EPJ Web of Conferences **24**, 04005 (2012)
DOI: 10.1051/epjconf/20122404005

Seasonal variations of $^{228}Ra/^{226}Ra$ and $^{228}Th/^{228}Ra$ ratios in surface water from the eastern East China Sea

K. Yoshida[1], M. Inoue[1], Y. Kiyomoto[2], M. Minakawa[3], H. Kofuji[4], S. Nagao[1], Y. Hamajima[1] and M. Yamamoto[1]

[1] Institute of Nature and Environmental Technology, Kanazawa University, Japan
[2] Fisheries Research Agency, Seikai National Fisheries Research Institute, Japan
[3] Fisheries Research Agency, National Research Institute of Fisheries Science, Japan
[4] Mutsu Marine Laboratory, Japan Marine Science Foundation, Aomori, Japan

Abstract

A total of 30 surface water samples were collected at 2 sites on the east side of the East China Sea (ECS) during 2008-2010, and ^{226}Ra, ^{228}Ra, and ^{228}Th activities were measured using low-background γ-spectrometry. The $^{228}Ra/^{226}Ra$ and $^{228}Th/^{228}Ra$ activity ratios of the samples exhibited notable seasonal variations ($^{228}Ra/^{226}Ra$ = 0.2-3.3; $^{228}Th/^{228}Ra$ = < 0.1-0.4) accompanying salinity changes (31.0-34.6). These changes are hypothesized to cause the change by altering the mixing ratio of ^{228}Ra-rich and particle-rich continental shelf water within the ECS.

1 Introduction

The East China Sea (ECS) is one of the largest marginal seas in the northwest Pacific Ocean with a broad continental shelf (more shallow than

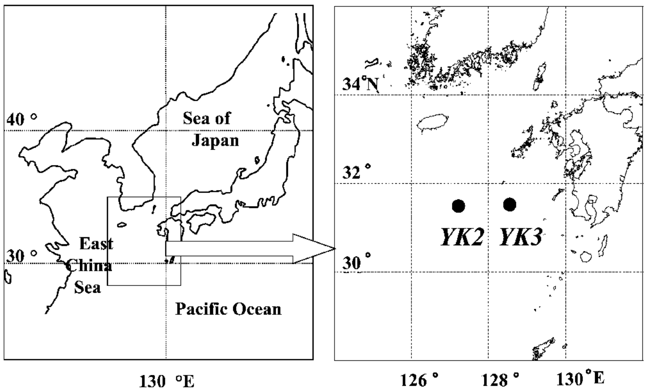

Figure 1: Sampling locations far seawater samples.

$\sim$ 200 m) in the western part. To reveal geochemical cycles and hydrographic features, many researchers have studied ocean current migration patterns over the ECS using various techniques (Yanagi *et al.*, 1997; Isobe, 2008), but clarifying water circulation characteristics in the ECS is difficult because of the region's markedly complicated seasonality (Ichikawa and Beardsley, 2002). Our previous study clarified that the ^{228}Ra (half-life $(t_{1/2})$ = 5.75 y)/^{226}Ra (1600 y) activity ratio of the surface water exhibited notable seasonal variation in the ECS. This is believed to be caused by a change of the mixing ratio of ^{228}Ra-rich continental shelf water and ^{228}Ra-poor Kuroshio water (Inoue *et al.*, 2010). The ECS is an area of high productivity and particle scavenging (Iseki *et al.*, 2003). Because of the particle-reactive property of thorium isotopes, the ^{228}Th (1.91 y) activity of seawater and the extent of its disequilibrium with its soluble parent ^{228}Ra (^{228}Th/^{228}Ra ratio) have often been utilized to estimate scavenging from seawater (Bacon, 2004). In the present study, we examined the seasonal change in the ^{228}Th/^{228}Ra ratio of surface waters in the ECS together with that of ^{228}Ra/^{226}Ra, and assessed seasonal circulation of water and particles.

2 Samples and experimental method

Seawater sampling locations on the east side of the ECS are shown in Fig. 1. A total of 30 surface water samples ($\sim$20 L) were collected from 2 sites (*YK2*, 31°45′N-127°15′E, 125 m; *YK3*, 31°45′N-128°45′E, 762 m) in 2-4 months interval during the *Yoko Maru* and *Soyo Maru* expeditions in 2008-2010. Detailed explanations of these experimental procedures were presented elsewhere (Nakano *et al.*, 2008). Briefly, the least Ra-contaminated Ba (0.7 mBq/g-Ba for ^{226}Ra; 0.2 mBq/g-Ba for ^{228}Ra) and Fe carriers were added to unfiltered water, and $BaSO_4$ and $Fe(OH)_3$ were precipitated with radium and thorium isotopes. Low-background γ-spectrometry was performed using Ge-detectors located at the Ogoya Underground Laboratory, Japan (Hamajima and Komura, 2004). The yields of radium isotopes and ^{228}Th were tentatively estimated to be 85% and 90%.

3 Results and discussion

3.1 Seasonal variations of ^{228}Ra activity and ^{228}Ra/^{226}Ra ratio

Seasonal Variations of ^{228}Ra activity and ^{228}Ra/^{226}Ra ratio are presented in Fig. 2. The ^{228}Ra/^{226}Ra ratio of waters exhibits a wide variation (0.2-3.3), mainly resulting from variation in the activity of short-lived ^{228}Ra (0.2-7.4 mBq L^{-1}) supplied from the shallow continental shelf (Fig. 2). In winter and spring, the ^{228}Ra/^{226}Ra ratios at *YK2* and *YK3* are similar to that of Kuroshio water. In summer, ^{228}Ra-rich shelf water appears to be brought to the east side, and the ^{228}Ra/^{226}Ra ratios at *YK2* and *YK3* reach higher levels, reflecting large contributions of shelf water. The ^{228}Ra/^{226}Ra

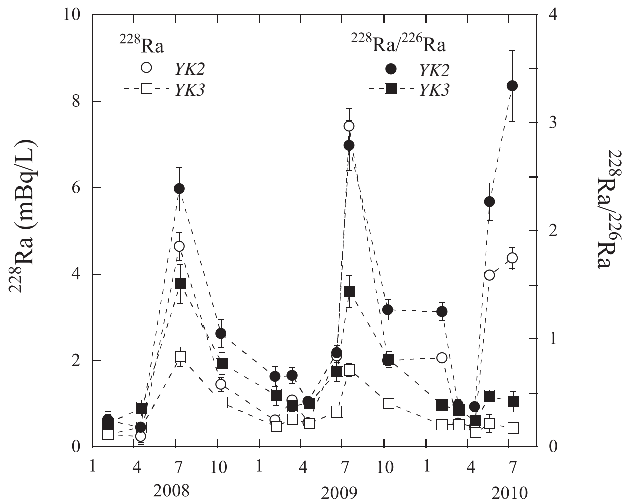

Figure 2: Seasonal variations of ^{228}Ra activity and ^{228}Ra/^{226}Ra ratio in surface waters of the ECS (sites *YK2* and *YK3*). Data in 2008 are from Inoue *et al.* (2010).

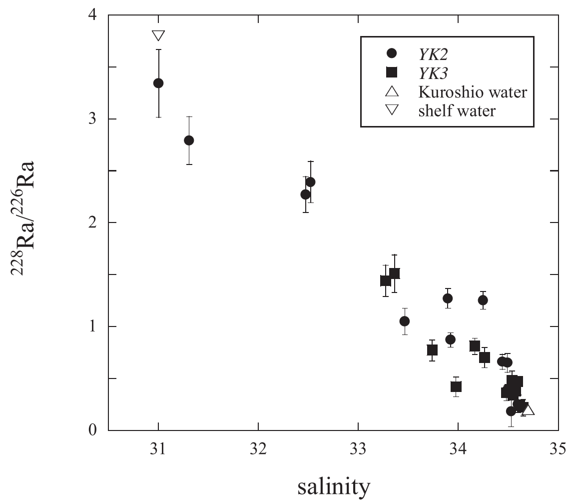

Figure 3: ^{228}Ra/^{226}Ra ratio *vs* salinity in surface waters of sites *YK2* and *YK3*.

ratio of water in the continental side (site *YK2*) is evidently higher compared with that in Japan side (site *YK3*). On the other hand, annual variations of ^{228}Ra/^{226}Ra ratio are also observed during this period (*e.g.*, high ^{228}Ra/^{226}Ra ratio at *YK2* and markedly low ratio at *YK3* in July 2010), which reflects the annual difference of the mixing pattern of shelf water.

The ^{228}Ra/^{226}Ra ratio is plotted against salinity in Fig. 3. The continental shelf water in the ECS exhibits a markedly higher ^{228}Ra/^{226}Ra ratio and lower salinity (^{228}Ra/^{226}Ra, ~ 3.8; salinity, ~ 31) than water from the Kuroshio-dominated area (~ 0.2; ~ 34.7) (Nozaki *et al.*, 1989). As shown in Fig. 3, the ^{228}Ra/^{226}Ra ratio in *YK* waters is fundamentally within the range between values observed in the Kuroshio and shelf waters. The wide range of the ^{228}Ra/^{226}Ra ratio at *YK* is explained by the seasonal and lateral variations of the mixing ratio of the shelf water.

3.2 Seasonal variations of ^{228}Th activity and ^{228}Th/^{228}Ra ratio

The seasonal variation of ^{228}Th activity is compared to that of ^{228}Th/^{228}Ra ratio in Fig. 4. The ^{228}Th activity is markedly high in summer ($0.5\,\mathrm{mBq\,L^{-1}}$ at *YK2*), which is due to the high activity of the parent nuclides ^{228}Ra. In Fig. 5, the ^{228}Th/^{228}Ra ratio at the ECS surface is plotted against salinity. The water with high ^{228}Th activity and low salinity (< 34) exhibits

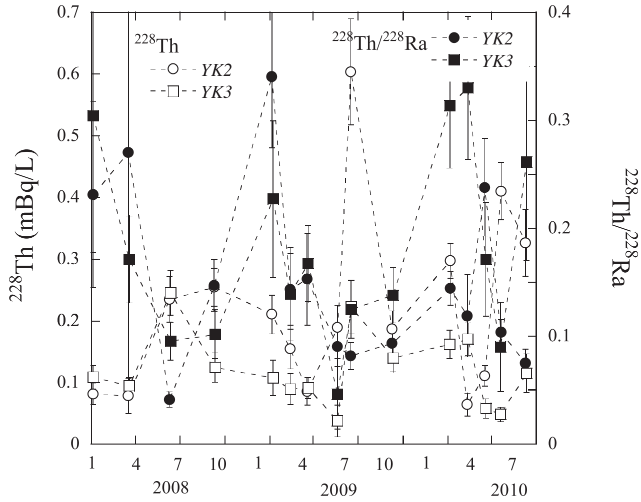

Figure 4: Seasonal variations of ^{228}Th activity and ^{228}Th/^{228}Ra ratio in surface waters of *YK2* and *YK3*.

a lower ^{228}Th/^{228}Ra ratio (0.05-0.15) relative to high salinity (34.0-34.6) Kuroshio-dominated water (^{228}Th/^{228}Ra = 0.15-0.35). Reactive nuclides (^{234}Th, ^{210}Pb, ^{210}Po, and ^{10}Be) in the surface water on the shelf are much lower than that on the Kuroshio area, being due to the vigorous scavenging

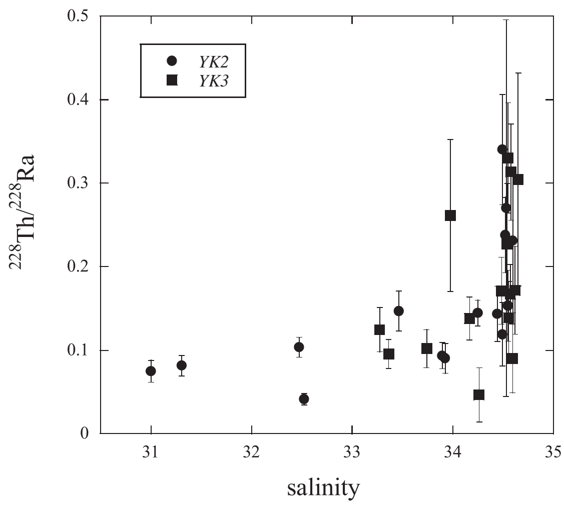

Figure 5: ^{228}Th/^{228}Ra ratio *vs* salinity in surface waters of *YK2* and *YK3*. Large deviation of ^{228}Th/^{228}Ra in the high salinity water is due to large statistical errors of low ^{228}Ra and ^{228}Th activities.

into sediments on the shelf area (Nozaki *et al.*, 1991; Aono and Yamada, 1999; Yang *et al.*, 2003). The lower ^{228}Th/^{228}Ra ratio of the shelf water in summer is considered to be caused by the removal of ^{228}Th by the scavenging (Yamada and Aono, 2006).

In order to elucidate the variation of ^{228}Th/^{228}Ra ratio, further experimentation and study (fine-resolution lateral and vertical measurements of these nuclides within the ECS) are needed.

Acknowledgements

We are indebted to the researchers, captain and crew of the R/Vs *Yoko Maru* and *Soyo Maru* for their assistance during sampling. This study was supported in partly by the Sasagawa Scientific Research Grant from The Japan Science Society (K. Yoshida) and a Grant-in-Aid for Scientific Research No. 21510011 from the Ministry of Education, Culture, Sports, Science and Technology of Japan (M. Inoue).

References

Bacon M. P., in *Marine Radioactivity*, edited by Livingston H. D. (Elsevier) 2004, pp. 139-165.

Hamajima Y. and Komura K., *Appl. Radiat. Isot.* **61** (2004) 179.

Ichikawa H. and Beardsley R. C., *J. Oceanogr.* **58** (2002) 77.

Inoue M., Nakano Y., Kiyomoto Y., Kofuji H., Hamajima Y. and Yamamoto M., *J. Oceanogr.* **66** (2010) 425.

Iseki K., Okamura K. and Kiyomoto Y., *Deep-Sea Res. II* **50** (2003) 457.

Isobe A., *J. Oceanogr.* **64** (2008) 569.

Nakano Y., Inoue M. and Komura K., *J. Oceanogr.* **64** (2008) 713.

Nozaki Y., Kasemsupaya V. and Tsubota H., *Geophys. Res. Lett.* **16** (1989) 1297.

Nozaki Y., Tsubota H., Kasemsupaya V., Yashima M. and Ikuta N., *Geochim. Cosmochim. Acta* **55** (1991) 1265.

Yamada M. and Aono T., *Mar. Geol.* **227** (2006) 1.

Yanagi T., Morimoto A. and Ichikawa K., *Continental Shelf Res.* **17** (1997) 655.

Yang Y.-L., Kusakabe M. and Southon J. R., *Deep-Sea Res. II* **50** (2003) 339.