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$^{228}Ra/^{226}Ra$ ratio and ^{7}Be concentration in the Sea of Japan as indicators for water transport: comparison with migration pattern of Fukushima Dai-ichi NPP-derived ^{134}Cs and ^{137}Cs

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ABSTRACT

To assess the migration patterns of radiocesium emitted from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), we analyzed $^{228}Ra/^{226}Ra$ ratios and ^{7}Be concentrations and compared them with ^{134}Cs and ^{137}Cs concentrations in seawater samples collected within the Sea of Japan before and after the FDNPP accident (i.e., during the period 2007–2012) using low-background γ-spectrometry. The $^{228}Ra/^{226}Ra$ ratios in surface waters exhibited lateral and seasonal variations, reflecting the flow patterns of surface water. This indicates the transport patterns of the FDNPP-derived radiocesium by surface water. Cosmogenic ^{7}Be (half-life: 53.3 d) exhibited markedly high concentrations (5–10 mBq/L) at depths shallower than 50 m, with concentrations decreasing steeply (0.2–2 mBq/L) at depths of 50–250 m. The distribution of ^{7}Be concentrations suggests that the downward delivery of the FDNPP-derived radiocesium to below 50 m depth was negligible for a few months prior to its removal from the Sea of Japan.

1. Introduction

The Tsushima Warm Current (TWC), a major feature of flow patterns in the Sea of Japan, is thought to be composed of three branch currents (Toba et al., 1982). To understand the transport of pollutants and nutrients released by human-induced accidents, many previous studies have investigated the flow pattern of the TWC using various techniques (Hase et al., 1999; Morimoto and Yanagi, 2001). However, owing to the considerable complexity of this circulation pattern, its characteristics remain poorly understood.

Various radionuclides in seawater have been used as powerful tracers to investigate geochemical cycles in the Sea of Japan (e.g., Inoue et al., 2007) and in other oceans globally (e.g., Cochran and Masqué, 2004). A lateral profile of ^{228}Ra (half-life: 5.75 y)/^{226}Ra (half-life: 1600 y) concentration (activity) ratio in surface water is illustrated in Fig. 1, together with the main water currents acting within the Sea of Japan and the East China Sea (ECS). At the Japanese Archipelago side of the Sea of Japan, $^{228}Ra/^{226}Ra$ ratios in surface water are useful for investigating mass water movement for several reasons. 1) The $^{228}Ra/^{226}Ra$ ratios of the Kuroshio water (~0.2) and the ECS continental shelf water (~3.5), the two dominant sources of the TWC in the Sea of Japan, are markedly different (Nozaki et al., 1989; Inoue et al., 2012a). 2) In the Tsushima Strait, the $^{228}Ra/^{226}Ra$ ratios in surface water exhibit large seasonal changes (~0.5–3), reflecting the mixing ratio of the two currents (Inoue et al., 2010). 3) In contrast to the pattern typical of open oceans in which $^{228}Ra/^{226}Ra$ ratios decrease from coastal to offshore

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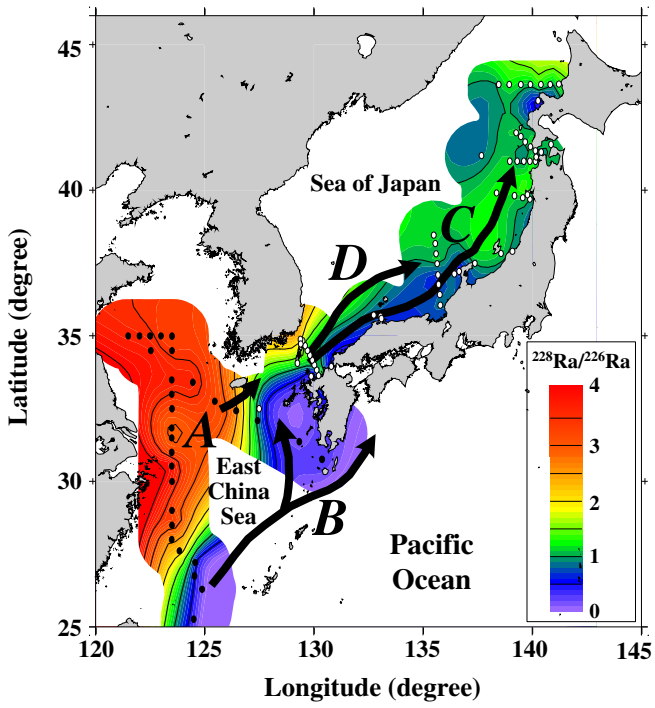

Fig. 1. Map showing the distribution of the $^{228}Ra/^{226}Ra$ ratios of surface waters within the East China Sea (Nozaki et al., 1989) and the Sea of Japan (Inoue et al., 2007) including the data of present study (see text) during May–June 1987–2011; the main water currents around the Japanese Archipelago are also illustrated (A: ECS continental shelf water current, B: Kuroshio current, C: coastal branch current of TWC, D: offshore branch current of TWC).

areas, such as on the Pacific Ocean side of Honshu, the main island of Japan (Yamada and Nozaki, 1986), the $^{228}Ra/^{226}Ra$ ratios of coastal water in this region are dominantly preserved, representing a conservative component during the migration of the TWC along Honshu Island and reflecting minimal input of radium isotopes (Inoue et al., 2007).

Furthermore, cosmogenic ^{7}Be (half-life: 53.3 d) is delivered continually to the sea surface through atmospheric dry and wet deposition. If ^{7}Be is transported by the circulation of seawater, together with cesium isotopes, the establishment of a vertical profile of ^{7}Be concentration would be beneficial to the understanding of the vertical transport of soluble components supplied to the sea surface. The residence time of the TWC within the Sea of Japan has been estimated to be a few months (Inoue et al., 2007); therefore, ^{7}Be is particularly suitable for studying this phenomenon owing to its much shorter half-life compared to ^{228}Ra (5.75 y). However, few studies have attempted to characterize vertical profiles of ^{7}Be in marine environments (Andrews et al., 2008). Thus, one of the primary aims of the present study is to characterize the spatial migration of soluble components in the surface and mixing layers within the Sea of Japan using $^{228}Ra/^{226}Ra$ ratios and ^{7}Be concentrations.

The massive East Japan earthquake and subsequent tsunami hit northeastern Japan from the Pacific Ocean side on March 11, 2011. These unprecedented disasters induced the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, which resulted in the widespread release of large amounts of ^{134}Cs (half-life: 2.06 y) and ^{137}Cs (half-life: 30.2 y) to the atmosphere and to land and sea surfaces over a wide region of eastern Japan (e.g., Chino et al., 2011; Hirose, 2012). In a previous study, we examined ^{134}Cs and ^{137}Cs concentrations in surface water samples from the Sea of Japan and evaluated the lateral distribution and migration pattern of low-levels of radiocesium (<1 mBq/L for ^{134}Cs) derived from the FDNPP (Inoue et al., 2012b,c, 2013). In the present study, we investigated vertical profiles of radiocesium concentrations in the mixing layer, adding new data in surface waters; here, we discuss their spatial migration patterns within the Sea of Japan. The present study also aims to compare water circulations implied from $^{228}Ra/^{226}Ra$ ratios and ^{7}Be concentrations with the spatial migration patterns of the FDNPP-derived radiocesium. The obtained datasets will be useful for future studies of radiocesium and other soluble contaminants released by human-induced accidents within the Sea of Japan and for the investigation of water circulation in the region.

2. Samples and experimental procedures

The seawater sampling sites used in the present study are illustrated in Fig. 2.

We collected seven filtered water samples (approximately 50 L) from the water column of the Japan Basin (site *SY07*), and 33 unfiltered water samples (20–60 L) from the Yamato Rise (site *SY08A*), the Tsushima Basin (site *SY08B*), the Yamato Basin (site *SY09A*), and the Japan Basin (site *SY09B*) on the *Soyo Maru* expeditions (July 2007–2009). Seawater samples of ~60 L were collected from water columns in the northeastern Japan Basin (*SY12C* and *D*), the southern Yamato Basin (*SY12E*), and the Yamato Rise (*SY12F*) on the *Soyo Maru* expedition in July 2012 (Fig. 2a).

We collected surface seawater samples of ~20 L within the Sea of Japan on three expeditions; the *Dai-7 Kaiko Maru* in May 2011 (*KK11*) (Fig. 2b), the *Oshoro Maru* in June 2011 (*OS11*) (Fig. 2c), and the *Asuka-II* in October 2011 (*AS11*) (Fig. 2d). We also collected nine *FP11* (Fig. 2b) and two *IP11* (Fig. 2d) water samples along the coastline of Honshu Island in April 2011 and August 2011, respectively.

All water samples were unfiltered, except those from the *SY07* site passed through 0.5 μm median pore size for filter cartridges with 2 L/min. The ^{134}Cs and ^{137}Cs concentrations for the *OS11* and *AS11* samples have been already published in Inoue et al. (2012c), and those for the *KK11* samples were reported in Inoue et al. (2013).

Detailed descriptions of the experimental procedures have been presented elsewhere (Nakano et al., 2008; Inoue et al., 2013). Briefly, after adjustment to pH ~1 with concentrated HNO_3, cesium isotopes in seawater samples were separated quantitatively by coprecipitation with ammonium phosphomolybdate (AMP/Cs), by adding 0.52 g of CsCl and 8.0 g of AMP to a ~20–60 L aliquot of water (partly 0.26 g CsCl and 4.0 g AMP for ~20 L water). After the removal of AMP/Cs, a minimally radium-contaminated Ba carrier was added and $BaSO_4$ was precipitated with the radium isotopes. Then, Fe carrier was added, and $Fe(OH)_3$ was precipitated with ^{7}Be and ^{228}Th by adjusting the sample pH to ~7–8.

During the chemical procedures, we covered the plastic bucket containing the water sample with a vinyl sheet to prevent input of ^{7}Be-bearing dry deposits, except at site *SY07*, where most of the chemical procedures were conducted in the open air on the research vessel during the expedition. For these samples, we estimated the contribution of ^{7}Be from dry deposition to be 20.5 mBq, based on our experimental procedures on ^{7}Be-free deep seawater collected at 5500 m depth in the North Pacific Ocean (37°35′N, 143°30′E; 5524 m depth) in July 2008.

The chemical yields of radiocesium isotopes (88–100%) and ^{7}Be (92–100%) were determined from the yield of AMP/Cs and $Fe(OH)_3$ (or by the mean values obtained in our laboratory: 95% of AMP/Cs in *OS11* and *AS11* waters and 95% of $Fe(OH)_3$ in *SY12* water samples), respectively.

The filter cartridges used for filtration of *SY07* waters were ashed at 450 °C for γ-spectrometry.