Studies of Radioactivity Produced by the Hiroshima Atomic Bomb:

2. Measurements of Fallout Radioactivity

HIROMI HASAI*, MASAHARU HOSHI** AND KENJIRO YOKORO**

*Faculty of Engineering, Hiroshima University
Higashi-Hiroshima 724, Japan
**Research Institute for Nuclear Medicine and Biology,
Hiroshima University, Hiroshima 734, Japan

Fallout/Hiroshima atomic bomb/Fission product/Dosimetry/Internal exposure

Three studies of fallout measurements were reviewed for the discussion of possible radioactivity intake from the Hiroshima atomic bomb. The first study discussed correlations between enriched $^{234}$U and $^{137}$Cs specific activities from the measurement of soil samples collected in the "black rain" area. The second study measured $^{137}$Cs activity on the rock and roof tile samples collected in the hypocenter area immediately after the explosion. Some of the rock and roof tile samples collected near the hypocenter had a small but detectable amount of $^{137}$Cs activity. However, it has been determined that $^{137}$Cs exposure, for example, was negligible compared with DS86 dose estimates, since these activity levels were low. The third study detected $^{90}$Sr activity in some of the specimens of human bones exhumed on Ninoshima Island. This study compared the difference in activity between the bone head and shaft, with higher activities obtained in the bone head. This fact suggests a short intake period for this activity, however, the levels of $^{90}$Sr contamination were too low to allow a discussion of the exposure risks.

INTRODUCTION

New atomic bomb (A-bomb) doses for Hiroshima and Nagasaki have been proposed as the dosimetry system 1986 (DS86)\(^1\).\(^2\). Estimated DS86 organ doses have been used to evaluate radiation risks among survivors based on epidemiological data. In the DS86 study, Okajima et al.\(^3\) discussed fallout and neutron-induced radioactivity in the "black rain" and hypocenter areas, respectively. Doses from induced radioactivity in the hypocenter area were at most several tens of rads\(^3\).\(^4\). However, these doses were not included in DS86, because of the difficulties involved in making dose estimations for each survivor. There are two reasons for such difficulties: (1) to estimate external exposure from induced radioactivities, it would be necessary to check the precise history of the routes an individual followed in such an area after the A-bomb and (2) to estimate internal exposure, it is necessary to project the radioactive intake due to inhalation and injection.

Many scientists have attempted to estimate fallout radioactivity due to the Hiroshima A-bomb...
in order to discuss intakes by the A-bomb survivors or residents in such areas. Takeshita\textsuperscript{4)} summarized such studies done before 1975, and estimated cumulative doses due to induced and fallout radioactivity. Hashizume et al.\textsuperscript{5)} collected soil samples in the black rain area, which is far away from the hypocenter, and measured $^{137}$Cs levels. However, he could not verify $^{137}$Cs contamination due to the much higher levels of contamination resulting from global fallout attributable to nuclear weapons tests elsewhere in the world. These collected soil samples were measured again by Takada et al.\textsuperscript{6)}. He measured enriched $^{234}$U, which is assumed to be enriched simultaneously according to the procedure used for $^{235}$U. Yamamoto et al.\textsuperscript{7)} measured Am and Pu isotopes in these samples, but he could not detect any fallout from the Hiroshima A-bomb. Shizuma et al.\textsuperscript{8)} used rock and roof tile samples that had been collected immediately after the A-bomb and detected $^{137}$Cs in some of these samples. Hoshi et al.\textsuperscript{9)} tried a method of statistical treatment of $^{137}$Cs data obtained by Hashizume et al.\textsuperscript{5)} and discussed the possibility of $^{137}$Cs fallout in the black rain area. Kawamura et al.\textsuperscript{10)} measured $^{90}$Sr activities from human bone exhumed on Ninoshima Island and discussed the possibility of $^{90}$Sr intake.

This review summarizes recent developments in studies concerning fallout activity measurement of $^{234}$U\textsuperscript{6)} in soil samples collected in the black rain area, $^{137}$Cs\textsuperscript{8)} in rock samples collected immediately after the A-bomb and $^{90}$Sr\textsuperscript{10)} measured in human bones exhumed on Ninoshima Island. Other possible exposures resulting from these activities are also discussed. For further details, these individual papers should be consulted.

**MATERIALS AND METHODS**

**Soil samples in the black rain area**

Soil samples were obtained at distances from 2 to 30 km from the Hiroshima hypocenter in 1976\textsuperscript{5)}. Soil samples which seemed likely to have been in a stable condition since the A-bomb were selected and collected from the surface to 10 cm in depth\textsuperscript{5)}. Takada et al.\textsuperscript{6)} used the alpha-ray detecting method for their measurements because of its convenience. This method is more efficient at detecting $^{234}$U than $^{235}$U. Because $^{235}$U is a fuel component, however, $^{234}$U is also assumed to be enriched by the gas diffusion method used. For these reasons, $^{234}$U rather than $^{235}$U was measured. Soil samples were shaken in 0.1 N nitric acid for 5 hours to remove surface uranium thought to be attached to small soil particles. After this, the residue was treated with 8 N nitric acid at 120°C for 1 hour to remove any inner uranium. The former was termed "S-uranium" and the latter "I-uranium". After these chemical treatments, the uranium was extracted and alpha rays were measured using silicon surface barrier detectors\textsuperscript{6)}.

**Stone and roof tile samples from the hypocenter area**

Stone and roof tile samples exposed to the Hiroshima A-bomb were found in December 1987 in a storehouse belonging to the Faculty of Science at Hiroshima University. These samples were collected within about 1.5 km ground range from the hypocenter from October 27 to December 3, 1945\textsuperscript{8)} for analyses of the heat ray intensity of the A-bomb. A field notebook found with the samples precisely recorded the dates and locations for the samples collected\textsuperscript{8}). Thus 44 samples from 30 locations were obtained. These samples were not affected by fallout due to global
weapons tests, hence they were quite valuable for the estimation of fallout in the Hiroshima hypocenter area using $^{137}$Cs measurements. They were prepared for Ge measurement.

**Human bone samples**

A number of bones of the dead were exhumed on Ninoshima Island in 1955 and in 1971 as detailed in Kawamura et al.\textsuperscript{10}. According to a comprehensive document, approximately 10,000 injured citizens fled by boat from Port Ujina, Hiroshima, to Ninoshima Island between 6 August and 25 August. Many of the victims died there and were cremated or buried without cremation by the end of the month\textsuperscript{10}. Thus the exhumed bones were from unidentified individuals exposed to the A-bomb who took refuge and died on the island during the latter part of 1945. The bone samples measured consisted of five femurs, six tibiae, three humeri, two sets of ribs and five sets of vertebrae. From the surface condition of the bones, it was determined that the individuals had not been cremated, and the samples were ashed for measurement of $^{90}$Sr\textsuperscript{10}.

**RESULTS**

The results of the alpha-ray measurements obtained as a ratio of $^{234}$U to $^{238}$U are compared between S-uranium (surface) and I-uranium (inner). The results are shown in Fig. 1. The closed and open circles indicate samples from the black rain and control areas, respectively. Plots of S-uranium in the black rain area are slightly higher than the unity level. The ratios of $^{234}$U/$^{238}$U

![Fig. 1. Activity ratio $^{234}$U/$^{238}$U of S-Uranium and I-uranium. (From Takada et al. 1983; Ref. 6)](image-url)
were compared with $^{137}$Cs results from Hashizume et al. The relationship between the $^{234}$U/$^{238}$U ratio and the $^{137}$Cs value is shown in Fig. 2. This figure indicates an increase in the $^{234}$U/$^{238}$U ratio with increasing $^{137}$Cs activity.

From the results of the measurement of $^{137}$Cs in the hypocenter area of Hiroshima, it is found that there was very little $^{137}$Cs fallout in the hypocenter area. Only five samples in 44 had measured values higher than the detectable limit. Observed $^{137}$Cs activities in these five samples corrected for the decay of $^{137}$Cs from the time of the A-bomb were <1.3 mBq·cm$^{-2}$. These results are shown in Table 1. Table 2 indicates external exposures estimated by Shizuma et al. and compares their values with other estimates.

Table 3 shows five examples of $^{90}$Sr concentration data in human bone samples exhumed on Ninoshima Island. These bone samples were from individuals who died during a period

Fig. 2. Correlation of $^{234}$U/$^{238}$U activity ratios and $^{137}$Cs specific activity. (From Takada et al. 1983; Ref. 6)
Table 1. Cesium-137 concentration in samples collected near the hypocenter area of the Hiroshima atomic bomb. Samples were collected from October to December 1945. Tabulated values are decay-corrected to the time of the bomb. (From Shizuma et al. 1989; Ref. 8)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Location</th>
<th>Material</th>
<th>Ground range (m)</th>
<th>$^{137}$Cs (mBq·cm$^{-2}$)</th>
<th>Detection limit (mBq·cm$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ginkokyokai</td>
<td>C</td>
<td>A</td>
<td>$178 \pm 23$</td>
<td>$0.48 \pm 0.25$</td>
<td>0.31</td>
</tr>
<tr>
<td>Yasuda seimei</td>
<td>C</td>
<td></td>
<td>$257 \pm 23$</td>
<td>$0.31 \pm 0.20$</td>
<td>0.25</td>
</tr>
<tr>
<td>Sei Hospital</td>
<td>R</td>
<td>B</td>
<td>$55 \pm 29$</td>
<td>$0.32 \pm 0.21$</td>
<td>0.27</td>
</tr>
<tr>
<td>Kaminaka</td>
<td>R</td>
<td></td>
<td>$253 \pm 29$</td>
<td>$0.45 \pm 0.21$</td>
<td>0.28</td>
</tr>
<tr>
<td>Sorazawa shrine</td>
<td>R</td>
<td></td>
<td>$584 \pm 29$</td>
<td>$0.43 \pm 0.23$</td>
<td>0.28</td>
</tr>
<tr>
<td>Upper limit</td>
<td></td>
<td></td>
<td></td>
<td>$0.48 \pm 0.25$</td>
<td>(1.3 ± 0.7)$^c$</td>
</tr>
</tbody>
</table>

$^a$Concrete.
$^b$Rooffile.
$^c$Immediately after the fallout.

Table 2. Estimates of cumulative external exposure in the hypocenter and fallout areas. The unit of the numbers in this table is C/kg. (From Shizuma et al. 1989; Ref. 8)

<table>
<thead>
<tr>
<th>source of radiation</th>
<th>Hiroshima</th>
<th>Nagasaki</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Hypocenter</td>
<td>Koi-takesu</td>
</tr>
<tr>
<td>Neutron-induced activity</td>
<td>21$^a$</td>
<td>0</td>
</tr>
<tr>
<td>(80 R)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fallout</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Radiation rate</td>
<td></td>
<td>0.26–0.77$^a$</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>$\lesssim 0.026 \pm 0.014$$^b$</td>
<td>N$^c$</td>
</tr>
<tr>
<td>( $\lesssim 0.010 \pm 0.05$ R)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$Okajima et al. (1987)$^3$.
$^b$Shizuma et al. (1989)$^8$.
$^c$No measurement.
approximately three weeks after the detonation on August 6. Not all of the samples showed $^{90}\text{Sr}$ contamination exceeding the detection limit (Table 3). Three samples which had levels of contamination greater than the detection limit also show differences between levels in the bone heads and shafts. This difference suggests an intake of $^{90}\text{Sr}$ resulting from the Hiroshima A-bomb\textsuperscript{10}.

### DISCUSSION

The $^{234}\text{U}/^{238}\text{U}$ activity ratios of S-uranium are plotted against $^{137}\text{Cs}$ specific activities in soil samples as shown in Fig. 2. In this figure, data for $^{137}\text{Cs}$ specific activities as measured by Hashizume et al.\textsuperscript{5} with a Ge(Li) detector were used. Takada et al.\textsuperscript{6} obtained two different lines as drawn in this figure. The $^{234}\text{U}/^{238}\text{U}$ ratios in the control area were nearly constant against $^{137}\text{Cs}$ specific activities. However, the ratios in the black rain area tended to increase proportionally with the $^{137}\text{Cs}$ activities. Although the data for the fallout area scatter around the line, the features of the data are different from those from the control area. These facts can be understood as follows.

In the control area, the $^{137}\text{Cs}$ is largely due to fallout from nuclear tests worldwide, and
enriched uranium has not always been the main raw material used in nuclear weapons. Therefore, the $^{234}\text{U}/^{238}\text{U}$ activity ratios are nearly constant and do not increase with the specific activity of $^{137}\text{Cs}$. On the other hand, in the black rain area, the $^{137}\text{Cs}$ fallout due to the Hiroshima A-bomb explosion contributed the remaining $^{137}\text{Cs}$ found in the soil. Since the $^{234}\text{U}/^{238}\text{U}$ activity ratio was larger than unity in the enriched uranium used for the Hiroshima A-bomb, the data for the black rain area show an increasing uranium isotope ratio in relation to the increasing specific activity of $^{137}\text{Cs}$. These results suggest that fallout nuclides from the Hiroshima A-bomb are still detectable in the black rain area and that its fissile raw material was enriched uranium.

The $^{137}\text{Cs}$ data obtained from the measurements of the 44 stones and roof tiles were almost below the detection limit of the germanium detector. Only five values exceeded the detection limit. This fact means that almost all fission products fell outside of the hypocenter area.

To estimate the upper limit of the cumulative exposure due to this fallout, upper limits for these data were assumed from the data shown in Table 1. The cumulative exposure estimated from this upper limit was $0.026 \pm 0.014$ mC$\cdot$kg$^{-1}$ ($0.10 \pm 0.05$ R), based on a conversion factor of $^{137}\text{Cs}$ concentration to exposure of $3.7 \times 10^7$ Bq$\cdot$km$^{-2}$ = $0.077$ mC$\cdot$kg$^{-1}$ (1 mCi$\cdot$km$^{-2}$ = 0.3 R) as discussed by Shizuma et al.$^9$ This calculation and comparisons with other estimates are indicated in Table 2. The estimate of the cumulative exposure in the hypocenter area corresponds to $<0.13\%$ of that from the induced radioactivity in the hypocenter area and $<10\%$ of that in Koi Takasu district.$^3$ This result indicates that fallout contributed very little to the radiation exposure experienced in the hypocenter area of Hiroshima. This data also corresponds to $<4\%$ of the $^{137}\text{Cs}$ deposition due to fallout from atmospheric nuclear testing.$^8$ Thus, it is hard to detect excess amounts of $^{137}\text{Cs}$ deposition from later samples. Only the samples collected before the global fallout began enable us to determine $^{137}\text{Cs}$ deposition levels.

$^{90}\text{Sr}$ deposition data (Table 3) showed different intensities between the head and shaft in two tibial and three femoral samples. In order to determine whether the detected $^{90}\text{Sr}$ bone activity was incorporated before death and to allow a discussion of whether the activity was incorporated over a short period or a single instance, it is useful to measure the intra-bone distribution of $^{90}\text{Sr}$. Experimental animal data show that administered alkaline earths including $^{90}\text{Sr}$ are concentrated in active growth sites of bone, but that the distribution tends to be more even with a continuous uptake.$^{11,12}$ The pattern of intra-bone distribution of $^{90}\text{Sr}$ presently detected in the proximal half of one tibia and two femurs appears consistent with a single instance or relatively short period of intake of the radionuclide.

Although the contamination levels are too small to allow a discussion of radiation risks due to internal exposure$^{10}$, it is possible that the $^{90}\text{Sr}$ activity in the exhumed bone samples might have been internally deposited via inhalation of fission debris and/or ingestion of contaminated water$^{10}$.

Experiments on the detection of fallout and induced radioactivity are proceeding. A large-scale study of the epidemiological effects of internal exposure has not yet been made on the same scale as those for A-bomb survivors in Hiroshima and Nagasaki, and the risks of such internal exposure have not yet been confirmed by the epidemiological method. As the present study reports measurements of the intake of such radioactivity, even if it is negligibly small, is necessary to allow us to estimate or dismiss any risks. Large-scale epidemiological studies related to the
Chernobyl nuclear accident are proceeding. If a relationship between fallout and risks of internal exposure is obtained in the Chernobyl studies, these fallout measurements will be used to estimate risks for A-bomb survivors in Hiroshima, risks which are not included in DS86.

REFERENCES