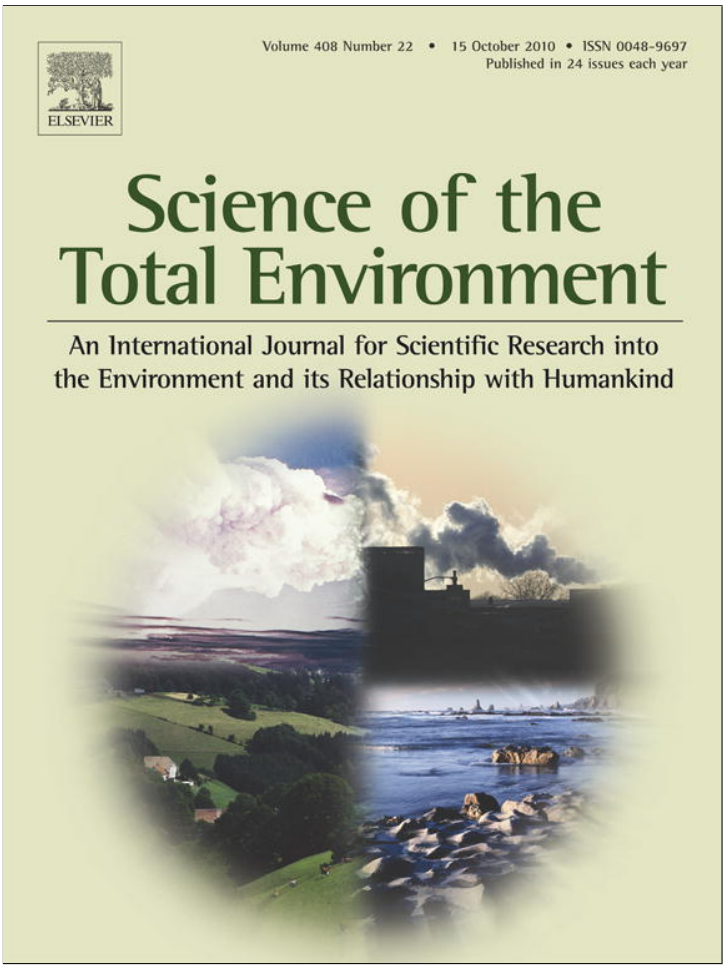

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Feasibility of using ^{236}U to reconstruct close-in fallout deposition from the Hiroshima atomic bomb

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ABSTRACT

The first results on the feasibility of using ^{236}U to reconstruct the level and spatial distribution of close-in fallout deposition from the Hiroshima A-bomb are reported, coupled with the use of global fallout ^{137}Cs and $^{239+240}$Pu. The results for global fallout ^{236}U in soil samples (0–30 cm) from Ishikawa prefecture showed that the deposition density of ^{236}U from the global fallout can be accurately evaluated using AMS. All deposited ^{236}U, ^{137}Cs and $^{239+240}$Pu appeared to have been recovered using 30-cm cores. It was also noted from the depth profiles for ^{236}U/$^{239+240}$Pu and ^{236}U/^{137}Cs ratios that the downward behavior of ^{236}U in the soil was apparently similar to that of $^{239+240}$Pu, while the ^{137}Cs was liable to be retained in upper layers compared with ^{236}U and $^{239+240}$Pu. The accumulated levels were 1.78×10^{13} atoms m^{-2} for ^{236}U, 4340 Bq m^{-2} for ^{137}Cs and 141 Bq m^{-2} for $^{239+240}$Pu. The ratios of ^{236}U/^{137}Cs and ^{236}U/$^{239+240}$Pu were $(4.10\pm0.12)\times10^{9}$ and $(1.26\pm0.04)\times10^{11}$ atoms Bq^{-1}, respectively. Results of ^{236}U, ^{137}Cs and $^{239+240}$Pu measurements for the seven soil cores (0–30 cm) from Hiroshima were discussed on the basis of ratios of ^{236}U/^{137}Cs and ^{236}U/$^{239+240}$Pu by comparing with those from the background area in Ishikawa, indicating that the global fallout dominates the current level of ^{236}U accumulation in soil in the Black-rain area around Hiroshima after the Hiroshima bomb, and the contribution of the close-in fallout ^{236}U produced by the Hiroshima A-bomb seems difficult to observe.

1. Introduction

The Hiroshima atomic bomb (A-bomb) exploded over Hiroshima city on 6 August 1945. Since then, researchers in Japan and USA have been extensively engaged in radiation dose assessment related to the A-bombs deployed in Hiroshima as well as in Nagasaki (Ritchie and Hurst, 1959; Auxier, 1977; Roesch, 1987). Recently, a new dose system DS02 (Dosimetry System 2002) for survivors of the Hiroshima and Nagasaki A-bombs has been developed through a joint effort by Japanese and US scientists (Young and Kerr, 2005). DS02 reconstructs radiation fields of neutrons and gamma-rays released at the time of explosion, and is used to estimate the individual dose of A-bomb survivors who are included in the epidemiological cohort maintained at the Radiation Effects Research Foundation (RERF) (Preston et al., 2004). In addition to radiation at the time of explosion, it is known that the so-called "Black-rain" fell over the north and northwest areas from the epicenter, from 20 to 30 min after the explosion. Radiation exposure due to this close-in fallout was not taken into consideration in DS02 because its contribution was considered to be small for RERF cohort members who were mainly inside the city at the time of the bombing (Okajima et al., 1987). Recently, in relation with an enlargement of social compensation for A-bomb survivors, concerns on radiation exposure due to close-in fallout have been raised among the people who experienced the Black-rain.

Soon after the Hiroshima A-bomb exploded, fission products such as ^{95}Zr and ^{140}Ba were identified by Japanese scientists in sand samples collected in September 1945 on a rain gutter of a house at about 3 km from the hypocenter (Yamazaki, 1953). The investigated area at that time, however, was very limited and the radioactive contamination in most of the Black-rain area was not studied. In 1976–1978, with a concern of possible radiation exposure due to the Black-rain, the long-lived fission product ^{137}Cs was actively measured in surface soil samples collected within the 30-km circle area around Hiroshima city (Japan Public Health Association, 1976, 1978). Excess ^{137}Cs activity from the close-in fallout could not be clearly recognized due to the interference of global fallout ^{137}Cs deposition originating from atmospheric nuclear tests in 1950s and 1960s. However, Takada et al. (1983), by analyzing a correlation of ^{137}Cs contents and ^{234}U/

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^{238}U activity ratios in soil samples, suggested that manifestations of close-in fallout from the Hiroshima atomic bomb could be still detectable in the Black-rain. Thus, radioactive characteristics as well as spatial distribution of the close-in fallout area by the Hiroshima A-bomb have not been specified even 60 years after the A-bombing.

By taking into consideration that enriched ^{235}U was used in Hiroshima A-bomb, it was considered that the deposition of ^{236}U ($T_{1/2} = 2.342 \times 10^7$ y, α-decay), which are formed by nuclear reaction ^{235}U(n, γ), might be a useful indicator for clarifying the close-in fallout level and deposition area. Due to its low specific activity, ^{236}U is not measured with sufficient sensitivity by conventional counting techniques, but it can be determined by thermal ionization mass spectrometry (TIMS) and accelerator mass spectrometry (AMS).

In this paper, we report the first results on the feasibility of using ^{236}U to reconstruct the level and spatial distribution of close-in fallout deposition from the Hiroshima A-bomb. Emphasis was placed on (1) the evaluation of the level and depth distribution of global fallout ^{236}U by using soil samples from Ishikawa prefecture, coupled with the use of global fallout ^{137}Cs and $^{239+240}$Pu, and (2) an attempt to reconstruct the spatial distribution ^{236}U based on the ratios of ^{236}U/^{137}Cs and ^{236}U/$^{239+240}$Pu in Hiroshima, comparing with those from the background area in Ishikawa.

2. Materials and methods

2.1. Soil samples

2.1.1. Soil samples from Ishikawa as background area

In order to evaluate the background level due to global fallout ^{236}U, in total 8 cores of 0–10, 0–20 and 0–30 cm in depth (4.7 cm in diameter) were previously collected within an area of ca. 4 m^2 in August 2008. This sampling site is a forest near LLRL (Low Level Radioactivity Laboratory, Ishikawa prefecture) which has been monitored for a long time by our group (see Fig. 1 in Sakaguchi et al. (2009)). Fallout nuclides such as ^{137}Cs, Pu isotope, ^{241}Am and ^{237}Np have been measured, yielding ratios in agreement with reported values for global fallout. The results of ^{236}U, ^{137}Cs and $^{239+240}$Pu measurements on these samples were already reported by Sakaguchi et al. (2009). In February 2009, in order to study in further detail the depth profile of ^{236}U in soil together with those of ^{137}Cs and $^{239+240}$Pu, a 30 cm core with 4.7 cm in diameter was taken at the site named T3 in Sakaguchi et al. (2009). The core was subdivided into 6 parts of 0–5, 5–10, 10–15, 15–20, 20–25 and 25–30 cm depth. The sub-samples were air-dried, and sieved through a 2-mm mesh to remove pebbles and big plant remains, and pulverized in an agate mortar to obtain homogeneous samples. The top layer (0–5 cm) of this core was soil of fresh–organic appearance and contains only a small amount of inorganic soil compared with the other layers.

2.1.2. Soil samples from Hiroshima

In January 2009, soil cores up to a depth of 30 cm (4.7 cm in diameter) were collected from 7 locations around Hiroshima city by considering the previously measured deposition patterns of ^{137}Cs performed in 1976 and 1978. Sampling points were selected which seemed in an undisturbed condition since the time of the bombing (Fig. 1). Each core sample was divided into three parts: 0–10, 10–20 and 20–30 cm. The samples were treated in the same manner as the soil core from Ishikawa (Sakaguchi et al., 2009).

2.2. Measurements of radionuclide

2.2.1. ^{137}Cs and Pu measurements

Each soil sample was packed into a plastic vessel with a diameter of 6 cm and a height of 2 cm. ^{137}Cs was determined by γ-ray spectrometry using a Ge-detector (Princeton Gamma Tech, relative efficiency 40%) with measurement times of 172,000 s for each sample.

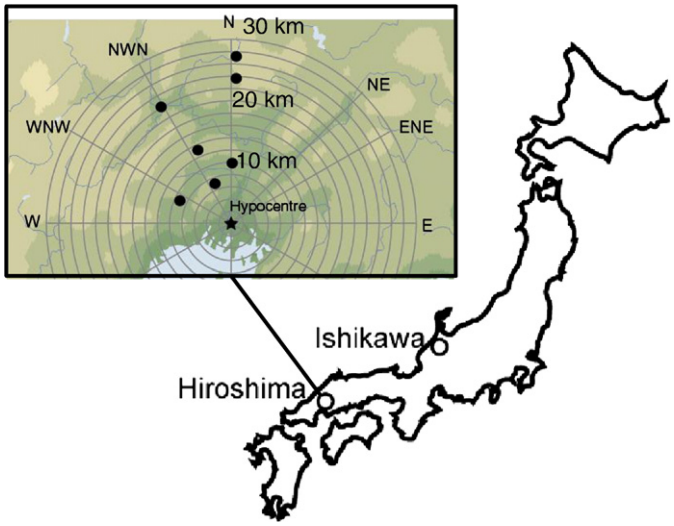

Fig. 1. Map of the sampling stations: Ishikawa and Hiroshima, Japan. The hypocenter of the A-bomb is shown as a star sign. The shading represents altitude above sea level.

The spectrometer was calibrated with standards prepared by the New Brunswick Laboratory (NBL) reference material No. 42-1 (4.04% uranium) and analytical grade KCl. The detection efficiency for ^{137}Cs (662 keV) was 5%.

For Pu analysis, aliquots (20–30 g) of soil samples were treated twice with 8 M HNO_3 for 3–5 h on a hot plate (150 °C) with continual agitation after the addition of ^{242}Pu as a yield tracer. Plutonium was radiochemically separated and purified by using the method reported by Yamamoto et al. (2002, 2008). The purified Pu was electroplated onto polished stainless steel disc, and the activities were determined by α-spectrometry.

2.2.2. U measurements

A portion (3–5 g) of soil samples was leached with 8 M HNO_3 (10 ml soln. per 1 g soil sample) for 3 h on a hot plate (150 °C) with continual agitation. This method was confirmed by Sakaguchi et al. (2009) as the optimal way to treat Hiroshima and Ishikawa surface soil samples in accordance with the aim of this study. The total amount of solution leached was weighed and separated into two aliquots: 1) one third for determining the total amount of leached ^{238}U by α-spectroscopy and 2) two thirds for determining the ^{236}U/^{238}U atom ratio by AMS. For the measurement of the ^{238}U contents, after adding a known amount of ^{232}U to the solution, the uranium was radiochemically separated and purified using an anion-exchange resin column method (Sakaguchi et al., 2004). The purified U was electroplated onto polished stainless steel discs, and the activities were determined by α-spectrometry in the same manner as Pu. For the AMS measurements of ^{236}U/^{238}U at the VERA facility at the University of Vienna, U in the solution was separated and purified in the same manner as above, but without the addition of ^{232}U tracer, since we could not rule out a ^{236}U contamination in the spike. The sputter targets for the AMS measurement were prepared with an iron oxide matrix by calcination at 800 °C for 2 h after co-precipitation with $Fe(OH)_3$. These targets were prepared with ^{238}U concentration of 2–3 μg mg^{-1} Fe oxide. The resultant $^{238}U^{7+}$ currents ranged from 20 to 100 pA, compared to typically 50 nA observed for pure U_3O_8 samples (the currents are typically proportional to the uranium concentration within a factor of 3). However, these currents were well in the range of the current amplifiers connected to our Faraday cups (SR570, Stanford Research Systems, Inc, Sunnyvale, California, USA). The process blank samples gave currents of 1 pA or below. To achieve sufficient precision of the current measurement, the integration time for $^{238}U^{7+}$ was extended to up to 1 s. More information on the

measurement of ^{236}U by AMS is given by Steier et al. (2010). VERA reaches an abundance sensitivity of $^{236}U/^{238}U = 10^{-13}$.

3. Results and discussions

3.1. Core from Ishikawa as background site

Results on ^{137}Cs, $^{239+240}Pu$ and ^{236}U in the Ishikawa core sample are presented in Tables 1 and 2. Depth profiles of the inventory for these nuclides are shown in Fig. 2, together with ratios of these nuclides. In this case, the plotted data are Bq m^{-2} for ^{137}Cs and $^{239+240}Pu$, and atoms m^{-2} for ^{236}U of soil measured in each of the six depth layers.

As seen from Fig. 2 and Table 1, 2 ^{36}U was detected even in the deepest layer of 25–30 cm depth, and ^{236}U concentration was found to be in the ranges from 7.62×10^{6} to 7.62×10^{8} (atoms g^{-1}), and $(3.85\pm0.63)\times10^{11}$ to $(9.24\pm0.43)\times10^{12}$ (atom m^{-2}) for all of the layers measured. The depth profiles of ^{137}Cs, $^{239+240}Pu$ and ^{236}U had a subsurface maximum in the 5–10 cm depth layer below the top fresh–organic soil layer (0–5 cm), and then decreased steeply deeper than this depth (Fig. 2). These depth profiles demonstrate that 80% of the ^{137}Cs, 62% of the $^{239+240}Pu$ and 58% of the ^{236}U remained in the 0 to 10 cm layer out of the total radioactivity up to 30 cm depth. One hundred percent of ^{137}Cs and 98% of ^{236}U and $^{239+240}Pu$ were in the top 25 cm. Considering these depth profiles, it can be assumed that substantially all deposited ^{137}Cs, $^{239+240}Pu$ and ^{236}U have been recovered using the 30 cm core. It is also noted from the depth profiles of the $^{236}U/^{239+240}Pu$ and $^{236}U/^{137}Cs$ ratios that the downward migration behavior of ^{236}U in the soil is apparently similar to that of $^{239+240}Pu$. The ^{137}Cs is liable to be retained in upper layers compared with ^{236}U and $^{239+240}Pu$. The accumulated areal inventories of ^{137}Cs, $^{239+240}Pu$ and ^{236}U were 4340 Bq m^{-2} for ^{137}Cs and 141 Bq m^{-2} for $^{239+240}Pu$ and 1.78×10^{13} atoms m^{-2} for ^{236}U. The ratios of $^{236}U/^{137}Cs$ and $^{236}U/^{239+240}Pu$ were $(4.10\pm0.12)\times10^{9}$ and $(1.26\pm0.04)\times10^{11}$ atoms Bq^{-1}, respectively. These values are consistent with previous measurements (Sakaguchi et al., 2009).

3.2. Cores from Hiroshima

Results of ^{137}Cs, $^{239+240}Pu$ and ^{236}U measurements for the 30-cm soil cores collected at Hiroshima in 2009 are listed in Tables 3 and 4. All of the data are shown in terms of activity or atom per unit soil weight (mBq g^{-1} or atoms g^{-1}) and as activity or atom per unit area (Bq m^{-2} or atoms m^{-2}). Comparison using the accumulated areal inventories seemed to be more meaningful than using concentrations (activity or atoms per unit soil weight), because the latter tended to vary depending on various parameters (soil type, topography, water content, mineralogy, erosion rate, rain fall, etc.). As shown in Table 3 and Fig. 3, the depth profiles of accumulated levels show that 70–95% of the ^{137}Cs and 60–95% of both the ^{236}U and $^{239+240}Pu$ were retained in the 0 to 10 cm layer, while in the deepest 20–30 cm layer, 0–4.2% of the ^{137}Cs, 1.3–5.1% of the ^{236}U and 1.9–6.0% of the $^{239+240}Pu$ were contained, except for the point of 4 km north–northwest from the hypocentre (NWN4-2) (Fig. 3) where 10–12% of the ^{137}Cs, $^{239+240}Pu$ and ^{236}U were found in the deepest layer. These data shows that also ^{137}Cs is accumulated closer to the surface compared with Pu. We observe that ^{236}U behaves in similar to $^{239+240}Pu$, but not like ^{137}Cs, both in soils from Hiroshima and as well in the soil from Ishikawa mentioned above. It has been well known that Cs is positively exchanged with alkaline metals such as K within the clay interlayer, resulting in a significant reduction of ^{137}Cs downward mobility in the surface soil environment (e.g. Bunzl et al., 1998; Lee and Lee, 2000; Seaman et al., 2001; Everett et al., 2008). On the other hand, although Pu is recognized as a particle reactive element, it has been reported that global fallout Pu is trapped in organic phases and/or forms complexes with organic material which are responsible for the vertical diffusion or migration in environmental surface soil (e.g. Bunzl et al., 1998; Lee and Lee, 2000; Seaman et al., 2001). Uranium-236 might also form stable organic complexes and behaves like Pu in soil. As shown in Fig. 3, nearly identical values of $^{236}U/^{239+240}Pu$ ratios are found through the whole depths. Overall, substantial differences are observed in the $^{236}U/^{137}Cs$ ratios for the soil cores measured.

Although there is a high uncertainty due to the small surface area of soil collected, we want to compare our areal inventories of ^{137}Cs, $^{239+240}Pu$ and ^{236}U with the expected global fallout levels. The ranges of areal inventories of ^{137}Cs and $^{239+240}Pu$ up to a depth of 30 cm were from 1100 to 2500 and from 36 to 77 Bq m^{-2}, respectively (Table 4). Average values for ^{137}Cs and $^{239+240}Pu$ were 1790 and 59 Bq m^{-2}, respectively. These accumulated levels for ^{137}Cs and $^{239+240}Pu$ are within the ranges expected for global fallout for the latitude band between 30° and 40° N (e.g. Hardy et al., 1973; Yamamoto et al., 1983; Aoyama et al., 2006). The areal inventory of ^{236}U was in the range 4.30×10^{12}–1.14×10^{13} atoms m^{-2} with an average value of 7.44×10^{12} atoms m^{-2}. This is about half the value of that in the Ishikawa prefecture. This trend can be observed for other global fallout nuclides such as ^{137}Cs and Pu isotopes, which might be mainly due to the differences of precipitation between Hiroshima (1500 mm y^{-1}) and Ishikawa (2200 mm y^{-1}).

The $^{239+240}Pu/^{137}Cs$ activity ratios ranged from 0.030 to 0.042 (0.033 on average) within the ranges which were already reported for

Table 2
Depth profile of activity/atom ratios among ^{137}Cs, $^{239+240}Pu$ and ^{236}U in a 30 cm soil core from Ishikawa in 2009.

Depth (cm)	Weight (g)	$^{239+240}Pu/^{137}Cs$ (A.R.)	$^{236}U/^{137}Cs$ (atoms Bq)	$^{236}U/^{239+240}Pu$ (atoms Bq)
0–5	14.8	0.0197 ± 0.0008	$(2.89\pm0.17)\times10^{9}$	$(1.46\pm0.10)\times10^{11}$
5–10	56.5	0.0260 ± 0.0008	$(3.00\pm0.14)\times10^{9}$	$(1.15\pm0.06)\times10^{11}$
10–15	75.6	0.0481 ± 0.0014	$(6.21\pm0.38)\times10^{9}$	$(1.29\pm0.09)\times10^{11}$
15–20	69.8	0.0579 ± 0.0023	$(8.16\pm0.61)\times10^{9}$	$(1.41\pm0.11)\times10^{11}$
20–25	125.9	0.0867 ± 0.0027	$(13.1\pm0.98)\times10^{9}$	$(1.52\pm0.12)\times10^{11}$
25–30	87.6	N.A.	N.A.	$(1.31\pm0.24)\times10^{11}$
Total	430.2	0.0326 ± 0.0006	$(4.10\pm0.12)\times10^{9}$	$(1.26\pm0.04)\times10^{11}$

N.A.: not analyzed.
A.R.: activity ratio.

Table 1
Depth distribution of concentration and inventory for ^{137}Cs, $^{239+240}Pu$ and ^{236}U in a 30 cm soil core from Ishikawa in 2009.

Depth (cm)	weight (g)	Concentration			Inventory				
		^{137}Cs (mBq g)	$^{239+240}Pu$ (mBq g)	^{236}U (atoms g)	^{137}Cs (Bq m^2)	%	$^{239+240}Pu$ (Bq m^2)	%	^{236}U (atom/m^2)
0–5	14.8	45.40 ± 0.23	0.90 ± 0.04	$(1.31\pm0.07)\times10^{8}$	387 ± 2	8.9	7.6 ± 0.3	5.4	$(1.12\pm0.06)\times10^{12}$
5–10	56.5	94.54 ± 0.84	2.46 ± 0.07	$(2.84\pm0.13)\times10^{8}$	3076 ± 27	70.9	80.0 ± 2.4	56.5	$(9.24\pm0.43)\times10^{12}$
10–15	75.6	12.39 ± 0.11	0.60 ± 0.02	$(7.69\pm0.47)\times10^{7}$	540 ± 5	12.4	26.0 ± 0.7	18.4	$(3.35\pm0.20)\times10^{12}$
15–20	69.8	3.73 ± 0.09	0.22 ± 0.01	$(3.04\pm0.22)\times10^{7}$	150 ± 3	3.5	8.7 ± 0.3	6.1	$(1.22\pm0.09)\times10^{12}$
20–25	125.9	2.58 ± 0.04	0.22 ± 0.01	$(3.39\pm0.25)\times10^{7}$	187 ± 3	4.3	16.3 ± 0.5	11.5	$(2.46\pm0.18)\times10^{12}$
25–30	87.6	N.D.	0.058 ± 0.003	$(7.62\pm0.12)\times10^{6}$	N.D.	0	2.9 ± 0.2	2.1	$(3.85\pm0.63)\times10^{11}$
Total	430.2				4340 ± 28		141 ± 3		$(1.78\pm0.05)\times10^{13}$

N.D.: Not detected.

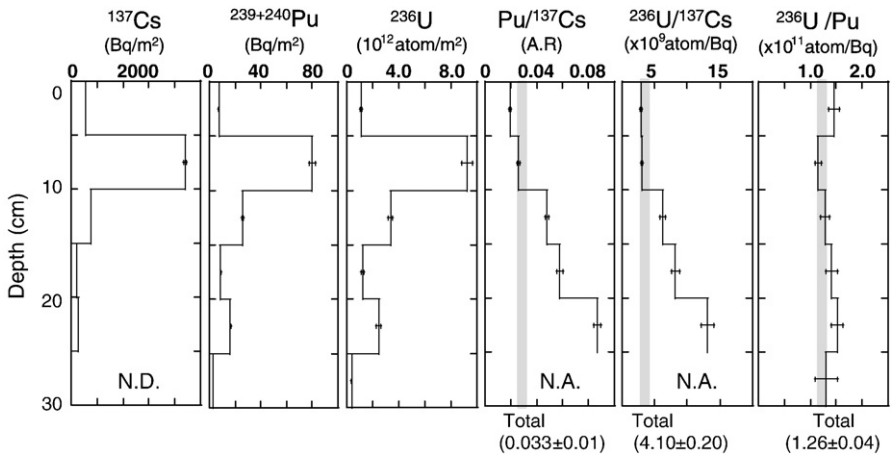

Fig. 2. The depth distributions of ^{137}Cs, $^{239+240}$Pu and ^{236}U, and their activity and/or atom–activity ratios in the 30 cm core from Ishikawa. N.D.: not detected, N.A.: not analyzed.

global fallout (Yamamoto et al., 1983; UNSCEAR, 2000; Ohtsuka et al., 2004). The ^{236}U/^{137}Cs and ^{236}U/$^{239+240}$Pu ratios were 3.40×10^{9}–5.34×10^{9} atoms Bq^{-1} with an average value of $(4.18\pm0.69)\times10^{9}$ atoms Bq^{-1} and 1.10×10^{11}–1.48×10^{11} atoms Bq^{-1} with an average value of $(1.25\pm0.12)\times10^{11}$ atoms Bq^{-1}, respectively. These values indicate that ^{236}U observed in the Hiroshima core samples can be explained by global fallout origin.

3.3. Feasibility using ^{236}U to reconstruct close-in fallout area

Twenty to 30 min after the explosion of the Hiroshima A-bomb, rain started falling in and around Hiroshima city. That rain, the so-called Black-rain, might have transported fission products and components of the A-bomb to the ground, and may have contributed to radiation exposure as residual radiation. According to the report by Uda et al. (1953) which was based on inquiry of affected people in the city, the area of heavy rain was an oval shape with axes of 11×19 km (66 km^2), extending to the northwest and north direction from the hypocenter (Fig. 4). Later, a wider estimate of the Black-rain area was proposed by Masuda (1989).

The occurrence of ^{236}U originating from the uranium A-bomb in Hiroshima is mainly due to fast neutron reaction of ^{235}U(n, γ). Although the detailed structure and composition of the Hiroshima A-bomb are still not officially released, it is assumed based on the best information currently available that 64.15 kg uranium with an average enrichment of 80% was loaded in the bomb (Sublette, 2007), corresponding to 51.3 kg of ^{235}U. The amount of ^{235}U that underwent fission was calculated to be 910 g, by considering the bomb yield of 16 kton TNT (Young and Kerr, 2005) together with a consumption rate of 57 g of fissionable material per kton TNT (Glasstone and Dolan, 1977). The amount of ^{236}U produced through the ^{235}U(n, γ) reaction can be estimated from the cross section ratio of the ^{235}U(n, γ)^{236}U reaction to the ^{235}U(n, fission) reaction. According to JENDL3.3 neutron cross section library (Shibata et al., 2002), for a

Table 3
Depth distribution of concentration and inventory for ^{137}Cs, $^{239+240}$Pu and ^{236}U in a 30 cm soil core from Hiroshima in 2009.

	Distance from hypocentre (km)	Depth (cm)	Weight (g)	Concentration			Inventory				
				^{137}Cs (mBq g)	$^{239+240}$Pu (mBq g)	^{236}U (10^7 atoms g)	^{137}Cs (Bq m^2)	%	$^{239+240}$Pu (Bq m^2)	%	^{236}U (atom/m^2)
WNW10-2	10	0–10	89.0	15.38 ± 0.26	0.495 ± 0.035	6.55 ± 0.42	789 ± 14	71	25.4 ± 1.8	55	3.36 ± 0.22
		10–20	223.5	2.32 ± 0.05	0.155 ± 0.017	1.91 ± 0.16	299 ± 6	27	20.0 ± 2.2	43	2.46 ± 0.21
		20–30	105.6	0.32 ± 0.07	0.018 ± 0.004	0.20 ± 0.05	19 ± 4	2	1.1 ± 0.2	2	0.12 ± 0.03
		Total	418.0				1107 ± 15		46.5 ± 2.9		5.94 ± 0.30
NWN4-2	4	0–10	141.9	16.01 ± 0.19	0.461 ± 0.043	5.36 ± 0.39	1310 ± 16	79	37.7 ± 3.5	66	4.38 ± 0.32
		10–20	202.1	1.49 ± 0.07	0.109 ± 0.014	1.23 ± 0.17	174 ± 8	11	12.7 ± 1.7	22	1.44 ± 0.20
		20–30	269.1	1.11 ± 0.05	0.045 ± 0.006	0.49 ± 0.08	172 ± 8	10	7.0 ± 1.0	12	0.76 ± 0.13
		Total	613.0				1655 ± 19		57.4 ± 4.0		6.58 ± 0.39
NWN8-2	8	0–10	137.6	16.01 ± 0.19	0.461 ± 0.043	7.14 ± 0.48	2366 ± 22	94	48.5 ± 3.6	63	5.67 ± 0.38
		10–20	257.3	1.49 ± 0.07	0.109 ± 0.014	2.49 ± 0.23	153 ± 6	6	26.3 ± 2.2	34	3.70 ± 0.34
		20–30	115.5	1.11 ± 0.05	0.045 ± 0.006	0.21 ± 0.07	1 ± 3	0	1.6 ± 0.4	2	0.14 ± 0.05
		Total	510.4				2521 ± 23		76.3 ± 4.2		9.51 ± 0.52
NWN14-2	14	0–10	138.3	29.83 ± 0.27	0.611 ± 0.046	6.16 ± 0.33	1509 ± 48	90	43.1 ± 1.2	84	4.91 ± 0.26
		10–20	201.9	1.03 ± 0.04	0.177 ± 0.015	0.56 ± 0.08	161 ± 29	10	7.4 ± 0.7	14	0.65 ± 0.09
		20–30	257.8	0.02 ± 0.04	0.023 ± 0.005	0.09 ± 0.03	5 ± 31	0	1.1 ± 0.2	2	0.14 ± 0.04
		Total	598.0				1675 ± 64		51.6 ± 1.4		5.70 ± 0.28
N10-2	10	0–10	154.3	16.54 ± 0.14	0.483 ± 0.023	5.31 ± 0.32	1472 ± 13	74	43.0 ± 2.1	65	4.73 ± 0.29
		10–20	243.0	3.38 ± 0.06	0.143 ± 0.011	2.37 ± 0.21	474 ± 8	24	20.0 ± 1.5	30	3.32 ± 0.29
		20–30	221.3	0.36 ± 0.04	0.029 ± 0.004	0.54 ± 0.08	46 ± 6	2	3.7 ± 0.5	6	0.69 ± 0.10
		Total	618.7				1992 ± 16		66.6 ± 2.6		8.74 ± 0.42
N24-2	24	0–10	83.7	39.59 ± 1.47	1.061 ± 0.047	17.99 ± 1.00	1909 ± 47	80	51.2 ± 2.3	67	8.68 ± 0.48
		10–20	146.3	4.55 ± 0.52	0.249 ± 0.018	2.49 ± 0.20	383 ± 22	16	21.0 ± 1.6	27	2.10 ± 0.17
		20–30	122.0	1.42 ± 0.31	0.066 ± 0.006	0.83 ± 0.10	100 ± 14	4	4.6 ± 0.4	6	0.58 ± 0.07
		Total	351.9				2392 ± 53		76.7 ± 2.8		11.36 ± 0.51
N28-2	28	0–10	96.8	20.26 ± 0.97	0.578 ± 0.018	7.26 ± 0.41	1130 ± 54	96	32.2 ± 1.0	89	4.05 ± 0.23
		10–20	176.2	0.43 ± 0.13	0.033 ± 0.003	0.19 ± 0.04	44 ± 13	4	3.3 ± 0.3	9	0.19 ± 0.04
		20–30	96.7	0.12 ± 0.13	0.012 ± 0.002	0.10 ± 0.03	7 ± 8	1	0.7 ± 0.1	2	0.06 ± 0.02

Table 4
Depth profile of activity/atom ratios among ^{137}Cs, $^{239+240}$Pu and ^{236}U in a 30 cm soil core in 2009.

	Depth (cm)	Weight (g)	$^{239+240}$Pu/^{137}Cs (A.R.)	^{236}U/^{137}Cs (10^9 atoms Bq)	^{236}U/$^{239+240}$Pu (10^{11} atoms Bq)
WNW10-2	0–10	89.0	0.0322 ± 0.0023	4.24 ± 0.28	1.32 ± 0.13
	10–20	223.5	0.0669 ± 0.0073	8.20 ± 0.73	1.23 ± 0.17
	20–30	105.6	0.0576 ± 0.0180	6.31 ± 2.14	1.10 ± 0.35
	Total	418.0	0.0420 ± 0.0026	5.34 ± 0.28	1.27 ± 0.10
NWN4-2	0–10	141.9	0.0288 ± 0.0027	3.35 ± 0.25	1.16 ± 0.14
	10–20	202.1	0.0729 ± 0.0100	8.27 ± 1.21	1.13 ± 0.21
	20–30	269.1	0.0406 ± 0.0059	4.39 ± 0.76	1.08 ± 0.23
	Total	613.0	0.0347 ± 0.0024	3.97 ± 0.24	1.15 ± 0.10
NWN8-2	0–10	137.6	0.0205 ± 0.0015	2.39 ± 0.16	1.17 ± 0.12
	10–20	257.3	0.1721 ± 0.0156	24.2 ± 2.44	1.41 ± 0.17
	20–30	115.5	1.1880 ± 2.7018	107 ± 246	0.90 ± 0.37
	Total	510.4	0.0303 ± 0.0017	3.77 ± 0.21	1.24 ± 0.10
NWN14-2	0–10	138.3	0.0285 ± 0.0012	3.25 ± 0.20	1.14 ± 0.07
	10–20	201.9	0.0462 ± 0.0093	4.04 ± 0.92	0.88 ± 0.15
	20–30	257.8	0.2412 ± 1.6491	30.6 ± 209	1.27 ± 0.46
	Total	598.0	0.0308 ± 0.0014	3.40 ± 0.21	1.10 ± 0.06
N10-2	0–10	154.3	0.0292 ± 0.0014	3.21 ± 0.20	1.10 ± 0.08
	10–20	243.0	0.0422 ± 0.0032	7.01 ± 0.62	1.66 ± 0.19
	20–30	221.3	0.0790 ± 0.0137	14.9 ± 2.86	1.89 ± 0.36
	Total	618.7	0.0334 ± 0.0013	4.39 ± 0.21	1.31 ± 0.08
N24-2	0–10	83.7	0.0268 ± 0.0013	4.54 ± 0.28	1.70 ± 0.12
	10–20	146.3	0.0547 ± 0.0050	5.47 ± 0.53	1.00 ± 0.11
	20–30	122.0	0.0462 ± 0.0074	5.80 ± 1.03	1.26 ± 0.18
	Total	351.9	0.0321 ± 0.0013	4.75 ± 0.24	1.48 ± 0.08
N28-2	0–10	96.8	0.0285 ± 0.0016	3.58 ± 0.27	1.26 ± 0.08
	10–20	176.2	0.0761 ± 0.0241	4.46 ± 1.69	0.59 ± 0.14
	20–30	96.7	0.1038 ± 0.1210	8.71 ± 10.4	0.84 ± 0.30
	Total	369.6	0.0307 ± 0.0017	3.64 ± 0.26	1.19 ± 0.07

A.R.: activity ratio.

fission neutron energy, 0.091 and 1.2 b are given for ^{235}U(n, γ)^{236}U and ^{235}U(n, fission), respectively. Thus, 69 g of ^{236}U production could be estimated for the Hiroshima explosion.

If it is assumed that all ^{236}U produced by the Hiroshima bomb deposited uniformly on the heavy Black-rain area of 66 km^2, a deposited density is calculated to be about 1 μg m^{-2}, or 2.7 × 10^{15} ^{236}U atoms m^{-2}. These values are much higher than that of global fallout inventory, which are on the order of 10^{12}–10^{13} atoms m^{-2}. In addition, if the produced ^{236}U is assumed to distribute uniformly in the surface soil up to a depth of 10 cm, in a soil of density 1.6 g cm^{-3} containing 3 ppm of natural uranium, then the ^{236}U/^{238}U atom ratio in this layer is expected to be 2×10^{-6}.

Results in the present study indicate, however, that the level of ^{236}U observed in soil from the Black-rain area around Hiroshima was

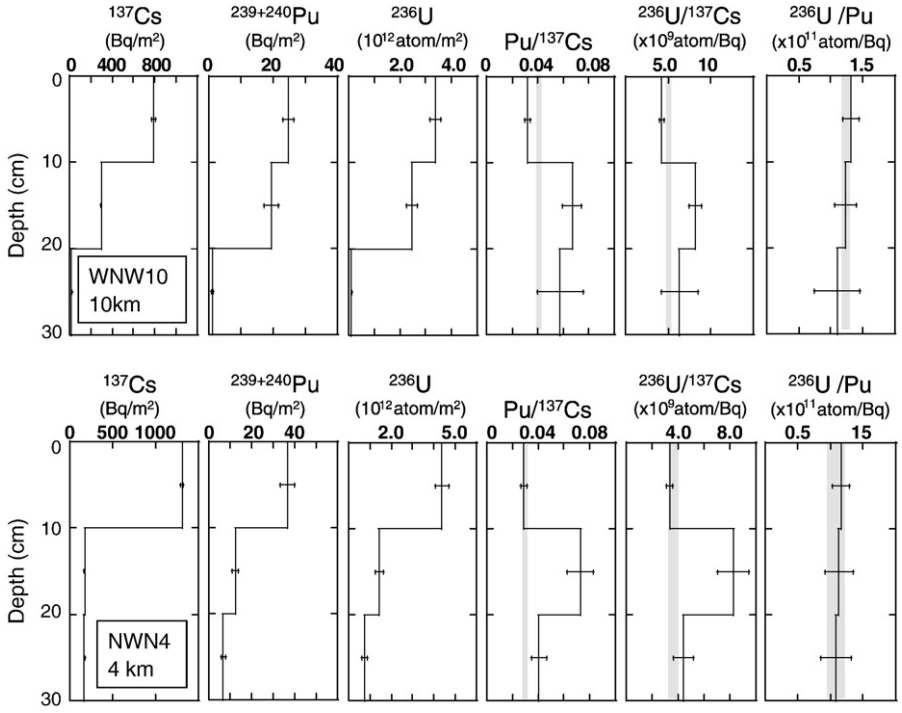

Fig. 3. The representative depth distributions of ^{137}Cs, $^{239+240}$Pu and ^{236}U, and their activity and/or atom–activity ratios in the 30 cm cores from Hiroshima city. WNW10: 10 km northwest–north from hypocentre; NWN4: 4 km north from hypocentre.

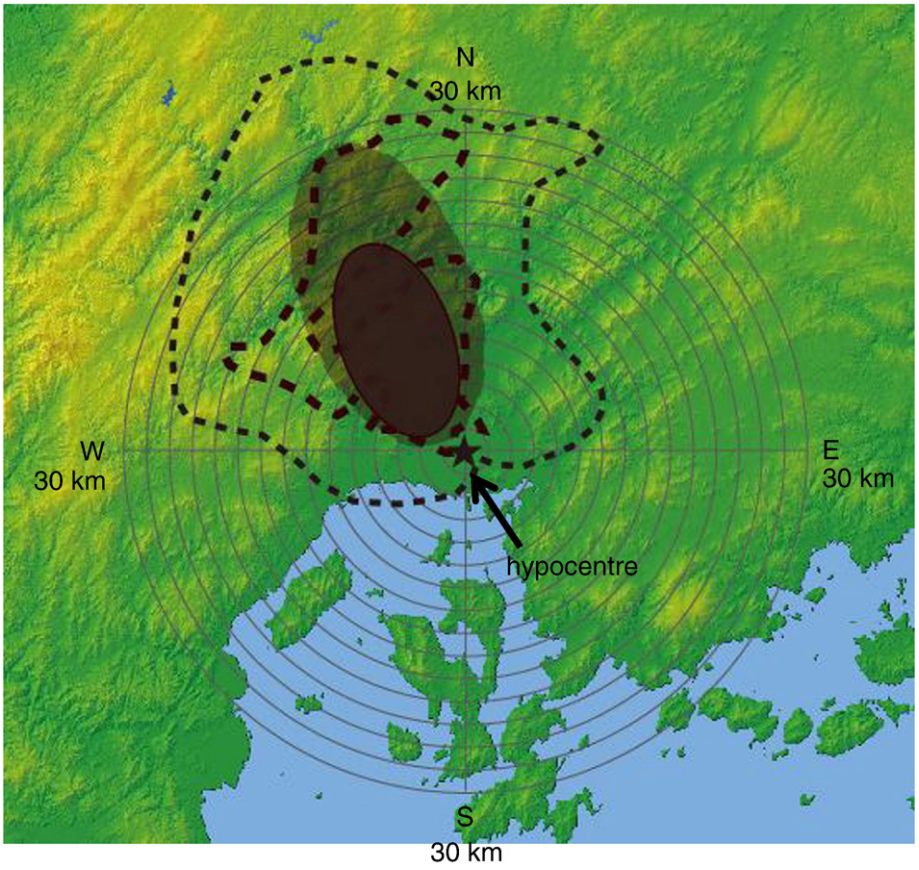

Fig. 4. The Black-rain areas as estimated by Uda et al. (1953) and Masuda (1989). Dark color: heavy rain (Uda et al.); light color: weak rain (Uda et al.); large/thick dashed line: heavy rain (Masuda); small dashed line: weak rain (Masuda).

by two orders of magnitude lower than the level obtained by above calculation. We have to assume that less than 1% of the total ^{236}U produced is deposited on the Black-rain area. The ^{236}U levels measured for the Hiroshima cores are even lower than those observed in the Ishikawa prefecture where only global fallout is considered to contribute to ^{236}U accumulation. It should also be noted that a similar $^{236}U/^{239+240}Pu$ ratio was found in Hiroshima to that observed in Ishikawa, in which $^{239+240}Pu$ could not be expected from the Hiroshima bomb's fallout. These observations suggest that the global fallout dominates the current level of ^{236}U accumulation in soil in the Black-rain area around Hiroshima, and that the contribution of the close-in fallout ^{236}U produced by the Hiroshima A-bomb might be small. Considering that our data are limited, further work is needed to come to a conclusion on the close-in fallout deposition.

4. Conclusions

The feasibility of using ^{236}U which is formed by the nuclear reaction $^{235}U(n,\gamma)$ was examined in order to reconstruct the level and spatial distribution of close-in fallout deposition from the Hiroshima A-bomb.

Our results on global fallout ^{236}U obtained from soil samples (0–30 cm) from Ishikawa prefecture demonstrate that the deposition density of ^{236}U from the global fallout can be accurately determined now using AMS to measure ^{236}U in soil cores. Substantially all deposited ^{137}Cs, $^{239+240}Pu$ and ^{236}U have been recovered using 30 cm cores. The depth profiles of the $^{236}U/^{239+240}Pu$ and $^{236}U/^{137}Cs$ ratios show that the downward migration behavior of ^{236}U in the soil was similar to that of $^{239+240}Pu$, while the ^{137}Cs was more liable to be retained in upper layers when compared with ^{236}U and $^{239+240}Pu$. Results of ^{137}Cs, $^{239+240}Pu$ and ^{236}U measurements for the seven soil cores (0–30 cm) from Hiroshima indicated on the basis of ratios of $^{236}U/^{137}Cs$ and $^{236}U/^{239+240}Pu$ that the global fallout dominated the current level of ^{236}U accumulation in soil in the Black-rain area around Hiroshima after the Hiroshima bomb, and that the contribution of the close-in fallout ^{236}U produced by Hiroshima A-bomb seems difficult to observe. Further work is needed.

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