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Temporal variation of $^{240}Pu/^{239}Pu$ atom ratio and $^{239+240}Pu$ inventory in water columns of the Japan Sea

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ABSTRACT

The $^{239+240}Pu$ concentrations and $^{240}Pu/^{239}Pu$ atom ratios were determined by alpha spectrometry and double-focusing SF-ICP-MS for seawater samples obtained in 1984 and 1993 from the Yamato and Tsushima Basins of the Japan Sea in the western North Pacific margin. The total $^{239+240}Pu$ inventories in the whole water columns were approximately doubled during the period from 1984 to 1993 in the two basins. The increasing rates were estimated to be 5.1 Bq m^{-2} yr^{-1} in the Yamato Basin and 4.2 Bq m^{-2} yr^{-1} in the Tsushima Basin and they corresponded to ~0.02% of the annual $^{239+240}Pu$ inflow rate into the Japan Sea through the Tsushima Strait. The mean $^{240}Pu/^{239}Pu$ atom ratios were ~0.240 and significantly higher than the mean global fallout ratio of 0.18. Furthermore, there were no temporal or spatial variations of $^{240}Pu/^{239}Pu$ atom ratios during this period in the Japan Sea. The total $^{239+240}Pu$ inventories originating from the close-in fallout increased from 17.6 Bq m^{-2} to 34.6 Bq m^{-2} in the Yamato Basin and from 20.1 Bq m^{-2} to 34.6 Bq m^{-2} in the Tsushima Basin; however, the relative percentage of ~40% from the close-in fallout was unchanged during this period. A likely mechanism for the increasing Pu inventory would be the continuous inflow of the Tsushima Current from the western North Pacific, and the removal of Pu from surface waters by scavenging onto the settling particles, followed by regeneration of Pu from the settling particles during the downward transport.

1. Introduction

The Japan Sea is a typical marginal sea located in the North Pacific Ocean with a surface area of 1.0×10^6 km^2 and it is connected to the western North Pacific Ocean and the Sea of Okhotsk by four shallow straits with a maximum depth of less than 130 m (Fig. 1). The seawater inflow to the Japan Sea is mainly by the Tsushima Warm Current, which is a branch of the Kuroshio Current, from the western North Pacific Ocean through the Tsushima Strait. The deep water of the Japan Sea is completely isolated from the Pacific deep water and there is no inflow of deep water from the surrounding seas to the Japan Sea. Its water mass below 300 m depth, which is called the Japan Sea Proper Water, is characterized by low and homogenous salinity (34.0–34.1), low temperature (0.0–1.0 °C) and highly dissolved oxygen concentration (220–230 μmol kg^{-1}); the deep water below 1000 m depth is separated into the Deep Water and the Bottom Water by a transition layer at 2000 m depth (Gamo and Horibe, 1983). The turnover time of the Japan Sea deep water has been estimated to be ca. 100 yr (Watanabe et al., 1991; Tsunogai et al., 1993; Kumamoto et al., 1998).

Anthropogenic radionuclides such as ^{239}Pu (half-life: 2.41×10^4 yr) and ^{240}Pu (half-life: 6.56×10^3 yr) have been released into the environment as the consequence of atmospheric nuclear weapons testing (UNSCEAR, 2000). In the North Pacific Ocean, two distinct sources of Pu isotopes can be identified; i.e., the global stratospheric fallout and close-in trophospheric fallout from the Pacific Proving Grounds in the Marshall Islands (Bertine et al., 1986; Buesseler, 1997). The Pu input from the close-in trophospheric fallout from the Pacific Proving Grounds mainly took place in the early 1950s whereas the maximum deposition of the global stratospheric fallout occurred in 1963. Plutonium isotopic signature is very useful for understanding the transport process in the oceans and identifying the sources of Pu. The $^{240}Pu/^{239}Pu$ atom ratios from the global stratospheric fallout are 0.176 ± 0.014 (Krey et al., 1976) or 0.180 ± 0.014 (Kelley et al., 1999) while those from close-in tropospheric fallout from nuclear weapons testing at the Pacific Proving Grounds are 0.33–0.36 (Diamond et al., 1960; Komura et al., 1984; Koide et al., 1985; Buesseler, 1997).

The vertical distributions of $^{239+240}Pu$ concentration in the water column have been reported in the Japan Sea and adjacent seas (e.g., Nagaya and Nakamura, 1987; Yamada et al., 1996; Hirose et al., 1999; Lee et al., 2003; Ito et al., 2003); however, only limited data in seawater have been reported to date on $^{240}Pu/^{239}Pu$ atom ratios (Kim et al., 2004; Yamada and Zheng, 2008). A study on $^{240}Pu/^{239}Pu$ atom ratios in seawater around the Korean Peninsula indicated the presence of the close-in tropospheric fallout Pu from the Pacific Proving Grounds (Kim et al., 2004). Comparing the distribution of

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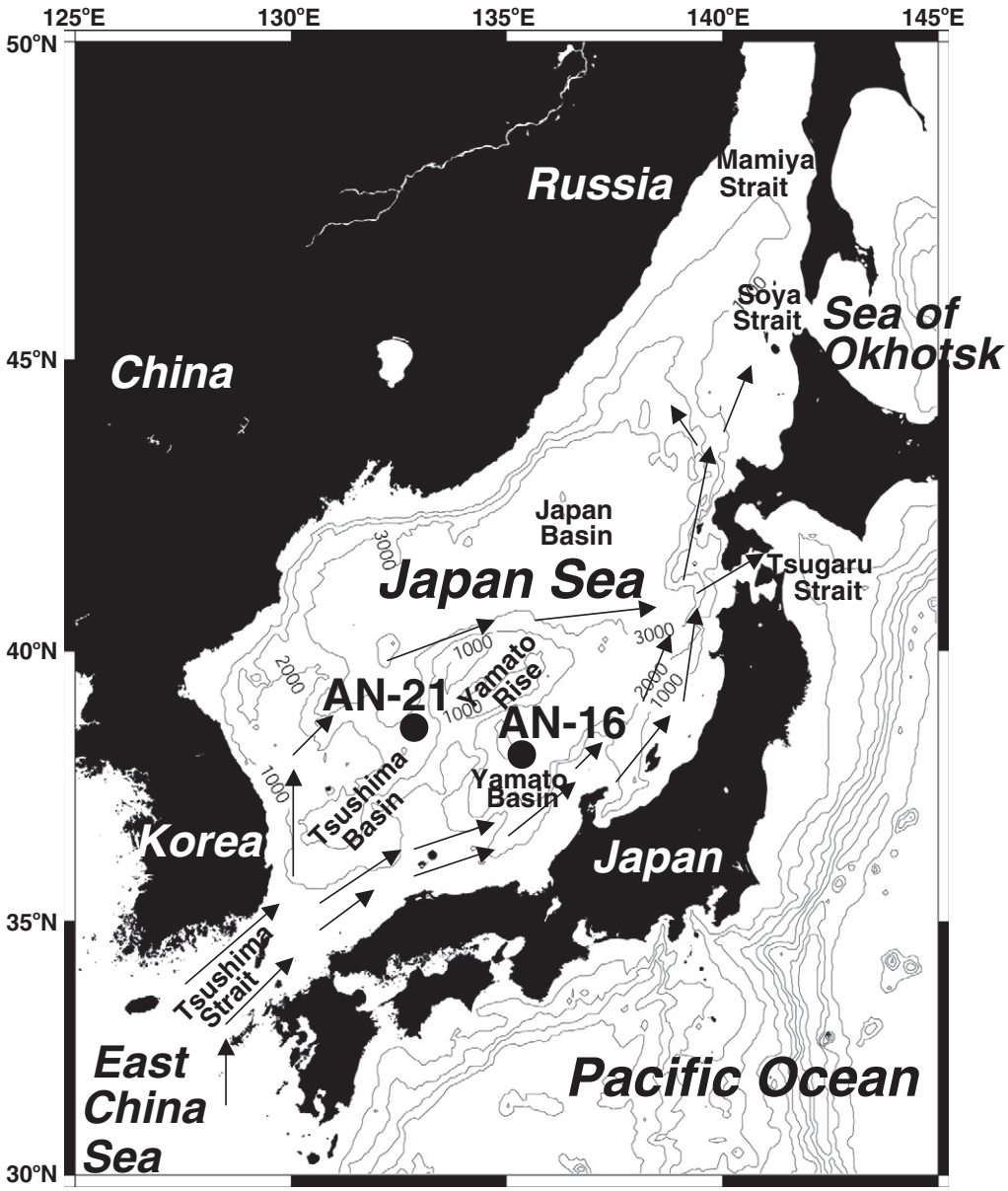

Fig. 1. Map of the Japan Sea, including the sampling station locations. The black arrows indicate the schematic flow patterns of the Tsushima Warm Current in the Japan Sea.

$^{240}Pu/^{239}Pu$ atom ratios and $^{239+240}Pu$ concentrations on a time scale of one decade would allow researchers to trace temporal variations of Pu sources in the Japan Sea. Then, the objectives of this study were to measure the vertical distributions of $^{239+240}Pu$ concentration and $^{240}Pu/^{239}Pu$ atom ratio in seawater samples obtained in 1984 and 1993 from the Japan Sea, to trace temporal changes of $^{239+240}Pu$ inventories in the water columns by data comparison between the two sampling years, to estimate temporal changes of the relative contributions of the global stratospheric fallout Pu and close-in tropospheric fallout Pu, and to discuss the transport and scavenging processes of Pu in the Japan Sea.

2. Materials and methods

2.1. Sample collection

The sampling procedure used in 1984 was described by Nagaya and Nakamura (1987) and Nozaki and Yamada (1987). Seawater samples were collected at two sampling stations, AN-16 (38°18′N, 135°29′E; water depth: 2931 m; sampling date: September 1984) in the Yamato Basin and AN-21 (38°41′N, 132°48′E; water depth: 2829 m; sampling date: September 1984) in the Tsushima Basin (Fig. 1) during the KH-84-3 Cruise (Antares Expedition) of the R/V Hakuho-Maru. A double barrel PVC large-volume sampler was used. The samples for surface water were collected from the ship's deck by using a pumping system. Once samples were brought aboard, the Pu, Th and Pa isotopes in them were coprecipitated using iron hydroxide. Data on $^{239+240}Pu$ concentrations at AN-16 (Nagaya and Nakamura, 1987) and on ^{230}Th and ^{231}Pa concentrations at AN-16 and AN-21 (Nozaki and Yamada, 1987) are available in the literature. Data on $^{239+240}Pu$ concentrations at AN-21 are reported in this study.

The sampling and analytical procedures used in 1993 for Pu were described by Yamada et al. (1996). Seawater samples were collected during the N-93-02 Cruise of the R/V Natsushima. One objective of the 1993 Natsushima Cruise was to re-visit stations AN-16 and AN-21 of the 1984 Hakuho-Maru Cruise. Samples were collected at AN-16 (water depth: 2937 m; sampling date: May 1993) and AN-21 (water depth: 2825 m; sampling date: May 1993) with Niskin bottles that were