Deposition of ^{241}Am and $^{239+240}\text{Pu}$ at Tokai, Ibaraki Prefecture

Aiji YAMATO

Tokai-works, Power Reactor and Nuclear Fuel Development Corporation

Tokai-mura, Naka-gun, Ibaraki Pref. 319-11, Japan

Received January 25, 1982

In order to estimate the fallout amounts of transuranium elements, monthly deposits have been collected at Tokai, Ibaraki Prefecture, during the period from October 1978 to March 1980, and were analyzed both on ^{241}Am and $^{239+240}\text{Pu}$ also on major surface soil components, SiO_2 and Al_2O_3 . Major soil matrices contents in the deposits suggested that 5.1 % of the observed $^{239+240}\text{Pu}$ deposition was contributed by resuspension from ground deposit with wind-blow, on the other hand, much greater contribution of 10.3 % of ^{241}Am was by the resuspension.

Fallout amounts exclude resuspension of ^{241}Am and $^{239+240}\text{Pu}$ throughout the period were estimated to be 11.0 and 212 mBq/m^2 (0.30 and 5.73 pCi/m^2) respectively and annual values were 10.0 and 166 $\text{mBq/m}^2\cdot\text{y}$ (0.27 and 4.49 $\text{pCi/m}^2\cdot\text{y}$), respectively in 1979. Activity ratios of $^{241}\text{Am}/^{239+240}\text{Pu}$ changed widely. The average value of 0.066 was calculated from total amounts of each nuclides.

Key Words: deposition, americium-241, plutonium-239+240, fallout, resuspension, environmental radioactivity

1. Introduction

Among fallout transuranium elements on the earth's surface ^{241}Am is not induced directly by nuclear detonations but it has been ingrown *in situ* through the decay of ^{241}Pu which has been induced and dispersed widely by nuclear explosion tests with other Pu isotopes. The worldwide fallout amount of Pu isotopes has been measured at different latitudinal stations by Environmental Measurements Laboratory of U.S.A.^{1),2)}.

In Japan, the cumulative value up to 1977 in Tokyo was reported to be 44 Bq/m^2 (1.2 mCi/km^2) for $^{239+240}\text{Pu}$, 2.0 Bq/m^2 (0.055 mCi/km^2) for ^{238}Pu and 630 Bq/m^2 (17 mCi/km^2) for ^{241}Pu by Miyake, et al.³⁾ Consequently, deposited ^{241}Pu decays with half life of 14.4 years and bear ^{241}Am , half life of 433 years, *in situ*. Also in stratosphere, ^{241}Am ingrowth has been taking place following the earlier detonations and born ^{241}Am may have been supplied to the earth. Bennett⁴⁾ calculated the annual delivery to the earth's surface and also the ingrowth in the soil. Thein, et al.⁵⁾ measured the concentration of transuranics in rain and the annual delivery by rain in Monaco was computed

to be $0.30 \pm 0.037 \text{ Bq/m}^2$ ($8.1 \pm 0.1 \text{ pCi/m}^2$) for $^{239+240}\text{Pu}$ and $0.021 \pm 0.0074 \text{ Bq/m}^2$ ($0.58 \pm 0.02 \text{ pCi/m}^2$) for ^{241}Am . To date, no ^{241}Am direct delivery data was reported in Japan.

Krey, et al.⁶⁾ reported the activity ratio of ^{241}Am to $^{239+240}\text{Pu}$ to be about 0.25 in 1975. The present author and his co-workers⁷⁾ found recently that the ratio is much higher than that in undisturbed surface layers of soil as around 0.40 at some locations in Japan. In the same time from this profile study on soil, occurrence of translocation of these nuclides was pointed out, too. The nuclides deposited on land are partly translocated by wind-blow resuspension, by rain wash out or run-off and by other human or natural activities. As the amount of run-off by rain and transportation from land to oceans through rivers was estimated to be several percents by Miyake, et al.⁸⁾, the most of them have been remained on land after deposition. According to Yamagata, et al.⁹⁾ resuspension of ^{90}Sr by wind-blow occurs in significant quantities and 10~15% of observed fallout deposition was contributed by resuspension from soil.

In this paper, the author intends to measure the direct delivery amount of ^{241}Am and $^{239+240}\text{Pu}$, activity ratio of the nuclides and the

Table 1 Deposition of $^{239+240}\text{Pu}$, ^{241}Am and resuspended soil components at Tokai, Ibaraki Pref.

Sampling period	Observed deposition*			Resuspended		Ingrown
	$^{239+240}\text{Pu}$ (mBq/m ²)	^{241}Am (mBq/m ²)	SiO_2 (g/m ²)	$^{239+240}\text{Pu}$ (mBq/m ²), (%)**	^{241}Am (mBq/m ²), (%)**	
Oct. '78	11.1±0.59	0.89±0.30	0.42	0.33 (3.0)	0.052 (5.8)	0.35
Nov.	5.0±0.99	0.25±0.18	0.48	0.37 (7.4)	0.059 (23)	0.15
Dec.	20.7±3.0	1.04±0.33	0.56	0.41 (2.0)	0.067 (6.6)	0.59
Jan. '79	3.0±0.74	0.70±0.22	0.42	0.33 (13)	0.052 (7.3)	0.079
Feb.	—	—	—	— (—)	— (—)	—
Mar.	17.8±2.2	1.63±0.26	0.74	0.58 (3.5)	0.089 (5.5)	0.42
Apr.	35.5±7.4	3.63±0.41	2.44	1.89 (5.6)	0.30 (8.3)	0.79
May	48.8±5.2	3.89±0.74	1.18	0.93 (1.9)	0.14 (5.3)	1.02
Jun.	11.1±2.2	1.26±0.18	1.06	0.81 (7.9)	0.13 (10.3)	0.22
Jul.	15.5±4.4	0.56±0.24	0.55	0.44 (2.8)	0.067 (11.9)	0.28
Aug.	5.2±0.74	N. D.	0.90	0.70 (13.5)	0.11 (—)	0.088
Sep.	17.8±3.7	0.89±0.35	0.54	0.41 (2.3)	0.067 (7.5)	0.28
Oct.	15.5±3.0	1.41±0.52	0.68	0.52 (3.4)	0.039 (2.8)	0.22
Nov.	2.2±0.74	0.37±0.27	0.46	0.36 (16.4)	0.056 (15.1)	0.028
Dec.	N. D.	N. D.	0.84	0.67 (—)	0.10 (—)	—
Jan. '80	2.2±1.5	N. D.	1.16	0.89 (40)	0.14 (—)	0.020
Feb.	N. D.	N. D.	0.86	0.67 (—)	0.10 (—)	—
Mar.	10.4±1.5	0.30±0.17	1.42	1.11 (10.7)	0.17 (56.7)	0.069
Total	222	16.8	14.2	11.4 (5.1)	1.74 (10.3)	—

*: Uncertainties given are 1 propagated errors —: Not analyzed or not calculated

**: Percentage of resuspension to observed deposition N.D.: Not detected (1Bq=27.0 pCi)

translocated amounts by resuspension from soil of the nuclides.

2. Materials and Analytical Methods

Total deposition (wet and dry) samples were collected by a 0.5 m² stainless steel collector placed on the roof top of a building (15 m height) at Tokai works, Power Reactor and Nuclear Fuel Development Corporation, Tokai, Ibaraki Pref. (36°27'N, 140°36'E), and the samples were dried up with 2~3 ml of nitric acid by heating. The dried samples were subjected for analysis of ^{241}Am , $^{239+240}\text{Pu}$, SiO_2 and Al_2O_3 . Methods employed were mentioned elsewhere^{(10)~(12)} for ^{241}Am and $^{239+240}\text{Pu}$ and a classical method for SiO_2 and Al_2O_3 .

3. Results and Discussion

The observed results of the determination of transuranium elements and SiO_2 in these samples

are presented in Table 1. The sample No. 5, February 1979 has not been subjected for the analysis. Since the all samples contain measurable amounts of SiO_2 or Al_2O_3 , it may be said that some fractions of the transuranium elements in the samples had been resuspended from the ground accompanied with fine surface soil and the rest consists from originally delivered amounts by rain and dry fallout and ingrown ^{241}Am from ^{241}Pu decay. To account the fraction of transuranium elements accompanied with resuspended material, the determination of transuranium elements, SiO_2 and Al_2O_3 on fine surface soil samples (below 100 mesh) collected at 4 locations vicinity of the fallout measuring station. The results are set in Table 2. The average ratio of ^{241}Am and $^{239+240}\text{Pu}$ to SiO_2 in surface soil samples was found to be 0.12 mBq/g (3.24 fCi/g) - SiO_2 for ^{241}Am and 0.79 mBq/g (21.4 fCi/g) - SiO_2 for $^{239+240}\text{Pu}$, respectively. The

Table 2 Contents of major elements and transuranic elements in surface fine soil collected near fallout measuring point

St.	Location*	Composition of soil** (%)				Transuranics per unit weight of soil matrices			
		SiO ₂	Al ₂ O ₃	Fe ₂ O ₃ , TiO ₂	Ign. loss	$^{239+240}\text{Pu}$		^{241}Am	
						(mBq/g-SiO ₂)	(mBq/g-Al ₂ O ₃)	(mBq/g-SiO ₂)	(mBq/g-Al ₂ O ₃)
1	S 1.0	48.5	23.5	9.0	14.8	0.78	1.61	0.091	0.19
2	SSW 2.1	45.3	23.3	8.0	19.2	1.00	1.94	0.21	0.41
3	W 1.9	46.2	26.6	10.5	12.9	0.76	1.32	0.089	0.15
4	NNW 1.5	48.7	22.1	9.0	13.4	0.61	1.36	0.092	0.20
	Average	47.2	23.9	9.1	15.1	0.79	1.56	0.12	0.24

*: Direction and distance from fallout measuring point

**: Surface soil finer than 100 mesh was collected from cultivated farm

(1 Bq=27.0 pCi)

average $^{241}\text{Am}/^{239+240}\text{Pu}$ activity ratio in surface soil was 0.15. If one assumes that ① the resuspension of transuranics are associated with that of fine surface soil, and ② all SiO₂ or Al₂O₃ in the collector are originated only from surface soil, then one can estimate the contribution by resuspension to the observed values.

The resuspended amount of transuranics are calculated from the average values of soil assay and also of SiO₂ in the collector. These estimated contributions of resuspension are shown in 5th and 6th columns of Table 1.

Ingrown activity of ^{241}Am in the samples during the period from sample collection to the analysis was computed on the basis of ^{241}Pu activity using the following equation:

$$^{241}\text{Am}^t = \frac{\lambda_{\text{Am}}}{\lambda_{\text{Pu}} - \lambda_{\text{Am}}} ^{241}\text{Pu}^0 (e^{-\lambda_{\text{Am}} \cdot t} - e^{-\lambda_{\text{Pu}} \cdot t})$$

$$= \frac{^{241}\text{Pu}^0}{29.06} (e^{-0.0016t} - e^{-0.0481t})$$

where,

t : time in years elapsed between the sample collection and the analysis of ^{241}Am

$^{241}\text{Am}^t$: activity of ingrown ^{241}Am

$^{241}\text{Pu}^0$: activity of ^{241}Pu immediately after the sample collection

λ_{Am} : ^{241}Am decay constant

λ_{Pu} : ^{241}Pu decay constant

The half-lives of 14.4 years and 433 years respectively for ^{241}Pu and ^{241}Am were used in the computations. The contents of ^{241}Pu in the samples immediately after the collection were estimated as the ratios of ^{241}Pu to $^{239+240}\text{Pu}$ were 11.2, 10.8 and 10.0 in 1978, in 1979 and in 1980, respectively⁴⁾. The computed ingrown activity

of ^{241}Am were shown in the 7th column of Table 1.

By subtracting the resuspended amount and the ingrown activity of ^{241}Am from observed deposition, fallout origin transuranics are calculated. The results are illustrated in Fig. 1.

*: Not detected (1Bq=27.0 pCi)

Fig. 1 Monthly delivery of $^{239+240}\text{Pu}$ and ^{241}Am and precipitation at Tokai, Ibaraki Pref.

The average resuspended percentages to observed deposition are 10.3% for ^{241}Am and 5.1% for $^{239+240}\text{Pu}$, respectively. Thein, et al.⁵⁾ mea-

sured the direct delivery of these nuclides by rain into the Mediterranean, and pointed out that ① the percentage contribution of resuspended solids does not exceed 0.8% for $^{239+240}\text{Pu}$ and 2.8% for ^{241}Am , and ② consistently higher resuspended values were observed on ^{241}Am than on $^{239+240}\text{Pu}$, reflecting higher $^{241}\text{Am}/^{239+240}\text{Pu}$ ratio in resuspended matter than in rain water. They did not subjected the solid materials in the collector for analysis. In present study, all the solids were subjected for analysis and the resuspended solids which consist mostly from fine surface soil of surrounding farms contains higher activity ratio of the nuclides by *in situ* build-up of ^{241}Am through the decay of ^{241}Pu which had been deposited on the soil at some earlier time. From the differences of geographical features between Monaco and Tokai, it is not difficult to say that the resuspended amount of soil at Tokai is greater than that of Monaco and presumably the resuspended transuranics, too. Yamagata, et al.⁹⁾ reported that 10~15% of ^{90}Sr deposition was contributed by the translocation of ground deposit through wind-blow dust from determination of monthly collection of 25 locations in Japan in 1968~1969. This value on ^{90}Sr is close to that for ^{241}Am in the present results.

It has already known that the amounts of fallout radionuclides depend on the amount of precipitation e.g., in case of ^{90}Sr , rain brought down more than 90% of total delivery in Tokyo¹³⁾. Comparison of the delivery data of $^{239+240}\text{Pu}$ and ^{241}Am in Fig. 1, they are closely correlated with the amounts of precipitation except in December 1978. A nuclear explosion test was performed by Peoples Republic of China in this month.

The Fig. 1 indicates that the delivery of both nuclides was higher relative to the precipitation during the period of March~May 1979 reflecting spring peak phenomena and also high in autumn relating to much precipitation in this period of September~October 1979.

Total amounts of direct fallout of $^{239+240}\text{Pu}$ and ^{241}Am through the period of observation is estimated to be 212 and 11.0 mBq/m² (5.73 and 0.30 pCi/m²). In 1979 the annual values of the nuclides were 166 and 10.0 mBq/m² (4.49 and

0.27 pCi/m²) respectively. These values are almost a half of those reported by Thein, et al. in Monaco. This difference may be caused by differences of meteorological and geographical conditions between Tokai and Monaco. Bennett¹⁴⁾ calculated the deposition rates of $^{239+240}\text{Pu}$ and ^{241}Am to be 144 and 8.1 mBq/m²·y (3.9 and 0.22 pCi/m²·y), respectively in the New York region in 1979. These data are close to the present results.

Based on the annual delivery values as shown in Fig. 1, the activity ratio for $^{241}\text{Am}/^{239+240}\text{Pu}$ is found to be 0.066. This is close with Bennett's¹⁴⁾ calculation of 0.056 in 1979. Concentration of ^{241}Am , ^{238}Pu and $^{239+240}\text{Pu}$ in surface air in Monaco was measured to be 48.1 ± 7.4 , 18.5 ± 3.7 nBq/m³ and 1.18 ± 0.17 $\mu\text{Bq}/\text{m}^3$ (1.3 ± 0.2 , 0.5 ± 0.1 and 31.7 ± 4.7 aCi/m³), respectively in January~March 1977¹⁴⁾, therefore activity ratio of $^{241}\text{Am}/^{239+240}\text{Pu}$ was $4.0 \pm 0.9\%$. This activity ratio is also not much different from the present data.

The variations in the $^{241}\text{Am}/^{239+240}\text{Pu}$ ratios for monthly delivery values with time are illustrated in Fig. 2. The ratios vary irregularly over a wide range of 0.005~0.21. The precise reasons for the variation is not perceived. It is suggested by Thein, et al.⁵⁾, however, that the variation may be related to the different mean age of the fallout brought down to the earth's surface in different seasons. They pointed also that the primary peak of the activity ratio appeared towards the end of the dry season. In present work, two peaks in January and November 1979 were found, but in months of December in 1978 and in 1979 no high ratios were found. They have dry climate in this season in the Pacific coast of Japan. In Decem-

Fig. 2 Activity ratio of ^{241}Am to $^{239+240}\text{Pu}$ in fallout at Tokai, Ibaraki Pref.

ber 1978, the delivery of $^{239+240}\text{Pu}$ increased much comparing with that of ^{241}Am and the activity ratio decreased. In December 1979, both activities for $^{239+240}\text{Pu}$ and for ^{241}Am were too small to be measured. So that the activity ratio could not be calculated.

It is observed, however, that the ratios do not vary in duration of spring peak season of March~June 1979 showing the rather constant values of 0.057~0.086.

Acknowledgement

The present work was done as a part of Scientific Research Fund (A), Prof. N. Ikeda group, supported by the Ministry of Education. The author wishes to express his thanks to Prof. N. Ikeda, Tsukuba Univ. for his advices on the manuscript and to the members of Environmental Protection Section, P.N.C. for their technical assistance.

References

- 1) Harley, J.H.: "Proc. Environ. Plutonium Symp.", LA-4756 (1971)
- 2) Hardy, E.P., et al.: *Nature*, **241**, 444~45 (1973)
- 3) Miyake, Y., et al.: "Proc. 6th Int. Congr. Rad. Res.", ed by S. Okada, et al., pp. 940~48, Toppan Printing Co., Tokyo (1979)
- 4) Bennett, B. G.: "Environmental Aspects of Americium", Thesis New York Univ. (1979)
- 5) Thein, M., et al.: *Geochim. Cosmochim. Acta*, **44**, 1091~97 (1980)
- 6) Krey, P.W., et al.: "Transuranium Nuclides in the Environment", pp. 671~78, IAEA, Vienna (1976)
- 7) Yamato, A., et al.: "Int. Symp. Migration in the Terrestrial Environment of Long-Lived Radionuclides from the Nuclear Fuel Cycle", IAEA-SM-257/139, Knoxville Tenn. U.S.A. (1981)
- 8) Miyake, Y., et al.: *Pap. Meteorol. Geophys.*, **24**, 75~78 (1973)
- 9) Yamagata, N., et al.: *Bull. Inst. Pub. Health*, **18**, 165~86 (1969)
- 10) Yamato, A.: *Radioisotopes*, **30**, 104~08 (1981)
- 11) Yamato, A., et al.: *J. Radioanal. Chem.* (to be published)
- 12) Ballestra, S., et al.: Radioactivity in Sea Series, No. 66, IAEA, Vienna (1979)
- 13) Miyake, Y., et al.: *J. Geophys. Res.*, **67**, 189~93 (1962)
- 14) M. Thein: Private Communication (1979)

要 旨

茨城県東海村における ^{241}Am および $^{239+240}\text{Pu}$ の降下量

大 和 愛 司

動力炉・核燃料開発事業団東海事業所

319-11 茨城県那珂郡東海村村松

超ウラン元素のフォールアウト量を知るために、茨城県東海村で1978年10月から1980年3月までの月間降下物を集め、 ^{241}Am および $^{239+240}\text{Pu}$ ならびに表土の主要成分である SiO_2 および Al_2O_3 について分析した。降下物中の表土主成分含量から、観測された $^{239+240}\text{Pu}$ 降下量の5.1%が、地面からの風による再浮遊の寄与であり、一方、 ^{241}Am の場合、10.3%とより大きな寄与を受けていることが示された。再浮遊の寄与を除いたフォールアウト量は、観測期間全体で ^{241}Am が 11.0mBq/m^2 (0.30pCi/m^2)、 $^{239+240}\text{Pu}$ が 212mBq/m^2 (5.73pCi/m^2) で、1979年の年間降下量はおおの10.0および $166\text{mBq/m}^2\cdot\text{y}$ (0.27 および $4.49\text{pCi/m}^2\cdot\text{y}$) であった。 $^{241}\text{Am}/^{239+240}\text{Pu}$ の放射能比は広範囲に変化した、それぞれの全降下量から平均値として0.066が求まった。