

What is the origin of ^{137}Cs detected in under-floor soil samples of houses built in 1-3 years after the Hiroshima atomic bomb ?

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1. Introduction

Other than direct radiation by Hiroshima atomic bomb (A-bomb), from twenty to third minutes after the explosion, so-called "black rain" fell down over the north and northwest areas of Hiroshima City. Radiation exposure due to close-in fallout by this event was not taken into consideration in DS02 (Young and Kerr, 2005) because its contribution was considered to be small for RERF cohort members who were mainly inside the city at the time of the bombing. Recently, in relation with enlargement of social compensation for A-bomb survivors, concern on radiation exposure due to close-in fallout has been raised among the people who experienced the black-rain.

Until now, to evaluate the possible radiation exposure by the close-in fallout related to the black rain, long-lived fission product ^{137}Cs has been actively measured in surface soil samples, accompanied by much smaller amounts of data on ^{90}Sr (JPHA, 1976, 1978). Excess ^{137}Cs activity from the close-in fallout, however, could not be clearly recognized due to much larger quantity of global fallout ^{137}Cs deposition originating from atmospheric nuclear tests in 1950s and 1960s. Thus, radioactive characteristics as well as spatial distribution of the close-in fallout by the Hiroshima A-bomb have not been specified even after 60 years from the A-bombing.

Therefore, measurements of ^{137}Cs and Pu isotopes in under-floor soil samples from about 20 houses built in 1-3 years after 1945 have been attempted since 2008, in order to evaluate the close-in fallout deposition at the time of Hiroshima atomic explosion (Yamamoto et al. 2010). The $^{239,240}\text{Pu}$ was used as indicator to evaluate the contamination from global fallout ^{137}Cs other than Hiroshima A-bomb derived ^{137}Cs . If a fission product ^{137}Cs is detected in soil samples under houses, this finding will give us convincing evidence that the close-in fallout fell down, indicating the possibility that details of level and spatial distributions of the close-in fallout become apparent. As a result, ^{137}Cs (several to several 100 Bq/m², mostly being 10-50 Bq/m²) and trace of $^{239,240}\text{Pu}$ (0.1-24 Bq/m², mostly being around 1Bq/m²) were detected for all samples measured, although their ^{137}Cs levels were very low compared with their levels (1000-2500 Bq/m²) in undisturbed forest soils around Hiroshima. To elucidate the origin of ^{137}Cs (and

$^{239,240}\text{Pu}$) detected in the under-floor soil samples, attempts were also made to determine the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in some of those soil samples. These results will be presented and discussed from the standpoint of the evaluation of Hiroshima atomic bomb derived ^{137}Cs fallout level.

2. Materials and methods

2.1. Soil sampling

Sampling locations of 20 houses where under-floor soil samples were collected are shown in Fig.1. The samples were mainly collected from the under-floor of old houses built in 1-4 years after A-bomb at the north and northwest areas. Most of the locations

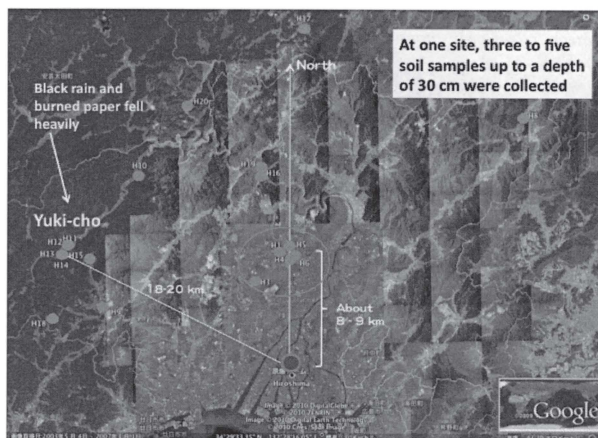


Fig. 1 Sampling locations of under-floor soil samples of houses built in 1-3 years after the Hiroshima atomic bomb explosion.

where houses were built at that time were in the paddy and dry fields. At one house, three to five under-floor soil samples up to a depth of 30 cm were taken by using stainless steel pipe (5.0 cm ϕ x 30 cm). Around the Yuki-cho far away from about 20 km northwest from the hypocenter, it is well known that the black rain and burned paper heavily dropped at that time.

2.2. Measurement

The obtained samples were air-dried, sieved through a 2-mm mesh screen to remove pebbles, and pulverized using an agate mortar to obtain samples as homogeneous as possible. A soil sample (60-80g) was at first subjected to non-destructive γ -ray spectrometry using a Ge detector to determine ^{137}Cs concentration. However, ^{137}Cs was not clearly detected for most of the samples, except for several samples. To measure accurately low level ^{137}Cs in all soil samples, chemical separation of Cs (^{137}Cs) using about 100 g soil sample was carried out by Kyushu Environmental Evaluation Agency (Fukuoka, Japan), and the obtained AMP samples were measured using extremely low-background Ge detector installed at the

Ogoya underground laboratory at LLRL. Ogoya underground laboratory is about 20 km southeast of LLRL. The laboratory is in a 550 m tunnel of the former Ogoya copper mine where closed in 1971. It is located 270 m from the tunnel entrance, where the overburden is 270 mwe. The muon intensity is 1/100 of the above ground value. Measurement time is more than one week or further long. Low-level ^{137}Cs was clearly detected for all soil samples examined.

After γ -ray spectrometric analysis, plutonium analysis was carried out radiochemically (Yamamoto et al. 1983, 2008). In brief, an aliquot of 70-100 g of soil sample was leached twice with conc. HNO_3 containing a small amount of H_2O_2 on a hot plate, with the addition of a known amount of ^{242}Pu as a yield tracer. The Pu in the leached fraction was then separated and purified carefully by passing through an anion exchange resin column (Dowex 1-X8, 100-200 mesh). The purified Pu was electroplated onto a polished stainless steel disc and its α -ray activities (^{238}Pu , $^{239,240}\text{Pu}$ and ^{242}Pu) were measured by a surface barrier Si detector coupled with 1 k channel pulse height analyzer. The measurements were continued for more than 3-4 weeks to minimize the statistical error by counting.

Other than α -spectrometry, Pu fraction separated and purified from some of soil samples with the non-addition of ^{242}Pu as a yield tracer was subjected to determination of $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio by means of thermal ionization mass spectrometry (TIMS), which is installed at JAEA.

3. Results and discussion

The results of ^{137}Cs measurements for under-floor soil samples are shown in Fig. 2. As can be seen from this figure, ^{137}Cs was more or less detected for all of samples measured. Sample numbers H1-H6, H16-H17 and H19 are from areas in a northern direction, numbers H7-H14 and H20 from a northwestern direction, mainly from Yuki-cho, and number H18 from a northeastern direction. Among them, higher values ranging from 150-800 Bq/m^2 were observed at H2, H3, H6, H8 and H14 locations. The ^{137}Cs levels found are some changeable even in under-floor samples from the same house. Other locations showed relatively low values of less than 50 Bq/m^2

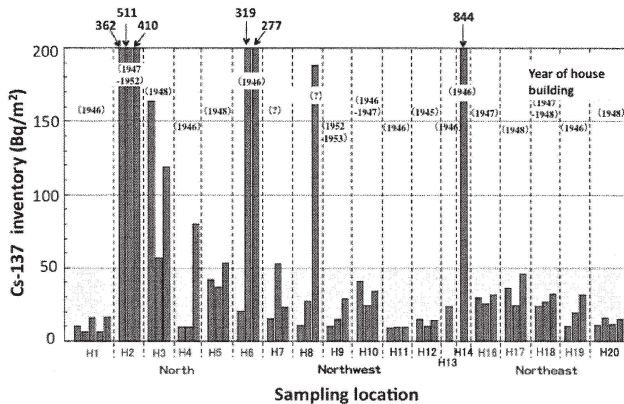


Fig. 2 Results of ^{137}Cs inventories in under-floor soil samples of houses built mainly in 1-3 years after 1945.

In Fig. 3, the results for samples which $^{239,240}\text{Pu}$ was measured are shown, together with ^{137}Cs levels. Plutonium-239,240 was detected with low levels for all samples examined. The $^{239,240}\text{Pu}$ levels were higher in samples where ^{137}Cs was observed in higher level. Except for the samples detected with higher $^{239,240}\text{Pu}$ levels, other most of the samples showed the values of less than 1 Bq/m².

In addition to those $^{239,240}\text{Pu}$ concentrations, this time, measurement of $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio was examined for four soil samples by using TIMS. Detected atomic ratios ranged from 0.13 to 0.19, as shown in Fig. 3. For global fallout Pu, usually, value of 0.18 has been observed in soil samples. Although detected values for three samples seems to be a little lower than 0.18, these findings seem to indicate that these soil samples received some influence from global fallout. Current ^{137}Cs and $^{239,240}\text{Pu}$ levels in forest and plat areas in Hiroshima are in the range from 1,000 to 2,500 Bq/m² and from 40 to 80 Bq/m², respectively, and the activity ratios of $^{239,240}\text{Pu}/^{137}\text{Cs}$ is around 0.03-0.04. These levels should be compared with ^{137}Cs and $^{239,240}\text{Pu}$ levels found in the under-floor soil samples.

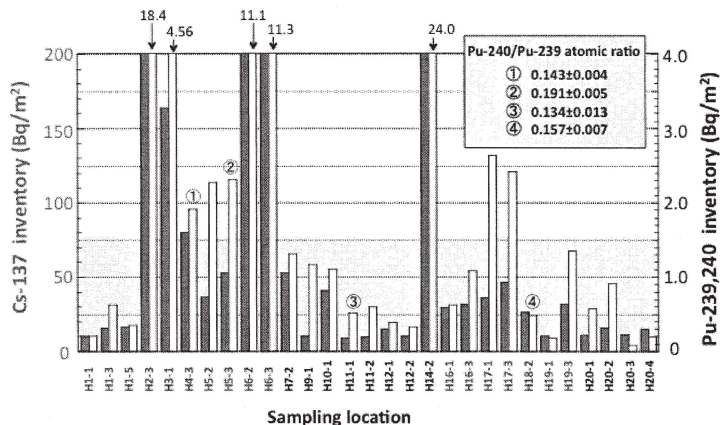


Fig. 3 Results of $^{239,240}\text{Pu}$ inventories and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in some of under-floor soil samples of houses built in 1-3 years after 1945

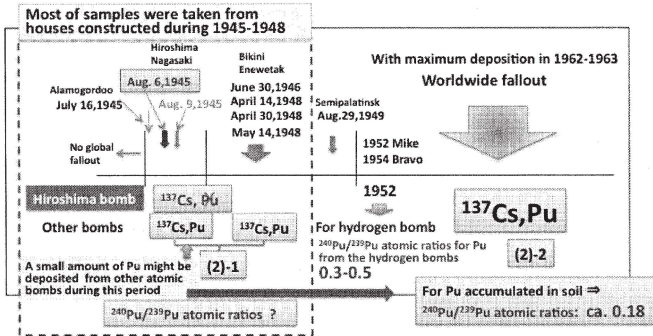
Then, let me refresh your memory on Hiroshima and other worldwide atomic bombs.

As shown in Fig.4, under-floor soil samples were mostly collected from houses built during the period of 1945-1949. During this period, other than Hiroshima atomic bomb, some nuclear atomic bombs were exploded in the atmosphere. Pu originating from Hiroshima A-bomb is assumed to be negligibly small, but it seems likely that global fallout Pu from atomic bombs conducted at Almgordoo, Nagasaki and Bikini and Enewetak during this period deposited more or less. Furthermore, soil samples may be affected by global fallout in 1962-1963 with maximum fallout level.

In all of the samples measured, $^{239,240}\text{Pu}$ was detected more or less.

What does the finding of Pu detection mean, with the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio of 0.13-0.19 measured in some soil samples?

Let me refresh your memory on fallout from Hiroshima and/or worldwide atomic bombs



So, by considering the above situation, It seems likely that that ^{137}Cs detected in the under-floor is more or less contaminated with the global fallout ^{137}Cs

Fig. 4 Relationship between Hiroshima atomic bomb and other atomic bombs.

In Fig. 5, the result of $^{239,240}\text{Pu}$ and ^{137}Cs depositions found in ice cores at Canada by Kudo et al. (1998) is shown. The $^{239,240}\text{Pu}$ originating from Nagasaki A-bomb is detected with extremely low level, indicating that Pu had been scattered globally from 1945.

As for global fallout from atomic bombs before 1950

Kudo et al. measured $^{239,240}\text{Pu}$ and ^{137}Cs in 10 ice cores on the Agassiz ice cap, Ellesmere Island, Canada.

A. Kudo et al., *J. Environ. Radioactivity*, 40,289-298(1998)

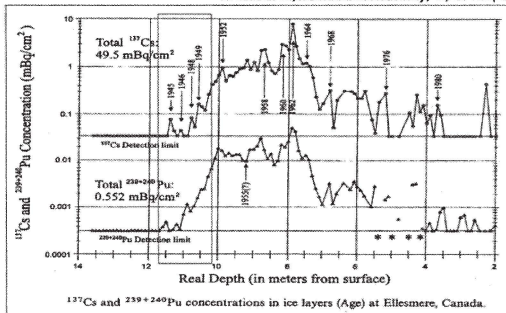


Fig. 5 ^{137}Cs and $^{239,240}\text{Pu}$ levels in ice core at Ellesmere, Canada by Kudo et al. (1998).

An occurrence of global fallout Pu deposition from atomic bombs other than Nagasaki atomic bomb after 1945 is also recognized.

By taking into account the above-mentioned findings, Hiroshima A-bomb derived ^{137}Cs at that time may be roughly estimated by using ^{137}Cs level found in the under-floor soil. Here, we considered two approaches for discrimination: one is based on assumption that all of the $^{239,240}\text{Pu}$ detected was derived from global fallout. In this case, Hiroshima derived ^{137}Cs can be calculated simply by subtracting the global fallout ^{137}Cs estimated by $^{239,240}\text{Pu}$ from total amount of measured ^{137}Cs , as shown in Fig.6. The second approach is the assumption that $^{239,240}\text{Pu}$ detected was only due to fallout from atomic bombs conducted during the periods of 1945-1949. The cumulative deposition of ^{137}Cs in Tokyo is reported by Aoyama (2006). Its deposition is about 10 Bq/m^2 up to the year of 1949. By assuming that this value is the case for Hiroshima area, today's level is around 2.3 Bq/m^2 by considering decay, and further by assuming that current $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratio of 0.033 is the case for both fallout nuclides during this period, Pu deposition can be roughly estimated to be around 0.1 Bq/m^2 . This value is consistent with the estimated value in 1945-1949 by Hirose et al. (2001). Then, in case that $^{239,240}\text{Pu}$ found is less than at least 0.5 Bq/m^2 , it seems to be reasonable to assume that its Pu was already contaminated by fallout Pu during 1945-1949, although it is speculative. In this case, ^{137}Cs contamination from global fallout during this period seems to be neglected.

Discrimination of ^{137}Cs between Hiroshima A-bomb and global fallout

- 1) By assuming that All of the Pu detected was derived from global fallout,
Hiroshima A-bomb derived ^{137}Cs =
 $^{137}\text{Cs} (\text{Bq/m}^2) - ^{239,240}\text{Pu} (\text{Bq/m}^2) / 0.033$ (current global $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratio)
- 2) By assuming that Pu detected was only derived from fallout during 1945-1948

^{137}Cs fallout levels in Tokyo (Tsukuba) up to the year of 1949

Year	(A)		(B)		M. Aoyama, Doctor thesis (1999)
	Cs-137 deposition Bq/m^2	Cummulative deposition Bq/m^2	Cs-137 deposition Bq/m^2	Cummulative deposition Bq/m^2	
1945	5.78	5.78	2.60	2.60	
1946	2.20	7.85	1.92	4.46	
1947	0.00	7.87	0.00	4.36	
1948	6.53	14.0	4.75	9.02	
1949	1.01	14.7	1.91	10.7	

(A): From Canadian Arctic ice cores, (B): From each nuclear test's energy

Accumulated ^{137}Cs : about $10 \text{ Bq/m}^2 \Rightarrow$ decay to 2010: 2.3 Bq/m^2

$^{239,240}\text{Pu}$ at that time : 2.3×0.033 (?) = ca. 0.1 Bq/m^2

Pre-1959: $^{239,240}\text{Pu}/^{137}\text{Cs} = 0.018$ (Koide et al.) \rightarrow $^{239,240}\text{Pu}$ at that time: $10 \times 0.018 = 0.2 \text{ Bq/m}^2$

Pre-1959: $^{239,240}\text{Pu}/^{137}\text{Cs} = 0.012$

In case that $^{239,240}\text{Pu}$ found is less than at least 0.5 Bq/m^2 (it is speculative), it seems to be reasonable to think that its Pu in under-floor soil was already contaminated by fallout Pu during 1945-1948. \Rightarrow ^{137}Cs contamination from global fallout is neglected.

Fig. 6 Discrimination of ^{137}Cs between Hiroshima A-bomb and global fallout.

Table 1 Estimation of the Hiroshima atomic bomb derived ¹³⁷Cs deposition.

¹³⁷Cs deposition expected from the Hiroshima A-bomb						(1)	(2)	
Sample No.	Sampling date		240/239 atomic ratio	Global origin CsCs from Hiroshima		Elapsed time (year)	Cs-137 at 1945	Cs-137 at 1945
	Pu-238/240 (Bq/m ²)	Cs-137 (Bq/m ²)		Pu/Cs=0.033 (Bq/m ²)	A-bomb (Bq/m ²)		from A-bomb (Bq/m ²)	without correction (Bq/m ²)
H1-1	0.20 ± 0.04	10.54 ± 1.52		6.18	4.36	62.92	18.5	45.1
H1-3	0.83 ± 0.14	15.78 ± 1.83		19.23	(3.45)	62.92		67.5
H1-5	0.35 ± 0.17	16.57 ± 1.71		10.54	6.03	62.92	25.8	70.9
H2-3	18.40 ± 1.06	410.3 ± 8.0		557.46	(147.18)	63.55		
H3-1	4.56 ± 0.27	163.7 ± 7.8		138.28	25.39	63.70	110.6	
H4-3	1.92 ± 0.35	79.91 ± 3.10	0.143 ± 0.004	58.18	21.72	63.70	94.6	
H5-2	2.28 ± 0.38	37.04 ± 4.33		69.09	(32.05)	63.70		
H5-3	2.32 ± 0.41	53.33 ± 2.88	0.191 ± 0.005	70.30	(16.97)	63.70		
H6-2	11.12 ± 0.80	318.9 ± 5.1		336.82	(17.97)	63.70		
H6-3	11.26 ± 0.49	276.6 ± 9.8		341.28	(64.66)	63.70		
H7-2	1.31 ± 0.21	53.02 ± 3.02		39.71	13.31	83.70	58.0	230.9
H9-1	1.17 ± 0.14	10.49 ± 1.15		35.57	(25.06)	64.27		46.3
H10-1	1.11 ± 0.17	40.89 ± 1.98		33.62	7.27	64.27	32.1	180.5
H11-1	0.52 ± 0.13	9.38 ± 1.01	0.134 ± 0.013	15.80	(6.42)	64.27		41.4
H11-2	0.60 ± 0.13	9.61 ± 1.28		18.20	(8.59)	64.27		42.4
H12-1	0.39 ± 0.17	14.95 ± 1.56		11.79	3.16	64.50	14.0	66.3
H12-2	0.33 ± 0.08	10.37 ± 1.55		9.91	0.46	64.50	2.0	46.0
H14-2	24.02 ± 0.52	843.8 ± 6.5		727.88	115.95	64.50		
H16-1	0.63 ± 0.11	29.27 ± 1.45		19.08	10.19	64.82	45.5	130.8
H16-3	1.09 ± 0.20	31.76 ± 2.56		32.99	(1.23)	64.82		142.0
H17-1	2.64 ± 0.32	36.27 ± 1.75		80.00	(42.73)	64.82		
H17-3	2.42 ± 0.36	46.15 ± 3.55		73.33	(27.18)	64.82		
H18-2	0.48 ± 0.06	26.62 ± 3.28	0.157 ± 0.007	14.53	12.09	64.82	54.0	119.0
H19-1	0.18 ± 0.06	10.44 ± 1.1		5.46	4.98	64.86	22.3	46.7
H19-3	1.35 ± 0.28	31.91 ± 1.7		40.88	(9.97)	64.86		142.7
H20-1	0.58 ± 0.13	10.94 ± 1.7		17.61	(6.66)	64.94		49.0
H20-2	0.92 ± 0.17	15.70 ± 1.1		27.99	(12.28)	64.94		70.4
H20-3	0.08 ± 0.03	11.38 ± 1.1		2.36	9.02	64.94	40.4	51.0
H20-4	0.19 ± 0.08	14.96 ± 1.6		5.77	9.19	64.94	41.2	67.1
						Max	ca. 50	100

The result of calculation is listed in Table 1. The global fallout derived ¹³⁷Cs levels estimated by using both detected ^{239,240}Pu and current ^{239,240}Pu/¹³⁷Cs activity ratio of 0.033 are given in the fifth column of this Table. The value given in the sixth column is the estimated Hiroshima derived ¹³⁷Cs level. In this case, many samples give negative value. Although further consideration is required, maximum value of 50 Bq/m² is tentatively more likely. The values listed in the last column in Table 1 show the result of second assumption for samples, which Pu levels are less than 0.5 Bq/m², probably these values seem to correspond to the upper limit. In this case, maximal deposition of 100 Bq/m² is more likely.

Thus, as a whole, we estimated roughly the Hiroshima atomic bomb derived ¹³⁷Cs deposition by using the under-floor soil samples of houses built in 1-3 years after the Hiroshima atomic bomb. However, most important issue depends on the soil preparation under houses at that time. Usually, before building a new house, soil preparation such as dipping out surface soil, clearing the land and so on are carried out. According to carpenter, most of the wooden houses at that time were built without causing large disarrangement of surface soil. Further information and data are needed.

4. Summary

Since 2008, we have measured ¹³⁷Cs and ^{239,240}Pu isotopes in about 60 soil samples from the under-floor of 20 houses built within 1-3 years after 1945, in order to evaluate the close-in fallout deposition due to the “Black rain” at the time of Hiroshima atomic explosion. ^{239,240}Pu was used as indicator to evaluate the

contamination from global fallout ^{137}Cs other than Hiroshima A-bomb derived ^{137}Cs .

As a result, it seems likely that ^{137}Cs deposition at that time due to the Hiroshima A-bomb is 50-100 Bq/m^2 .

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