量の比は1.6%である。核実験にともなう ^{239}Pu と ^{238}Pu の全放出量は，^{239}Pu と ^{90}Sr の比を1.6%と仮定すれば，各々約0.22MCi と <10kCi と推定される。

1959年～1961年と1963年～1966年の期間の ^{239}Pu の平均滞留時間は両者とも1.7年である。この値は ^{90}Sr より求められた1.4年より若干長い。