Journal of Environmental Radioactivity 182 (2018) 142–150

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Temporal and spatial variations of ^{134}Cs and ^{137}Cs levels in the Sea of Japan and Pacific coastal region: Implications for dispersion of FDNPP-derived radiocesium

M. Inoue[a,*], Y. Shirotani[a], S. Yamashita[a], H. Takata[b], H. Kofuji[a], D. Ambe[c], N. Honda[d], Y. Yagi[d], S. Nagao[a]

[a] Low Level Radioactivity Laboratory, Institute of Nature and Environmental Technology, Kanazawa University, Nomi, Ishikawa 923-1224, Japan
[b] Marine Ecology Research Institute, Isumi, Onjuku Chiba 299-5105, Japan
[c] National Research Institute of Fisheries Science, Fisheries Research Agency, Fukuura, Kanazawa, Yokohama 236-8648, Japan
[d] Japan Sea National Fisheries Research Institute, Japan Fisheries Research and Education Agency, Suido-cho, Chuou-ku, Niigata, 951-8121, Japan

ARTICLE INFO

Keywords:
Radiocesium
Fukushima Dai-ichi Nuclear Power Plant accident
Surface seawater
Sea of Japan
Kuroshio Current

ABSTRACT

To investigate the dispersion of Fukushima Dai-ichi Nuclear Power Plant (FDNPP)-derived radiocesium in the Sea of Japan and western Pacific coastal region and determine the sources of radiocesium in these areas, we examined the temporal and spatial variations of ^{134}Cs and ^{137}Cs concentrations (activities) during 2011–2016 in seawaters around the western Japanese Archipelago, particularly in the Sea of Japan. In May 2013, the surface concentration of ^{134}Cs was ~0.5 mBq/L (decay-corrected to March 11, 2011), and that of ^{137}Cs exceeded the pre-accident level in this study area, where the effects of radiocesium depositions just after the FDNPP accident disappeared in surface waters in October 2011. Subsequently, radiocesium concentrations gradually increased during 2013–2016 (~0.5–1 mBq/L for ^{134}Cs), exhibiting approximately homogeneous distributions in each year. The temporal and spatial variations of ^{134}Cs and ^{137}Cs concentrations indicated that FDNPP-derived radiocesium around the western Japanese Archipelago, including the Sea of Japan, has been supported by the Kuroshio Current and its branch, Tsushima Warm Current, during 2013–2016. However, in the Sea of Japan, the penetration of ^{134}Cs was limited to depths of less than ~200 m during three years following the re-delivery of FDNPP-derived radiocesium.

1. Introduction

The massive East Japan Earthquake and the tsunami that subsequently hit the Pacific Ocean side of northeastern Japan on March 11, 2011 severely damaged the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), releasing large amounts of ^{134}Cs (half-life: 2.06 y) and ^{137}Cs (half-life: 30.2 y) over an extensive ocean and land area, particularly in eastern Japan (Hirose, 2016). To assess the migration pattern of radiocesium, the surface seawater concentrations of ^{134}Cs and ^{137}Cs have been extensively examined for highly contaminated regions (i.e., those around the FDNPP and the transition area between the subtropical Kuroshio Current and the subarctic Oyashio Current) (Tsumune et al., 2012; Buesseler et al., 2012; Povinec et al., 2013; Takata et al., 2016) and wide areas of the Northern Pacific Ocean (Kumamoto et al., 2014, 2015). Previous studies of radiocesium concentrations in surface water samples from the Sea of Japan during 2010–2012 indicated that low level ^{134}Cs (< 1.4 mBq/L) dispersed in waters of the Sea of Japan and rapidly decreased to the pre-FDNPP accident level until October 2011 (Inoue et al., 2012a, 2012b; 2013a).

In general, concentration levels and distribution pattern of radiocesium in seawater are drastically affected by motion of water mass and mixing, necessitating continuous analyses to assess the fate of FDNPP-derived radiocesium in global marine environments following the FDNPP accident. Although statistical analysis (Takata et al., 2017) revealed that the ^{137}Cs concentrations in seawater of the Sea of Japan started to increase in 2013, detailed oceanographic evidence for the transport of FDNPP-derived radiocesium to the Sea of Japan requires the detection and quantitation of ^{134}Cs. Herein, low-background γ-spectrometry was used to examine the temporal and spatial variations of low ^{134}Cs and ^{137}Cs concentrations in the Sea of Japan and other areas around the western Japanese Archipelago during 2011–2016. Radiocesium concentrations in water (*KK* series water samples; see details in section 2.1.) have also been systematically monitored by the Marine Ecology Research Institute (contracted by the Japanese Ministry

* Corresponding author.
E-mail address: i247811@staff.kanazawa-u.ac.jp (M. Inoue).

https://doi.org/10.1016/j.jenvrad.2017.11.032
Received 8 August 2017; Received in revised form 15 November 2017; Accepted 24 November 2017
Available online 08 December 2017
0265-931X/ © 2017 Elsevier Ltd. All rights reserved.

of Education, Culture, Sports, Science and Technology (MEXT)) from May 2011 to March 2013 and by the Secretariat of the Nuclear Regulation Authority of Japan (NRA) from April 2013 to the present time. Thus, we also analyzed MEXT and NRA (2017) monitoring data on radiocesium (^{134}Cs, ^{137}Cs) concentrations in seawater collected from 2011 to 2016 and evaluated the radiocesium inventory in seawater.

In our previous studies, we used the variation of the ^{228}Ra/^{226}Ra ratio (and ^{228}Ra concentration) to clarify surface water migration in the Sea of Japan and the East China Sea (ECS) (Inoue et al., 2007, 2017a). After the FDNPP accident, soluble radium isotopes were often employed to study the migration pattern of radiocesium in and around the Sea of Japan (Inoue et al., 2013a, 2014) and the northwestern North Pacific Ocean (Inoue et al., 2016). Herein, we build on the previously obtained distributions of ^{228}Ra and ^{226}Ra concentrations in the ECS and the Sea of Japan (Inoue et al., 2007, 2017a) and further characterize the processes controlling the variations of radiocesium during five years after the FDNPP accident.

2. Materials and methods

2.1. Seawater samples

The locations of seawater sampling sites used in the present study are shown in Fig. 1. Surface water samples (50 L) were collected at five coastal (sites *SM*, *FK*, *IS*, *NI*, and *HK*) and two offshore sites (site *YB* in the Yamato Basin and site *JB* in the Japan Basin) in the Sea of Japan, and at five sites in the ECS–Pacific coastal areas (sites *SG, KG, EH, SZ,* and *IB*) in May and June 2011–2016 during the research vessel expeditions of Kaiyo Engineering Co., Ltd. (*KK11–16* expeditions) (Fig. 1a). At all coastal sites, near-bottom waters (10–20 m above the bottom) were collected together with surface waters. Exceptionally, surface and near-bottom water samples (20 L) collected at sites *SG*, *SM*, *FK*, *IS*, *NI*, *HK*, *YB*, and *JB* in May 2011 were for our previous study (Inoue et al., 2013a). Herein, sample name is denoted by the combination of the expedition abbreviation, expedition year and sampling site name, e.g. *KK12-HK* indicates site *HK* in the expedition by the vessel of Kaiyo Engineering in 2012.

Ten (~20 L) and six (~40 L) coastal–offshore surface water samples were also collected in September 2015 and 2016 during the expeditions of *Hokko Maru* (*HKK15* samples) and *Tenyo Maru* (*TY16* samples) (Fig. 1b). A total of fourteen coastal surface water samples (~20 L) were collected in August 2012, May 2013, and May 2016 during the expeditions of *Mizuho Maru* (*MZ12, MZ13,* and *MZ16* samples) (Fig. 1c). In July 2016, ~60 L seawater samples (surface, ~20 L) were collected from water columns in the Sea of Japan (*SY16-C2* and *SY16-E2*) together with three surface water samples during the expedition of *Soyo Maru* (*SY16* samples) (Fig. 1d). All water samples were unfiltered.

2.2. Chemical separation and γ-spectrometry

Cesium-134 and ^{137}Cs were quantitatively separated by co-precipitation with ammonium phosphomolybdate (AMP) by adding CsCl and AMP to water samples (~20–60 L), and 4, 8, or 14 g AMP/Cs fractions were prepared (Inoue et al., 2013b). For *KK11-FK, -KG, -EH, -SZ,* and *-IB* samples and *KK12–16-YB* and *-JB* samples, radiocesium was purified using a cation-exchange column and precipitated as Cs/chloroplatinate after AMP separation.

The obtained AMP samples were characterized by low-background γ-spectrometry using specially designed Ge detectors at the Ogoya Underground Laboratory in Japan (Hamajima and Komura, 2004). The ^{134}Cs (605 keV) and ^{137}Cs (662 keV) concentrations in AMP/Cs fractions were calibrated using a mock-up sample prepared using radiocesium leached out from contaminated soil collected around Fukushima (see Inoue et al., 2013b). The detection limits of ^{134}Cs for ~3 days counting were ~0.08 mBq/L and ~0.03 mBq/L by using 20 L and 50–60 L seawater sample, respectively. We defined the detection limit of γ-spectrometry as the concentration having a counting statistics error of < 1/3 of value. The counting statistics errors of data in this report are in ranges of 5–43% for ^{134}Cs and as 3–10% for ^{137}Cs (Table S1). Although some data for ^{134}Cs are below the detection limit, those are used in considering their important role given in our discussion.

The ^{134}Cs and ^{137}Cs concentrations of *KK11-HK, -NI, -IS, -SM, -SG, -YB,* and *-JB* water samples were reported previously (Inoue et al., 2013a). We also quoted previously obtained data for 2010–2012 (*SY10*, *OS11*, *AS11*, and *MZ11* waters; Inoue et al., 2012a, 2012b; 2013a) providing additional insights.

3. Results and discussion

3.1. Temporal variation of radiocesium levels

The results of γ-spectrometric seawater sample analyses are shown in Table S1 in supporting data. The temporal variations of ^{134}Cs and ^{137}Cs concentrations between 2010 and 2016 observed in surface water samples collected in coastal and offshore areas in the Sea of Japan and the ECS–Pacific coastal areas, excluding *KK11–16-IB* waters and low-salinity (25–32) coastal waters (*MZ13-1–4*), are presented in Fig. 2.

Around the Japanese Archipelago, the concentrations of global fallout–derived ^{137}Cs gradually decreased from the 1960s, except for the sharp anomaly in 1986 caused by the Chernobyl reactor accident (Ikeuchi, 2003; Oikawa et al., 2013a; Takata et al., 2017). In July 2010, just before the FDNPP accident, the surface concentration of ^{134}Cs around the western Japanese Archipelago was below the detection limit (< 0.1 mBq/L), and that of ^{137}Cs was 1.3–1.6 mBq/L, with a mean of 1.47 mBq/L (*SY10-s20–s36*; Inoue et al., 2012a, 2012b).

In May–June 2011, the surface concentrations of ^{134}Cs and ^{137}Cs on the sampling date showed wide variations in the ranges of < 0.1–1.4 and 1.5–3.3 mBq/L, respectively, being maximal in the northeastern Sea of Japan (Inoue et al., 2012a, 2012b) (Fig. 2a). Based on the radioactive depositions distribution within the Sea of Japan (Yasunari et al., 2011), the radiocesium input was ascribed to inhomogeneous dispersion of FDNPP-related radioactive depositions. Subsequently, the surface concentrations of ^{134}Cs and ^{137}Cs in the Sea of Japan steeply decreased, reaching pre-accident levels in October 2011. This result was attributed to the exchange of contaminated water with the uncontaminated water of the Tsushima Warm Current (TWC) migrating northeastward along Honshu Island after passing the Tsushima Strait (Inoue et al., 2012b). Afterwards, the ^{134}Cs and ^{137}Cs concentrations in surface waters kept pre-accident values in May 2012 (*KK12* samples) and August 2012 (*MZ12* samples).

In May 2013, a low surface concentration of ^{134}Cs (< ~0.2 mBq/L on sampling date) was detected in the Sea of Japan (Fig. 2a), corresponding to a decay-corrected value of ~0.5 mBq/L on the FDNPP accident date (Fig. 2b). The ^{137}Cs concentration (~2 mBq/L) in 2013 exceeded the pre-accident level (~1.5 mBq/L), being equivalent to that recorded in the 1990s in the coastal areas of the Sea of Japan (Oikawa et al., 2013a; Takata et al., 2017). Herein, the temporal variations of radiocesium concentrations over five years were compared after being decay-corrected to the FDNPP accident date (March 11, 2011) (Fig. 2b). The thus obtained values exhibited a slight gradual increase from 2013 to 2016, being ~0.5–1 mBq/L for ^{134}Cs and 2–2.5 mBq/L for ^{137}Cs. On the other hand, as discussed below, a few sites in the Sea of Japan (sites *KK13–16-YB*, *-JB*, and *-HK*) displayed lower radiocesium concentrations than other sites.

3.2. Source of radiocesium delivered to the sea of Japan

The correlation between ^{134}Cs and ^{137}Cs levels in surface seawater samples decay-corrected to the FDNPP accident date is presented in Fig. 3, indicating that the level of ^{134}Cs in 2010–2016 is positively correlated to that of ^{137}Cs (r = 0.939, n = 145). Regression analysis

Download English Version:

https://daneshyari.com/en/article/8080875

Download Persian Version:

https://daneshyari.com/article/8080875

Daneshyari.com