

Investigation of the Fate of U-235 from the Hiroshima A-bomb

Yoko Fujikawa^{*1}, Kiyoshi Shizuma^{*2} and Satoru Endo^{*2}

¹Research Reactor Institute, Kyoto University

Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan.

E-mail: fujikawa@rri.kyoto-u.ac.jp

² Graduate School of Engineering, Hiroshima University, Kagamiyama 1-4-1, Higashi-Hiroshima
739-8527, Japan

Abstract. The Hiroshima A-bomb was considered to have discharged nearly 50 kg of unexpended U-235 into the environment on August 6, 1945. In order to clarify the fate of the discharged U-235, we collected and analyzed U-235/U-238 isotope ratios of some environmental samples from Hiroshima using ICP-QMS (inductively coupled plasma – quadrupole mass spectrometry). Excess U-235 and Cs-137 was detected from a plastered wall that was tainted with the streaks of the *black rain* from the Hiroshima A-bomb, thus proving that at least part of the unexpended U-235 fell with the *black rain*. Because the distribution of radioactive fallout is not known well compared with the distribution of direct radiation and induced radioactivity, the usage of U-235/U-238 ratio as a fingerprint unique to fallout from the bomb was considered promising. In the course of analyses of the *black rain* samples, it was shown that only the rain that fell shortly after the detonation probably contained the detectable amount of U-235 from the bomb. Analysis of the U-235/U-238 ratios of soil samples collected from various places in Hiroshima-city three days after the A-bomb detonation, however, showed that the ratio was generally that of natural uranium. An intercomparison study, then, was conducted to investigate the analytical precision of the U-235/U-238 isotope ratio measurement. As the average relative bias between ICP-QMS and multi-collector ICP-MS was 0.07%, it was considered that the trace amount of excess U-235 from the A-bomb could be detected by repetitive analysis with ICP-QMS. So far, however, not much U-235 from the A-bomb was found from the contemporary topsoil samples from Hiroshima.

1. Introduction

1. 1. Background Information

Extensive study was conducted to estimate the radiation dose of gamma rays and neutrons received by survivors of the Hiroshima A-bomb, as summarized in Dosimetry System 1986 [1] and Dosimetry System 2002 (publication currently under progress by Radiation Effects Research Foundation, Hiroshima, Japan). According to DOE [2], DS02 provides reduced uncertainties in dose estimation and increased confidence in the validity of the risk assessments, while the estimates of risk is not much changed from DS86. The dose estimation problem was therefore basically solved albeit some uncertainty remained regarding individual dose governed by the combination of behaviour and shielding provided at the time of the detonation, and the effect of radioactive fallout.

Less is known about the distribution of the radioactive fallout in case of the Hiroshima A-bomb (*Little Boy*) compared to the Nagasaki A-bomb. Because amounts of FP (fission products) generated by *Little Boy* were much smaller than those deposited in Hiroshima from the other atmospheric nuclear explosions, analysis of FP contents of contemporary soil samples does not show the distribution of the bomb fallout. The high fallout district in Hiroshima was reportedly Koi and Takasu, located approximately 3 km west of the hypocenter [3]. The cumulative dose there was considered to be 1 – 3 R (reviewed in Okajima *et al.* [3]). But due to the limitation in instrumentation at that time, detailed distribution of the fallout radionuclides could not be known. Shizuma *et al.* [4] measured Cs-137 contents of the soil (collected three days after the detonation and was stored without being exposed to the nuclear fallouts in 1960s and 1970s), and reported a slightly higher value of 4 R at Koi [4]. But

because the the number of suitable samples was limited, the exact distribution of the fallout still remained unclear.

1. 2. Purpose and Scope of the Research

The ultimate purpose of our study is to clarify the distribution and fate of radioelements discharged from *Little Boy* based on the U-235/U-238 ratio of environmental samples. The Hiroshima A-bomb was the uranium bomb, utilizing approximately 50 kg of U-235 [5]. Because elevated U-235/U-238 ratios in stratospheric (global) fallout were reportedly scarce except for Cosmos-954 and Cosmos-1714 burn-up, excess U-235 can be utilized as a fingerprint characteristic of the local fallout from *Little Boy*. The excess U-235 from the bomb was first detected by our group [6]. The investigations conducted with respect to this subject includes the following:

- a) measurement of U-235/U-238 in contemporary surface soil from Hiroshima [7],
- b) assessment and development of pre-treatment procedure for uranium separation from soil samples [8],
- c) measurement of U-235/U-238 ratio and Cs-137 in the *black rain* from the Hiroshima A-bomb [6],
- d) measurement of U-235/U-238 ratio in the soil collected from Hiroshima three days after the detonation [9],
- e) quality assurance of the U-235/U-238 measurement by intercomparison study [10],
- f) measurement of U-235/U-238 in undisturbed soil profile from Nissyoen, Hiroshima [11].

In the present report, we summarize the result of the above investigations and propose the direction to the future research utilizing U-235 to clarify the distribution of the fallout

2. Materials and Methods

2.1. Analytical Procedure and Instruments

Analytical instrument used for the U-235/U-238 ratio measurement in our investigation was inductively coupled plasma - quadrupole mass spectrometry (ICP-QMS) model HP-4500 from Yokogawa Analytical Systems (Tokyo, Japan), equipped with Nickel sampling and skimmer cones, a shielded torch, quartz double-pass spray chamber, and micro-flow nebulizer (Agilent Technologies, CA, USA). The instrument was calibrated daily with a mixture of 10 ppb Tl, Y and Li solution for instrumental sensitivity, and with natural uranium solution (SRM4321B from National Institute of Standards and Technology) with known isotope ratio for mass bias correction. The apparatus was housed in a class-10,000 clean room. Uranium was separated from acid – digested sample with U-TEVA resin prior to analysis [8].

Because ICP-QMS is not very suitable for high - precision isotope ratio measurement, the analytical precision of uranium isotope ratio measurement was examined by an intercomparison between our ICP-QMS and multi-collector inductively coupled plasma mass spectrometry (MCICP-MS) in Lawrence Livermore National Laboratory for 14 soil samples collected from a former nuclear tests site in the Marshall Islands.

2.2. Samples

Samples analyzed were as follows:

- a) soil samples from Japan without the chance of pollution with excess U-235, i.e., a brown forest soil (hereafter soil-b) from Ibaragi, Japan; an Ando soil (soil-d hereafter) from Ibaragi, Japan; and a red and yellow soil (hereafter soil-KUR), from Osaka, Japan (Detailed description of the soil is given in [7] and [12]),
- b) contemporary soil samples collected from various place from Hiroshima [7],
- c) soil samples collected from a former nuclear tests site in the Marshall Islands [10],
- d) plastered wall that was tainted with the streaks of the *black rain* from *Little Boy* [6],

- e) soil samples from Hiroshima collected by Nishina *et al.* [13] three days after the A-bomb detonation there, which later were used by Shizuma *et al.* [4] to estimate cumulative exposure from Cs-137 fallout from *Little Boy*,
- f) soil samples from dried-up pond from Nissyoen, Hiroshima where the *black rain* reportedly fell [11].

Shown in Fig. 1 is the map of Hiroshima city with the area of *black rain* proposed by Masuda [14].

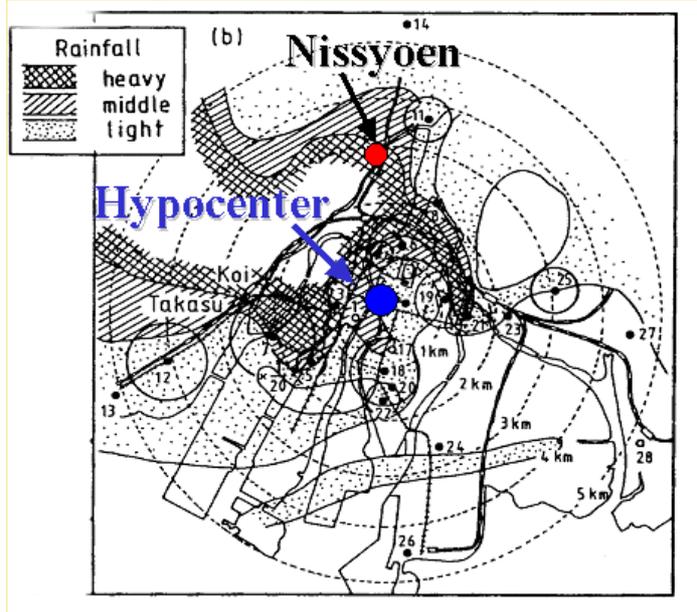


FIG.1 The rain area proposed by Masuda [14] in Hiroshima

3. Results and Discussion

3-1 Analytical Precision of Uranium Isotope Ratio Measurement by ICP-QMS

Based on the investigation conducted so far, careful and repetitive measurement using ICP-QMS was considered effective to detect trace amount of ^{235}U originated from *Little Boy*.

The comparison between ICP-QMS and MC-ICP-MS analysis showed that ICP-QMS can be used to detect less than 1 % (relative) variation in the U-235/U-238 ratio when repetitive measurement (16 times or more) was conducted by ICP-QMS. Namely, the average relative bias on U-235/U-238 and U-234/U-238 atom ratios between the

instruments was only 0.07% and 0.6 % for the 14 soil samples from Marshall Islands with varying uranium isotope ratio, respectively. The maximum relative bias of U-235/U-238 and U-234/U-238 atom ratios observed between the instruments was ca. 0.2 % and 2.0 %, respectively. The bias between the two instruments was smaller than the internal precision of ICP-QMS itself [10].

If the unexpended uranium from the Hiroshima A-bomb was deposited uniformly within the 5 km radius from the hypocenter to the depth of 15 cm in soil, the increase in U-235 content is 0.0028 mg/kg-soil assuming bulk soil density of 1500 kg/m³. The indigenous U-235 content in soil, on the other hand, is 0.0072 mg/kg-soil if natural uranium content of soil is 1 mg/kg-soil, which is the commonly observed uranium concentration in Japanese soil. This suggests that more than 10 % (relative) increase in U-235/U-238 ratio can be expected for the soil that received the fallout containing the uranium from the bomb. Considering the analytical precision achievable by ICP-QMS, we considered that if excess uranium was present in Hiroshima, it should be detectable by our instrument.

3.2. Analysis of the Black Rain

For the *black rain* samples obtained from the rain-tainted wall of a house 3.7 km west of the hypocenter in Hiroshima, repetitive measurement exceeding 5 times was not done because of the limitation in the amount of sample. Careful blank control was conducted in order to prevent the chance of the cross-contamination of the sample based on our experience on ultra-trace analysis [15].

The results obtained clearly showed elevated amount of U-235 relative to U-238 (U-235/U-238 atom ratio 0.00741 to 0.00779) and Cs-137 where black streaks of rain was left on the wall [6]. It was certain that at least part of the U-235, the bomb component, and fission products produced by nuclear reactions fell with the *black rain*.

It was said that heavy rain began to fall 20 to 30 minutes after the detonation of Hiroshima. During the first hour or two of this rain was huge black drops containing dirt, dust, and soot sucked up into the air at the time of the explosion and during the fire. The rainwater later became more clear with less amount of solid materials. While the wall surface with black rain streaks contained both excess U-235 and Cs-137, the surface with white rain streaks and reddish rain streaks did not contain detectable amount of excess U-235 but contained small amount of Cs-137 [16]. The behaviour of FP including Cs-137 and bomb material could have been different as suggested by its distribution among the different fractions of rain.

3.3. Analysis of the Soil Samples Collected Shortly after the Detonation

Soil collected from various places from Hiroshima city (sampling locations shown in Fig. 1) three days after the detonation did contain Cs-137 that was originated from Little Boy [3]. The soil later was subjected to HNO₃ digestion in open vessels at 180 °C for 30 minutes, and the supernatant was subjected to analyses by ICP-QMS, after separation of uranium. The observed U-235/U-238 ratio, however, was that of natural uranium considering the analytical error, except for sample No. 4 which showed slightly higher ratio than natural ratio. The possible reasons that excess U-235 was not much detected in our soil samples were that U-235 in black rain was diluted out by indigenous uranium in the soil, or that U-235 in black rain was transported out of the topsoil by infiltration or by surface runoff by the time the samples were collected.

It is also probable that the procedure of the digestion of sample was inadequate. By using concentrated HNO₃ for digestion, indigenous uranium other than bomb-originated U-235 can be leached from the sample, rendering the detection of the variation in U-235/U-238 ratio difficult. Judging from the amount of U-235 discharged from the A-bomb, however, the effect of the chemical extraction procedure was not likely to be significant, if significant amount of the bomb material fell as local fallout.

U-234/U-238 activity ratio (A. R. hereafter) in HNO₃ digest of some Hiroshima soil were higher than 1.0 (1.21 at maximum), indicating a deviation from radiological equilibrium. Takada *et al.* [17] also reported high U-234/U-238 A. R. values (1.37 at maximum) in 0.1N HNO₃ digest of soil collected from Hiroshima fallout area, and discussed the possible effect of enriched uranium that was considered to have fallen there. High A. R. values in HNO₃ digest, however, may also be caused by natural mechanisms. A. R. value in HNO₃ digest of soil and that in digest of totally decomposed soil, for example, was 1.63 and 0.95, respectively, in case of soil from Kumatori, Osaka, Japan. Occurrence of high U-234/U-238 A. R. in HNO₃ leached fraction by natural mechanisms was also possible in case of Hiroshima soil samples [9].

3.4. Analysis of the Contemporary Soil Samples from Hiroshima and the Other Places

Soil samples collected from various places from Japan outside Hiroshima showed natural isotope ratio of U-235/U-238 [7].

Shown in Figs. 2 and 3 are the depth profiles of U-235/U-238 ratio and Cs-137 contents in soil obtained for the two (core No.2 and No. 4) of the nine soil cores obtained from the dried-up ponds in Nissyoen, Hiroshima, Japan, where the black rain fell. The soil samples were totally decomposed by fusion and acid digestion prior to separation of uranium for ICP-QMS analyses. Approximately 10 to 20 times of repetitive analysis by ICP-QMS was done for each sample, changing time and date of the measurement to rule out the random error of the measurement.

The core no. 2 (Fig.2) shows the undisturbed Cs-137 distribution profile with depth, and slightly increased U-235/U-238 ratio at the depth of 7 cm where the Cs-137 content approaches to zero. Because the previous experiment showed that the average relative bias of our uranium isotope ratio measurement was better than the internal precision of ICP-QMS itself [10], maximum relative bias of 0.2 % (U-235/U-238) was also shown in the figures to indicate the confidence limit of our data.

Comparison between Cs-137 profiles in Figs.2 and 3 shows that the soil from sampling station no.4 was disturbed probably due to anthropogenic activity while the soil from station no.2 was not much

disturbed. The U-235/U-238 ratio in core no.2 was slightly higher in the deeper layer (from the depth of 4 cm to 9 cm) where the concentrations of fallout Cs-137 from atmospheric nuclear test were smaller. The observed U-235/U-238 atom ratios in core no.4, on the other hand, were relatively constant to the depth of 12 cm.

Because the detonation of Hiroshima (1945) was well before the 1960s when the deposition of Cs-137 from atmospheric nuclear explosion peaked, the slight increase in U-235/U-238 ratio in core no. 2 may be the effect of the Hiroshima A-bomb. The elevation of the U-235/U-238 ratio, however, was very small. If most of the U-235 from the bomb was deposited as local fallout and was retained in topsoil layers, more significant increase in the isotope ratio is expected.

The U-235/U-238 ratio showing no significant elevation in the core no. 4 (Fig. 3) with disturbed profile of Cs-137 supports our assumption that a very small increase in U-235/U-238 ratio may exist at the certain depth of soil in this site. The small elevation in the uranium isotope ratio could be effectively extinguished by physically disturbing the soil layers.

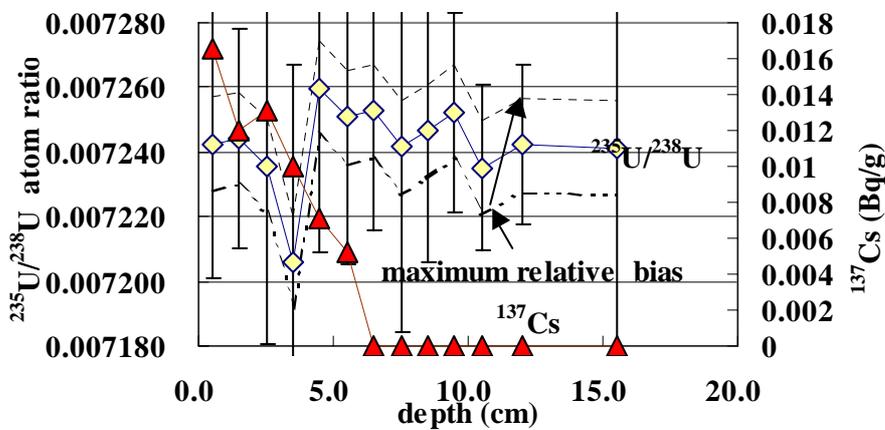


FIG. 2. Cs-137 content and U-235/U-238 atom ratio in core no.2 from Nissyoen, Hiroshima.

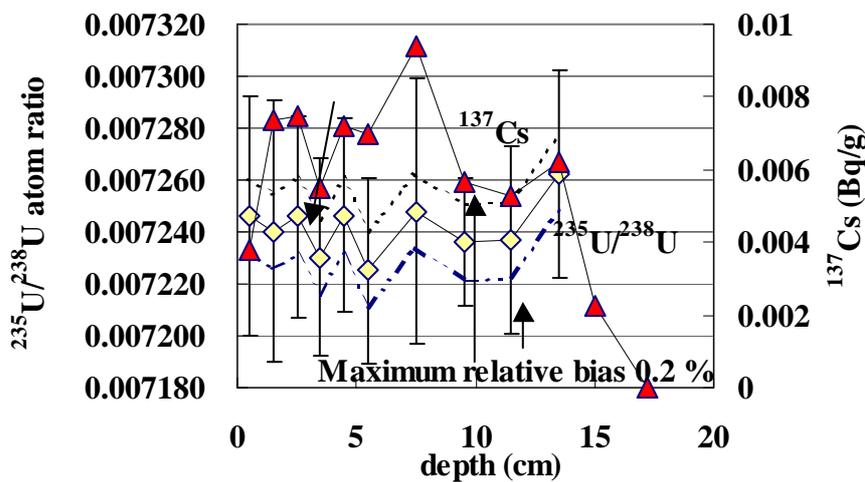


FIG. 3 Cs-137 content and U-235/U-238 atom ratio in core no.4 from Nissyoen, Hiroshima.

4. Conclusions

Excess U-235 was detected in the black rain from the Hiroshima A-bomb. Part of U-235 therefore was locally deposited on the ground, but the amount of excess U-235 found in soil samples from Hiroshima was much less than the amount expected if most of the unexpended U-235 fell there.

Further investigation is necessary to clarify the fate of U-235 from the A-bomb. The optimal sample treatment procedure to solubilize bomb-originated U-235 in soil need to be pursued. The extent of fractionation of U-235 and the other fission products regarding their deposition on the ground also needs to be investigated.

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