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Baseline

Spatial and vertical distribution of radiocesium in seawater of the East China Sea

Lijun Zhao[a], Dantong Liu[a], Jinlong Wang[a,*], Jinzhou Du[a], Xiaolin Hou[b], Yifei Jiang[a]

[a] State Key Laboratory of Estuarine and Coastal Research, East China Normal University, Shanghai 200062, China
[b] Xi'an AMS Center and KLLQG, Institute of Earth Environment, Chinese Academy of Science, Xi'an 710075, China

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ABSTRACT

The ^{137}Cs activity in surface water of the East China Sea (ECS) was 0.66–1.36 Bq m^{-3} during May of 2011. The low activities were observed in the Changjiang Estuary and Zhejiang-Fujian coast and high activities were observed in the south offshore and Kuroshio Current pathway, suggesting that the influence from the current system in the ECS. The ^{134}Cs were undetectable (< 0.03 Bq m^{-3}) and the contribution of the Fukushima accident to ECS is estimated to be below 3%; hence it is negligible during the investigation period. Using the vertical profiles of ^{137}Cs in the ECS, the mass balance is obtained, which suggests that the oceanic input dominates the ^{137}Cs source in the ECS. ^{137}Cs is potentially useful to trace water mass movement in the ECS. Our study provides comprehensive baseline of ^{137}Cs in the ECS for evaluation of the possible influence of the nuclear power plants in the future.

Radiocesium (^{134}Cs and ^{137}Cs) is derived from nuclear explosions since the world's first atomic bomb explosion in 1945, nuclear accidents, nuclear reprocessing facilities and nuclear power plants. The thermonuclear bomb testing mainly contributed to the global fallout ^{137}Cs (~900 PBq) (Hu et al., 2010). The nuclear accidents also released a bunch of ^{137}Cs to the earth environments (e.g., 85 PBq for Chernobyl) (UNSCEAR, 2008). Most recently, the accident happened in the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) has released large amount of ^{137}Cs to the atmosphere (12 PBq) (Steinhauser et al., 2014). ^{137}Cs entered into the stratosphere was finally deposited on the earth globally, while those entered troposphere mainly distribute locally. Aarkrog (2003) estimated that around 603 PBq ^{137}Cs was input to ocean since 2000. Thus, ^{137}Cs is used for monitoring the nuclear contaminant around the nuclear reprocessing facilities and nuclear power plants (NPPs). The FDNPP derived radionuclides have dispersed and deposited almost all over the north hemisphere (Buesseler, 2012; Wang et al., 2012; Evrard et al., 2015). Among them, substantial fraction of them deposited to the ocean (Buesseler et al., 2012; Inoue et al., 2012; Suzuki et al., 2013). Investigations have shown the significantly increased level of ^{134}Cs and ^{137}Cs in the seawater samples in the offshore of Fukushima, as well as a large area in the Northwest Pacific Ocean (Honda et al., 2012; Povinec et al., 2013; Tumey et al., 2013).

Due to the relative conservative feature of ^{137}Cs in the ocean (Hirose et al., 1992), it has being utilized to trace the water mass and currents, and is useful to inspect and modify the ocean circulation model (Garraffo et al., 2016). East China Sea (ECS) is a broad and flat-continental shelf marginal sea of the Northwest Pacific. The complex current system and high concentration of suspended particles makes the ECS one of most important “sinks” of radionuclides and nutrients transported from the Northwest Pacific (Wang et al., 2017b). Besides, the surface of this area indicates a signature of “recirculated” seawater originates from intermediate layers of subtropical North Pacific (Miyazawa et al., 2009). However, the observation of ^{137}Cs around the ECS after accident was very limited. Wu et al. (2013) reported that the seawater ^{137}Cs was 1.08 ± 0.09 Bq m^{-3} in a few seawater samples (n = 8) and they suggest that FDNPP's accident might significantly enhance the ^{137}Cs level in the ECS, however, this database is too limited to assess the influence of FDNPP's accident associated with the transport of ^{137}Cs with water mass in the ECS. Wang et al. (2012) reported that ^{137}Cs activity in aerosols in Shanghai (about 50 km of the coast in the ECS) during 28th–29th March 2011 was 0.12 ± 0.09 mBq m^{-3}, which is significantly higher than the background level (0.04 mBq m^{-3}) before accident. These results urge a comprehensive observation of spatial and vertical distribution of ^{137}Cs in the ECS.

This work aims to investigate the level, distribution and source of radioactive cesium (^{134}Cs and ^{137}Cs) in the ECS by analyzing surface and profile seawater samples collected in the ECS during June 2011 in order to explore the impact of Fukushima accident in the ECS. Meanwhile, it can be also used to trace pathway of different water masses, and to supply a baseline for monitor the potential risk from 14

* Corresponding author.
E-mail address: jlwang@sklec.ecnu.edu.cn (J. Wang).

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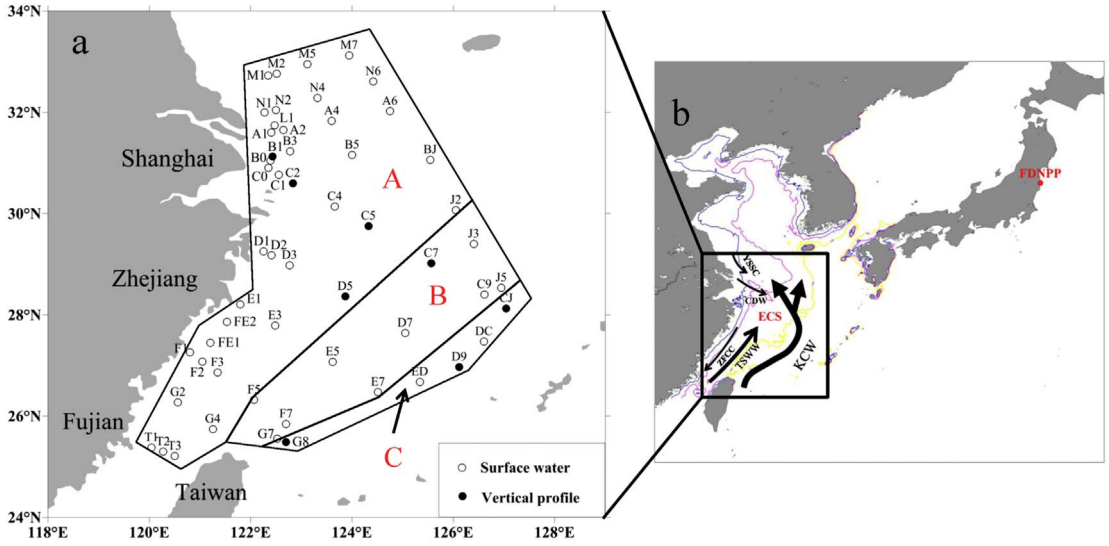

Fig. 1. (a) Sampling stations and water masses in the Changjiang Estuary and adjacent seas, solid circles represent vertical profile stations and hollow circles represent surface water stations, (b) Geographical thumbnail of water masses from the East China Sea (ECS) and the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). Changjiang Diluted Water (CDW, summer), Taiwan Strait Warm Water (TSWW), Kuroshio Current Water (KCW) and Yellow Sea Surface Current (YSSC, summer) (modified from Su and Huh, 2002; Huang et al., 2013). The study area is divided into three parts, A: the water depth below 100 m, B: the water depth between 100 m and 1000 m, C: the water depth over 1000 m.

operating NPPs (as of May 2011) along the Chinese coast in the ECS as well as from the other nuclear facilities, nuclear accidents and other sources (e.g., oceanic input).

The ECS is a typical marginal sea and locates in the west of the Northwest Pacific Ocean (Fig. 1), receiving large amount of fresh water and sediment discharge from the Changjiang (Yangtze River), which is the largest river over the Euro-Asia continent (Dai et al., 2014). The ECS could be divided into the shallow western continental shelf and deep eastern continental shelf on the basis of the terrain of northwest high, southeast low. ECS owns complex water masses that interact with each other, such as the Changjiang Diluted Water (CDW), which extends at first along the estuary to the southeast until 122.50°E and then turns southward to form a tongue of diluted water in winter. The Zhejiang-Fujian Coast Current (ZFCC) is strong in winter and relatively weak in spring. The Kuroshio Current Water (KCW), a major western boundary current along the outer edge of the continental shelf, carrying a lot of high temperature and high salt water. The northward Taiwan Strait Warm Water (TSWW), with its upper water formed by the mixing of Kuroshio Current Surface Water and the Taiwan Strait Water, is originating from Kuroshio Current Subsurface Water (KCSSW) east of Taiwan (Lee et al., 2001; Lie and Cho, 2002; Lee and Chao, 2003; Lee and Takeshi, 2007). In the north part of the ECS shelf, the Yellow Sea Surface Current (YSSC) also participates in the structure of the ECS water masses (Su, 2001).

Seawater samples, including 55 surface seawater samples and 8 water profiles, were collected from 55 stations in the ECS (Fig. 1a) during the R/V "Shiyan 3" cruise between 11th May and 5th June 2011. An aliquot of 100 L seawater sample was collected from surface (0–0.5 m) and different depths of water columns by a submersible pump and a CTD rosette system, respectively. And then the samples were filtered by 0.5 μm pore-size pre-cleaned special polypropylene cartridge (Wang et al., 2017a).

Cesium in seawater sample was separated by adsorption using ammonium phosphomolybdate (AMP) at pH ~1.5 (Hirose et al., 2005; Yamada et al., 2006). Twenty five mg of CsCl was first spiked to 100 L seawater as the carrier and used for chemical yield tracer. The spiked seawater was stirred and equilibrated for 6 h. Forty grams AMP power (seawater) was added to each sample, the samples was then stirred for 30 min. After 24 h settlement the supernatant was siphoned out. The slurry with AMP/Cs precipitation was transferred to a 2 L plastic bottle and transported to the lab for further treatment. In the laboratory, the slurry was centrifuged to separate AMP-Cs precipitate, which was finally freeze-dried by a lyophilizer for radioactivity measurement.

Cesium-134 and Cesium-137 in the prepared samples were measured using a gamma spectrometer (GWL-120210-S, ORTEC) 604.7 keV (branch ratio = 97.6%) and 661.7 keV (branch ratio = 85.1%) gamma rays were used to calculate the concentration of ^{137}Cs and ^{134}Cs, respectively. The detection limits (LLD) of the gamma spectrometry system for both radionuclides were estimated to be 0.03 Bq m^{-3} for 100 L seawater. The chemical recoveries of about 78% for cesium were obtained by measuring of the stable Cs in the AMP precipitation after separation using atomic absorption spectrophotometer (America PE, AA-800). For analytical quality control, the IAEA standard source (IAEA-447) was analyzed for ^{137}Cs, the measurement results agreed with the certificated value.

The salinity and temperature in surface water of the ECS are plotted in Fig. 2. The temperature was 14.8–26.2 °C with a mean value of 20.1 °C, and the salinity was 24.2–34.6 with a mean value of 32.2. The temperature decreased from northwestern to southeastern and increased from the coast to offshore. The lowest temperature was observed in the north offshore of the ECS, and the highest value was observed in the south part of the ECS, especially in KCW pathway. The salinity increased from north to south and from west to east. The ^{137}Cs activities in all surface seawater samples from the ECS are shown in Table 1, and the range was 0.66–1.36 Bq m^{-3} with a mean value of 1.07 ± 0.09 Bq m^{-3} (n = 55). Lower ^{137}Cs activities were observed in the Changjiang Estuary, Zhejiang-Fujian coast and section C (from C0 to CJ) (Fig. 3). Higher values were observed in the northeast offshore and south offshore of the ECS (Fig. 3). Generally, the ^{137}Cs observed in this study is lower than the Yellow Sea (1.30–3.04 Bq m^{-3} with a mean value of 2.35 ± 0.44 Bq m^{-3}, Kim et al., 1997), Japan Sea (0.3–20.0 Bq m^{-3} with a mean value of 1.9 ± 0.1 Bq m^{-3}, Povinec et al., 2005) and the Western Pacific Ocean (0.5–23.7 Bq m^{-3} with a mean value of 1.8 ± 0.1 Bq m^{-3}, Povinec et al., 2005), and meanwhile is similar to the South China Sea (1.14–1.41 Bq m^{-3} with a mean value of 1.28 ± 0.08 Bq m^{-3}, Wu et al., 2013).

The ^{137}Cs in five vertical profiles (B1, C2, C5, C7 and D5) are slightly higher in bottom layer compared to the surface layer and generally showed the stable vertical distribution. These distribution patterns are similar with the profiles of temperature and salinity distribution (Fig. 4). This should be attributed to the sufficiently mixing derived by the complex current system. The vertical profiles of ^{137}Cs