Estimation of close-in fallout ^{137}Cs deposition level due to the Hiroshima atomic bomb from soil samples under houses built 1–4 years after the explosion

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Abstract

Cesium-137 and Pu isotopes in under-floor soil samples from about 20 houses built 1–4 years after 1945 were measured in an attempt to evaluate the close-in fallout deposition at the time of the Hiroshima atomic explosion. ^{239,240}Pu was used to distinguish global fallout ^{137}Cs from Hiroshima A-bomb derived ^{137}Cs. In all samples measured, low levels of ^{137}Cs (range: 6.4–843 Bq/m^2, but mostly in the range 10–50 Bq/m^2) and traces of ^{239,240}Pu (0.1–24 Bq/m^2, but mostly less than 1 Bq/m^2) were detected. The results by considering the contribution form the global fallout suggest that ^{137}Cs deposition due to the Hiroshima A-bomb was in the range of 50–100 Bq/m^2 in the areas studied.

1. Introduction

In addition to the direct radiation caused by the detonation of the Hiroshima atomic bomb (A-bomb), a "black rain" of radioactive dust and debris fell over the north and northwest areas of Hiroshima City for a period of twenty to thirty minutes after the explosion. Radiation exposure due to the close-in fallout caused by this event was not estimated in DS02, the new dosimetry system developed by Young and Kerr (2005), because the contribution of this fallout to the radiation doses received by cohort members of the Radiation Effects Research Foundation who were inside the city at the time of the bombing was considered to be small. Because compensation for A-bomb survivors has increased in recent times, concern over radiation exposure due to the close-in fallout has been raised by the people who were exposed to the black rain.

Until now, potential radiation exposure from close-in fallout related to the black rain has been evaluated by measuring the amount of ^{137}Cs in surface soil samples, accompanied by much smaller amounts of data on ^{90}Sr (JPHA, 1976, 1978). A problem with this approach, however, is that additional ^{137}Cs activity from the close-in A-bomb fallout is difficult to detect over and above the much larger quantity of global deposition of ^{137}Cs originating from atmospheric nuclear tests in the 1950s and 1960s. Thus, the characteristics of the radioactivity and the spatial

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distribution of the close-in fallout from the Hiroshima A-bomb have not been determined even 60 years after the event.

Yamamoto et al. (2010) measured ^{137}Cs and Pu isotopes in under-floor soil samples from about 20 houses built 1–4 years after 1945 in an attempt to evaluate the close-in fallout deposition at the time of the Hiroshima atomic explosion. ^{239,240}Pu was used to distinguish global fallout ^{137}Cs from Hiroshima A-bomb derived ^{137}Cs. The presence of ^{137}Cs in soil under these houses, which was protected from global deposition, would provide convincing evidence of contamination by close-in fallout, providing the ability to reveal details of the levels and spatial distribution of the close-in fallout. Low levels of ^{137}Cs (range: 6.4–843 Bq/m^2, but mostly in the range 10–50 Bq/m^2) and traces of ^{239,240}Pu (0.1–24 Bq/m^2, but mostly around 1 Bq/m^2) were detected in all samples measured. The ^{137}Cs levels were very low compared with those (1000–2500 Bq/m^2) found in undisturbed forest soils around Hiroshima. To elucidate the origin of the ^{137}Cs and ^{239,240}Pu detected in the under-floor soil samples, attempts were also made to determine the ^{240}Pu/^{239}Pu atomic ratios in a subset of the soil samples. Those results are presented here and all data measured so far were discussed with respect to the evaluation of close-in fallout ^{137}Cs levels by the Hiroshima atomic bomb.

2. Materials and methods

The locations of the 20 houses at which under-floor soil samples were collected are shown in Figure 1. The samples were mainly collected from under the floors of houses built 1–4 years after the detonation of the Hiroshima A-bomb in areas of up to 25 km north and northwest of the hypocenter. Around the area of Yuki-cho, some 20 km northwest of the hypocenter, heavy falls of black rain and charred paper are known to have occurred after the explosion. Most of the locations were paddy or dry land fields at the time the house was constructed. At each house, three to five under-floor soil samples were taken as a complete core by driving a stainless steel pipe (diameter = 5.0 cm × length = 30 cm) into the ground to a depth of about 30 cm.

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 that were as homogeneous as possible. A soil sample (60–80 g) was at first subjected to non-destructive γ-ray spectrometry using a Ge detector to determine the ^{137}Cs concentration. However, ^{137}Cs was only clearly detected from a few samples. To accurately measure these low levels of ^{137}Cs in all soil samples, chemical separation of ^{137}Cs using a soil sample of about 100 g was carried out by Kyushu Environmental Evaluation Agency (Fukuoka, Japan). The obtained samples by coprecipitation with ammonium phosphomolybdate (AMP/Cs) were measured using an extremely low-background Ge detector installed at the Ogoya underground laboratory owned by and located about 20 km southeast of the Low Level Radioactivity Laboratory of Kanazawa University, Ishikawa Prefecture, Japan. The laboratory is in a 550-m tunnel of the former Ogoya copper mine, which was closed in 1971. The Ge detector is located 270 m from the tunnel entrance, where the overburden is 270 m water equivalent. The muon intensity is 1/100th that of the above-ground value. The measurement time was 1 week or more. Low-level ^{137}Cs was clearly detected in all soil samples examined.

After γ-ray spectrometric analysis, radiochemical plutonium analysis was carried out (Yamamoto et al., 1983, 2008). In brief, an aliquot of 70–100 g of soil sample was leached twice with concentrated 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 1X8 [100–200 mesh], The Dow Chemical Company, Midland, MI). The purified Pu was electroplated onto a polished stainless steel disc and its α-ray activities (^{238}Pu, ^{239,240}Pu, and ^{242}Pu) were measured by a surface barrier Si detector coupled with a 1000-channel pulse height analyzer. The measurements were continued for more than 3 weeks to minimize the statistical error of counting.

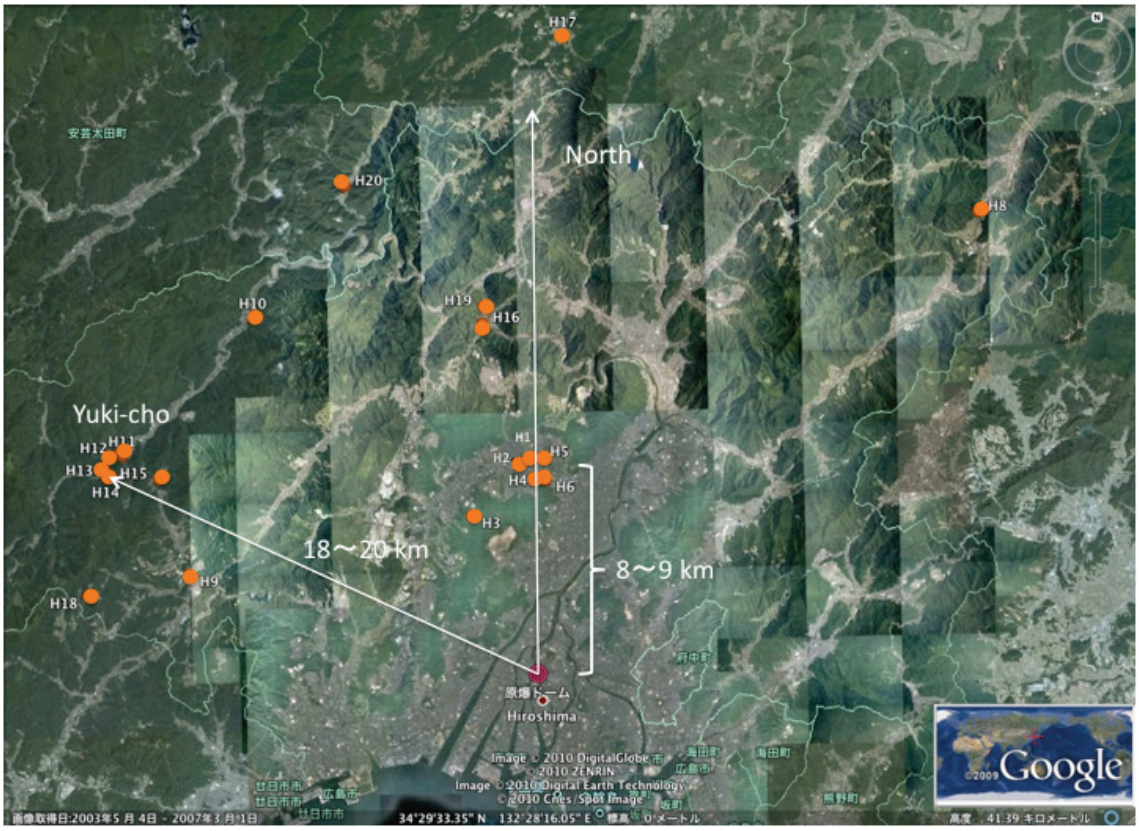

Figure 1 Locations of under-floor soil samples obtained from houses built 1–4 years after the Hiroshima atomic bomb explosion (for details of sampling points, see Appendix of this book).

In addition to α-spectrometry, the Pu fraction was separated and purified from four of the soil samples (about 50g) with the non-addition of ^{242}Pu as a yield tracer and then subjected to determination of the ^{240}Pu/^{239}Pu atomic ratio by means of thermal ionization mass spectrometry (TIMS) using an instrument installed at the Japan Atomic Energy Agency in Ibaraki, Japan.

3. Results and discussion

Cesium-137 was detected in all samples measured (Fig. 2). Samples H1–H6, H16, H17, and H19 were from the areas directly north, numbers H7–H14 and H20 were located northwest, mainly from Yuki-cho, and number H18 was located northeast of the hypocenter. The higher values, ranging from 119–843 Bq/m^2, were observed at H2, H3, H6, H8, and H14. The ^{137}Cs levels at each of these locations were variable, even between samples from the same house. Other locations showed relatively low values of less than 50 Bq/m^2.

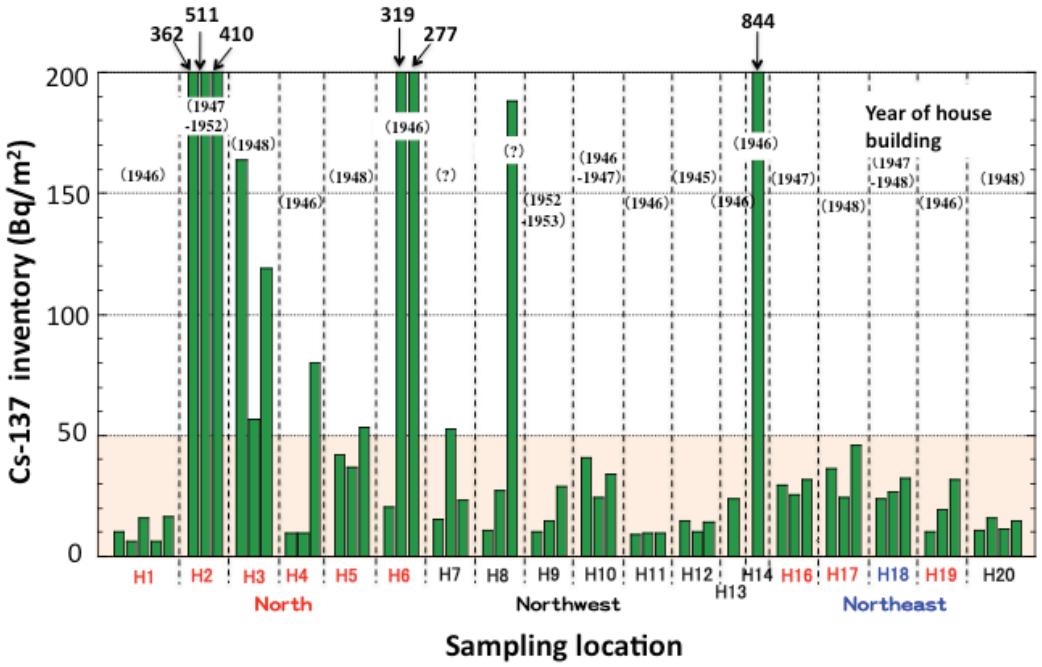

Figure 2 The results of ^{137}Cs inventories in under-floor soil samples of houses built mainly 1–4 years after 1945.

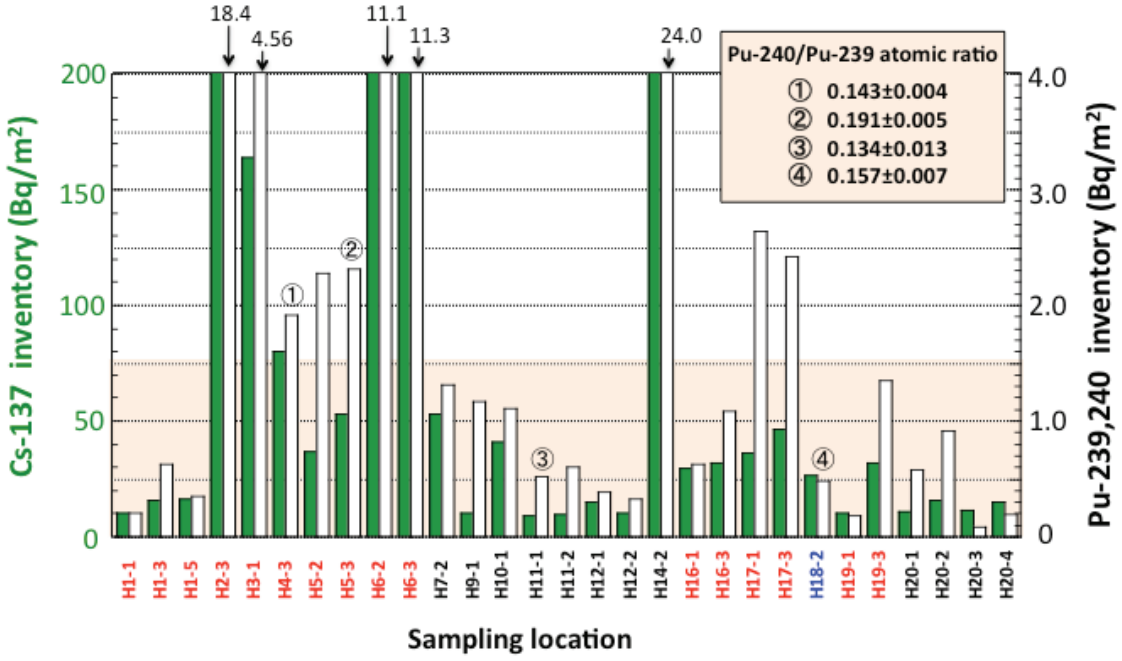

Figure 3 ^{239,240}Pu inventories, and ^{240}Pu/^{239}Pu atomic ratios in some under-floor soil samples of houses built 1–4 years after 1945.

Plutonium-239,240 was detected at low levels in all samples examined (Fig. 3). The ^{239,240}Pu levels were higher in samples containing higher levels of ^{137}Cs. Apart from seven samples with ^{239,240}Pu levels ranging from 2 to 24 Bq/m^2, most of the samples showed values of less than 1 Bq/m^2.

In the four soil samples that were analyzed using thermal ionization mass spectrometry, the ^{240}Pu/^{239}Pu atomic ratios ranged from 0.13 to 0.19 (Fig. 3). Global fallout Pu most typically yields a value of 0.18 from soil samples. Although the detected values for three of the four samples were lower than 0.18, the findings suggest that these samples were more or less influenced by global fallout. Current ^{137}Cs and ^{239,240}Pu levels in forested areas around Hiroshima range from 1000 to 2500 Bq/m^2 and 40 to 80 Bq/m^2, respectively, giving activity ratios of ^{239,240}Pu/^{137}Cs of around 0.03–0.04. These levels should be compared with those found in the under-floor soil samples.

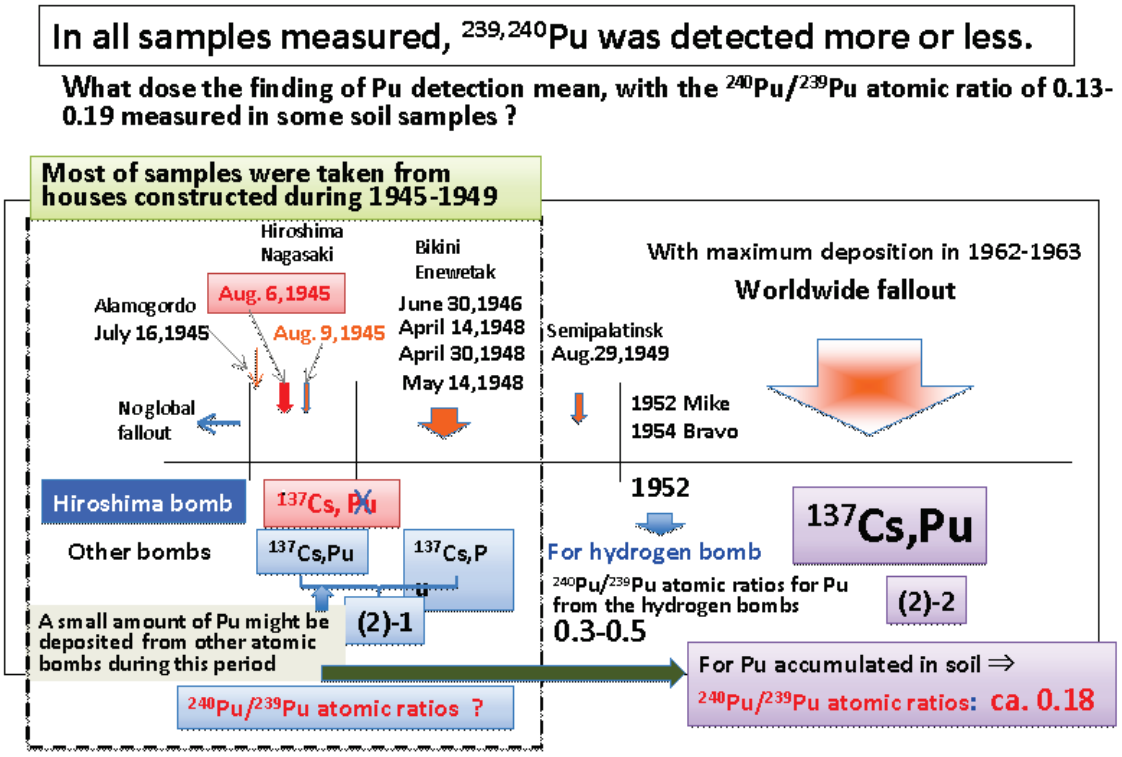

Figure 4 Relationship between Hiroshima atomic bomb and other atomic bombs.

As shown in Figure 4, the under-floor soil samples were mostly collected from houses built during the period 1945–1949 after the Hiroshima atomic bomb. During this period, five atomic bombs at Alamogordo, Nagasaki, Bikini Atoll, and Eniwetok were exploded in the atmosphere (Fig. 4). Pu originating from Hiroshima A-bomb is assumed to be negligibly small, but it seems likely that fallout Pu from atomic bombs conducted during the period 1945–1949 deposited more or less. Furthermore, the maximum global fallout of Pu occurred in 1962–1963 (Fig. 4), and this too could have potentially affected the soil samples.

Figure 5 shows the results of ^{239,240}Pu and ^{137}Cs depositions found in ice cores in Canada by Kudo et al. (1998). The ^{239,240}Pu originating from the Trinity as well as Nagasaki is detected at an extremely low level, indicating that Pu was distributed globally commencing in 1945. The

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

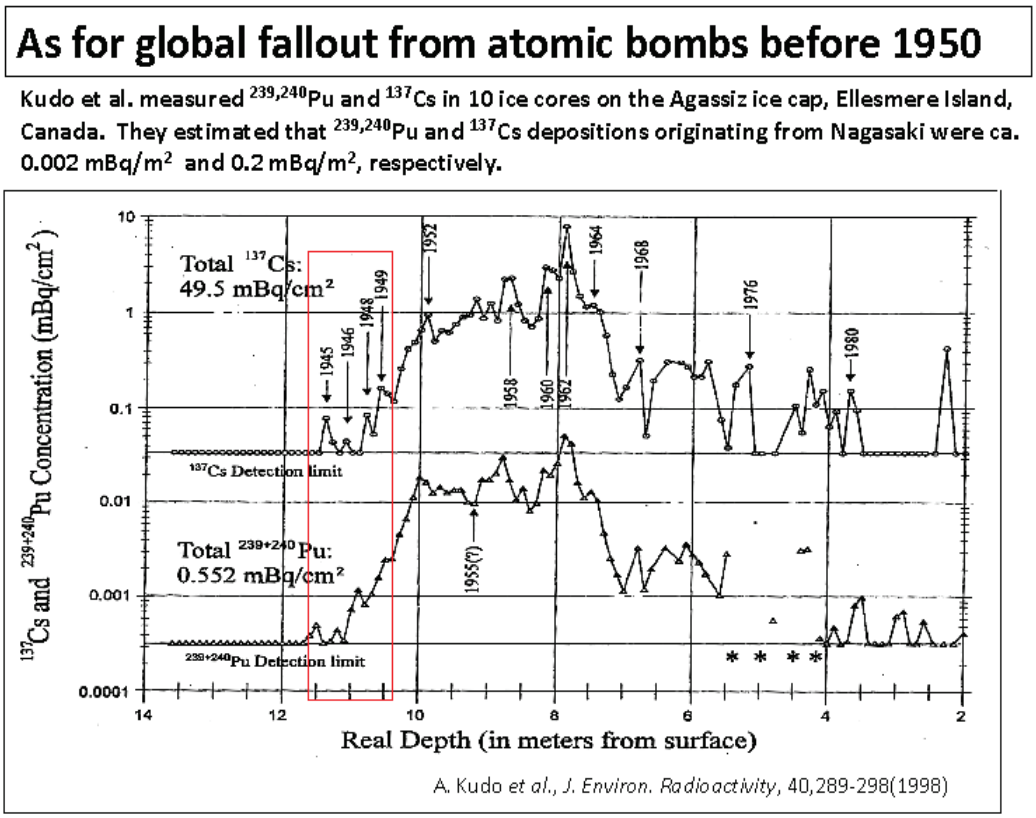

Figure 5 Levels of ^{137}Cs and ^{239,240}Pu in ice cores at Ellesmere, Canada reported by Kudo et al. (1998).

We considered two approaches for discrimination of the ^{137}Cs derived from the Hiroshima A-bomb using the ^{137}Cs levels found in the under-floor soil samples: One is based on the assumption that all of the ^{239,240}Pu detected was derived from global fallout. In this case, the Hiroshima-derived ^{137}Cs can be calculated simply by estimating the global-fallout ^{137}Cs from the relation with ^{239,240}Pu activity and subtracting this from the total amount of ^{137}Cs measured (Fig. 6). The second approach is based on the assumption that the ^{239,240}Pu detected was only due to fallout from atomic detonations during the period 1945–1949, the period in which the houses were built. The cumulative deposition of ^{137}Cs in Tokyo was reported by Aoyama et al. (2006). The cumulative deposition to the year 1949 was about 10 Bq/m^2. By assuming that this value was the same for the Hiroshima area, the present-day level would be 2.3 Bq/m^2 after accounting for decay. Further assuming that the current ^{239,240}Pu /^{137}Cs ratio of 0.033 has remained constant during the entire period, Pu deposition can be roughly estimated to be around 0.1 Bq/m^2. This value is consistent with that estimated in 1945–1949 by Hirose et al. (2001). If the level of ^{239,240}Pu is found to be less than at least 0.5 Bq/m^2, it is reasonable to assume that this Pu was derived only from fallout during 1945–1949. By this second method, ^{137}Cs contamination from global fallout during the 1945–1949 period is 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}Cs (Bq/m^2) –^{239,240}Pu(Bq/m^2)/ 0.033 (current global ^{239,240}Pu/^{137}Cs ratio)

2) By assuming that Pu detected was only derived from fallout during 1945-1949

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

Year	(A) Cs-137 deposition Bq/m^2	(A) Cummulative deposition Bq/m^2	(B) Cs-137 deposition Bq/m^2	(B) 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	7.67	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 Bq/m^2⇒ decay to 2010: 2.3Bq/m^2**
  ^{239,240}Pu at that time : 2.3x 0.033 (?)= ca. 0.1 Bq/m^2
- **Pre-1959: ^{239,240}Pu/^{137}Cs=0.018 (Koide et al.)→^{239,240}Pu at that time:10x0.018=0.2 Bq/m^2**
- **Pre-1959: ^{239,240}Pu/^{137}Cs=0.012**
- **In case that ^{239,240}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. The contamination ^{137}Cs from global fallout is neglected.**

Figure 6 Discrimination between ^{137}Cs derived from the Hiroshima A-bomb and that derived from global fallout.

Table 1 shows the full set of data from our study, including the determined values of ^{239,240}Pu, ^{137}Cs (at sampling date), and atomic ratio of ^{240}Pu/^{239}Pu from the under-floor samples (columns 2, 3, and 4); the global-fallout ^{137}Cs levels estimated from the detected ^{239,240}Pu and current ^{239,240}Pu/^{137}Cs activity ratio of 0.033 (column 5); the estimated Hiroshima-derived ^{137}Cs level (column 6); and the estimates of ^{137}Cs deposition at the time of 1945 derived from the Hiroshima atomic bomb using the two alternative assumptions outlined above (columns 8 and 9). In method-1, many of the samples returned a negative value for the estimates of Hiroshima-derived ^{137}Cs. We consider that these values are probably due to sampling error regarding site disturbance during construction (see below), and ^{137}Cs deposition values of about 50 Bq/m^2 are tentatively considered to be more likely. The values listed in the last column in Table 1 show the results of the second approach (method-2) in which Pu levels are less than 0.5 Bq/m^2. These values would appear to represent the upper limit and suggest that ^{137}Cs deposition of around 100 Bq/m^2 was most likely.

An important possible confounding influence in this study is the soil preparation that occurred on the house blocks prior to and during construction. Before building a new house, soil preparation such as clearing of vegetation or scraping off surface soil to level the block is common. This could have potentially resulted in strong mixing, removal, or change in the spatial variation of the radioisotope signatures. However, according to carpenters we interviewed, most of the wooden houses built around this time were built without causing major disturbance of the surface soil, so the indicative results provided by the range of values

measured can be considered valid. Further information and data are needed, especially in the Koi and Takasu areas where fallout from the Hiroshima A-bomb was deposited heavily.

Table 1 Estimation of ^{137}Cs deposition derived from the Hiroshima atomic bomb.

	^{137}Cs deposition expected from the Hiroshima A-bomb						(1) method	(2) method
Sample No.	Sampling date ^{239,240}Pu (Bq/m^2)	Sampling date ^{137}Cs (Bq/m^2)	^{240}Pu/^{238}Pu atomic ratio	Global origin ^{137}Cs Pu/Cs=0.033 (Bq/m^2)	^{137}Cs from Hiroshima A-bomb (Bq/m^2)	Elapsed time (year)	^{137}Cs at 1945 from A-bomb (Bq/m^2)	^{137}Cs at 1945 without corretion (Bq/m^2)
H1-1	0.20 ±0.04	10.54 ± 1.62		6.18	4.36	62.92	18.6	45.1
H1-3	0.63 ±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	63.70	58.0	230.9
H9-1	1.17 ±0.14	10.49 ± 1.15		35.57	(25.08)	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	(43.73)	64.82		
H17-3	2.42 ±0.36	46.15 ± 3.55		73.33	(27.18)	64.82		
H18-2	0.48 ±0.08	26.62 ± 3.28	0.157 ±0.007	14.53	12.09	64.82	54.0	119.0
H19-1	0.18 ±0.08	10.44 ± 1		5.46	4.98	64.86	22.3	46.7
H19-3	1.35 ±0.28	31.91 ± 1.7		40.88	(8.97)	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

4. Summary

We measured ^{137}Cs and ^{239,240}Pu isotopes in about 60 soil samples from under the floors of 20 houses built during the period 1–4 years after 1945 to evaluate the close-in fallout of ^{137}Cs deposition that accompanied the black rain that immediately followed the Hiroshima atomic explosion. The level of ^{239,240}Pu was used to evaluate the ^{137}Cs contamination derived from global fallout other than that derived from the Hiroshima A-bomb. In all samples measured, low levels of ^{137}Cs (range: 6.4–843 Bq/m^2, but mostly in the range 10–50 Bq/m^2) and traces of ^{239,240}Pu (0.1–24 Bq/m^2, but mostly less than 1 Bq/m^2) were detected. By considering the contribution from the global fallout, it is probable that ^{137}Cs deposition due to the Hiroshima A-bomb was in the range of 50–100 Bq/m^2 in the areas studied.

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amount of soil.

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