

Atomic Bomb Fallout and “Black rain” at Manose District (Hirama-cho) Located Northeast of Nagasaki City

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Abstract

Plutonium atomic bomb (A-bomb) was exploded at 503 meters height above Nagasaki City on August 9, 1945. Many studies on radioactive contamination, radiation dose, effect to people and so on have been conducted until now. Local fallout from the A-bomb mainly spread a lot in the east direction centering on Nishiyama area located about 4 km east of the hypocenter.

Like the Hiroshima A-bomb, black rain accompanied by radioactive materials fell shortly after the explosion, but relationship between local fallout fall zone and black rain fall area has not

been sufficiently elucidated, which has enhanced the concerns on radiation exposure to the A-bomb survivor. Concerning the black rain due to the Nagasaki A-bomb, it has been said that black rain did not fall across a wide area in a comprehensive way, except for the heavily contaminated Nishiyama area, but fell at various areas with conditions of light rain and/or soaking after the explosion. Among many villages or towns, especially in the Manose District where is a small intermountain village about 7.5 km northeast of the hypocenter, it had always been heard and known that the residents living there encountered black rain and experienced hair loss. In this district, the hair loss of inhabitants occurred actually with high probability, but its reason has not been elucidated well. Therefore, in order to re-evaluate the local fallout level in the Manose District by the A-bomb, residual long-lived radionuclides ^{137}Cs and Pu isotopes were measured for soil samples collected at the Manose District including the surrounding areas in July, 2011.

As a result, accumulated levels of ^{137}Cs and $^{239,240}\text{Pu}$ in the Manose District were not so high, each being nearly background (BG) level. The dispersion of local fallout Pu from the A-bomb was reconfirmed by $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios measured in soil samples at the same time, and was found to be primarily limited in the east direction, especially around the Nishiyama area. Those results were consistent with results obtained so far. Further researches are needed for understanding scientifically the cause of the high incidence rate of hair loss in this district.

1. Introduction

Regarding radiation exposure to residents by the A-bombs of Hiroshima and Nagasaki, the exposure dose received directly from neutrons and γ -rays emitted immediately after the explosion has been evaluated by the radiation dosimetry system called DS02 (Dose System 2002) (Stephen, 2012). Besides these direct radiation, "black rain" fell in Hiroshima and Nagasaki after the explosion, accompanied by the fission products generated by the atomic bomb and induced radionuclides as local fallout, and external and internal radiation exposures caused indirectly by them have been also feared. Some studies have been conducted on the black rain, but the information and knowledge obtained so far have been limited (Manhattan Study Team Final Report, 2002).

In 1976 and 1978, the Ministry of Health and Welfare conducted "Survey of residual radioactivity in Hiroshima and Nagasaki" in order to grasp the amount and distribution of the local fallout (Japan Society of Public Health, 1976, 1978). For this purpose, massive soil samplings were performed at the undisturbed areas in 16 directions of concentric circles at 2 km intervals, and residual radionuclide ^{137}Cs was mainly measured, accompanied by smaller amounts of data on ^{90}Sr .

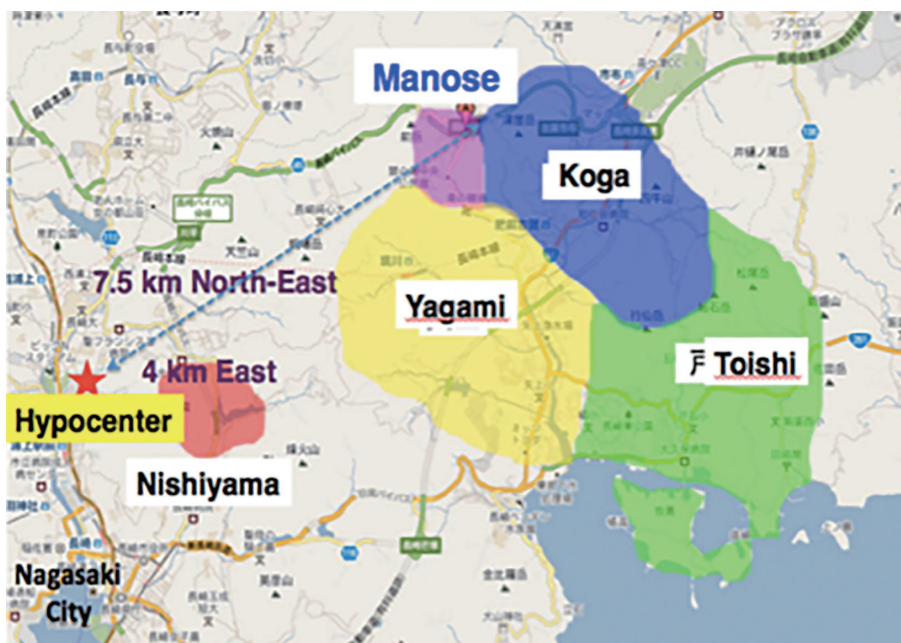


Fig. 1 Location map of Manose District located in the northeastern part of Nagasaki. The figure was quoted from the Black rain in Manose-Nagasaki Doctor & Dentist Association, Nagasaki Nishiyama area net.

As a result, in the ^{137}Cs survey of 1976, the averages were $2,530 \pm 1,390 \text{ Bq/m}^2$ at 107 points in Hiroshima and $5,200 \pm 1,970 \text{ Bq/m}^2$ at 98 points in Nagasaki. Except for the Nishiyama area of Nagasaki, the local fallout ^{137}Cs from the Hiroshima and Nagasaki A-bombs was not recognized clearly in each area inside and outside the black rain drop as a conclusion of statistical analysis. In 1978, additional survey was conducted at the areas where the accumulated amount of ^{137}Cs was higher in the survey of 1976, but no significant difference could be found because the influence of global fallout ^{137}Cs was too large. In 1991, Hiroshima and Nagasaki Cities launched an expert committee related to the black rain, and problems of residual radioactivity, weather simulation, human effect and so on were examined, but confirmed conclusion has not been obtained (Hoshi, 2010).

In order to grasp the deposition and distribution of the A-bomb fallout associated with black rain in Hiroshima, from around 2008 we focused attention on the under-floor soils of the house constructed within the periods of 1 ~ 4 years after the A-bomb as samples thought to be preserving the local fallout of the time without being influenced by the global fallout, and have measured residual radionuclides ^{137}Cs and Pu isotopes, in which the Pu was used as an indicator of contamination from the global fallout. The results by considering the contribution from the global fallout suggested that ^{137}Cs deposition due to the Hiroshima A-bomb was most likely in the range of $50 \sim 100 \text{ Bq/m}^2$ (at the time of detonation) in the areas from north to northwest direction studied (Yamamoto *et al.*, 2013).

In this study, focusing on the Manose District (a small intermountain village about 7.5 km northeast of the hypocenter) where black rain fell shortly after the Nagasaki A-bomb and the hair loss

occurred (Black rain in Manose,2011; Honda, 2012), ^{137}Cs and Pu isotopes in soil have been investigated with the aim of obtaining information on local fallout level and distribution from the Nagasaki A-bomb as a way to solve problems between radiation exposure and hair loss. Here, we report these results together.

2. Overview of the major fallout study of Nagasaki A-Bomb

At Nagasaki A-Bomb (August 9, 1945), Pu atomic bomb was used, unlike the U atomic bomb in Hiroshima, and exploded with the energy yield of 21 kt TNT at 503 m above Nagasaki City. Fission products and induced radionuclides produced by this explosion were basically dispersed by the wind and fell to the ground with rain and/or as particles. Therefore, in the survey on the deposition and spread of local fallout, the measurement results made on samples such as soil give direct evidence, although reproduction calculation using simulation can also be used.

Numerous residual radioactivity studies have been conducted so far in Nagasaki as well as Hiroshima. The largest project was the survey of residual radioactivity in soil carried out in 1976 and 1978 as mentioned above. Except for the Nishiyama area where ^{137}Cs was detected at a high concentration, it was very difficult to find out the traces of the local fallout ^{137}Cs from the Nagasaki atomic bomb due to the large amount of global fallout from the atmospheric nuclear tests from the 1950's to the early 1960's. After that, in addition to ^{137}Cs , some studies on deposition and distribution of Pu isotopes have been conducted (Sakanoue and Tsuji, 1971; Okajima *et al.*, 1980, 1990; Yamamoto *et al.*, 1983a, 1983b, 1985; Kudo *et al.*, 1991, 1995, 2001; Mahara *et al.*, 1994, 1995; Okumura *et al.*, 1999).

Table 1 shows the results of ^{137}Cs and $^{239,240}\text{Pu}$ accumulated levels measured by Yamamoto *et al.* (1985) using the soil samples collected in the survey of 1976 (Japan Society of Public Health, 1976). The amount of local Pu fallout in the Nishiyama area was several ten times higher than those of other areas (NNE, NE, SE, NW). The $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios (0.017 ~ 0.018) in the Manose direction (NE) were within the global fallout value (ca. 0.018), suggesting that the influence of fallout from the Nagasaki A-bomb was negligibly small.

Kudo *et al.* (2001) collected soil samples from a wide area (about 50 sites) in the vicinity of the hypocenter and eastward 100 km to evaluate the spatial distribution and accumulation of ^{137}Cs and Pu fallout (Fig. 2). For $^{239,240}\text{Pu}$, the maximum of 1,500 Bq/m² was observed in the Nishiyama area, and its accumulated amount decreased sharply with the distance toward the east (Fig. 3). The BG level of $^{239,240}\text{Pu}$ was around 60 Bq/m². On the other hand, the ^{137}Cs was high in the vicinity of the hypocenter, but did not show a clear distribution or trend compared to $^{239,240}\text{Pu}$ (Fig. 4). The maximum accumulated level of ^{137}Cs was about 5,000 Bq/m². The $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios are shown in Fig.

Table 1 Historical data-I: Pu and ¹³⁷Cs in soil samples collected at Nagasaki in 1976 (Yamamoto *et al.*, 1985).

Historical data-1 by Yamamoto <i>et al.</i> 1985					
Pu and ¹³⁷Cs in soil samples at Nagasaki collected in 1976					
Direction	Distance km	1976	1976	1976→2015	1976
		Pu-239,140 Bq/m ²	Cs-137 Bq/m ²	Cs-137 Bq/m ²	Cs-137/Pu
Nishiyama					
E	2.7	1668.7 ± 68.07	5365 ± 109	2179	0.311
E	3.5	521.7 ± 13.93	3667 ± 93	1489	0.142
E	3.8	1335.7 ± 39.81	5328 ± 120	2164	0.251
E	4.5	677.1 ± 13.19	3652 ± 88	1483	0.185
E	8	136.9 ± 4.74	2213 ± 71	899	0.062
E	32	77.7 ± 4.34	4995 ± 110	2029	0.016
Other area					
NNE	4	62.9 ± 2.13	2435 ± 87	989	0.026
NNE	10	66.6 ± 3.39	4366 ± 120	1773	0.015
NE	6	55.5 ± 2.62	3178 ± 83	1291	0.017
NE	10	59.2 ± 1.80	3308 ± 41	1344	0.018
SE	6	62.9 ± 2.20	2771 ± 72	1126	0.023
NW	26	37.0 ± 1.45	2383 ± 56	968	0.016

Contamination was primarily localized within 10 km toward the east direction of the hypocenter. In this time, contamination in the NE direction to Manose was not so high, being nearly BG level.

5. The global fallout ^{239,240}Pu/¹³⁷Cs activity ratio is currently around 0.02 to 0.03. The higher ratios of 0.1 or more were found to 2 ~18 km eastern direction. Fig. 6 shows 70 sampling points in soils collected within 15 km from the hypocenter and 5 sampling points collected as controls at more than 15 km by Okamura *et al.* (1999). As shown in Fig. 7, the same result as the results by Kudo *et al.* (2001) was observed. The maximum of ^{239,240}Pu was around 1,000 Bq/m² in the Nishiyama area, and its accumulated level decreased sharply with the distance toward the east. Regarding ¹³⁷Cs, the result did not also show any clear distribution of contamination. In any case, the higher deposition of ^{239,240}Pu was restricted to the Nishiyama area, and the accumulated level gradually decreased as going to the east, and the other areas were close to the BG level.

3. Outline of Manose District

According to the “Nuclear Bomb Record” by the Nagasaki Fire Department (Nuclear Bomb Record, 1945), following the flash of light by the A-bomb, a lot of dust was rolled up with the sound of object and the surrounding area became like a sunset. The dust included not only the black ash of instantaneous combustion that was also called black dust, but also various light weight objects such as paper pieces and cloth broken. Although the range of its spread is not precisely known, it is said that a large amount of dust flowed in the east direction on a gentle southwest wind. In Nagasaki,

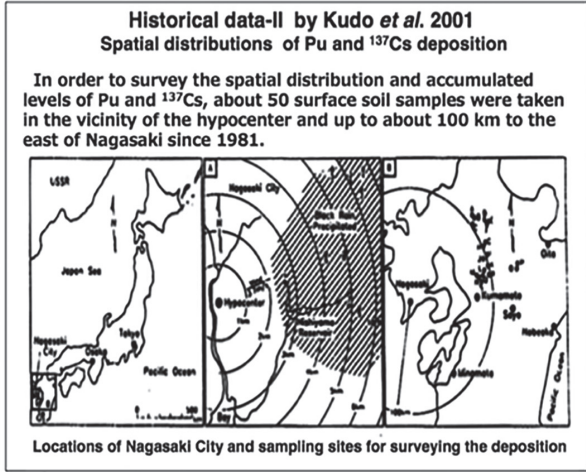


Fig. 2 Historical data-II: Soil sampling points in order to survey the spatial distribution and accumulated levels of Pu and ^{137}Cs (Kudo *et al.*, 2001).

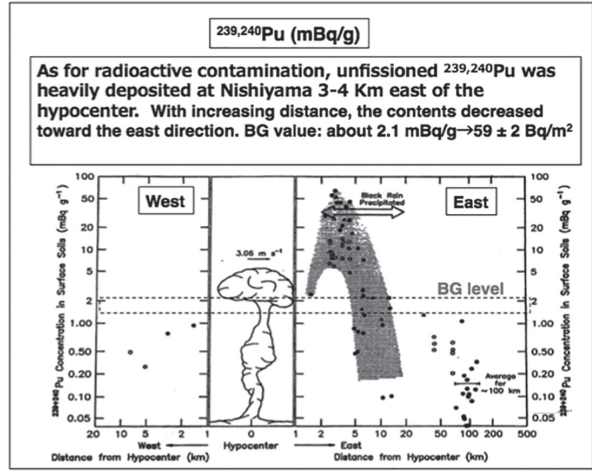


Fig. 3 Historical data-II: Distribution of $^{239,240}\text{Pu}$ inventories (Kudo *et al.*, 2001).

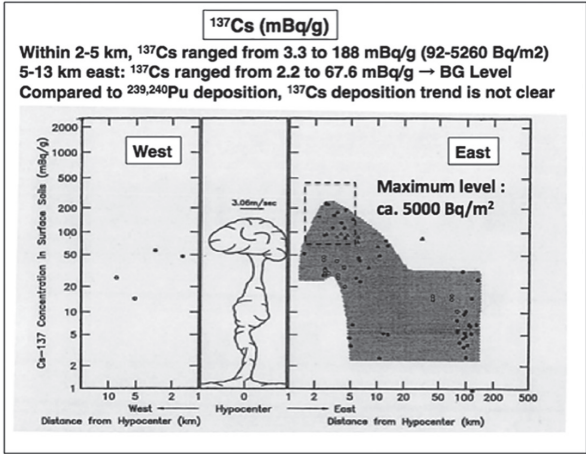


Fig. 4 Historical data-II: Distribution of ^{137}Cs inventories (Kudo *et al.*, 2001).

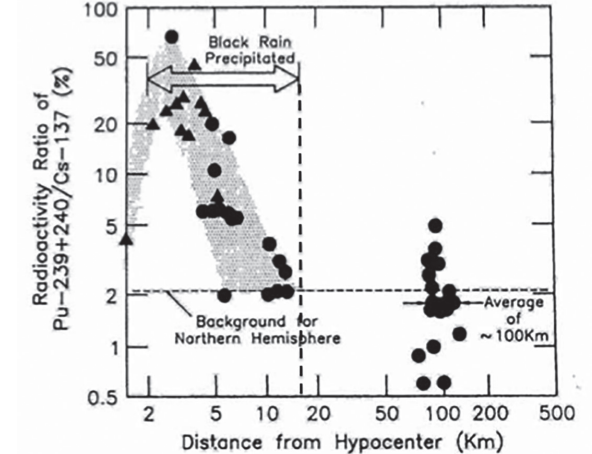


Fig. 5 Historical data-II: Distribution of $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios (Kudo *et al.*, 2001).

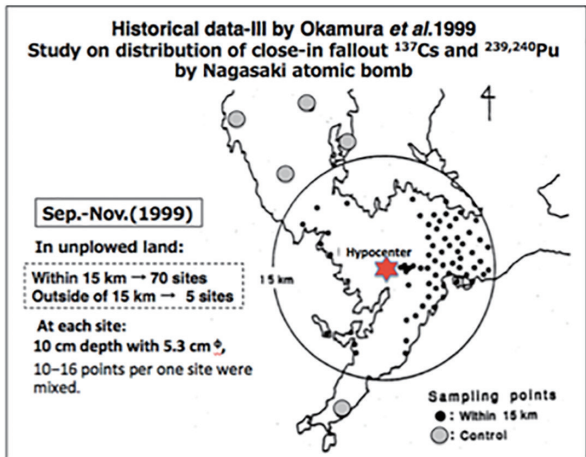


Fig. 6 Historical data-III: Soil sampling points in order to investigate the spatial distribution and accumulated levels of Pu and ^{137}Cs (Okamura *et al.*, 1999).

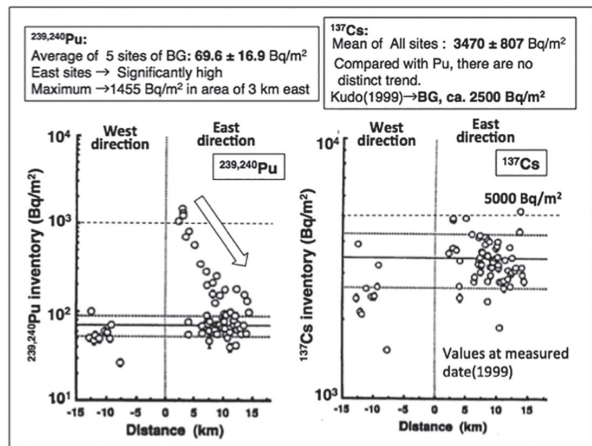


Fig. 7 Historical data-III: Distribution of $^{239,240}\text{Pu}$ and ^{137}Cs inventories (Okamura *et al.*, 1999).

there were few academic records about “black rain” accompanied by radioactive materials and soot of fire (black dust). In the wide area surrounding the hypocenter, it is said that it rained regardless of the amount of rainfall. But, except for the Nishiyama area located just behind the top of Mt. Konpira (altitude 366 m), large amount of rain did not fall. In the Nishiyama area, it rained 20 minutes after the explosion. Other than that, from patients who visited from the mountainous small village “Manose” District, it has been heard and known that black rain fell in the Manose District after the atomic bombing and the hair loss occurred. In the Manose area at the atomic bomb time, 320 residents were living, most of them were farming and were eating rice and vegetables contaminated by black rain. Also, there were no well, and the stream water or spring water had been used as living water (Black rain in Manose, 2011; Honda, 2012).

In March 2011, the Nagasaki Prefectural Insurance Medical Association conducted an interview with outpatients exposed in the eastern area from the hypocenter, including the Manose area, in order to consider the relationship between radiation exposure and hair loss, and the effects of low-dose radiation on the human body (Black rain in Manose, 2011; Honda, 2012). According to the investigation by Honda (2012), in 14 persons interviewed in 13 households with memories of black rain, persons who had hair loss were 6 (43%) out of 14 persons. When including family members, the rate of depilation was 15 (25%) out of 59 persons. On the contrary, in the adjacent Yagami, Koga and Toishi Districts, 3 (3%) out of 99 persons had “depilation”. Other acute symptoms were diarrhea 3 (21%) out of 14 persons, vomiting 2 (14%) out of 14 persons, both are said to be light.

3.1 Research in Manose District this time

3.1.1 Sampling of soil

Based on the above information, soil sampling was performed at 19 sites (M1~ M19) centering on the Manose District (Fig. 8, Appendix I). Sampling was conducted in collaboration with the radiation expert groups of the Prefectural Insurance Medical Association, Hiroshima University and Nagasaki University. There are 7 sites (M2 ~ M8) around Manose area, 2 sites (M9, M10) around Koga area, 2 sites (M11, M12) around Funaiishi area, 1 site (M13) around Hiramama area, 3 sites (M 14 ~ M 16) around Yagami area and 3 sites (M 1, M 17, M 18) around Nishiyama area. One to three soil core samples about 30 cm in depth (stainless steel pipe, diameter 4.7 cm) were collected at each site. All the collected soil samples were taken to the laboratory, removed from the pipe, dried at around 100 °C, sieved 2 mm, and pulverized and mixed so as to be as homogeneous as possible.

3.1.2 Measurement of ^{137}Cs and Pu isotopes

For the soil samples, ^{137}Cs and $^{239,240}\text{Pu}$ concentrations and $^{239}\text{Pu}/^{240}\text{Pu}$ atomic ratio were measured by the following methods:

[^{137}Cs]: Fission products (FP). Aliquots (60-80 g) of the samples were placed in a plastic

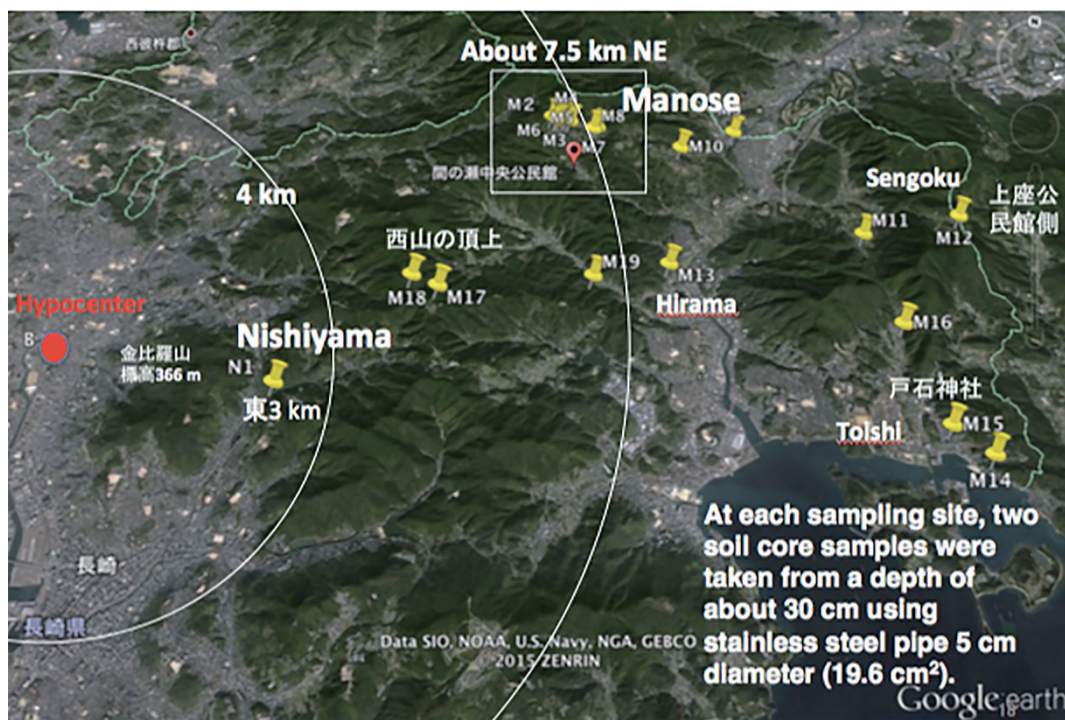


Fig. 8 Map of the soil sampling locations of the present study. Centering on the Manose District, some soil samples were also taken from the Nishiyama and other areas up to 10 km toward the east.

container (inner diameter 6 cm, height 2 cm), ^{137}Cs was determined directly by γ -ray spectrometry using an ordinary Ge semiconductor detector. The spectrometer was calibrated with standards prepared from the New Brunswick Laboratory (NBL) reference material No. 42-1 (4.04% uranium) and analytical grade KCl.

[$^{239,240}\text{Pu}$]: Atomic bomb material. After γ -ray spectrometry, radiochemical analysis of Pu in soil was carried out (Yamamoto *et al.*, 1983 b). After weighing 40-50 g of soil sample, it was calcinated at about 450°C overnight, and Pu was leached using HNO_3 with the addition of a small amount of H_2O_2 by heating for at least 3 ~ 4 h on a hot plate. ^{242}Pu was used as a chemical yield tracer. The supernatant and residue were separated by centrifugation. Thereafter, Pu in the supernatant was coprecipitated with $\text{Fe}(\text{OH})_3$ and Pu was separated and purified using an anion exchange resin column. The purified Pu was electroplated onto a stainless steel disc, and its activities were determined by α -ray spectrometry. Accuracy and precision of Pu analytical method used here was checked using some reference materials and shown to be satisfactory. Since it is difficult to distinguish α -ray energies of ^{239}Pu and ^{240}Pu in the α -ray spectrometer, the total of both is described as $^{239,240}\text{Pu}$.

[$^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio]: Furthermore, in order to clarify the Pu source of contamination, $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio was determined by HR-ICP-MS after chemical separation by the Japan Analytical Center.

3.2 Results and discussion

The results of ^{137}Cs and $^{239,240}\text{Pu}$ accumulated levels (inventory: Bq/m^2) and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios are listed in Table 2. All ^{137}Cs data were decay-corrected to the sampling date. Fig. 9 shows the distribution of ^{137}Cs and $^{239,240}\text{Pu}$ inventories. As can be seen in this figure, at the N 17-2 point of the Nishiyama area M 17, the maximum inventories of $3,000 \text{ Bq}/\text{m}^2$ and $808 \text{ Bq}/\text{m}^2$ were found for ^{137}Cs and $^{239,240}\text{Pu}$, respectively. Previously, in soil samples collected from the area of Nishiyama water

Table 2 Results of ^{137}Cs and $^{239,240}\text{Pu}$ inventories, $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios, and $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios in soil samples.

Sampling Site	Sample No.	Location	Inventory (Bq/m^2)		Atomic ratio	Activity ratio
			^{137}Cs	$^{239,240}\text{Pu}$	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{239,240}\text{Pu}/^{137}\text{Cs}$
M1	N1-2	Nishiyama	274.3 ± 18.2	132.0 ± 1.9	0.058 ± 0.001	0.481 ± 0.033
	N1-3		152.8 ± 19.8			
M2	N2-1	Manose	571.0 ± 83.0	52.7 ± 0.8	0.203 ± 0.007	0.092 ± 0.015
	N2-2		711.1 ± 52.2			
	N2-3		1789.9 ± 139.0			
M3	N3-1	Manose	1409.2 ± 36.6	52.6 ± 0.6	0.173 ± 0.004	0.037 ± 0.001
	N3-2		1469.6 ± 73.4			
M4	N4-1	Manose	180.2 ± 40.6	5.99 ± 0.13	0.157 ± 0.008	0.033 ± 0.008
	N4-2		903.8 ± 48.2			
M5	N5-1	Manose	58.8 ± 4.3	2.73 ± 0.14	0.187 ± 0.022	0.046 ± 0.004
	M5-2					
	N5-3		638.1 ± 38.5			
M6	N6-1	Manose		6.17 ± 0.27	0.200 ± 0.02	
	N6-2					
M7	N7-2	Manose	1718.7 ± 117.2	68.3 ± 2.7	0.157 ± 0.007	0.040 ± 0.003
M8	N8-1	Manose	2020.8 ± 170.1	89.0 ± 1.8	0.154 ± 0.006	0.044 ± 0.004
	N8-2		1121.8 ± 100.3			
M9	N9-1	Koga		27.7 ± 0.9	0.149 ± 0.01	
	N9-2					
M10	N10-1	Koga	111.7 ± 23.3	31.8 ± 1.6	0.154 ± 0.016	0.052 ± 0.006
	N10-2		612.4 ± 60.3			
M11	N11-1	Sengoku	91.6 ± 33.3	4.46 ± 0.24	0.151 ± 0.021	
	N11-2					
M12	N12-1	Sengoku	2117.8 ± 50.5	29.8 ± 0.9	0.171 ± 0.008	0.043 ± 0.006
	N12-2		698.5 ± 87.2			
M13	N13-1	Hirama	48.6 ± 3.8	28.7 ± 1.4	0.061 ± 0.004	0.590 ± 0.054
	N13-2					
M14	N14-1	Toishi	475.9 ± 48.3	42.1 ± 1.7	0.141 ± 0.006	0.088 ± 0.010
M15	N15-1	Toishi	633.3 ± 50.7	192.3 ± 3.7	0.040 ± 0.001	1.951 ± 0.248
	N15-2		98.5 ± 12.4			
M16	N16-1	Toishi	42.5 ± 3.2	35.5 ± 1.5	0.055 ± 0.003	0.834 ± 0.072
	N16-2		181.0 ± 23.3			
M17	N17-1	Nishiyama	835.5 ± 72.1	808.3 ± 25.1	0.039 ± 0.002	0.270 ± 0.015
	N17-2		2998.6 ± 120.5			
M18	N18-1	Nishiyama	1332.3 ± 103.9	1868.3 ± 127.1		
	N18-2					
M19	N19-1	Yagami	32.5 ± 4.8	5.81 ± 0.84	0.092 ± 0.016	0.179 ± 0.037

All data are as of the date of sampling.

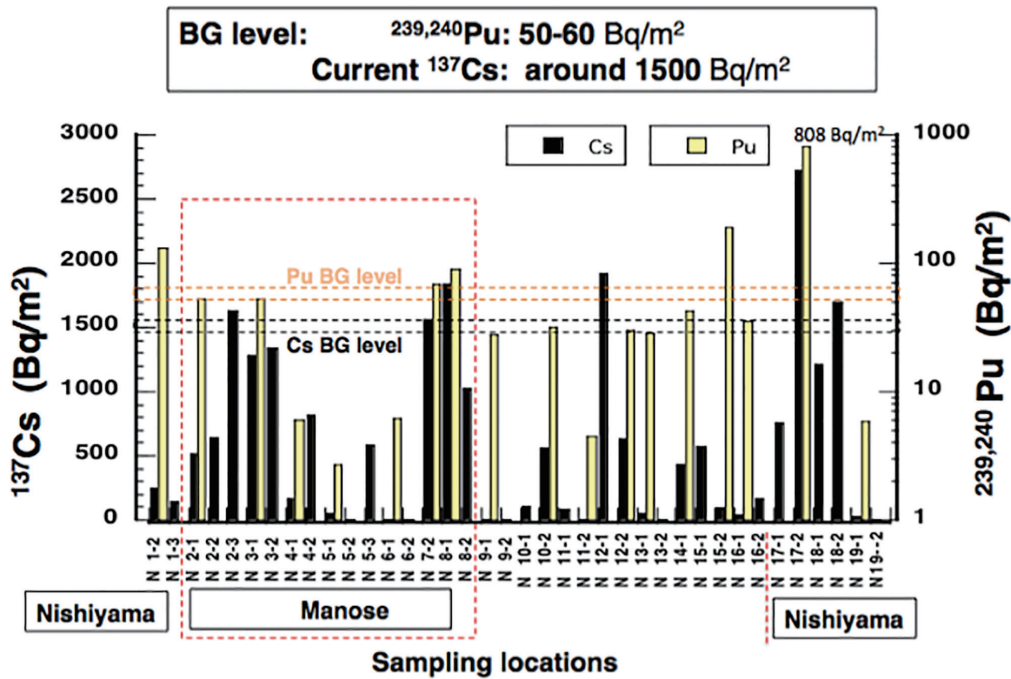


Fig. 9 Results of ^{137}Cs and $^{239,240}\text{Pu}$ inventories in soil samples.

source in 1976, higher values of 2,200 ~ 5,300 Bq/m² for ^{137}Cs and 78 ~ 1,670 Bq/m² for $^{239,240}\text{Pu}$ were found (Yamamoto *et al.*, 1985), but this time lower values of 150 ~ 270 Bq/m² for ^{137}Cs and 130 Bq/m² for $^{239,240}\text{Pu}$ were observed in the Nishiyama water source site M 1. There is considerable fluctuation depending on the sampling site even in the same area.

In the Manose District (M2 to M8) where we are paying attention, there were lower points where the ^{137}Cs inventories were around 1,000 Bq/m² or less, but a little higher points of 1,500 to 2,000 Bq/m² (N 2-3, N 3-1, N 3-2, N 7-2, N 8-1 and N 8-2) were also scattered. Most of the low values were taken from the under-floor of private houses. According to the data of Kudo *et al.* (1999), the BG level of the ^{137}Cs inventory was around 2,500 Bq/m² as of 1999. For comparison, when its value was decay-corrected to the date of sampling this time (2011/7), it is about 1,800 Bq/m². This value is close to the higher ^{137}Cs values found in the Manose. The $^{239,240}\text{Pu}$ levels were several 10 Bq/m², being in the range of the BG level (50 ~ 60 Bq/m²). As a whole, the Nishiyama area is high for both ^{137}Cs and $^{239,240}\text{Pu}$ and those of the other areas are BG level or less.

In Fig. 10, the relationship between $^{239,240}\text{Pu}$ inventory and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio is shown.

The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio for atomic bomb grade Pu is in general about 0.05, while the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in soils for global fallout from the atmospheric nuclear weapon tests is around 0.18. Since that ratios of both sources are largely different, the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio is often used as an isotopic fingerprint to clarify the source of Pu contamination. The values detected this time (Table 2) varied widely from 0.039 to 0.20. The lower values were observed in three areas; 0.039 ~ 0.055 in the Nishiyama area, 0.061 at the N13-1 point in the Hirma area and 0.092 at the N19-1

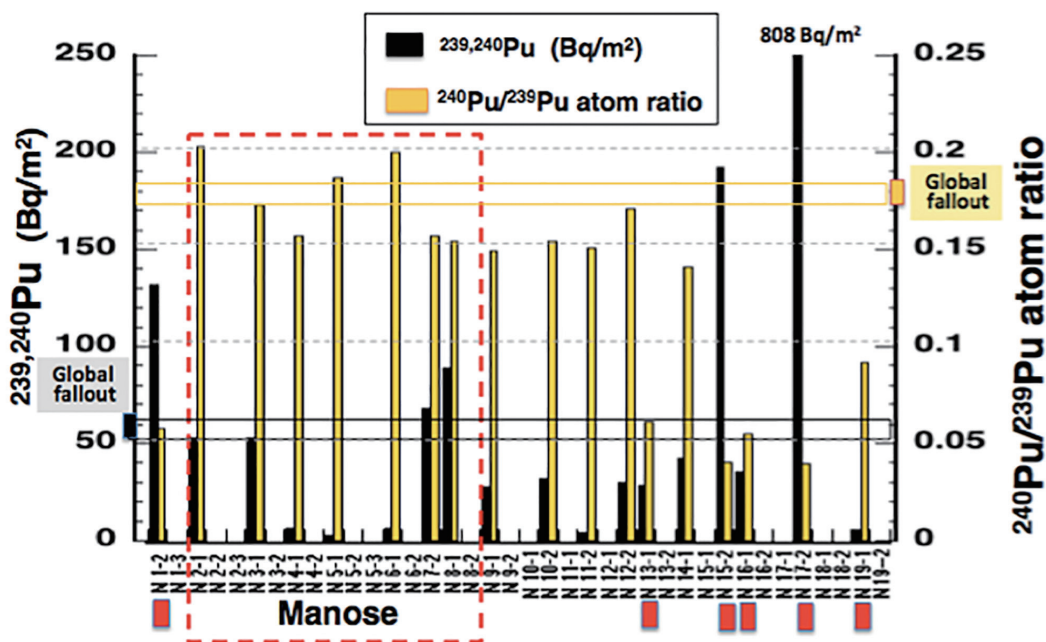


Fig. 10 Relationship between $^{239,240}\text{Pu}$ inventories and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in soil samples. A red square shows a point where the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio measured is less than 0.1.

point in the Yagami area, indicating that Pu found in these areas is mostly due to the local fallout Pu from the Nagasaki A-bomb. In other areas including the Manose, higher values (0.15 ~ 0.20) were observed, indicating that the influence of Pu derived from the Nagasaki A-bomb is small or negligible. Although a small amount of $^{239,240}\text{Pu}$ was detected in the under-floor soil samples of houses from the Manose area, we emphasize that these Pu are of global fallout origin from the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios. Overall, the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios were the lowest around the Nishiyama, and increased from Nishiyama area towards Toishi area in the east. In the northeast direction where Manose District is located, higher $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in the range of 0.16 ~ 0.20 were detected and were within the range of the global fallout value. The points where both 1,500 ~ 2,000 Bq/m² for ^{137}Cs and 5 ~ 70 Bq/m² for $^{239,240}\text{Pu}$ were observed are roughly equivalent to the BG levels.

The $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios varied in a wide range of 0.033 ~ 1.95. Given that the $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratio in the BG area contaminated only by the global fallout is currently 0.03 ~ 0.04, the points where higher values than those were found also indicate that these areas were preferentially contaminated with Pu compared with ^{137}Cs due to the Nagasaki A-bomb fallout. Those ratios were also high around the Nishiyama area. In the Manose district, the values of 0.03 ~ 0.05 were found at most points, being similar to the BG value. Although a little higher ratio of 0.092 was found at the N2-1 points in M2 site, the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio (0.2) was close to the global fallout value.

When looking at the sampling points centering on the Manose District with a little more detail, in the vicinity of the M2 point, it rained around 30 minutes from 11:30 at that time. The residents living in that area were eating leaf vegetables that a lot of burning residue adhered and became black. Furthermore, it was said that the river also became black and the area was hit by a windstorm like a typhoon that glasses and walls fly. The N2-1 and N2-2 points at area M2 are fields, and N2-3 point is at the ridge near the field, but the accumulated level of ^{137}Cs was low; 570 Bq/m² for N2-1 and 710 Bq/m² for N2-2, and disturbance and runoff of soil are conceivable. The ^{137}Cs value (1,800 Bq/m²) at N2-3 of the ridge is BG level. The M3 site is a field, and the ^{137}Cs and $^{239,240}\text{Pu}$ inventories and $^{240}\text{Pu}/^{239}\text{Pu}$ activity ratios were representative BG levels. At M4 site, samples were taken from the dirt floor of a hut and the inventories of ^{137}Cs and $^{239,240}\text{Pu}$ were low; 180 Bq/m² for ^{137}Cs and 6.0 Bq/m² for $^{239,240}\text{Pu}$ at N4-1 point, and 900 Bq/m² for ^{137}Cs at N4-2. The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio was around 0.16, being close to the value (ca. 0.18) of the global fallout. M5 site is under the floor of a private house, the level was also low; $^{240}\text{Pu}/^{239}\text{Pu}$ ratio found indicates the contamination from the global fallout. M6 site is the graded field, and the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio at N6-1 point was similar to the global fallout value. M7 site is close to undisturbed land, and both ^{137}Cs and $^{239,240}\text{Pu}$ show each BG level. The site M8 is a bamboo bush, this point was also almost equal to the BG level. For under-floor soil samples collected from the private house of the Manose District, we expected lower $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio than that of global fallout, but all the measured values were close to the BG level, indicating that those areas are unlikely to be contaminated with the Nagasaki A-bomb Pu. Therefore, the ^{137}Cs in the under-floor soil seem to be mostly due to the global fallout. In the Koga and Funaishi areas (M 9, M 10, M 11) in the east from the Manose district, the level of ^{137}Cs was as low as 1,000 Bq/m² or less, except for N12-1 point (forest) with relatively high ^{137}Cs value (2,100 Bq/m²). The $^{240}\text{Pu}/^{239}\text{Pu}$ ratios (0.15 ~ 0.17) indicate the contamination from the global fallout. In the Hirama (M13) and Toishi (M14 and M15) areas, the ^{137}Cs level was as low as 43 to 633 Bq/m², but $^{239,240}\text{Pu}$ of as high as 190 Bq/m² at N 15-2 point in the door stone was detected. The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio in this area was 0.14 at the N 14-1 point, but at other points it was low at 0.04 ~ 0.06, confirming the flying of atomic bomb Pu.

Overall, as can be seen from the ^{137}Cs and $^{239,240}\text{Pu}$ inventories measured this time, the Nishiyama District was the highest polluted, and it was also reconfirmed from $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio that pollution spread to the east. The influence of the atomic bomb fallout seems to be small in the northeast direction where there is the Manose District.

The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio measured this time was compared to the isodose distribution map of the final detailed measurement data of the Manhattan Study Team measured by the US after dropping the atomic bomb (Manhattan Study Team Final Report, 2002; Honda, 2013). As shown in Fig. 11, the doses are high at the areas of points where the low $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios (preferential

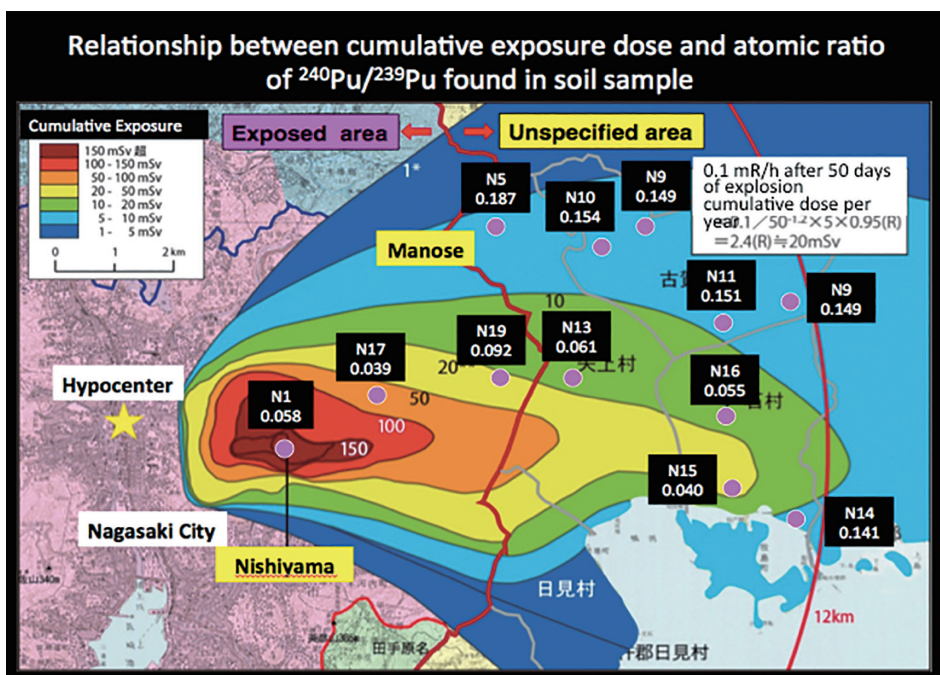


Fig. 11 Relationship between cumulative exposure dose and atomic ratio of $^{249}\text{Pu}/^{239}\text{Pu}$ found in soil samples. Based on the isodose distribution map of the final report of the Manhattan Study Team measured by the US after the atomic bomb and detailed measurement data (Manhattan Study Team Final Report, 1946), the result of superimposing the $^{249}\text{Pu}/^{239}\text{Pu}$ ratios measured this time is shown.

contamination from the atomic bomb grade Pu) were found, and conversely the doses are low at the areas of points where the high $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios (preferential contamination from the global fallout Pu) were found. The dose distribution and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio distribution are harmonic. In Nagasaki, in addition to the ^{137}Cs , the measurements of the Pu inventory and particularly the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio are also important to clarify the level and the dispersion of the local fallout in connection with black rain

4. Conclusion and future problems

This time, as an aid to clarify relationship between the local fallout from the Nagasaki A-bomb and hair loss at the Manose District located about 7.5 km northeast from the hypocenter, soil sampling was conducted in July 2011 at the areas centering on the Manose District, and ^{137}Cs and $^{239,240}\text{Pu}$ concentrations have been measured with $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio. As a result, the Nishiyama area was the highest polluted, and it was reconfirmed from $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio that pollution mainly spread to the east from the hypocenter. The inventories of ^{137}Cs and $^{239,240}\text{Pu}$ at Manose District were not so high, each being nearly BG level.

The problem is, as mentioned above, why hair loss had occurred frequently among residents of the Manose District. As far as the results of ^{137}Cs and Pu this time are concerned, the contribution from the atomic bomb fallout was few or negligibly small, mostly being from the global fallout. By according to the testimony of the residents, the black rain surely fell in the Manose region. Most of the fallout seems to have fell and scattered mainly to the eastern direction including the Nishiyama area before reaching the direction of Manose District. The deposition of radioactive materials accompanied by back rain seems, therefore, to be small in the Manose area located far from Nishiyama area. However, by considering the fact that hair loss thought to be caused by radiation exposure occurred frequently in reality, it is impossible to deny the possibility that some fallout dropped, it adhered to the head and so on, and brought radiation exposure. While paying attention to such things, in future it seems to be necessary to evaluate the deposited amounts of atomic bomb fallout, especially of short-lived radionuclides, at the early stage of the atomic bomb explosion by using simulation, and the relationship between radiation exposure dose by them and hair loss.

Acknowledgments

We would like to express our gratitude to the residents of the Manose area for their useful information and cooperation in collecting soil and so on. We are also grateful to Ms. M. SHIMOTE for her help in editing the paper. This research was supported by JSPS KAKENHI Grants Numbers 23406002 (April 2011-March 2014) and 26257501 (April 2014-March 2018).

References

- Hoshi, M., 2010: The current state of research on radioactive fallout associated with the Hiroshima Atomic Bomb "Black Rain", Hiroshima "Black Rain" Radiation Research Group, Hiroshima Atomic Bomb "Black Rain" - HiSOF.Rain" (in Japanese). <http://www.hisof.jp/01publication/0301BlackRain2010.pdf>
- Honda, T., 2012: Human body influence of residual radiation asked by ORNL-TM-4017, Japan Statistical Society Journal, 42 (1), pp. 103-117 (in Japanese).
- Honda, T., 2013: The atomic bomb "Experience" is an atomic bomb victim, a testimony, 2013 Voice of Hiroshima and Nagasaki 27th collection, p188-202 (in Japanese). <http://www2.nim.co.jp/images/shougen.pdf> ion (Relationship between body dose and Pu-240/Pu-239 ratio, Fig. 12, was prepared by overlaying the data in the figure prepared as a judicial opinion).
- Imanaka, T., Yamamoto, M., Kawai, K., Sakaguchi, A., Hoshi, M., Chaizhunusova, N., Apsalikov, K.,

- 2010: Reconstruction of local fallout composition and gamma-ray exposure in a village contaminated by the first USSR nuclear test in the Semipalatinsk nuclear test site in Kazakhstan, *Radiat. Environ. Biophys.*, 49, 673-684.
- Kudo, A., Mahara, Y., Santry, D. C., Miyahara, S., Garrec, J. -P., 1991: Geographical distribution of fractionated local fallout from the Nagasaki A-bomb, *J. Environ. Radioact.*, 14, 305-316.
- Kudo, A., Mahara, Y., Santry, D. C., Suzuki, T., Miyahara, S., Sugahara, M., Zheng, J., Garrec, J. -P., 1995: Plutonium mass balance released from Nagasaki A-bomb and the applicability for future environmental research, *J. Appl. Radiat. Isot.*, 46, 1089-1098.
- Kudo, A., Mahara, Y., Zheng, J., Sasaki, T., Yunoki, E., Sugahara, M., Santry, D. C., Garrec, J. -P., 2001: Global transport of plutonium from Nagasaki to Arctic: review of the Nagasaki Pu investigation and the future, in: *Plutonium in the Environment A*. Kudo (Editor), Elsevier Science Ltd., 2001, pp. 233-250.
- Manhattan Final Report, 2002: Translation - Hiroshima / Nagasaki Manhattan Province Final Report At April 19, 1946, Nagasaki University post-atomic bomb disaster medical research institute, data collection preservation and analysis department, March 31, 2002 (in Japanese). <http://www-sdc.med.nagasaki-u.ac.jp/abcenter/manhattan/index.html>
- Mahara, Y., Miyahara, S., 1994: Residual plutonium migration in soil of Nagasaki, *J. Geophysical Res.*, 89, 7931-7936.
- Mahara, Y., Kudo, A., 1995: Plutonium released by the Nagasaki A-bomb: Mobility in the environment, *J. Appl. Radiat. Isot.*, 46, 1191-1201.
- Black rain in Manose, 2011: Nagasaki Prefecture Insurance Medical Association.
<http://www2.vidro.gr.jp/manose/>
- Japan Public Health Association, 1976 & 1978: Report on residual radioactivity survey in Hiroshima and Nagasaki.
- Okajima, S., Aikawa, Y., Shimazaki, T., Kudo, 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).
- Okajima, S., Shimazaki, T., Kubo, T., 1990: Measurement of $^{239,240}\text{Pu}$ in soil and plants in the Nishiyama District of Nagasaki, *Health Phys.*, 58, 591-596.
- Okumura Hiroshi, Shimazaki Tatsuya, Yoshida Masahiro, 1999: Measurement and University of Nagasaki Atomic Bomb Plutonium (Regional Creation and University), *Regional Creation and University (Nagasaki University Public Lecture Series 1)*, 1999-03-01, pp. 179 -185. <http://hdl.handle.net/10069/6425>.
- The record of the atomic bomb, 1945: Black ash / black rain, Nagasaki city peace · Atomic bomb, Nagasaki Atomic Bomb Museum.
<http://nagasakipeace.jp/japanese/atomic/record/scene/1104.html>

- Sakanoue, M., Tsuji, T., 1971: Plutonium content of soil at Nagasaki, *Nature*, 234, 92-93.
- Stephen, D. E., 2012: The 2002 dosimetry system (DS02) and available fluence for organ dose calculations, *Radiation Protection Dosimetry*, 149 (1), 21-27.
<https://doi.org/10.1093/rpd/ncr297>
- Yamamoto, M., Komura, K., Sakanoue, M., 1983a: Distribution of the plutonium due to atomic explosion in 1945 from global fallout plutonium in Nagasaki soil, *J. Radiat. Res.*, 24, 250.
- Yamamoto, M., Komura, K., Sakanoue, M., 1983b: Am-241 and plutonium in Japanese rice-field surface soils, *J. Radiat. Res.*, 24, 237-240.
- Yamamoto, M., Komura, K., Sakanoue, M., Hoshi, M., Sawada, S., Okajima, S., 1985: Plutonium isotopes, ²⁴¹Am and ¹³⁷Cs in soils from the atomic bomb areas in Nagasaki and Hiroshima, *J. Radiat. Res.*, 26, 211-223.
- Yamamoto, M., Hoshi, M., Sakaguchi, A., Zhumadilov, K., Endo, S., Sakaguchi, A., Imanaka, T., Miyamoto, Y., 2013: Estimation of close-in fallout ¹³⁷Cs deposition level due to the Hiroshima atomic bomb from soil samples under houses built 1-4 years after the explosion, *Revisiting The Hiroshima A-bomb with a Database*, Vol. 2, (Eds. Aoyama, M. & Ochi, Y.), ISBN. 978-4-9905935-1-3, pp.35-43.

Appendix I Sampling locations in soil samples collected at the surrounding area including Manose District.

Sampling site	Sample No.	GPS position		Air dose (μSv/h)		Location	Note
		Latitude	Longitude	1m height	Ground		
M1	N1-2 N1-3	32°46'10.8"	129°53'35.7"	0.028	0.034	Nishiyama Water resource	Slope of water resource
M2	N2-1 N2-2 N2-3	32°48'37.9"	129°55'46.5"	0.033	0.037	Manose Upland field Upland field Ridge	At past 11:30, a passing shower fell for around 30 minutes. The residents ate vegetables which the ashes attached to. The river became black, too. A mountain became like a typhoon in a bomb blast. The leaf of the potato becomes black. Glass and a wall flew.
M3	N3-1 N3-2	32°48'36.6"	129°55'50.1"	0.028	0.037	Manose Upland field	
M4	N4-1 N4-2	32°48'36.6"	129°55'56.7"	0.057	0.06	Manose Dirt floor	House was built in about 1953.
M5	N5-1 M5-2 N5-3	32°48'37.5"	129°55'54.5"	0.059	0.063	Manose Under floor	Floor bottom of the private house
M6	N6-1 N6-2	32°48'36.4"	129°55'48.9"	0.038	0.048	Manose Terraced field	Originally it is terraced field. House was built in about 1955.
M7	N7-2	32°48'32.3"	129°56'11.9"	0.036	0.033	Manose Untilled ground	In front of guardian diety of children
M8	N8-1 N8-2	32°48'31.7"	129°56'9.6"	0.030	0.036	Manose Bamboo grove	Slope of the clump of bamboo
M9	N9-1 N9-2	32°48'32.6"	129°57'28.8"	0.056	0.063	Koga Under floor	Originally it is upland field. House was built in about 1959. There was a black cloud, but it did not rain.
M10	N10-1 N10-2	32°48'19.6"	129°56'57.5"	0.058	0.079	Koga Under floor	Floor bottom of the private house
M11	N11-1 N11-2	32°47'26.8"	129°58'26.6"	0.050	0.062	Sengoku Under floor	House was built in about 1955. It didn't rain, but wastpaper flew.
M12	N12-1 N12-2	32°47'34.7"	129°59'17.0"	0.050	0.056	Sengoku Forest	There was the black newspaper article which it rained. A foggy deep place.
M13	N13-1 N13-2	32°47'4.1"	129°56'45.6"	0.056	0.060	Hirama Under floor	It did not rain, but various things fell.
M14	N14-1	32°45'40.5"	129°59'3.2"	0.051	0.052	Toishi Under floor	Floor bottom of the private house
M15	N15-1 N15-2	32°45'53.5"	129°58'47.8"	0.053	0.060	Toishi Untilled ground	Side of the torii of Toishi-jinja Shrin.
M16	N16-1 N16-2	32°46'39.5"	129°58'36.1"	0.055	0.068	Toishi Under floor	Originally it is upland field. House was built in about 1955.
M17	N17-1 N17-2	32°46'52.2"	129°54'50.0"	0.037	0.040	Nishiyama Forest	Top of Nishiyama
M18	N18-1 N18-2	32°46'58.4"	129°54'36.8"	0.030	0.038	Nishiyama Forest	Top of Nishiyama
M19	N19-1	32°47'4.3"	129°56'6.3"	0.033	0.039	Yagami	Arakawa

Sampling date: 2011/7/9 for M1~M8, 2011/7/10 for M9~M19.