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Impacts of Fukushima Daiichi Nuclear Power Plant accident on the Western North Pacific and the China Seas: Evaluation based on field observation of ^{137}Cs

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ABSTRACT

After the Fukushima nuclear accident (FNA), ^{137}Cs activities of seawater in the Western North Pacific (WNP) and China Seas were determined in order to examine whether ^{137}Cs derived from FNA across the Kuroshio was still existed and if there were any indications of FNA's impact. High ^{137}Cs activities at 200–500 m were observed in the south of Kuroshio during 2015, indicating ^{137}Cs from FNA could cross the Kuroshio. Surface ^{137}Cs activities were on average 1.43 ± 0.42 Bq m^{-3} for WNP, 1.11 ± 0.14 Bq m^{-3} for South China Sea (SCS) and 1.10 ± 0.29 Bq m^{-3} for East China Sea (ECS) during 2011–2015. The activities and inventories of ^{137}Cs were almost identical before and after the FNA, indicating the impacts of FNA on the WNP and China Seas were minor. The effective environmental half-lives of ^{137}Cs in surface seawater were firstly estimated to be 14.4 ± 1.7 yrs for ECS and 16.9 ± 2.1 yrs for SCS.

1. Introduction

The devastating tsunami of March 2011 induced by the 9.0 magnitude Tohoku earthquake, resulted in unprecedented radioactivity releases from the Fukushima Nuclear Accident (FNA) to the oceans. Over 80% of the total radioactivity released eventually entered the oceans through the various routes, such as atmospheric deposition and the direct discharge and leaking of the contaminated coolant water (Buesseler et al., 2011; Morino et al., 2011; Tsumune et al., 2012; Kanda, 2013). While there reported various radionuclides released, after the initial decay of those contaminants with half-lives less than days to weeks, much of the attention still focused on ^{137}Cs ($t_{1/2}$ = 30.07 years) because there was released in larger amount relative to other radionuclides (Steinhauser, 2014). The total amount of ^{137}Cs released ranged from 4 to 90 PBq (1 PBq = 10^{15} Bq), but with most estimates of the combined releases in the range 15–30 PBq (Chino et al., 2011; Tsumune et al., 2013; Buesseler, 2014; Aoyama et al., 2016). Meanwhile, due to its radiotoxic and relatively long half-life, the evaluation of ^{137}Cs in the large scope marine environment is crucial in addressing risks concerning marine ecosystems and public health through consumption of seafood.

The Kuroshio, important western ocean boundary current, originates from the northward bifurcation of the North Equatorial Current (NEC) which flows westward to the Philippine Seas and then northeastward along the boundary of both the China Seas and the south and east coasts of Japan (Wang et al., 2011).The Fukushima nuclear power plant is located in the southern part in the Kuroshio-Oyashio transition area and close to the Kuroshio extension which is the strongest jet off the east coast of Japan (Mizuno and White, 1983). Therefore, the Kuroshio is deemed to be the greatest barrier to prevent the radioactive contaminants originating from the FNA from southward dispersion, but is well contributed to the dispersion of the contaminants far eastward in the North Pacific (Buesseler et al., 2012; Rypina et al., 2013). The model results also reveal the released ^{137}Cs into the Western North Pacific (WNP) dispersed eastward broadly flowing the Kuroshio and its extension (Kawamura et al., 2011; Tsumune et al., 2012). As such, the ^{137}Cs concentrated upon the north of Kuroshio in the WNP has been extensively investigated (e.g. Buesseler et al., 2011, 2012; Aoyama et al., 2013; Tsumune et al., 2013), but only sparsely for the south of the Kuroshio and adjacent marginal seas, such as the China Seas. More recently, several studies revealed the radioactive Cs derived from the FNA following the North Pacific Subtropical Mode Water (STMW) could cross the Kuroshio extension and southward spread in the WNP during the period 2012–2013 (Kaeriyama et al., 2014; Kumamoto et al., 2014; Men et al., 2014). Moreover, atmospheric deposition of radioactive Cs in the south of the Kuroshio was also strongly suggested by atmospheric

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and oceanic dispersion simulations (Kawamura et al., 2011; Kobayashi et al., 2013). However, the ^{137}Cs released from FNA via the STMW and atmospheric deposition has not been assessed for the basin area of the WNP and the China Seas.

The present study thus aimed to examine whether ^{137}Cs derived from FNA across the Kuroshio was still existed within 4 years after accident and if there were any indications of the FNA's impact on the WNP (15–40° N, 110–160° E, focusing on the Kuroshio zone and far southern region of the Kuroshio) and the China Seas based on the four cruises during the period 2011–2015. Moreover, the effective environmental half-life of ^{137}Cs in the China Seas was firstly estimated in order to further assess the future environment impact. Finally, the information on Cs activity level would also help in establishing a baseline for future environmental risk assessment related to nuclear power plant operations that see a dramatically increasing rate along the coast of the region.

2. Materials and methods

2.1. Study area

The China Seas, mainly including the South China Sea (SCS), East China Sea (ECS) and Yellow Sea (YS), are located in the Northwest Pacific Ocean. The surface circulation is primarily governed by the East Asian Monsoon with the strong northeast monsoon prevailing from September to April and the relatively weak southwest monsoon from May to August (Liu et al., 2003), and which is leading to a seasonal alternation, i.e., surface circulation in the SCS with an anti-cyclonic gyre in summer and a cyclonic gyre in winter (Shaw and Chao, 1994), coastal current in the ECS flowing to the southward direction in winter and the Taiwan Current flowing to the northward direction in summer (Lee and Chao, 2003) (Fig. 1a). In addition, the China Seas have a unique circulation pattern with dynamic exchanges with the WNP via the western ocean boundary current, i.e., Kuroshio Current. The Kuroshio extension pathway to the SCS through the Luzon Strait presents a seasonal pattern with the intrusion being stronger in winter than in summer (Shaw, 1991). Subsequently, the Kuroshio branch current intrudes into the ECS at the northeast corner of Taiwan, while the Kuroshio mainstream continues to flow along the Japan in a northeast direction (Ichikawa and Beardsley, 2002) (Fig. 1a).

Before the FNA, the major ^{137}Cs source in the WNP and the China Seas was mainly from the atmospheric nuclear weapon tests conducted in the late 1950s and early 1960s (Povinec et al., 2004; Aoyama et al., 2011). The ^{137}Cs activities in surface water exponentially decreased from 10 to 100 Bq m^{-3} in the 1960s to 2–2.5 Bq m^{-3} in 2000 (Hirose and Aoyama, 2003a). Temporal changes of ^{137}Cs activity are determined by the apparent residence time of ^{137}Cs (15–26 years), radioactive decay, lateral and vertical removal processes (Hirose and Aoyama, 2003a; Povinec et al., 2005). Before the FNA, the ^{137}Cs concentrations in surface seawater in the WNP and the China Seas were estimated to be 1–2 Bq m^{-3} (Buesseler et al., 2012; Aoyama et al., 2013).

2.2. Sample collection

Sampling was conducted onboard the R/V *Dongfanghong II* in spring 2011, 2014 and 2015, and the R/V *Kexue I* in winter 2014. Basic sampling information is presented in Table 1 and the sampling locations are shown in Fig. 1b. Profile samples at stations DH23 and K6 were collected from Niskin bottles on a conductivity-temperature-depth (CTD) rosette system in spring 2011 and 2015. The remaining samples were all collected from the surface using a built-in pumping system. Upon collection, seawater samples (about 100 L each) were acidified with concentrated nitric acid to a pH of approximately 1.6. They were then shipped to a shore-based laboratory in preparation for Cs isotope analysis.

2.3. Analysis of ^{137}Cs in seawater

As described in the previous works (Aoyama et al., 2000, 2013; Wu et al., 2013), ^{137}Cs in seawater was concentrated by adsorption onto ammonium molybdophosphate (AMP) using a modified method. In brief, a stable Cs^+ carrier (CsCl solution) was added to the seawater samples as a yield determinant and continuously stirred for ~30 min, and then an aliquot of AMP was added and continuously stirred again for ~2 h. After standing for 18–24 h, the supernatant was siphoned off and the precipitate was filtered on a quantitative membrane filter. The AMP/Cs compound was dried at 70–80 °C for > 48 h and then weighed. The weight yield of the AMP/Cs compound was 96.7 ± 1.8% (n = 35). Finally, the dried AMP/Cs precipitates were placed into a plastic container before gamma counting. The ^{137}Cs activities in the AMP/Cs compounds were determined by gamma spectrometry using a low background type high-purity germanium detector (GCW4022). The ^{137}Cs activities were measured by comparing the photopeak area (662 keV for ^{137}Cs obtained by > 432,000 s of counting of the sample) with that of the standard sample, which had the same geometry and was tagged with known activity. The average detection efficiency and detection limit of γ-spectrometry were 5.0% and 0.25 Bq m^{-3} (95% confidence level), respectively.

2.4. Data treatment and statistical analysis

The ^{137}Cs activities are calculated by counting time, net peak counts, detection efficiency, chemical recovery, and sample volume. They are all decay-corrected back to the date of collection and their uncertainties are at one sigma based on counting statistics and standard propagation of errors. In addition, to examine if ^{137}Cs activities have a significant difference in the WNP and the China Seas, we applied a simple F-test and *t*-test in detail described in elsewhere (Lettner et al., 2000). In brief, the probability values at the F-test ($p > 0.05$) and the student-test ($p > 0.05$) show the two sets of data having the homogeneous variance and no significant difference between them, respectively; the opposite is also true.

3. Results and discussion

3.1. Temporal and spatial distributions of ^{137}Cs activity in the WNP and China Seas surface waters

The ^{137}Cs activities of surface seawater in the WNP and the China Seas during the period 2011–2015 are presented in Table 1. The temporal and spatial distributions of surface ^{137}Cs activities in the WNP and the China Seas are shown in Fig. 2.

As shown in Fig. 2a, the surface ^{137}Cs activities in the WNP and the China Seas had no significant difference (student t-test, $p = 0.50 > 0.05$) during 2011–2015. Meanwhile, we observed that the difference in ^{137}Cs activities at two adjacent stations at P5 and N2 collected in spring and winter 2014 was < 8%, comparable to their relative standard deviations. The variation of ^{137}Cs activities at another two adjacent stations at LU63 and P3 collected in spring 2011 and 2014 was also below 9%. Therefore, we suggested the seasonal variation was minor in the investigation period. For the ease of presentation, we combined the four datasets resulting from four cruises over four consecutive years in spring and winter. This assumption was also based on the fact that the surface Cs distribution is rather homogenous and significant changes of Cs cannot be expected in a time scale of 1–4 years, because of the long residence time of Cs in the ocean and the long half-lives (Hirose and Aoyama, 2003b).

In the WNP, the ^{137}Cs activities ranged from 0.87 to 2.28 Bq m^{-3} with an average of 1.43 ± 0.42 Bq m^{-3} (n = 15). They were comparable to those reported for 2012 in relation to an area adjacent to our investigation area (1.81–1.90 Bq m^{-3}) (Kamenik et al., 2013). The spatial distribution of ^{137}Cs activities showed a slight increase trend