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Vertical distributions of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in sediment cores: implications for the sources of Pu in the Japan Sea

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Abstract

The vertical distributions of $^{239+240}$Pu-specific activities and ^{240}Pu/^{239}Pu atom ratios in sediment cores of the Japan Sea were investigated. For comparison, the $^{239+240}$Pu-specific activities and ^{240}Pu/^{239}Pu atom ratios in the surface layer of bottom sediments in the Sea of Okhotsk were also determined. The $^{239+240}$Pu-specific activities in the surface layer sediments varied from 0.43 to 2.65 mBq/g in the Japan Sea. The lowest value was seen in the eastern Japan Basin, while the highest value was found in a sample collected from a coastal area off Hokkaido in the northern Japan Sea. It was found that the ^{240}Pu/^{239}Pu atom ratios in the surface sediments for the Japanese side of the Japan Sea ranged from 0.15 to 0.22. The ^{240}Pu/^{239}Pu atom ratios in the surface sediments collected from the Yamato Basin and Japan Basin were typically close to the global fallout value of 0.18, but higher ^{240}Pu/^{239}Pu atom ratios (0.20–0.22) were seen in samples from the northern Japan Sea. Vertical profiles of ^{240}Pu/^{239}Pu atom ratios revealed that these atom ratios were higher than the global fallout value in the deeper layers in the sediment cores from the northern Japan Sea on the Hokkaido coast and the Yamato Basin. In addition, the sediment cores from the northern Japan Sea near the Soya Strait and in the Ishikari Estuary had a constant vertical distribution of ^{240}Pu/^{239}Pu atom ratios with a value around 0.20. The results obtained in this work support the hypothesis that the prevailing ocean currents transported Pu derived from the Pacific Proving Grounds to the Japan Sea, and this contributed to the general elevated Pu inventory in the Japan Sea.

Keywords: Sediment core; $^{239+240}$Pu; ^{240}Pu/^{239}Pu; ICP-MS; Japan Sea

1. Introduction

Plutonium was introduced into the environment mainly from the fallout of atmospheric nuclear weapons tests. Because about 70% of the Earth's

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surface is covered by oceans, over two-thirds of this fallout has entered them. Because of its high chemical toxicity and high concentration factor in marine organisms (Kishimoto et al., 2002), much attention has been paid to the environmental monitoring of Pu in order to obtain information about the current status of Pu contamination. On the other hand, in marine sciences, plutonium is widely used as a geochemical tracer to study sediment mixing rates (Cochran, 1985) and to evaluate scavenging and removal processes in the water column (Livingston and Anderson, 1983; Buesseler, 1997; Yamada and Nagaya, 2000a; Yamada and Aono, 2002; Yamada and Nagaya, 2000b).

The Japan Sea is a typical marginal sea of the western Pacific Ocean. It is open to outer seas only through four straits (Tsushima, Tsugaru, Soya, and Mamiya). Because their sill depths are less than 150 m, the deep water below that depth, which is often called the Japan Sea Proper Water (JSPW) in a broad sense (Uda, 1934), is isolated from the North Pacific. The Proper Water consists of at least two water masses, the upper portion and the deep water, and the former is produced by wintertime deep convection (Senjyu, 1999). Studies that addressed Pu distribution in the Japan Sea revealed a higher surplus of total inventories (seawater and sediment) than those derived from global fallout (Pettersson et al., 1999; Hirose et al., 1999; Hong et al., 1999; Ikeuchi et al., 1999; Nagaya and Nakamura, 1987; Yamada et al., 1996; Lee et al., 2003). It has been recognized that the $^{239+240}$Pu inventory in water showed a strong correlation with water depth (Lee et al., 2003), and that, in sediment, it was inversely related to water depth and correlated positively with the sediment accumulation rate (Hong et al., 1999). However, most studies were conducted solely based on the total Pu activity ($^{239+240}$Pu), and over small study areas with limited samples. Therefore, the reasons for the excess inventories of the anthropogenic radionuclide, Pu, and the overall facts are not well understood; in particular, the sources of Pu remain a puzzle.

Recent work on the ^{240}Pu/^{239}Pu isotope signature in bottom sediments (Kim et al., 2003) and in seawater around the Korean Peninsula (Kim et al., 2004) has provided a plausible explanation for the excess Pu in the Japan Sea: the transport of Pu originating from nuclear tests in the Pacific Proving Grounds to the Japan Sea by oceanic currents, e.g., the North Equatorial Current, the Kuroshio Current and its branch, the Tsushima Current, flowed by advection and mixing. More researchers have an interest in understanding close-in fallout in the western North Pacific in terms of radiological assessment and tracing oceanographic processes occurring in the region. Because stratospheric (global) fallout has been drastically reduced since the 1980s (Hirose et al., 2003), the close-in fallout signature in recent decades has become more significant. This was evident from our recent study on the vertical profiles of Pu activity and Pu atom ratio (^{240}Pu/^{239}Pu) in sediments collected from Sagami Bay, western Northwest Pacific margin (Zheng and Yamada, 2004). We found a sedimentary record of global fallout and Bikini close-in fallout Pu, clearly indicating that Pu contamination which originated from Bikini and

Table 1
Sediment samples investigated in this study

Station	Area	Sampling date	Location	Core length (cm)	Water depth (m)
CM-06	Okhotsk Sea	22 July 1998	45°25′N; 145°05′E	32	3053
CM-07	Okhotsk Sea	23 July 1998	44°45′N; 145°10′E	37	1696
CM-08	Okhotsk Sea	24 July 1998	44°30′N; 145°00′E	41	1214
CM-09	Japan Sea	24 July 1998	45°38′N; 140°20′E	18.5	587
CM-13	Japan Sea	27 July 1998	43°40′N; 140°30′E	24	809
CM-15	Japan Sea	28 July 1998	43°34′N; 140°48′E	22	748
CM-16	Japan Sea	28 July 1998	43°32′N; 140°53′E	17	123
CM-17	Japan Sea	28 July 1998	45°29′N; 141°00′E	36	91
CM-18	Japan Sea	03 Aug. 1998	42°45′N; 138°16′E	23.5	3670
CM-19	Japan Sea	04 Aug. 1998	41°21′N; 137°20′E	3	3637
CM-20	Japan Sea	05 Aug. 1998	37°44′N; 135°14′E	52.5	2978