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# Distribution of Plutonium Isotopes and $^{137}Cs$ found in the Surface Soils of Nagasaki, Japan

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**Abstract**

The $^{240}Pu/^{239}Pu$ atom ratio and the concentrations of $^{239+240}Pu$ and $^{137}Cs$ were measured in soil samples collected within a 10 km radius of the hypocenter of the atomic bomb that was detonated over Nagasaki in 1945. It was found that samples collected in the eastern area of the hypocenter yielded data affected more by this bomb than those of other areas.

The $^{240}Pu/^{239}Pu$ atom ratio in this eastern area, especially in the area of Nishiyama (about 3 km east of the hypocenter), was lower than that of global fallout (0.176). This finding clearly indicated that the plutonium was originated from the atomic bomb exploded over Nagasaki. In the eastern area, it was found that, although the $^{240}Pu/^{239}Pu$ atom ratios were found to increase with distance from Nishiyama area, the atom ratio in the sample collected as far as 8 km east from the hypocenter was lower than that of global fallout. This indicated that the plutonium from the atomic bomb was dispersed in the eastern area. We also found that the atom ratio is more effective in identifying the origin of plutonium, than the method which seeks to understand the activity ratio and concentrations of $^{239+240}Pu$ and $^{137}Cs$.

**Key-words** : Plutonium, atom ratio, $^{137}Cs$, atomic bomb, Nagasaki

## 1. Introduction

The plutonium atomic bomb exploded above Nagasaki city on the 9th of August 1945. Following the explosion, plutonium and the fission products were dispersed over the area of Nagasaki.

Identification of the radionuclides deposited over this area provides important information on the effects of the atomic bomb explosion, for example the radiation dose to the population. Sakanoue and Tsuji (1971) measured $^{239+240}Pu$ concentrations in soil samples collected from northern, eastern and southern areas within 4 km of the hypocenter. It has been found that samples taken from the Nishiyama area (about 3 km east of the hypocenter) had the highest concentration. Following these findings, investigations were expanded over a wider area. Yamamoto et al. (1985) made determinations in soil samples collected from up to 30 km from the hypocenter and Kudo et al. (1991) studied soil samples collected within 100 km east and 3 km west of the hypocenter. Both these studies confirmed high concentrations of $^{239+240}Pu$ in the area of Nishiyama.

Since 1945, more than 500 of nuclear tests have been carried out worldwide, resulting in additional plutonium inventory as global fallout. Thus plutonium in Nagasaki area soil is a mixture of the plutonium from the Nagasaki atomic bomb and that from the global fallout. The values of $^{239+240}Pu/^{137}Cs$ and $^{238}Pu/^{239+240}$ Pu activity ratios were generally used to discriminate Pu that came from the Nagasaki atomic explosion and that which came from global fallout. However it has been realized that the $^{239+240}Pu/^{137}Cs$ activity ratio is affected by the composition

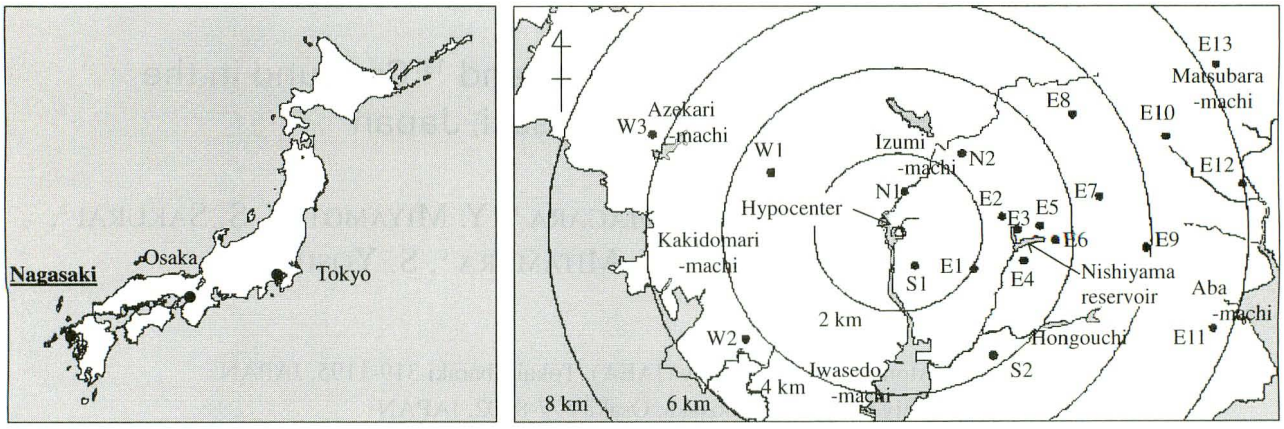


Fig. 1 Location of the sampling points in Nagasaki area.

of soil and the migration of these elements in the soil over a long period of time. (Mahara et al., 1984, Mahara et al., 1995).

The activity ratio of $^{238}Pu/^{239+240}Pu$ was reported to be 0.05 − 0.06 in the Nishiyama area (Yamamoto et al., 1985), and 0.02 − 0.04 for global fallout, that is, radionuclides released from nuclear tests into the air since 1945 and have spread worldwide (Hardy et al., 1973). The difficulty of distinguishing the difference of these activity ratios has been recognized in its experimental reliability by $\alpha$ - spectrometry in case of measuring sample which have low plutonium concentrations. Thus the $^{238}Pu/^{239+240}Pu$ activity ratio is not effective in the clarification of plutonium origin.

Inductively coupled plasma mass spectrometry (ICP-MS) has been more recently used for the determination of $^{240}Pu/^{239}Pu$ atom ratio in the environmental samples (Yamamoto et al., 1996, Boulyga et al., 2002). It has been reported that the $^{240}Pu/^{239}Pu$ atom ratio in weapons-grade plutonium is approximately 0.05 (Tayor et al., 2001), while that obtained from the global fallout of nuclear testing is 0.176 (Krey et al., 1976). Therefore, $^{240}Pu/^{239}Pu$ atom ratio is a useful tool for the identification of the source of plutonium.

In a previous paper, we reported the $^{240}Pu/^{239}Pu$ atom ratio taken from samples in the sediment of the Nishiyama reservoir. Based on the average measured value of 0.03, it has been concluded that plutonium isotopes in the sediment originated from the Nagasaki atomic bomb explosion (Saito et al., 2004). To date, no extensive data has been reported on the $^{240}Pu/^{239}Pu$ atom ratio in the soil around the hypocenter at Nagasaki, except three separate reports on soil samples taken from a limited area surrounding Nishiyama. (Komura et al., 1984, Yamamoto et al., 1985, Muramatsu et al., 2003).

In 2002, Noritake et al. collected surface soils within 10 km of the hypocenter and reported the distribution of $^{137}Cs$ (Noritake et al., 2002). Higher concentrations were found in the areas east and northeast of the hypocenter.

In this work, we report the $^{240}Pu/^{239}Pu$ atom ratio measured in the surface soil samples collected by Noritake and the authors within 10 km of the hypocenter. These data were applied to identify the detailed geological distribution of plutonium released from the Nagasaki atomic bomb.

## 2. Experiment

Two sets of surface soil samples were used in the experiments: (1) fifteen samples (E1, E7 − E13, N1, N2, W1 − W3, S1, S2) collected by Noritake within a 10 km radius of the hypocenter in 2002 (Noritake et al., 2002) and (2) five samples (E2 − E6) collected by the authors around the Nishiyama reservoir in 1999. Figure 1 shows the location of sampling points. First set of samples was collected from flatland that has not been disturbed by human activity or natural processes since the detonation. The samples were taken at depths from several to 20 cm below the surface level. The soils were of sandy silt and clay with dark gray or dark brown organic matter inclusions. After drying at 110 ℃ for 24 hours, these samples were sieved through a 2 mm mesh. Second set of samples was collected at depths between 2 and 5 cm, from forest land whose soil was mostly silt and clay. After drying until constant weight at 70 ℃, large sand particles and fragments of plants were removed. These soils then were ground with a pestle and mortar

### 2.1 Determination of $^{137}Cs$ by gamma-ray spectrometry

The activities of $^{137}Cs$ were measured with high-purity

Ge detectors (Model LOAX60450/30P or GWL-220230-S both from ORTEC, US) using about 1 - 2 g of the soil sample. The counting efficiency of $^{137}$Cs was evaluated using two certified reference materials (SRM4350B, SRM4354, National Institute of Standard and Technologies, US). All measured $^{137}$Cs activities were decay corrected to the sampling date.

### 2.2 Determination of $^{239+240}$Pu concentration and $^{240}$Pu/$^{239}$Pu atom ratio by ICP-MS

The concentration of $^{239+240}$Pu and the $^{240}$Pu/$^{239}$Pu atom ratio were measured for both sets of samples. A $^{242}$Pu spike (4 pg) was added into a dried sample (1 − 2 g). This was then placed in a Teflon beaker, after 8M $HNO_3$ (about 6 ml) were added, the mixture was heated on a hot plate to leach plutonium. The leaching procedure was repeated three times using a fresh portion of the acid (Muramatsu et al., 1999). After separating plutonium by using an anion exchange resin, concentration of $^{239+240}$Pu and $^{240}$Pu/$^{239}$Pu atom ratio were measured by use of double-focusing inductively coupled mass spectrometer (ELEMENT, Thermo Electron, UK). Details of the procedure were described elsewhere (Saito et al., 2004).

## 3. Results and Discussion

The measured values of the $^{240}$Pu/$^{239}$Pu atom ratio and the concentrations of $^{239+240}$Pu and $^{137}$Cs in soil samples, together with the concentrations of $^{137}$Cs measured by Noritake et al. (Noritake et al., 2002), are given in Table 1. The ratios were found to be between 0.032 and 0.22, while concentrations of $^{239+240}$Pu and $^{137}$Cs were 0.03 − 23 Bq/kg and from below 1.9 to 84 Bq/kg, respectively.

Figure 2 shows the $^{240}$Pu/$^{239}$Pu atom ratios at the sampling points. The ratios in the soils of the area east of the hypocenter (E1 − E12 except E8 and E11) were significantly lower than that of global fallout (0.176). The mean value of the ratios in the soils around the Nishiyama reservoir (E1 − E7 and E9) was 0.038 ± 0.007. This value is close to 0.042 ± 0.014, 0.020 − 0.034 and 0.037 ± 0.002 reported by Komura et al. (1984), Yamamoto et al. (1985) and Muramatsu et al. (2003), respectively, for soils in the Nishiyama area. The $^{240}$Pu/$^{239}$Pu atom ratio in weapons-grade plutonium before 1960 was approximately 0.01 (Rokop et al., 1996). The $^{240}$Pu/$^{239}$Pu atom ratio in plutonium released from the atomic bomb after the detonation became higher due to the production of $^{240}$Pu during the detonation. It is suggested, therefore, that plutonium which had a significantly lower atom ratio than that of global fallout was derived from the Nagasaki atomic bomb. The atom ratios in the soils were almost constant up to the distance of 6 km (0.03 − 0.04), and then increased with the distance. Even at 8 km from the hypocenter, the $^{240}$Pu/$^{239}$Pu atom ratio was lower than the global fallout value. This indicates that the plutonium from the atomic bomb was dispersed still further away. Obviously, it is necessary to analyze samples from areas further east to

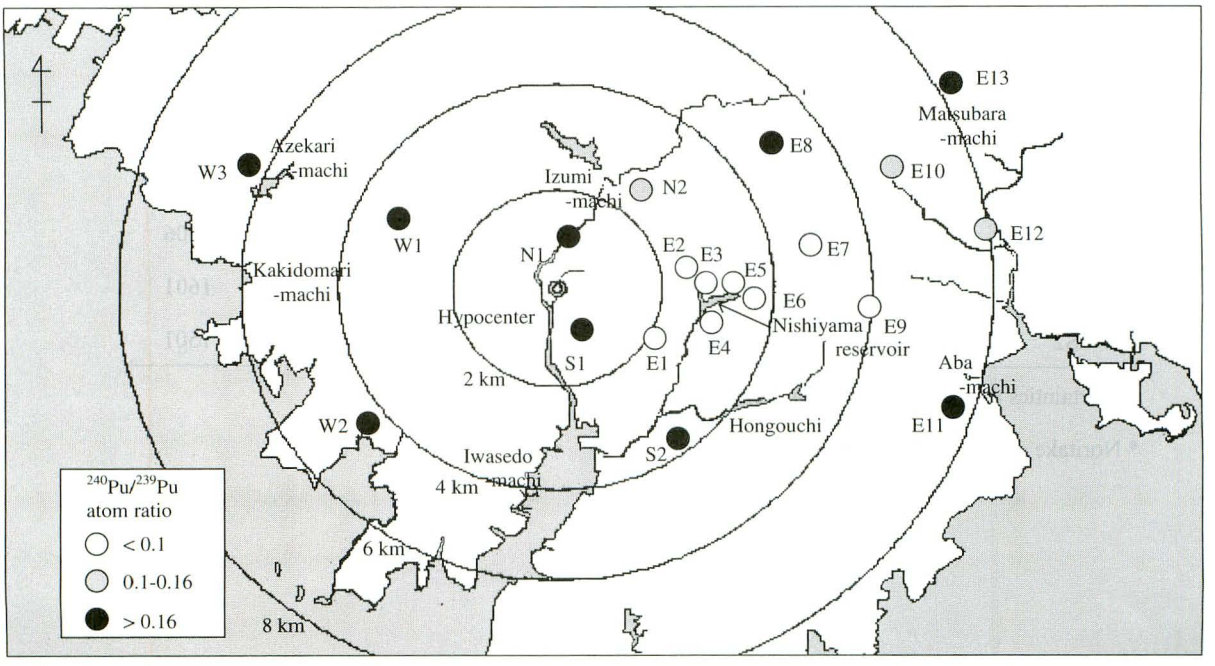


Fig. 2 Distribution of $^{240}$Pu/$^{239}$Pu atom ratio in soils collected in Nagasaki area.

Table 1 Concentrations of $^{239+240}Pu$ and $^{137}Cs$, and $^{240}Pu/^{239}Pu$ atom ratio in soils collected within 10 km of the hypocenter

| Location Site No. | Distance from hypocenter (m) | $^{239+240}Pu$ (Bq/kg dry) | $^{137}Cs$ (Bq/kg dry) | $^{240}Pu/^{239}Pu$ atom ratio | Sample No.* |
|---|---|---|---|---|---|
| E1 | 1975 | 1.3 ± 0.2 | 29 ± 1* | 0.049 ± 0.005 | 1510 |
| E2 | 2340 | 22 ± 4 | 73 ± 2 | 0.032 ± 0.003 | |
| E3 | 2760 | 16 ± 3 | 32 ± 2 | 0.032 ± 0.001 | |
| E4 | 3025 | 2.0 ± 0.4 | 13 ± 1 | 0.039 ± 0.009 | |
| E5 | 3300 | 23 ± 4 | 50 ± 2 | 0.033 ± 0.001 | |
| E6 | 3625 | 4.2 ± 0.7 | 15 ± 1 | 0.033 ± 0.001 | |
| E7 | 4700 | 9.8 ± 2 | 84 ± 2* | 0.046 ± 0.002 | 1405 |
| E8 | 4850 | 1.0 ± 0.2 | 43 ± 2* | 0.17 ± 0.02 | 1409 |
| E9 | 5650 | 1.9 ± 0.3 | 12 ± 1* | 0.042 ± 0.004 | 1404b |
| E10 | 6225 | 0.50 ± 0.09 | 16 ± 1* | 0.11 ± 0.02 | 1407 |
| E11 | 7550 | 0.21 ± 0.04 | 10 ± 1* | 0.17 ± 0.05 | 1402 |
| E12 | 7925 | 1.7 ± 0.3 | 78 ± 2* | 0.13 ± 0.01 | 1401 |
| E13 | 8175 | 1.5 ± 0.2 | 60 ± 2* | 0.17 ± 0.02 | 1403 |
| N1 | 900 | 0.025 ± 0.006 | < 1.9** | 0.18 ± 0.08 | 1603 |
| N2 | 2375 | 0.46 ± 0.07 | 18 ± 1* | 0.14 ± 0.02 | 1410 |
| W1 | 3275 | 0.13 ± 0.03 | 7 ± 1* | 0.17 ± 0.05 | 1604 |
| W2 | 4300 | 0.60 ± 0.09 | 23 ± 1* | 0.17 ± 0.01 | 1514 |
| W3 | 6100 | 0.12 ± 0.03 | 4 ± 1* | 0.22 ± 0.07 | 1606 |
| S1 | 850 | 0.12 ± 0.03 | 6 ± 1* | 0.17 ± 0.06 | 1601 |
| S2 | 3675 | 0.9 ± 0.1 | 28 ± 1* | 0.17 ± 0.01 | 1501 |

Uncertainties on the measured data are 2 standard deviation.

* Noritake et al., 2002. **Yamazaki, 2005.

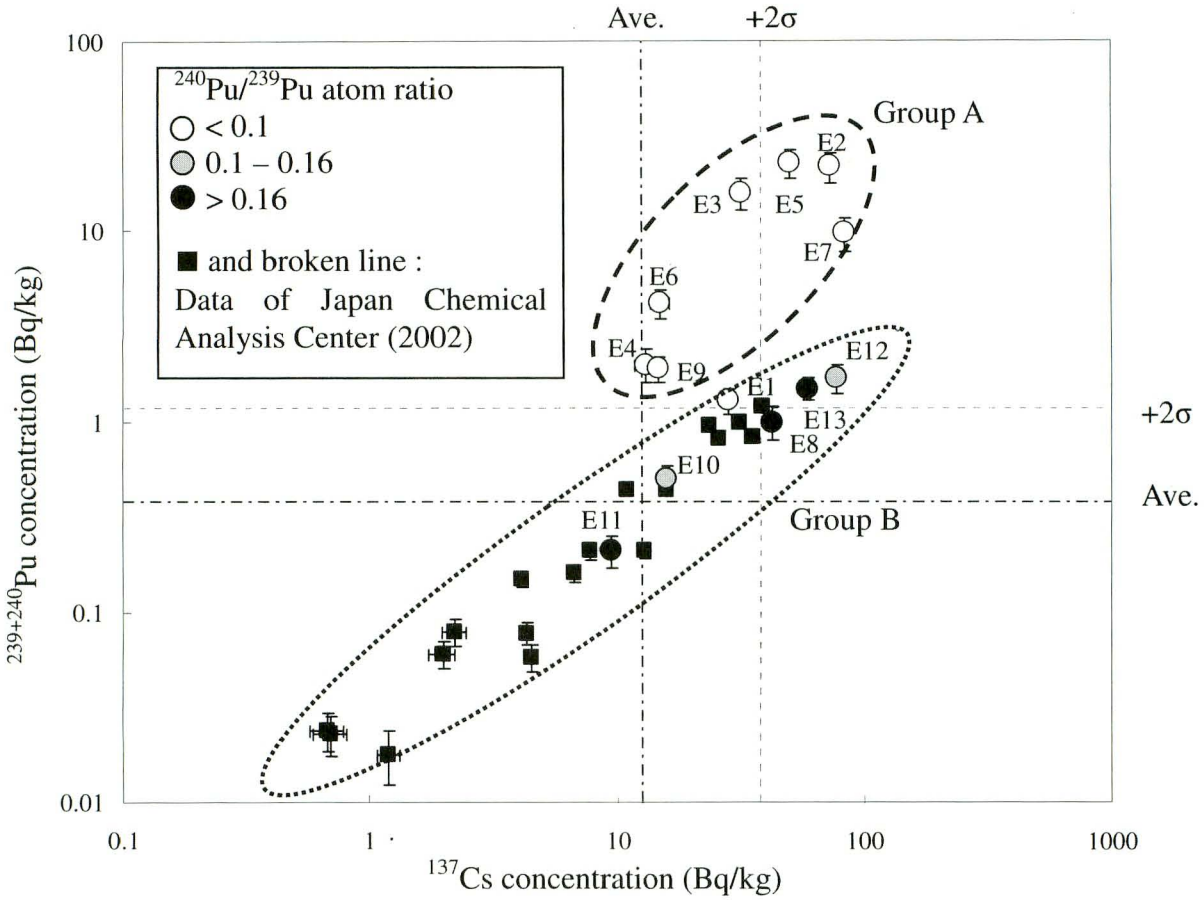


Fig. 3 Correlation of $^{137}Cs$ concentration to $^{239+240}Pu$ concentration in samples collected at the eastern area. (error: 2 $\sigma$ )

identify more precisely the extent of the area which plutonium released from the atomic bomb was deposited. On the other hand, the atom ratio which was found in soil samples taken from around the hypocenter and in the northern, western and southern areas of the hypocenter ranged from 0.14 to 0.22 close to the corresponding global fallout value (0.176). The results of this work, therefore, indicate that the plutonium from the atomic bomb was dispersed over the eastern area and then deposited in the soils there, especially in the area of Nishiyama.

Figure 3 shows correlation between concentrations of $^{239+240}Pu$ and $^{137}Cs$ in the samples collected at the eastern area. Besides our data, the concentrations and the average values for 18 soil samples taken from uncultivated lands at the depth of 0 − 5 cm all over Japan (Japan Chemical Analysis Center, 2002) are also plotted. The mean activity ratio of $^{239+240}Pu/^{137}Cs$ in the soils was 0.028 ± 0.08 (± SD) corresponding to a typical global fallout level of 0.031 (decay corrected to 2002) reported by Hardy (Hardy et al., 1973). The data were divided into two groups. Seven samples, E2 - E7 and E9, form 'Group A' and the rest of the samples form 'Group B'. Statistical analysis was carried out at 95% significance level to investigate the differences between variables of the study groups. Statistical significance was concluded when p-value was < 0.05. With Welch's t-test (p=0.0056), significant difference was found in the $^{239+240}Pu/^{137}Cs$ activity ratios of the studied groups. The $^{240}Pu/^{239}Pu$ atom ratios in the samples of 'Group A' were about one-fifth as low as that in the global fallout. Therefore, the $^{239+240}Pu/^{137}Cs$ activity ratio in the samples also indicates that the origin of plutonium is the Nagasaki atomic bomb. The $^{239+240}Pu/^{137}Cs$ activity ratios (0.022 − 0.045) in other samples of 'Group B'corresponded to those in the soils collected in the other areas of Japan by Japan Chemical Analysis Center. However, samples E1, E10 and E12 had lower $^{240}Pu/^{239}Pu$ atom ratios. If the $^{239+240}Pu/^{137}Cs$ activity ratio alone is used to identify the origin of plutonium, then it is concluded that the radionuclides in the samples of 'Group B' are originated from global fallout. However, the $^{240}Pu/^{239}Pu$

atom ratio in these samples is lower than that of global fallout. It clearly indicates that the soils contain plutonium isotopes from the Nagasaki atomic bomb. Thus, the $^{240}Pu/^{239}Pu$ atom ratio is more effective in the identification of plutonium origin than the $^{239+240}Pu/^{137}Cs$ activity ratio or the concentrations of $^{239+240}Pu$ and $^{137}Cs$ itself.

## 4. Conclusions

We obtained data of $^{240}Pu/^{239}Pu$ atom ratio and $^{239+240}Pu$ and $^{137}Cs$ concentrations in surface soils collected within 10 km from the hypocenter of atomic bomb explosion at Nagasaki. Lower $^{240}Pu/^{239}Pu$ atom ratio than that of global fallout was found in the area east of the hypocenter, especially in the Nishiyama area. This indicates that plutonium was originated from the atomic bomb explosion. Even if the $^{240}Pu/^{239}Pu$ atom ratio increased with distance from the hypocenter in the eastern area,. it was still lower than that of global fallout found at 8 km from the hypocenter. Based on these findings it was concluded that the plutonium from the atomic bomb was dispersed further away.

The $^{239+240}Pu/^{137}Cs$ activity ratios in samples with low $^{240}Pu/^{239}Pu$ atom ratios were comparable to the values measured in soils collected from different areas of Japan. The $^{240}Pu/^{239}Pu$ atom ratio is a more powerful parameter that can be used to identify the origin of plutonium than the $^{239+240}Pu/^{137}Cs$ activity ratio and/or the concentrations of $^{239+240}Pu$ and $^{137}Cs$ alone.

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