J. RADIAT. RES., 24, 229-236 (1983)

Uranium Isotopes in Hiroshima "Black Rain" Soil

JUN TAKADA, MASAHARU HOSHI, SHOZO SAWADA and MASANOBU SAKANOUE*

Research Institute for Nuclear Medicine and Biology, Hiroshima University, Kasumi 1-2-3, Minami-ku, Hiroshima, 734, Japan
*Low Level Radioactivity Laboratory, Nomi-gun, Ishikawa, 923-12, Japan

(Received April 19, 1983)

Uranium isotopes/Soil/Hiroshima "black rain"

Uranium isotopes were studied to clarify the fallout from the Hiroshima atomic bomb. The $^{234}U/^{238}U$ activity ratios of uranium leached from the soil in the Hiroshima fallout area were significantly higher than those of other areas according to alpha ray spectrometry. The correlation of specific activity of the fission product ^{137}Cs and the $^{234}U/^{238}U$ activity ratios was studied. Two different correlations were established inside and outside the fallout area. The results of this study suggest that manifestations of fallout from the Hiroshima atomic bomb are still detectable in the "black rain" area

INTRODUCTION

Soon after the Hiroshima atomic bomb exploded, fission products in earth and sand collected on a rain pipe of a house 3 kilometers from the hypocenter, were identified by Kimura and Murakami[1a]. However, enriched uranium of the Hiroshima atomic bomb was not found. In Nagasaki, however, high level fission products and ^{239}Pu[2] of the atomic bomb were discovered in the Nishiyama district.

From twenty to thirty minutes after the explosion, so-called "black rain" fell over the northwest part of Hiroshima City, as indicated in Fig. 1[1b]. Heavy rain continued for one hour or more over an elliptical area, 19 x 11 kilometers toward the northwest which included the hypocenter. Light rain fell in a 29 x 15 kilometer area. Earlier measurements[1c, 1d, 3, 4] of residual radio activity were made at points at the southern edge of the fallout area. Takeshita[5] estimated external cumulative doses in the "black rain" area to be 1.8 to 44 rad using these data. Residual ^{137}Cs activity was actively studied in the fallout area in 1976 and 1978[6, 7]. Existence of residual ^{137}Cs activity from the Hiroshima bomb was not recognized as due to fallout from world-wide nuclear tests[8].

The energy yield of the Hiroshima atomic bomb was estimated to be equivalent 12.5 kilotons[9] of TNT, which corresponds to the fission of about 700 grams of ^{235}U, and nearly all enriched uranium must be dispersed without fission. If the enriched uranium fell on the earth, uranium isotopic composition of the

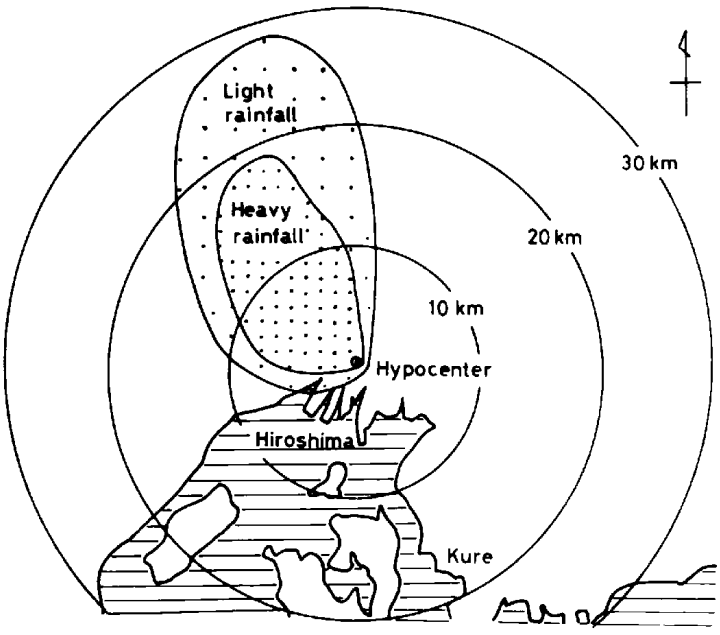

Fig. 1. "Black rain" fallout area in Hiroshima.

soil might be unusual. We measured the $^{234}U/^{238}U$ activity ratio of the uranium leached from soil using alpha ray spectrometry. The results obtained and the fallout from the Hiroshima bomb are discussed in this report.

MATERIALS AND METHODS

Soil samples were obtained at distances from 2 to 30 km from the Hiroshima hypocenter in 1976[6]. Sampling points which seemed likely to have been in stable condition since the time of bomb, were selected. The soil samples were collected from the surface to 10 cm depths after the removal of fallen leaves. The soil samples were dried at 100°C and passed through a 2 mm sieve. At each point, 20 grams soil was used for measurements. Uranium was separated according to the method shown in Fig. 2. Each soil sample was shaken in 60 ml 0.1 N nitric acid for 5 hours at room temperature. The soil suspended in the solution was subjected to centrifugation. The residue was treated with 60 ml 8 N nitric acid at 120°C for an hour, then removed by centrifugation. Thus, two kinds of solutions were prepared. We presume that the uranium in the former solution was leached from the surface of soil particles, and that the uranium in the latter solution was leached from the inside of soil particles. We term the former S-uranium, and the latter I-uranium. After the former solution was dried, it was dissolved in 30 ml of 7:93 nitric acid containing 60 g $Al(NO_3)_3$. Uranium was extracted using 30 ml ethyl acetate into the organic phase and was washed with 30 ml 7:93 nitric acid

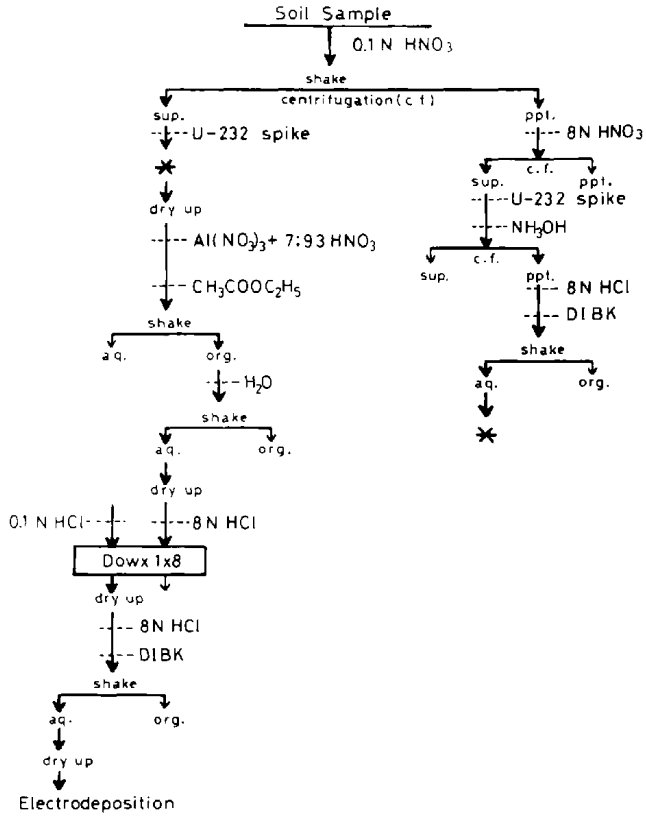

Fig. 2. Analytical scheme for the extraction of uranium.

containing 60 g $Al(NO_3)_3$. Then, uranium was back-extracted into the aqueous phase with three 20 ml portions of water. The aqueous phase was evaporated and made into 30 ml 8 N hydrochloric acid. The solution was passed through an anion exchange column (DOWEX 1 x 8) and washed with 40 ml 8 N hydrochloric acid. Uranium was eluted with 30 ml 0.1 N hydrochloric acid. The solution was dried, and made into 30 ml 8 N hydrochloric acid. Iron was extracted using 30 ml Di-*iso*-butyl keton into the organic phase. The aqueous phase containing uranium was dried and then dissolved in 10 ml 1:9 sulfuric acid at 100°C. The hydrogen exponent (pH) of the solution was adjusted to 2.0 by adding ammonium. Uranium was electrodeposited on a stainless steel plate over 2.5 hours at 0.5 amperes. Uranium leached by 8 N nitric acid was coprecipitated with iron hydroxide. After centrifugation, 50 ml of 8 N hydrochloric acid was added to the precipitate. Then, iron was extracted with 50 ml di-*iso*-butyl keton into the organic phase. The subsequent procedure was the same as for S-uranium. The alpha-spectrum of each source was measured using a 200 mm^2 silicon surface barrier detector for about 24 hours.

RESULTS AND DISCUSSION

The results of alpha-spectrometry for uranium leached from soil by 0.1 N nitric acid are shown in Table 1. These were classified according to geological features, because behavior of ^{234}U differs geochemically from that of ^{238}U[10]. Comment B and C indicate that the sampling points were inside and outside the fallout area. Moreover, comments G, P, R, A indicate the points of granite, palaeozoic formations, rhyolitic rocks, alluvium respectively. Comment blank indicates that geological features of that point are ambiguous. The results show that the data for palaeozoic formations are different from those for granite;

Table 1. Activity ratio $^{234}U/^{238}U$ of uranium leached from soil samples using 0.1 N nitric acid

No.	Distance* (km)	Comments	$^{234}U/^{238}U$ (Ci/Ci)	No.	Distance Km	Comments	$^{234}U/^{238}U$ (Ci/Ci)
	East-Southeast				North-Westnorth		
1	4	C , G	0.97 ± 0.04	16	4	B , G	1.03 ± 0.06
2	6	C , G	0.96 ± 0.04	17	6	B , G	1.09 ± 0.05
3	10	C , G	0.98 ± 0.04	18	8	B ,	1.00 ± 0.05
4	24	C , R	1.02 ± 0.05	19	10	B , G	1.01 ± 0.05
5	26	C , R	1.11 ± 0.04	20	12	B , G	1.08 ± 0.04
	South			21	14	B ,	1.11 ± 0.07
6	8	C , G	0.97 ± 0.06	22	16	B , P	1.17 ± 0.08
	West			23	22	B , P	1.37 ± 0.07
7	4	B , G	1.09 ± 0.04	24	24	B ,	1.32 ± 0.07
8	6	B , G	0.99 ± 0.04		North		
	West-Northwest			25	3	B , A	1.30 ± 0.05
9	4	B , G	0.96 ± 0.04	26	4	B ,	1.04 ± 0.08
10	6	B , G	1.13 ± 0.04	27	8	B , G	1.06 ± 0.12
11	8	B ,	0.96 ± 0.04	28	10	B , G	1.01 ± 0.04
12	10	B ,	0.88 ± 0.05	29	14	B , P	1.29 ± 0.08
13	18	B , P	1.31 ± 0.17		East-Northeast		
14	24	C , G	0.98 ± 0.06	30	4	C , G	0.98 ± 0.04
15	26	C , P	1.42 ± 0.08	31	14	C , P	1.24 ± 0.06

*: Distance from hypocenter

namely, the $^{234}U/^{238}U$ activity ratios of the latter are about 1.0, while those of the former are about 1.3. Therefore, to study the effects of the Hiroshima bomb, we must compare the data for the same geological features. Since granite is distributed widely about Hiroshima City, we compared the data for granitic soil, inside and outside the fallout area.

Examples of the alpha ray spectrum in both areas are shown in Fig. 3. In the observed spectrum, there was no strong ^{235}U peak such as was expected from enriched uranium from the Hiroshima bomb. Although the $^{234}U/^{238}U$ activity ratios in the control area are below 1.00, the ratios in the fallout area are above 1.00. The $^{234}U/^{238}U$ activity ratios of S-uranium are plotted against the ratios of I-uranium in Fig. 4. The data show that $^{234}U/^{238}U$ activity ratios of I-uranium in both areas are below 1.00. However, many $^{234}U/^{238}U$ activity ratios of S-uranium in the fallout area are above 1.00. This may be caused by the enriched uranium, which is thought to have fallen over the fallout area. If we assume that the enriched uranium adhered to soil and a portion of this has remained, the remaining enriched uranium is calculated to be about 1% of the natural uranium contained in granite.

The $^{234}U/^{238}U$ activity ratios of S-uranium are plotted against ^{137}Cs specific activities in soil samples in Fig. 5. We used data for ^{137}Cs specific activities as measured by Hashizume *et al*[6] with a Ge(Li) detector. The error in these data are within 3%. We obtained two different lines using a least squares method for

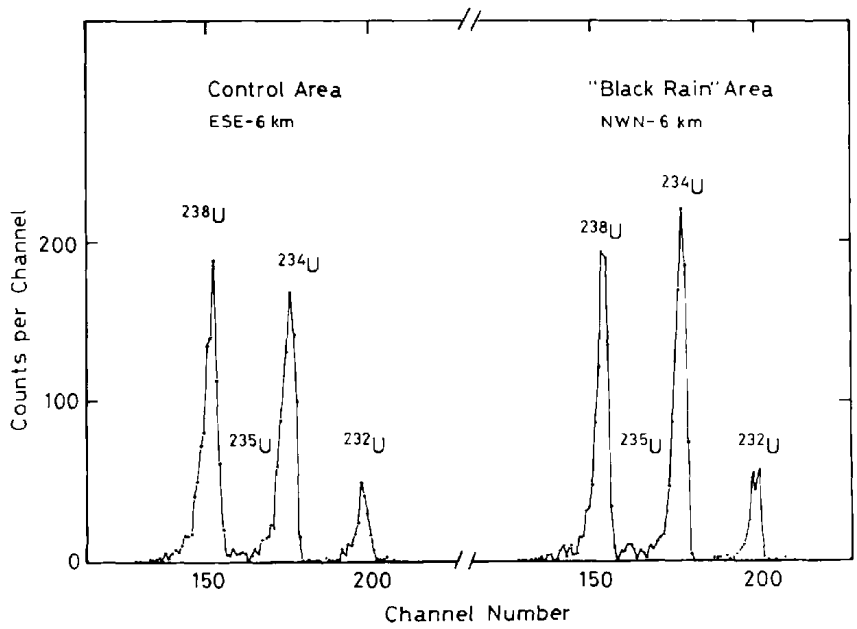

Fig. 3. Alpha ray spectra of uranium extracted from soil samples using 0.1 N nitric acid

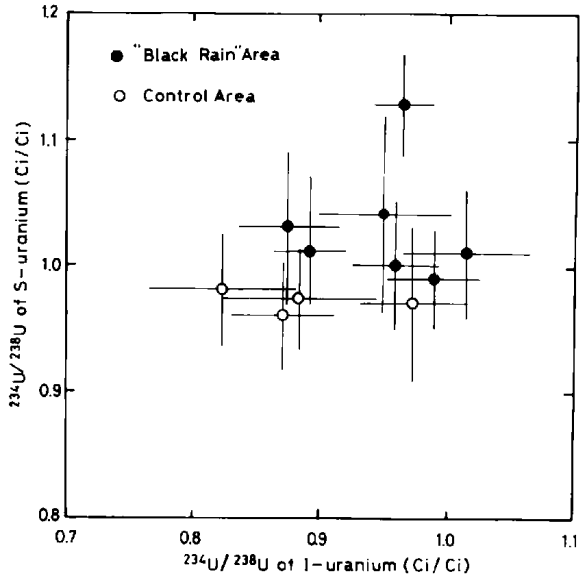

Fig. 4. Activity ratio $^{234}U/^{238}U$ of S-uranium and I-uranium.

each area's data. The $^{234}U/^{238}U$ activities ratios in the control area were nearly constant against ^{137}Cs specific activities. However, the ratios in the black rain area tended to increase proportionally with the ^{137}Cs specific activities. Although the data for the fallout area scatter around the line, the features of the data are different from those of the control area. These facts can be understood as follows.

In the control area, ^{137}Cs is largely brought by the fallout due to world-wide nuclear tests, for which enriched uranium has not always been used as the main raw materials of nuclear bombs. Therefore, the $^{234}U/^{238}U$ activity ratios are nearly constant and do not increase with the specific activity of ^{137}Cs.

On the other hand, in the "black rain" area, the ^{137}Cs fallout due to the Hiroshima atomic bomb explosion contribute considerably to the remaining ^{137}Cs in soils. Since the $^{234}U/^{238}U$ activity ratio is larger than unity in the enriched uranium used for the Hiroshima atomic bomb, the data for the "black rain" area show the increase of uranium isotope ratio according to the increase of the specific activity of ^{137}Cs.

From these results, it is suggested that the fallout nuclides from Hiroshima atomic bomb are still detectable in the "black rain" area and its fissile raw material was the enriched uranium.

ACKNOWLEDGEMENTS

We would like to thank Mr. Mikio Yanagisawa, Mr. Seiji Takeoka, Mr. Isao Okui and Mr. Tomotaka Tsujimura in our laboratories for their considerable

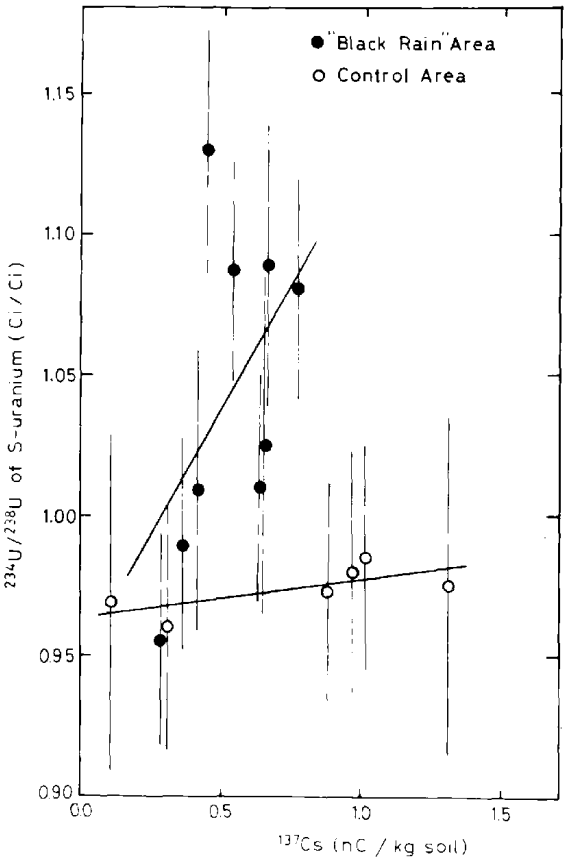

Fig. 5. Correlation of $^{234}U/^{238}U$ activity ratios and ^{137}Cs specific activity.

assistance. We also would like to thank Dr. Koreyuki Shiba, Mr. Tetsuo Shiratori of Japan Atomic Energy Research institute for telling us useful papers on enriched uranium. We are grateful to Dr. Walter J. Russell of the Radiation Effects Research Foundation for editing this manuscript. We offer this paper in honor of the decreased Dr. Kinji Takeshita who made this study possible.

REFERENCES

1. Science Council of Japan (1953) Collection of the Reports on the Investigation of the Atomic Bomb Causalties. Vol. 1. Japan Society for the Promotion of Sience, Tokyo.
a) Kimura, K. and Kamimura, U. Artificial Radioactivity found in soil Hiroshima Takasu. pp. 54-58.
b) Uda, M. et al. Meteorogical conditions related to the atomic explosion in Hiroshima. pp. 98-135.

c) Fujiwara, T. and Takeyama, H. Residual radiation around Hiroshima. pp. 75-83.
d) Miyazaki, Y. et al. A-bomb radiation in Hiroshima and vicinity (Parts 1 and 2). pp. 34-38.
2. Sakanoue, M. and Tsuji, T. (1971) Plutonium content of soil at Nagasaki. Nature 234 November 12: 92-93.
3. Arakawa, E. T. (1962) Residual radiation in Hiroshima and Nagasaki. ABCC Technical Report 2-62.
4. Pace, N. and Smith, R. E. (1959) Measurement of the residual radiation intensity at the Hiroshima and Nagasaki bomb sites. ABCC Technical Report 26-59.
5. Takeshita, K. (1975) C. Dose estimation from residual and fallout radioactivity. 1. Areal surveys. J. Radiat. Res., 16 Supplement: 24-31.
6. Hashizume, T. et al. (1977) Report of residual radioactivity surveys in Hiroshima and Nagasaki 1976, supported by the Ministry of Health and Welfare, Japan Public Health Foundation.
7. Hashizume, T. et al. (1979) Report of residual radioactivity surveys in Hiroshima and Nagasaki 1978, supported by the Ministry of Health and Welfare, Japan Public Health Foundation.
8. Hashizume, T. et al. (1978) Estimation of residual radiation from the Atomic Bombs in Hiroshima and Nagasaki. The Journal of the Hiroshima Medical Association. Vol. 31, No. 4: 455-458.
9. Auxier, J. A. et al. (1966) Free-field radiation-dose distributions from the Hiroshima and Nagasaki bombings. Health Physics 12: 425-429.
10. Cherdyntsev, V. V. (1969) Uran-234. Atomizdat Moskva. pp. 198-199.