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Plutonium isotopes derived from Nagasaki atomic bomb in the sediment of Nishiyama reservoir at Nagasaki, Japan

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Abstract

The source of plutonium in sediments deposited at Nishiyama reservoir at Nagasaki was characterized by their $^{240}Pu/^{239}Pu$ atom ratio. The average ratio was approximately 0.03, except in two layers. The main source of the plutonium was the Nagasaki atomic bomb. The plutonium continues to flow into the reservoir even now. The $^{240}Pu/^{239}Pu$ atom ratios in two layers were higher than the average, which showed that plutonium in these layers were made of those of nuclear tests added to those of the atomic bomb.

Keywords: Plutonium; Nagasaki; Atomic bomb; Isotopic ratio; Sediment

1. Introduction

Plutonium isotopes and ^{137}Cs are artificial nuclides and have long half-lives (^{239}Pu: 2.411×10^4 yr, ^{240}Pu: 6.564×10^3 yr, ^{137}Cs: 30.07 yr). When these nuclides are released, these are kept in the environment. Their environmental behavior is, therefore, of interest from the viewpoint of radioecology. Nagasaki city is one of the oldest areas where plutonium isotopes and ^{137}Cs were released when the plutonium atomic bomb was exploded on August 9, 1945.

Sakanoue (1987) and Mahara et al. (1988) found high levels of $^{239+240}Pu$ in sediments of Nishiyama reservoir at Nagasaki (Fig. 1(a)), about 3 km east of the hypocenter, which experienced the 'black rain' formed from the detonation of Nagasaki atomic bomb. They reported that the plutonium isotopes were derived from the atomic bomb. However nuclear tests conducted after the detonation of the atomic bomb scattered plutonium isotopes and fission products in the world. The nuclides fell in the reservoir and were added into the sediment. Thus, the concentrations in the sediments cannot identify whether the origin of the nuclides is the atomic bomb or nuclear tests.

As the isotopic composition of plutonium depends on its origin, isotopic ratios, especially $^{240}Pu/^{239}Pu$, are used to understand the origin of plutonium. The $^{240}Pu/^{239}Pu$ ratios in source materials of nuclear weapons are different according to the date when the weapons were made. The ratio in the fallout depends on the design of the weapons and the scale of explosion; the ratios during the nuclear tests ranged from 0.09–0.34 (Koide et al., 1985). At present, the integrated ratio in the fallout is 0.176 (Krey et al., 1976). The determination of the ratio would be a powerful tool for understanding sources of plutonium in the sediments.

This paper is intended to report sources of plutonium isotopes in the sediment of the Nishiyama reservoir. We determined $^{240}Pu/^{239}Pu$ ratio in core sediments and also measured concentrations of $^{239+240}Pu$ and ^{137}Cs. These

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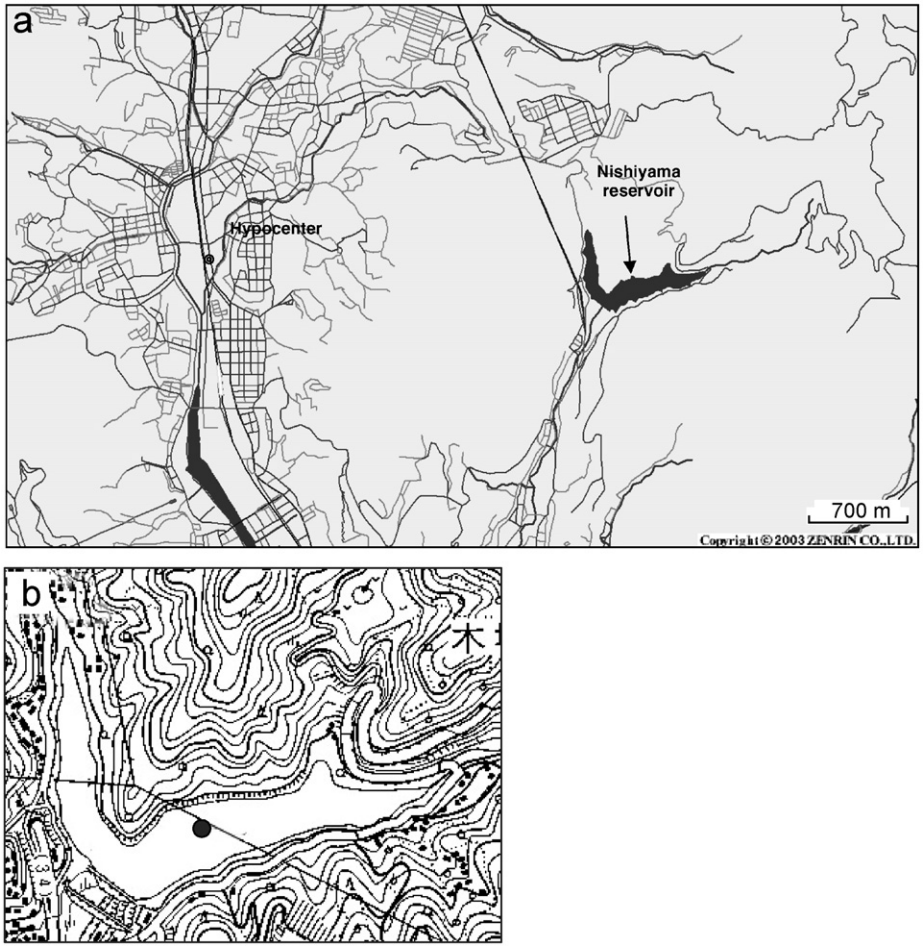

Fig. 1. Sampling point in Nishiyama reservoir at Nagasaki, Japan: (a) hypocenter of the Nagasaki atomic bomb and Nishiyama reservoir, (b) sampling point of core in Nishiyama reservoir.

results make it possible to determine the depth profile of these nuclides released from the atomic bomb.

2. Experiment

2.1. Samples

Sediment core was collected with a Mackereth-type core sampler at Nishiyama reservoir as shown in Fig. 1(b) in December 1999. After observation of the horizon in the core, the core was cut into slices 2 cm each in thickness. The sediment was dried and homogenized.

2.2. Determination of ^{137}Cs by γ-spectrometry

The sediment samples (20 g) were analyzed for their ^{137}Cs content by using HP–Ge detector (LOAX60450/30P, Ortec, USA), the counting efficiency of which were calibrated by measuring two standard reference materials (NIST SRM4350B: River sediment, SRM 4354: Freshwater lake sediment). The measured activity of ^{137}Cs in the sediments samples was decay corrected to the sampling date (December 20, 1999).

2.3. Chemical separation of Pu

Before ICP-MS measurements, plutonium was chemically isolated from soil matrix. The chemical separation and the measurement by ICP-MS were operated in the clean rooms (ISO class 5 and 6, respectively) of the CLEAR in JAEA. This facility has the ability to measure the isotopic ratio of trace amounts of uranium and plutonium in environmental samples (Magara et al., 2000). The procedure used for Pu leaching was based on the method reported by Muramatsu et al. (1999). The supernatants resulting from four steps of leaching with 8 M HNO_3 were dried to salt and then the salt was dissolved in 3 ml of 8 M HNO_3, which was followed by filtration through a Teflon membrane filter (pore size: 0.45 μm, DISMIC, Advantec, Japan) to remove any residue. The beaker was rinsed twice with 1 ml of 8 M HNO_3. All the three filtrates were mixed and sodium nitrite (200 mg) was added. The solution was left overnight to adjust Pu to the tetravalent state.

Plutonium purification was carried out with anion-exchange resin (75–150 μm, MCI GEL CA08P, Mitsubishi Chemical, Japan) in a 1 ml column. After resin was pre-treated with