Pu Isotopes, ²⁴¹Am and ¹³⁷Cs in Soils from the Atomic Bombed Areas in Nagasaki and Hiroshima

MASAYOSHI YAMAMOTO, KAZUHISA KOMURA*, MASANOBU SAKANOUE*, MASAHARU HOSHI**, SHOZO SAWADA** and SHUNZO OKAJIMA***

School of Pharmacy, Hokuriku University, Kanagawa-machi, Kanazawa-shi, Ishikawa 920-11, Japan (Received June 7, 1984; Revised version accepted January 23, 1985)

Plutonium isotopes/Americium/Soil/Atomic bomb/Nagasaki/Hiroshima

Plutonium Isotopes, ²⁴¹Am and ¹³⁷Cs in soil samples collected from Nagasaki and Hiroshima were measured to evaluate the contribution of residual radioactivity derived from the atomic bombs dropped in 1945.

In the soils from Nishiyama area of Nagasaki City, activity ratios different from those of global fallout were found for the ²³⁸Pu/²³⁹, ²⁴⁰Pu (0.05-0.06), ²³⁹, ²⁴⁰Pu/¹³⁷Cs (0.2-0.3), ²⁴¹Am/²³⁹, ²⁴⁰Pu (0.04-0.05) and ²⁴⁰Pu/²³⁹Pu (0.08-0.13), activity ratios indicating that a fair amount of plutonium derived from the atomic bomb had accumulated in this area. On the other hand, in the soils from the "Black-rain" area of Hiroshima, no significant difference from the global fallout was found for the activity ratios among the Pu isotopes, ²⁴¹Am and ¹³⁷Cs except for the soils collected at 10 km and 12 km in the NNW direction from the epicenter, where a factor 2 to 3 lower ²⁴¹Am/²³⁹, ²⁴⁰Pu activity ratio was observed. However, no evidence of the contribution of the atomic bomb was found by the measurement of ²⁴⁰Pu/²³⁹Pu isotopic ratio.

INTRODUCTION

Forty years have elapsed since the atomic bombs (hereinafter A-bombs) were exploded over Hiroshima and Nagasaki Cities in Japan. During this period, a number of studies^{1, 2)} have been made to analyze and document the radiation effects of the A-bombs on the survivors and their offsprings. On the other hand, studies^{1, 3)} on the residual and fallout radioactivities in the A-bombed areas have been made only a little as compared with those on radiation

^{*}Low-Level Radioactivity Laboratory, Kanazawa University, Tatsunokuchi-machi, Nomi-gun, Ishikawa 923-12, Japan.

^{**}Research Institute for Nuclear Medicine and Biology, Hiroshima University, Kasumi 1-2-3, Minami-ku, Hiroshima 734, Japan.

^{***}Department of Radiation Biophysics, Atomic Disease Institute, Nagasaki University, School of Medicine, Sakamoto-machi 12-4, Nagasaki 852, Japan.

山本政儀: 北陸大学薬学部放射薬品学教室, 金沢市金川町ま3, 〒920-11

小村和久、阪上正信: 金沢大学理学部附属低レベル放射能実験施設, 石川県能美部辰ロ町字和気オ24, 〒923-12

星 正治;澤田昭三:広島大学原爆放射能医学研究所,広島市南区霞1丁目2番3号,〒734

岡島俊三:長崎大学医学部原爆後障害医療研究施設,長崎市坂本町12番4号,〒852

effects. The field survey performed soon after the A-bomb explosions³⁾ has shown that the Nishiyama area of Nagasaki and the "Black-rain" area of Hiroshima (Fig. 1) had received the heaviest fallout from the A-bombs.

In order to investigate the residual radioactivities in the Hiroshima and Nagasaki A-bombed areas, in 1976 and 1978 several hundred soil samples were systematically collected in the two A-bombed areas as a national project conducted by the Ministry of Public Health⁴⁾. The measurement of long-lived ¹³⁷Cs was made for these soil samples because this nuclide is well adsorbed in surface layer of the soil⁵⁾ and can be used as a good measure for the evaluation of the fallout accumulation. The measurement of ⁹⁰Sr was also performed for some samples. It has been reported that the accumulation levels of ¹³⁷Cs in the soils collected from the Nishiyama area of Nagasaki are significantly higher than those from the areas other than Nishiyama, while no appreciable difference of ¹³⁷Cs accumulation from reference area was found even in the "Black-rain" area of Hiroshima.

Recently, the ²³⁴U/²³⁸U activity ratio instead of measuring ²³⁵U/²³⁸U has been measured to study the contribution of fallout uranium derived from the Hiroshima A-bomb. But, no evidence of ²³⁵U-enriched uranium was found for the surface soil collected from the "Blackrain" area⁶).

By contrast, in Nagasaki it has been demonstrated that the surface soil of the Nishiyama area is highly contaminated with the plutonium derived from the A-bomb^{7, 8)}. We have estimated that the plutonium derived from A-bomb is as much as 95–97% of the total plutonium in the soils collected from the Nishiyama area by the measurements of ²⁴¹Pu/^{239, 240}Pu and ²⁴¹Am/^{239, 240}Pu activity ratios⁹⁾. This finding was confirmed also by the measurement of ²⁴⁰Pu/²³⁹Pu isotopic ratio^{9, 10)}.

Since the measurements of Pu isotopes and ²⁴¹Am were found to be very useful for the discrimination of the plutonium due to A-bomb in 1945 from global fallout plutonium, this technique was applied for the soils collected from various locations in the vicinity of Nagasaki including Nishiyama area in order to study the contamination level and the spatial distribution of plutonium derived from A-bomb. This technique was applied also for the soils collected from Hiroshima, where systematic measurements of Pu isotopes and ²⁴¹Am have not been made yet.

In this paper, we report the measurements of Pu isotopes, ²⁴¹Am and ¹³⁷Cs for the Nagasaki and Hiroshima soils. Present data are believed to be useful for the evaluation of population dose from the fallout radionuclides due to A-bombs.

MATERIALS AND METHODS

Several hundred surface soils were collected in 1976 at 2 km interval from 2 km up to 30 km from the epicenter of A-bomb both in Hiroshima and Nagasaki⁴⁾. Undisturbed area was chosen and a 10 cm column of surface soil, in which more than 90% of the fallout plutonium and ¹³⁷Cs are known to be accumulated⁵⁾, was sampled to measure the accumulation levels of fallout nuclides. Sampling locations are shown in Fig. 1 for Nagsaki and Hiroshima. These samples are dried at 100°C, crushed and sieved through a 2-mm screen.

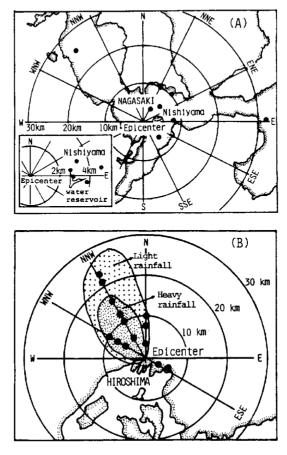


Fig. 1. Sampling points for soils from (A) Nagasaki and (B) Hiroshima.

Pu isotopes, ²⁴¹Am and ¹³⁷Cs were measured for some of these soils. About 50 g aliquot was taken and contents of ¹³⁷Cs and natural radionuclides were measured first by non-destructive gamma-spectrometry using a low background Ge(Li) detector. Then, 20–50 g aliquot was taken and Pu isotopes (²³⁸Pu and ²³⁹, ²⁴⁰Pu) and ²⁴¹Am were radiochemically analyzed by the method described by Yamato¹¹⁾, ²⁴²Pu and ²⁴³Am being used as tracers during chemical procedures. The plutonium and americium were electroplated on the stainless steel discs of 20 mm in diameter and their activities were measured by alpha-spectrometry using a Si(Au) surface barrier detector. For several soil samples, the isotopic ratio ²⁴⁰Pu/²³⁹Pu was further measured using a rather amount of soil (about 100 g for Nagasaki soils and about 1000 g for Hiroshima soils)¹⁰⁾.

Table 1. Concentrations of ^{239, 240}Pu, ²³⁸Pu, ²⁴¹Am and ¹³⁷Cs in soils (0-10 cm depth) collected from the Nagasaki atomic bombed area in 1976 and their activity ratios.

	Distance* (km)	Concentration (pCi/kg)						Activity ratio		
Direction		Pu-239, 2	40	Pu-238	Am-241	Cs-13	7**	Pu-238 Pu-239, 240	Am-241 Pu-239, 240	Pu-239, 240 Cs-137
[Nishiyama area]										
E	2.7	·760/31 (45	5.1) [#]	43.2/5.1	30.2/2.3*2	2.45/.05	(145) [#]	0.056/.006	0.040/.003	0.31/.01
E	2.8*1	1380/30		76.1/7.0	56.3/3.3*2	6.00/.05		0.055/.005	0.041/.003	0.23/.01
E	3.5	337/9 (14	4.1)	17.5/1.0	16.2/1.9	2.37/.06	(99.1)	0.052/.003	0.048/.003	0.15/.01
E	3.8	671/20 (36	6.1)	36.9/2.2	27.8/2.9*2	2.66/.06	(144)	0.055/.003	0.042/.005	0.25/.01
E	4.5	462/9 (18	8.3)	23.1/1.2	23.5/1.4	2.48/.09	(98.7)	0.050/.003	0.051/.003	0.19/.01
[Oth	er area]									
NNE	4	44.3/1.5 (1	1.7)	1.83/.16	11.2/.6	1.68/.06	(65.8)	0.041/.004	0.25/.02	0.027/.001
NNE	10	33.4/1.7 (1	1.8)	0.67/.14	9.0/.6	2.18/.06	(118)	0.020/.004	0.27/.03	0.016/.001
NE	6	40.3/1.9 (1	1.5)	1.12/.16	12.0/.9	2.29/.06	(85.9)	0.028/.004	0.30/.03	0.017/.001
NE	10	29.6/.9 (1	1.6)	0.91/.09	8.6/.7	1.63/.02	(89.4)	0.030/.003	0.29/.03	0.018/.001
E	8	57.8/2.0 (3	3.7)	2.87/.12	5.1/.5	0.93/.03	(59.8)	0.050/.003	0.09/.01	0.062/.003
E	32	35.8/2.0 (2	2.1)	1.14/.15	11.1/1.4	2.28/.05	(135)	0.032/.005	0.31/.04	0.016/.001
SE	6	42.9/1.5 ()	1.7)	1.77/.16	8.5/.6	1.93/.05	(74.9)	0.041/.004	0.20/.02	0.022/.001
NW	26	20.4/.8 (1	1.0)	0.73/.11	6.5/.5	1.27/.03	(64.4)	0.036/.005	0.31/.04	0.016/.001

^{*)}Distance from epicenter. **)pCi/g. #)mCi/km 2 .

The number after slash denotes the propagated counting error of one sigma: $760/31 = 760 \pm 31$. The ¹³⁷Cs values were decay corrected to the dates of sampling, while the ²⁴¹Am values are as of the dates of analysis from the end of 1982 to the beginning of 1983 (*2: values at January 1981).

^{*1)}Sampling date: July 1979 (0-5 cm depth).

Table 2. Concentrations of ^{239, 240}Pu, ²³⁸Pu, ²⁴¹Am and ¹³⁷Cs in soils (0-10 cm depth) collected from the Hiroshima atomic bombed area in 1976 and their activity ratios.

Direction	Distance* (km)	Concentration (pCi/kg)						Activity ratio			
		Pu-239, 240		Pu-238	Am-241	Cs-137**		Pu-238 Pu-239, 240	Am-241 Pu-239, 240	Pu-239, 240 Cs-137	
N	4	12.7/.5	(1.32)#	0.54/.08	3.7/.3	0.66/.02	(67.7)#	0.043/.007	0.29/.03	0.019/.001	
N	8	12.9/.4	(1.56)	0.48/.07	3.7/.3	0.75/.03	(91.0)	0.037/.006	0.29/.03	0.017/.001	
N	10	14.8/.7	(1.31)	0.47/.08	4.4/.3	0.80/.02	(70.7)	0.032/.006	0.30/.02	0.019/.001	
NNW	6	15.5/.7	(1.09)	0.33/.05	3.1/.3	1.00/.01	(70.3)	0.021/.003	0.20/.02	0.016/.001	
NNW	10	13.9/.7	(1.08)	0.42/.05	1.9/.2	0.53/.02	(41.4)	0.030/.004	0.13/.02	0.026/.002	
NNW	12	17.7/.8	(0.98)	0.50/.06	1.2/.1	1.07/.02	(59.2)	0.028/.004	0.07/.01	0.017/.001	
NNW	16	25.3/1.8	(0.65)	0.89/.08	7.5/.7	1.39/.03	(36.2)	0.035/.004	0.30/.03	0.018/.001	
NNW	22	40.3/2.0	(1.39)	1.75/.20	12.0/.9	2.47/.06	(85.5)	0.043/.005	0.30/.03	0.016/.001	
NNW	24	12.0/.4	(0.53)	0.40/.05	3.4/.3	0.52/.02	(22.9)	0.033/.004	0.28/.03	0.023/.001	
WNW	6	11.4/.5	(0.79)	0.34/.04	2.2/.2	0.79/.01	(54.6)	0.030/.004	0.19/.02	0.014/.001	
WNW	8	16.3/.7	(1.53)	0.54/.08	3.5/.4	0.77/.02	(72.0)	0.033/.005	0.21/.02	0.021/.001	
WNW	10	13.0/.6	(1.10)	0.51/.07	2.6/.3	1.03/.03	(87.3)	0.039/.006	0.20/.02	0.013/.001	
ESE	4	18.5/.7	(1.52)	0.61/.05	3.5/.3	1.36/.02	(112)	0.033/.003	0.19/.02	0.014/.001	
ESE	6	6.2/.3	(0.64)	0.18/.02	1.4/.2	0.46/.01	(47.8)	0.029/.004	0.23/.03	0.013/.001	

^{*)}Distance from epicenter. **)pCi/g. #)mCi/km².

The number after slash denotes the propagated counting error of one sigma: $12.7/.05 = 12.7 \pm 0.5$. The ¹³⁷Cs values were decay corrected to the dates of sampling, while the ²⁴¹Am values are as of the dates of analysis from the end of 1982 to the beginning of 1983.

Table 3. Atom ratio ²⁴⁰Pu/²³⁹Pu and activity ratios of some nuclide pairs in soils from Nagasaki and Hiroshima

			Pu-239, 240 concentration (pCi/g)		Atom ratio		
Location	Sample (Depth)	Sampling date		Pu-238 Pu-239, 240	Am-241 Pu-239, 240	Pu-239, 240 Cs-137	Pu-240 Pu-239
	E-2.8* (0-5 cm)	July 1979	1.380/.030	0.055/.005	0.041/.003*1	0.23/.01	0.031/.012
"Nishiyama" district (Nagasaki)	E-3.5 (0-10 cm)	June 1976	0.337/.009	0.052/.003	0.048/.003*2	0.15/.01	0.034/.010
	E-4.5 (0-10 cm)	June 1976	0.462/.009	0.050/.003	0.051/.003*2	0.19/.01	0.020/.009
"Black-rain"	NWN-10 (0-5 cm)	July 1983	0.050/.002	0.033/.003	0.090/.010*3	0.015/.001	0.179/.023
area (Hiroshima)	NWN-12 (0-5 cm)	July 1983	0.061/.004	0.036/.002	0.13/.01*3	0.020/.001	0.173/.023

^{*)}Direction and distnce from epicenter.

The number after slash denotes the propagated counting error of one sigma: 1.380/.030 = 1.380 ± 0.030. The ^{239, 240}Pu/¹³⁷Cs ratios were decay corrected to the dates of sampling, while the ²⁴¹Am/^{239, 240}Pu ratios are as of the dates of anlysis: *1) January 1981, *2) March 1983, *3) August 1983.

RESULTS AND DISCUSSION

The contents of Pu isotopes, ²⁴¹Am and ¹³⁷Cs, and their activity ratios are summarized in Tables 1 and 2 for Nagasaki and Hiroshima soils, respectively. The ²⁴⁰Pu/²³⁹Pu isotopic ratios are given in Table 3. The contents of natural radionuclides measured simultaneously with ¹³⁷Cs are given in Appendix. Errors given in these tables represent one standard deviation of counting due to alpha- and gamma-ray measurements. The ¹³⁷Cs values are corrected for decay to the dates of the soil sampling, while ²⁴¹Am are as of the dates of chemical analysis made from the end of 1982 to the beginning of 1983.

Nagasaki soils

By the previous works⁹⁾, it is well known that the surface soil of the Nishiyama area is highly contaminated by plutonium derived from the A-bomb in 1945. However, only a few data⁸⁾ are available for the area other than Nishiyama. A comparison was made for the contents of Pu isotopes, ²⁴¹Am and ¹³⁷Cs, and their activity ratios in the surface soils collected from Nishiyama and other areas.

The accumulations of ^{239, 240}Pu and ¹³⁷Cs per unit area are compared in Fig. 2. As shown in this figure, extremely high ^{239, 240}Pu accumulation was found in the soils of the Nishiyama area. The accumulation level of ^{239, 240}Pu ranging from 14.1 to 45.1 mCi/km² found in the Nishiyama area is higher on an average by a factor of about fifteen than the values of 1.0–3.7 mCi/km² in other areas. Whereas, the accumulation level of ¹³⁷Cs in the Nishiyama area (98.7 to 145 mCi/km²) is not so largely different from the levels in the soils collected from other areas (59.8 to 135 mCi/km²), indicating that most of ¹³⁷Cs is derived from global fallout and the contribution of the A-bomb in 1945 is relatively small.

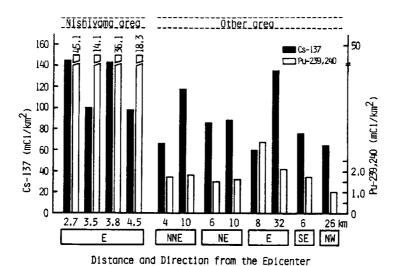


Fig. 2. Accumulations of ^{239, 240}Pu and ¹³⁷Cs in soils (0-10 cm depth) collected from the Nagasaki atomic bombed area in 1976.

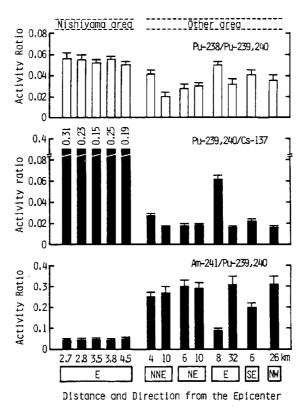


Fig. 3. Activity ratios of ²³⁸Pu/^{239, 240}Pu, ^{239, 240}Pu/¹³⁷Cs and ²⁴¹Am/^{239, 240}Pu in the Nagasaki soils. (The propagated counting error of one sigma is given by the symbol "T" as the highest value on the top of each bar.)

In order to clarify the contribution of the A-bomb, it seems much better to use the activity ratios instead of comparing only the accumulation levels of the fallout nuclides. Because the accumulation of fallout nuclides in a given area is influenced by many causes such as the difference of soil nature, configuration of the ground surface, local meterology such as the precipitation and so on. Fig. 3 shows the activity ratios of ²³⁸Pu/^{239, 240}Pu, ^{239, 240}Pu/¹³⁷Cs and ²⁴¹Am/^{239, 240}Pu. As shown in this figure, the activity ratios of ²³⁸Pu/^{239, 240}Pu (0.050–0.056), ^{239, 240}Pu/¹³⁷Cs (0.15–0.31) and ²⁴¹Am/^{239, 240}Pu (0.04–0.05) of the Nishiyama area are distinctly different from the corresponding global fallout values of 0.03–0.04¹²), about 0.017¹³) and about 0.3¹⁴), respectively. The ²⁴⁰Pu/²³⁹Pu isotopic ratios (0.02–0.04), shown in Table 3, deviate significantly from the commonly accepted value of 0.18 for global fallout ¹⁵). The difference of the activity ratios from global fallout values was found also for the soil collected from 8 km east of the epicenter of the A-bomb (see Fig. 1), though difference of activity ratios from the global fallout values is not so large as the case of the soils from Nishiyama area. Present data indicate that the contribution of the A-bomb in 1945 is detectable at as far as 8 km from the epicenter of the Nagasaki A-bomb. Measurement of ²⁴¹Pu or ²⁴⁰Pu/

²³⁹Pu isotopic ratio is desirable to evaluate quantitatively the contribution of the A-bomb. Within the statistical error of the measurement, the soils collected from other areas showed the same activity ratios as those of global fallout.

These results indicate that plutonium scatterd without fission reaction had fallen out mostly in the eastern direction centered at around Nishiyama area locating 2-4 km from the epicenter.

Hiroshima soil

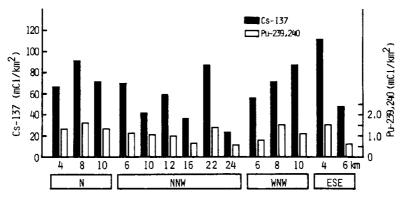
As is well-known, uranium enriched in ²³⁵U was used for the A-bomb dropped on Hiroshima. However, no evidence of enriched ²³⁵U has been obtained by the measurement of fallout radionuclides performed soon after the detonation³⁾, and radiochemical analysis of uranium isotopes in surface soils⁶⁾.

At the time of detonation, transuranium nuclides are expected to be formed by successive neutron capture reaction followed by beta-decay from uranium of the A-bomb. Such transuranics might have fallen out on the ground surface near epicenter of the A-bomb and some deviations in isotopic ratios of these nuclides from those of global fallout might be expected. Present study was made to examine the possibility of finding the contribution of A-bomb by the measurements of Pu isotopes and ²⁴¹Am together with ¹³⁷Cs in the soils collected from the "Black-rain" area where the heaviest fallout deposition had been observed soon after the detonation of the A-bomb. Besides the "Black-rain" area, some soils were collected from other area (at 4 and 6 km in the ESE direction from the epicenter shown in Fig. 1) and analyzed for comparison.

The accumulations of ^{239, 240}Pu and ¹³⁷Cs, and their activity ratios are shown graphically in Figs. 4 and 5, respectively. As shown in Fig. 4, accumulation of ^{239, 240}Pu in the Hiroshima soils ranged from 0.53 to 1.53 mCi/km² which were within the global fallout values (1.0–2.9 mCi/km²) in Japan⁴, and no appreciable difference was found even for the soils from the "Black-rain" area. On the other hand, accumulation of ¹³⁷Cs exhibited a wide range from 22.9 to 112 mCi/km² independent of the distance and the direction from the epicenter of A-bomb.

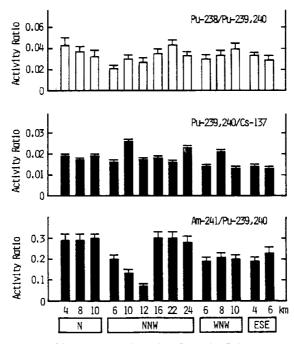
The activity ratios of ^{239, 240}Pu/¹³⁷Cs (0.013–0.023) and ²³⁸Pu/^{239, 240}Pu (0.021–0.043) of the Hiroshima soils are close to the corresponding global fallout values. Most of the soils showed the global fallout value of ²⁴¹Am/^{239, 240}Pu around 0.3, while two soils collected at 10 and 12 km in the NNW direction from the epicenter of the A-bomb showed rather low values of 0.13 and 0.07, respectively. In order to make clear the reason of this low value, the ²⁴⁰Pu/²³⁹Pu isotopic ratio was measured for the soils collected from these two points in 1983 and found to be 0.17–0.18 (Table 3). These values are just the same as the global fallout value of 0.18, indicating that the contribution of A-bomb is negligibly small.

The anomaly of ²⁴¹Am/^{239, 240}Pu activity ratio may be explained by the different mobilities of these nuclides in the soil. In the desert environment, mobility of ²⁴¹Am is known to be higher than that of ^{239, 240}Pu as reported by Essington et al. ¹⁶⁾ Most of the Hiroshima soils are sandy and coarse consisting of the weathered component of granite ⁶⁾. The soils collected at 16, 22 and 24 km in the NNW direction from the epicenter are rich in volcanic ash. Low



Distance and Direction from the Epicenter

Fig. 4. Accumulations of ^{239, 240}Pu and ¹³⁷Cs in soils (0-10 cm depth) collected from the Hiroshima atomic bombed area in 1976.



Distance and Direction from the Epicenter

Fig. 5. Activity ratios of ²³⁸Pu/^{239, 240}Pu, ^{239, 240}Pu/¹³⁷Cs and ²⁴¹Am/^{239, 240}Pu in the Hiroshima soils. (The propagated counting error of one sigma is given by the symbol "T" as the highest value on the top of each bar.)

 241 Am/ $^{239, 240}$ Pu activity ratios were found for the sandy soils consisting of coarse particles. Thus, the low value of 241 Am/ $^{239, 240}$ Pu activity ratio may be explained by the downward movement of 241 Am due to its higher mobility than plutonium.

Our approach to detect the contribution of Hiroshima A-bomb by the measurements of Pu isotopes and ²⁴¹Am was unsuccessful as described above. By the present study, the contribution of the transuranium elements derived from Hiroshima A-bomb was found to be negligibly small as compared with those derived from global fallout.

AKNOWLEDGEMENTS

The authors wish to thank Mr. T. Sunayashiki, Mr. S. Takeoka and Mr. K. Kato, Research Institute for Nuclear Medicine and Biology, Hiroshima University, for supplying the large amount of soils for the measurement of ²⁴⁰Pu/²³⁹Pu isotopic ratio.

REFERENCES

- A Review of Thirty Years Studies of Hiroshima and Nagasaki Atomic bomb Survivors (1975) J. Radiat. Res., 16 Suppl.
- Report of the United Nations Scientific Committee on the Effects of Atomic Radiation: UNSCEAR 1977.
- 3. Science Council of Japan (1953) Collection of the Reports on the Investigation of the Atomic Bomb Causalties. Vol. 1. Japan Society for the Promotion of Science, Tokyo.
- 4. Hashizume, T. et al. (1977, 1979) Report of Residual Radioactivity Surveys in Hiroshima and Nagasaki, supported by the Ministry of Health and Welfare, Japan Public Health Foundation (in Japanese).
- 5. Yamamoto, M., Yamamori, S., Komura, K. and Sakanoue, M. (1980) Behavior of Plutonium and Americium in Soils. J. Radiat. Res., 21: 204-212.
- Takada, J., Hoshi, M., Sawada, S. and Saknoue, M. (1983) Uranium Isotopes in Hiroshima "Black-rain" Soil. J. Radiat. Res., 24: 229-236.
- Sakanoue, M. and Tsuji, T. (1971) Plutonium Content of Soil at Nagasaki. Nature, 234(5324): 92-93.
- 8. Okajima, S., Aikawa, Y., Shimazaki, T. and Kubo, T. (1980) Measurement of Plutonium in Nagasaki Atomic Bombed Area. in: The Twenty-third Annual Meeting of the Japan Radiation Research Society, 10-p-D7, pp. 102 (in Japanese).
- 9. Yamamoto, M., Komura, K. and Sakanoue, M. (1983) Discrimination of the Plutonium due to Atomic Explosion in 1945 from Global Fallout Plutonium in Nagasaki Soil. J. Radiat. Res., 24: 250-258.
- Komura, K., Yamamoto, M. and Sakanoue, M. (1984) Determination of ²⁴⁰Pu/²³⁹Pu Ratio in Environmental Samples based on the Measurement of LX-/Alpha-ray Activity Ratio. Health Phys., 46(6): 1213-1219.
- 11. Yamato, A. (1982) An Anion Exchange Method for the Determination of ²⁴¹Am and Plutonium in Environmental and Biological Samples. J. Radioanal. Chem., 75: 265-273.
- 12. Hardy, E. P., Krey, P. W. and Volchok, H. L. (1973) Global Inventory and Distribution of Fallout Plutonium. Nature, 241(5390): 444-445.
- 13. Hardy, E. P. (1975) Regional Uniformity of Cumulative Radionuclide Fallout. USAEC Report HASL-288, pp. I-2 I-9.
- 14. Yamamoto, M., Komura, K. and Sakanoue, M. (1983) ²⁴¹Am and Plutonium in Japanese Rice-field Surface Soils. J. Radiat. Res., 24: 237-249.

- 15. Harley, J. H. (1980) Plutonium in the Environment A Review. J. Radiat. Res., 21: 83-104.
- 16. Essington, E. H., Fowler, E. B., Gilbert, R. O. and Eberhardt, L. L. (1976) Plutonium, Americium and Uranium Concentrations in Nevada Test Site Soil Profiles. in: Transuranium Nuclides in the Environment (IAEA, Vienna), pp. 157-173.

Appendix Uranium- and Thorium-series nuclides and 40K in soils from Nagasaki and Hiroshima measured by Gamm-spectrometry.

Sampling	Location*		Concentration (pCi/g)						
area			U-238*1	Ra-226*2	Pb-210*3	Ra-228*4	K-40		
"Nagasaki"	E	2.7	0.62/.07#	0.69/.08	2.93/.12	0.56/.06	6.1/.2		
	E	2.8	0.44/.06	0.37/.05	8.76/.20	0.36/.06	5.0/.2		
	E	3.5	0.99/.05	1.02/.07	4.66/.09	0.94/.06	6.6/.2		
	E	3.8	0.57/.07	0.58/.06	3.84/.13	0.54/.05	5.0/.2		
	E	4.5	0.83/.06	0.60/.04	5.03/.13	0.71/.04	2.2/.1		
	NNE	4	0.62/.03	0.74/.05	6.42/.12	0.53/.02	4.2/.2		
	NNE	10	0.96/.04	0.94/.06	5.32/.13	1.19/.07	5.0/.2		
	NE	6	0.96/.04	0.97/.07	6.71/.15	1.05/.06	6.7/.2		
	NE	10	0.51/.02	0.60/.06	4.29/.15	0.50/.03	4.5/.2		
	E	8	1.32/.04	1.36/.09	2.69/.17	1.25/.08	16.2/.5		
	E	32	1.56/.05	0.95/.09	5.58/.13	1.50/.09	8.0/.3		
	SE	6	1.11/.07	0.64/.04	6.29/.13	1.33/.08	3.0/.1		
	NW	26	0.79/.03	1.71/.35	3.79/.13	0.60/.04	10.4/.4		
	Ave	age	0.87/.32##	0.86/.36	5.10/1.69	0.85/.38	6.4/3.6		
"Hiroshima"	N	4	1.18/.10	2.64/.25	2.04/.15	1.09/.03	31.0/.1		
	N	8	1.63/.13	1.48/.15	2.27/.18	1.47/.05	30.0/.1		
	N	10	2.01/.12	0.76/.10	2.86/.15	2.07/.06	31.5/.1		
	NNW	6	1.91/.13	1.30/.15	3.71/.19	2.59/.08	20.8/.1		
	NNW	10	0.98/.09	1.75/.20	2.33/.15	1.59/.05	26.9/.1		
	NNW	12	1.42/.13	1.60/.19	3.74/.16	1.70/.05	19.6/.1		
	NNW	16	1.12/.19	1.29/.26	4.15/.30	1.27/.03	10.1/.1		
	NNW	22	1.02/.13	1.81/.30	4.89/.20	1.14/.03	16.1/.1		
	NNW	24	1.16/.12	1.58/.21	2.56/.15	1.22/.03	20.7/.1		
	WNW	6	1.46/.16	1.47/.20	2.33/.19	2.08/.06	24.7/.1		
	WNW	8	1.52/.18	1.35/.21	2.10/.19	1.98/.06	26.9/.1		
	WNW	10	1.32/.11	1.49/.15	2.88/.16	1.35/.04	29.9/.1		
	ESE	4	1.28/.14	2.14/.28	1.97/.14	1.26/.03	26.1/.1		
	ESE	6	2.10/.14	0.74/.07	3.52/.18	2.26/.07	27.1/.1		
	 Aver	age	1.44/.36	1.53/.49	2.95/.90	1.65/.47	24.4/6.2		

^{*)} Direction and distance from epicenter. #) or ##) The number after slash denotes the propagated counting error of one sigma or standard deviation of one sigma: $0.62/.07 = 0.62 \pm 0.07$.

^{*1)} Measured by ²³⁴Th.
*2) Measured by both ²¹⁴Pb and ²¹⁴Bi.

^{*3)} Corected to the dates of sampling.

^{*4)} Measured by ²²⁸Ac.