Preliminary results on soil core samples collected from the under-floors of houses built within 1-4 years after the Hiroshima Atomic Bomb

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Introduction

Twenty to 30 minutes after the explosion of the Hiroshima atomic bomb (A-bomb), rainfall occurred in and around Hiroshima city. That rain, the so-called Black-rain, might transport fission products, induced radionuclides and fissile materials of the A-bomb to the ground, and causes radiation exposure to people living there. According to the report by Uda et al. (1953), the heavy rain area extending to the north-west and north direction from the hypocenter was an oval shape with axes of 11 × 19 km (66 km^2) (Figure 1). This estimated area was based on the condition of the weather on the day and inquiry of affected people in the city. Later, a wider estimate of the black rain area was proposed by Masuda (1989) (Figure 1). Several studies have been conducted to estimate the rainfall area and radioactivity fallout caused by the Black-rain with the analysis of radionuclide composition. However, the Black-rain area and radionuclide deposition rate have not been clarilied, since global-fallout nuclides originated from atmospheric nuclear testing from 1950s to 1960s can disturb accurate determination of the rainfall area and amount of the radionuclides.

The purpose of this study is to clarify the nuclide composition and deposition area of Black-Rain by means of analyzing radionuclides (^{137}Cs, ^{236}U, and $^{239, 240}$Pu) in soil and wall samples collected from (i) under the floor of houses which were built between 1945 and 1948, (ii) around the hypocenter immediately after the explosion of the Hiroshima A-bomb (Nishina sample), and (iii) a wall which has streak-trails of Black-rain.

Material and method

Black-rain streaks on wall

A roof of a house at 3.7 km west of the hypocenter was blow off due to the strong wind of A-bomb at that time, and the Black-rain were come into the house and made the streaks. In 1967, the tainted part of the wall was excised by the residents when the house was renovated. And then, the wall had been preserved until the donation to the Hiroshima Peace Memorial Museum. It is expected that the streaks have not been affected by global-fallout. So this Black-rain streaks will give some of the information on fissile nuclide and bomb material, and will serve as an aid to clarify the Black-rain matter. Three black parts were shaved from the wall's surface and the black surface plaster was separated from the underlying white plaster layer. These were analyzed for ^{137}Cs by non-destructive gamma-ray spectrometry. After that, a portion of wall samples was digested with concentrated HNO_3 and H_2O_2 for 3 h on a hot place (160°C). The sample solution was filtrated (0.20 μm) and separated into two aliquots: (i) for determining the total amount of leached ^{238}U by inductively coupled plasma mass spectrometry (ICP-MS) and (ii) for chemical separation for uranium and plutonium isotopic analysis. Uranium and plutonium were purified from the latter sample using an anion exchange resin column method (Sakaguchi et al., 2004). Uranium fraction was separated into two aliquots (i) for determining the $^{235}U/^{238}U$ atom ratio by multi collector inductively coupled plasma mass spectrometry (MC-ICP-MS) and (ii) for determining the $^{236}U/^{238}U$ atom ratio by accelerator mass spectrometry (AMS). Plutonium concentration and $^{240}Pu/^{239}Pu$ atom ratio were measured with AMS.

Nishina soil samples

9th August 1945, Nishina research team conducted an early survey to find out the radioactive situation of Hiroshima city. The soil samples were collected within the area of 5 km from the hypocenter. Since these samples were collected just 3 days after the explosion, they are not affected by global-fallout. It is also expected that these samples contains radionuclides originated from Hiroshima A-bomb. The samples, which ^{137}Cs could be detected, were measured for uranium and plutonium isotopes with similar methods already mentioned above.

Soil core samples from under-floor

To evaluate the level and spatial distribution of close-in fallout nuclide related to Black-rain, soil core samples were taken from under floor of houses built within 1-3 years after detonation of the Hiroshima A-bomb. These soil samples might not be affected global-fallout so much. In May 2010, four under-floor

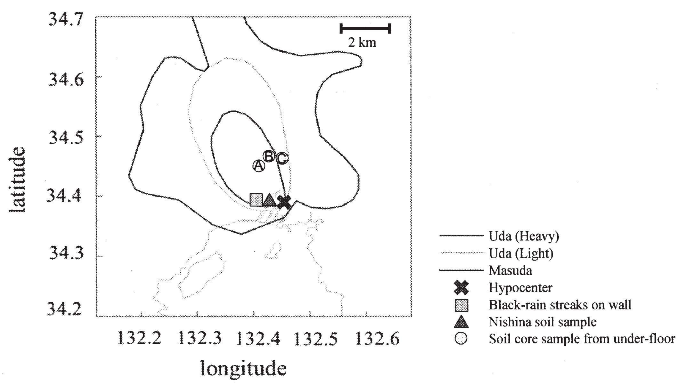

Figure 1 Black-rainfall area and sampling sites. The Black-rain areas were estimated by Uda et al. (1953) and Masuda (1989).

soil cores up to a depth of 30 cm (11 cm in diameter) were collected from 3 locations around Hiroshima City(A, B and C in Figure 1). Two cores were collected at Point A. Each core sample was divided into 5 or 6 parts: 0-3, 3-6, 6-9, 9-15, 15-20 and 20-30 cm in depth. The soil sample was leached with conc. $HNO_3+H_2O_2$ on hot plate (160°C) for three hours. The leaching solution of each sample was weighed and separated into three aliquots: (i) 1/200 for determining the total amount of leached ^{238}U by ICP-MS, (ii) 3/100 for determining the $^{235}U/^{238}U$ and $^{236}U/^{238}U$ atom ratios by MC-ICP-MS and AMS, and (iii) the rest was used for ^{137}Cs and Pu isotope measurements with Ge semiconductor detector and AMS.

The methods of sample treatment has been already reported in Sakaguchi et al. (2009 and 2010).

Results and discussion

Radionuclide composition in Black-rain from Hiroshima A-bomb

$^{239,240}Pu$ in Nishina and rain-streak samples were not detected due to the tiny amount of samples available for our analyses. The $^{236}U/^{137}Cs$ (atom/activity) ratios in the streaks and Nishina samples showed $(4.68\text{-}7.23) \times 10^7$ atom/Bq and 3.09×10^8 atom/Bq (decay corrected to 1945), respectively. These values were smaller compared with the production ratio from the explosion of the A-bomb (1.8×10^9 atom/Bq). It

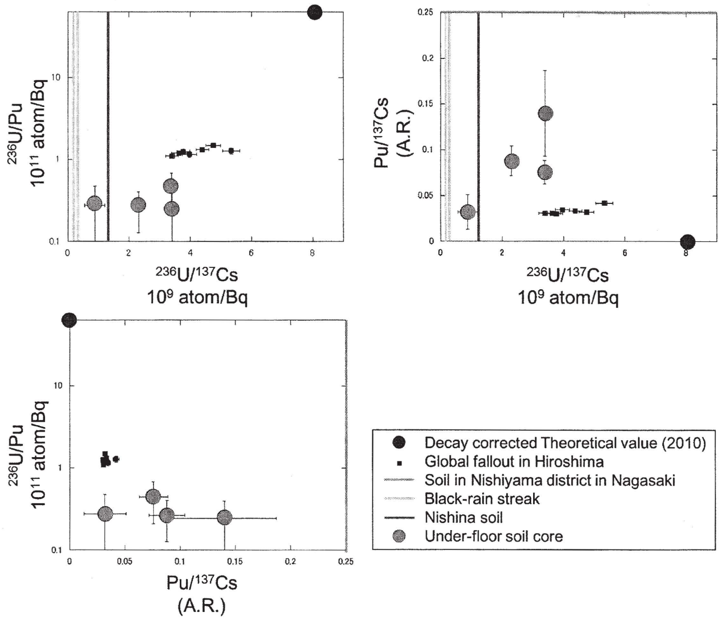

Figure 2 Result of $^{236}U/^{137}Cs$, $^{236}U/Pu$ and $Pu/^{137}Cs$ ratios in Nishina soil, streaks on wall and soil cores from under-floor.

is indicated that the radionuclides have been fractionated between refractory and volatile elements during the explosion and deposition of these nuclides. R/V (refractory/volatile) factors of Hiroshima A-bomb were calculated to be 0.17 and 0.026-0.040 from $^{236}U/^{137}Cs$ ratios of Nishina sample and the streaks on the wall. These values are nearly same value as 0.06-0.5 which was calculated by using $^{235}U/^{238}U$ and ^{137}Cs in a streaks on the wall (Imanaka, 2010). Thus, our values might be recognized as one of the representative value of radionuclide composition in Black-rain.

Radionuclide composition in soil cores from under-floor

The depth profiles of ^{137}Cs in terms of an accumulated level showed that 100% of the ^{137}Cs were retained within the layers up to the depth of 9 cm except for one core among the under-floor soil core samples. The ranges of areal inventories of ^{137}Cs were 19-62 Bq/m^2, and these levels were less than thirtieth of the global-fallout level (2000-3000 Bq/m^2) found in Hiroshima (Yamamoto et al., 2010; Sakaguchi et al., 2010). This suggests that significant amounts of radionuclides deposited locally due to close-in fallout, if soil samples used in this study were uncontaminated by global-fallout (Sakaguchi et al., 2011).

$^{236}U/^{137}Cs$, ^{236}U/Pu and Pu/^{137}Cs ratios in the under-floor soil cores were (0.88-3.40) × 10^9 atom/Bq, (2.42-4.44) × 10^{10} atom/Bq and 0.032-0.140 (activity ratio; A.R.), respectively (Figure 2). Some of these results from the soil samples are significantly different from the value of global-fallout composition, (3.40-5.34) × 10^9 atom/Bq, (1.10-1.48) × 10^{11} atom/Bq and 0.0303-0.0420 (A.R.). These values in the soil samples are also different from the values which were estimated from Nishina sample and the streaks on the wall. It can be said that the radiological composition from these soil core samples may have the mixture information, e.g. A-bomb composition in Nishina and streaks, global-fallout and some other end-member. Actually, Pu/^{137}Cs ratios in the under-floor soil cores show the higher values than that of global-fallout and expected value from the R/V factor. It might suggest that other origin contributed to these values. For example, Pu/^{137}Cs ratio in Nishiyama district in Nagasaki City was about 0.25 (calculated with the values from Kokubu et al., 2007, and decay corrected to 2011). That is, plutonium originated from Nagasaki A-bomb can be a one of the candidate which affected to the value from the under-floor soil samples. For other possibility, there is a specific R/V factor for each element (between Pu/Cs and U/Cs) due to the adsorbent of radionuclides in the atmosphere.

From these results, there is a possibility to identify the fingerprint of Black-rain and to clarify the area of the rain from these soil samples, because the value of our samples were different from the global-fallout values. However, further precise analysis, such as radionuclides in Nishina samples and other material which has the A-bomb radiological composition, are needed to identify the nuclide composition originated from Black-Rain.

Acknowledgements

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References

Imanaka T, External radiation exposure from deposited radionuclides with black rain by the Hiroshima, Current status of studies on radioactive fallout with "black rain" due to the Hiroshima atomic bomb. 89-100, (2010). (in Japanese)

Saito-Kokubu Y, Yasuda K, Magara M, Miyamoto Y, Sakurai S, Usuda S, Yamazaki H, Mitamura M, Yoshizawa S, Distribution of Plutonium Isotopes and ^{137}Cs found in the Surface Soils of Nagasaki, Japan. Journal of Geoscience, Osaka City University 50, 7-13, (2007).

Sakaguchi A, Kawai K, Steier P. Quinto F, Mino M, Tomita J, Hoshi M, Whitehead N, Yamamoto M, First results on ^{236}U levels in global fallout. Science of the Total Environment 407, 4238-4242, (2009).

Sakaguchi A, Kawai K, Steier P, Imanaka T, Hoshi M, Endo S, Zhumadilov K, Yamamoto M, Feasibility of using ^{236}U to reconstruct close-in fallout deposition from the Hiroshima Atomic Bomb. Science of the Total Environment 408, 5392-5398, (2010).

Sakaguchi A, Chiga H, Shizuma K, Hoshi M, Yamamoto M, Preliminary results on ^{137}Cs in soil core samples collected from the under-floors of houses built within 1-4 years after the Hiroshima Atomic Bomb. Revisit The Hiroshima A-bomb with a Database Latest Scientific View on Local Fallout and Black Rain, 93-96, (2011).

Shizuma K, Fukami K, Iwatani K, Hasai H, Low-background shielding of Ge detectors for the measurement of residual ^{152}Eu radioactivity induced by neutrons from the Hiroshima Atomic bomb. Nuclear Instruments and Methods in Physics Research B66, 459-464, (1992).

Uda M, Sugawara H, Kita I, Report on Hiroshima atomic bomb disaster concerning meteorology. In: The Committee for the Publication of the Investigation Reports, ed. Collection of investigation reports on the atomic bomb disaster (Vol. 1). Tokyo: Japanese Science Promotion Society, 98-135, (1953). (in Japanese)

Yamamoto M, Kawai K, Zhumadilov K, Endo S, Sakaguchi A, Hoshi M, Aoyama M, Trial to evaluate the spatial distribution of close-in fallout using ^{137}Cs data in soil samples taken under houses built 1-3 years after the Hiroshima atomic bombing, Current status of studies on radioactive fallout with "black rain" due to the Hiroshima atomic bomb. 79-88, (2010). (in Japanese).