EPJ Web of Conferences **24**, 04001 (2012)
DOI: 10.1051/epjconf/20122404001

Vertical distributions of ^{226}Ra, ^{228}Ra, and ^{137}Cs activities in the southwestern part of the Sea of Okhotsk

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Abstract

We collected 14 water column seawater samples in the southwestern part of the Sea of Okhotsk and employed low-background γ-spectrometry with convenient minimal radiochemical processing to determine the activities of ^{226}Ra (half-life $t_{1/2} = 1600$ y), ^{228}Ra (5.75 y), and ^{137}Cs (30.2 y) of the samples. All of these nuclides exhibited unique vertical profiles; ^{226}Ra, ^{228}Ra, and ^{137}Cs activities exhibit small variations from 50–500 m depth (^{226}Ra, $\sim 2\,\mathrm{mBq\,l^{-1}}$; ^{228}Ra, $\sim 0.4\,\mathrm{mBq\,l^{-1}}$; ^{137}Cs, $\sim 1\,\mathrm{mBq\,l^{-1}}$). These profiles can be explained by convective mixing of surface water to this layer.

1. Introduction

The Sea of Okhotsk, one of the largest marginal seas of the northwestern North Pacific, is composed of various distinct layers, such as dense shelf water (DSW) and Okhotsk Sea Intermediate water (OSIW), because of unique

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vertical water circulation (Shcherbina *et al.*, 2004). In the southwestern Sea of Okhotsk, circulation of water masses is markedly complicated and variable. In winter, the convective mixing of surface water is believed to form an intermediate layer (Kitani, 1973). However, contrary to recent studies of surface water migration, the vertical circulation characteristics have not yet been well determined. Various radionuclides in seawater have been used as tracers for studying geochemical cycles in marine environments (Cochran and Masqué, 2004). In particular, activities of ^{226}Ra, ^{228}Ra, and ^{137}Cs are good tracers of water mass transport. However, the standard γ-spectrometry technique for measuring these radionuclides usually requires very large volumes (hundreds to tens of thousands of liters) of deep seawater because of the radionuclides' low activities. Therefore, existing data on the distributions of these nuclides in the Sea of Okhotsk are not sufficient (Kawakami and Kusakabe, 2008), particularly beneath the surface, to allow us to investigate the details of water circulation. Low-background γ-spectrometry combined with minimal radiochemical processing (Nakano *et al.*, 2008) enabled the determination of low levels of ^{226}Ra, ^{228}Ra, and ^{137}Cs, using only 60 L water samples from the Sea of Okhotsk, providing enough data for this purpose. In this study, we applied this method to conduct a preliminary investigation of vertical profiles of these nuclides in the southwestern part of the Sea of Okhotsk, so we could elucidate the flow patterns of water masses in this area.

2. Samples and experimental methods

We collected 14 seawater samples ($\sim$ 60 L) at depths of 5, 10, 50, 100, 150, 200, 250, 500, 750, 1000, 1250, 1500, 1650, and 1800 m on the southwestern slope of the Kuril Basin in the Sea of Okhotsk (site *SY09C*; N45°01′, E145°01′; 1850 m depth). Sampling was conducted during the *Soyo Maru* expedition (Jul-Aug 2009), and all water samples were unfiltered. Detailed explanations of experimental procedures are presented elsewhere (Nakano *et al.*, 2008). Briefly, after adjustment to pH 1, ^{137}Cs was quantitatively separated by coprecipitation with ammonium phosphomolybdate (AMP). After removal of AMP, the least Ra-contaminated Ba carrier was added and barium sulfate was precipitated with radium isotopes. Then, an Fe carrier was added, and $Fe(OH)_3$ was precipitated by adjusting the sample pH to $\sim$ 7–8. The chemical yield of ^{137}Cs was 95% (the mean value for our laboratory), based on the ratio of stable Cs in untreated and treated water samples. The yield of radium isotopes (83–100%) was determined from

the yield of $BaSO_4$. Low-background γ-spectrometry of AMP and $BaSO_4$-$Fe(OH)_3$ mixture samples was performed using Ge-detectors, most of which are located at the Ogoya Underground Laboratory, Japan (Hamajima and Komura, 2004). γ-Peaks of ^{228}Ra (338 and 911 keV of ^{228}Ac) are ambiguous in deep waters (below 1000 m depth), indicating that levels were below our detection limit for water samples ($< 0.1\,\mathrm{mBq\,l^{-1}}$ for 60 L water).

3. Results and discussion

The vertical profiles of activities of nuclides studied are presented in fig. 1 together with salinity, temperature, and density. The water column in this area is considered to consist of intermediate cold water (ICW) (potential temperature of -1.8–$+2\,^{\circ}$C, salinity of 32.8–33.4) (50–300 m), transient layer water (TLW) (1–2 °C, 33.4–34.3) (300–1200 m), and deep layer water (below ~ 1200 m to bottom) beneath the surface water (above ~ 50 m) (Takizawa, 1982; Oguma *et al.*, 2008). The variation of ^{226}Ra activity in surface and ICW is small ($\sim 2\,\mathrm{mBq\,l^{-1}}$) (fig. 1c). In the transient and deep layers, ^{226}Ra activity gradually increases from 2–5 $\mathrm{mBq\,l^{-1}}$, showing a higher value than that of the deep water at the same depth in the northwestern North Pacific (*KH71-3N*; 2–4 $\mathrm{mBq\,l^{-1}}$; Tsunogai and Harada, 1980). These features are considered to reflect the reserve of ^{226}Ra that is continuously supplied from bottom sediment (and settling particles) because of its long life and/or the long residence time of deep layer water (Cochran, 1980). Differing steep gradients of ^{228}Ra activity typically observed in other oceans (vertical eddy coefficients of 0.5–7 $\mathrm{cm^2\,s^{-1}}$; surface to < 300 m depth; Kaufman *et al.*, 1973; Moore, 1972; Li *et al.*, 1980; Tanaka *et al.*, 2006), ^{228}Ra activity in the ICW exhibits small variation ($\sim 0.4\,\mathrm{mBq\,l^{-1}}$) after the decrease from the surface ($\sim 0.6\,\mathrm{mBq\,l^{-1}}$) (fig. 1d). ^{228}Ra activity steeply decreases in TLW and is markedly low in the deep layer water ($< 0.1\,\mathrm{mBq\,l^{-1}}$). The profile of ^{228}Ra/^{226}Ra ratio mainly results from large variation in the activity of short-lived ^{228}Ra, exhibiting a constant value (0.15) in ICW (fig. 1e). Differing the gradients from surface or subsurface in other oceans (Ito *et al.*, 2003; Povinec *et al.*, 2003), the activity of fallout ^{137}Cs in the *SY09C* waters exhibits small variation in ICW ($\sim 1\,\mathrm{mBq\,l^{-1}}$), remaining similar to the surface level, and it continues to decrease in TLW and the deep layer water (0.5 to 0.1 $\mathrm{mBq\,l^{-1}}$) (fig. 1f). The inventory of ^{137}Cs shows marked variation in each ocean, reflecting water migration after the initial supply of ^{137}Cs from atmospheric nuclear test explosions in the middle of the 1950's and early 1960's, the Chernobyl reactor accident and later temporal changes

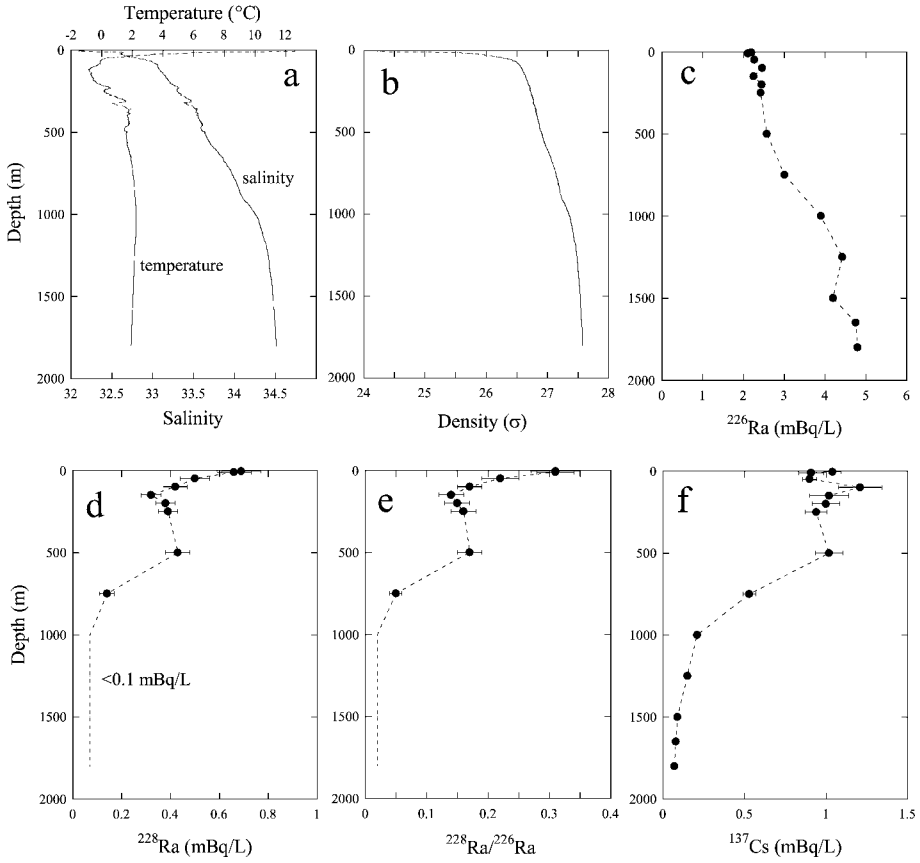

Figure 1: Vertical profiles of a) salinity and potential temperature, b) density, c) ^{226}Ra and d) ^{228}Ra activities, e) ^{228}Ra/^{226}Ra ratio, and f) ^{137}Cs activity of the *SY09C* water samples.

(Aoyama and Hirose, 2003). The inventory of ^{137}Cs (0–2000 m depth) in the Sea of Japan ($> 40°$N, 3.0 kBq m^{-2}; $< 40°$N, 2.5 kBq m^{-2}) is higher than that of the northwest Pacific (40–60°N) (1.6 kBq m^{-2}) (decay-corrected to 2001), reflecting the winter convection of surface water (Ito *et al.*, 2003; Aoyama and Hirose, 2003). ^{137}Cs activity of the *SY09C* waters is lower than that of waters from the Japan Basin in the Sea of Japan (the site *SY09B*; N43°00′, E138°00′; 3700 m depth; our unpublished data from water samples collected during 2009 *Soyo Maru* expedition) at all depths (particularly below $\sim$ 500 m depth) and, therefore, the estimated inventory of *SY09C* waters (0.7 kBq m^{-2}; 0–1850 m depth) is approximately 1/3 times that of the *SY09B* waters (2 kBq m^{-2}; 0–2000 m). The low ^{137}Cs inventory at the site

SY09C indicates that the movements of the Soya Warm Current Water from the Sea of Japan (Itoh and Ohshima, 2000) do not contribute to effective accumulation of ^{137}Cs in the southwestern Sea of Okhotsk. In the northwestern Okhotsk, rapid sea-ice formation leads to production of DSW ($26.9\sigma_\theta$ maximum density), which directly joins to OSIW (26.8–$27.4\sigma_\theta$; 250–1000 m depth) in the off-shore area. Also, the vertical distribution of artificially produced chlorofluorocarbons (CFCs) indicates the downward delivery of CFCs from the surface to intermediate layer during this process (Yamamoto-Kawai *et al.*, 2004; Shcherbina *et al.*, 2004). ICW at site *SY09C* (26.5–$26.8\sigma_\theta$; fig. 1b) is considered to be formed by a similar mechanism (*i.e.*, convection of surface water that reflects cold winter temperatures): Nuclide features observed in ICW are plausibly explained by downward delivery of ^{228}Ra and ^{137}Cs from the surface accompanying the convective mixing of surface water, which starts to migrate from the northwest Okhotsk to this area, before/during the homogenization by vertical mixing, though the convection areas of surface water are not clarified. Activities of all nuclides in the water sample from 500 m depth remain at similar levels as those of ICW, suggesting that the convection of surface water affects the upper part of TLW. On the other hand, at $\sim$ 500–1000 m depth, the vertical profiles of these nuclides change markedly (*e.g.*, steep decreases of ^{228}Ra and ^{137}Cs activities), reflecting the presence of boundary layers of these water masses and little vertical mixing below $\sim$ 500 m depth. Further clarification of the origin, convection area, and migration pattern of ICW will be facilitated by fine-resolution spatial measurements of these nuclides within the Sea of Okhotsk.

Acknowledgements

We thank the researchers, captain, and crew of the R/V *Soyo Maru* for their assistance during sampling. This study was supported partly by a Grant-in-Aid for Scientific Research No. 21510011 (M. Inoue) from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

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