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Spatial and vertical distribution of $^{129}$I and $^{127}$I in the East China Sea: Inventory, source and transportation

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Abstract:

Iodine-129 is useful to trace water mass movement in the ocean. In this study, the concentration of iodine isotopes in seawater of the East China Sea (ECS) in October 2013 were analyzed to investigate the spatial and vertical distribution of $^{129}\text{I}$ and $^{127}\text{I}$ so as to indicate the water mass exchange in the ECS. The results showed that the $^{129}\text{I}/^{127}\text{I}$ atomic ratios varied with the water mass flowing, with higher values of $(10-20)\times10^{-11}$ in the coastal regions and lower values of $<8\times10^{-11}$ in the offshore. The inventories of $^{129}\text{I}$ were estimated to be $(0.23-1.7)\times10^{12}$ atoms m$^{-2}$ ($n=18$) in upper 100 m waters, which is comparable to those of other regions without being contaminated by the nuclear accidents or nuclear reprocessing facilities. The total amount of $^{129}\text{I}$ in the ECS water column was estimated to be 88 g in which over 90% is attributed to the oceanic input (e.g., West Pacific) via the Kuroshio Current (KC), etc. The contributions of $^{129}\text{I}$ from the Changjiang input ($<7.5\%$) and atmospheric fallout ($<2.7\%$) were small. Those from the Fukushima accident was negligible during this investigation. The $^{129}\text{I}/^{127}\text{I}$ ratios vs. salinity distribution showed the distribution range and stratification of the Changjiang, Yellow Sea and KC waters in the ECS. Our study shows that the Changjiang fresh water could be transported to the North Jiangsu coast in October; the Taiwan Warm Current water could intrude to Northern part of the Changjiang Estuary (32 °N). Besides, our results suggest that the $^{129}\text{I}/^{127}\text{I}$ profile is useful to indicate the seawater mixing process in ocean marginal systems.

Key words: $^{129}\text{I}/^{127}\text{I}$; Vertical profile; Water mass; Inventory; the East China Sea
1. Introduction

Iodine-129 ($^{129}$I) (with a half-life of 15.7 Myrs) in the environment is derived from natural processes (250 kg) through cosmic ray reactions with xenon and the fission of uranium, thermonuclear bomb testing (50-150 kg), nuclear accidents (e.g., Chernobyl (1.3-6 kg) and Fukushima (1.2 kg)), nuclear power plants and nuclear reprocessing facilities (6500 kg, mainly contributed by La Hague, France and Sellafield, UK) (Aldahan et al., 2007a; Hou et al., 2009; 2013; He et al. 2013). Considering its long half-life and relatively low beta-energy (with a maximum beta-emitted energy of 154 keV), $^{129}$I is not very radiologically harmful in common situation. Instead, $^{129}$I is widely utilized as a tracer for the biogeochemical cycle of iodine, the movement and exchange of water masses, and the interactions between atmosphere and seawater in marine environments (Hou et al., 2007; Zhang and Hou, 2013; Xing et al., 2017). Due to its high solubility (K$_{d}$: 5-30 L kg$^{-1}$, Takata et al., 2013) and long residence time in the ocean (~300 kyrs) compared to the turnover time of ocean water (~1000 kyrs, Broecker and Peng, 1982), it can be utilized to trace the cycling of dissolvable organic carbon and phytoplankton blooming associated with reductive conditions (Schwehr et al., 2005; Hou and Hou, 2012; Liu D. et al., 2016).

Iodine-127 ($^{127}$I) is a stable isotope, and $^{129}$I/$^{127}$I atomic ratios (described as ratios in the following text) largely vary depending on the source of iodine in the marine environment. These ratios range from $3 \times 10^{-12}$ to $2 \times 10^{-6}$, which are higher than the prenuclear level ($1.5 \times 10^{-12}$) (Fehn et al., 2000; Xing et al., 2017). So, $^{129}$I/$^{127}$I ratio is very useful for identifying the source of $^{129}$I and for water mass movements in marine environments. For example, in the Irish Sea and the North Sea, the extremely high $^{129}$I/$^{127}$I ratios in seawater ($\sim 10^{-6}$) were attributed to discharge from two nuclear fuel reprocessing plants at La Hague (France) and Sellafield (UK) (Raisbeck et al., 1995;
Hou et al., 2007; Michel et al., 2012). Meanwhile, iodine isotopes have also been used to trace the movement of sea currents from the coast of Northern Europe to the Arctic Ocean (Hou et al., 2000; Alfimov et al., 2004a). In the coastal ocean and marginal sea, water masses usually vary seasonally, especially in coastal areas strongly affected by monsoons, and they show different features in vertical profiles, e.g., the Kuroshio Current has different temperature and salinity characteristics in its surface and subsurface (Ichikawa and Beardsley, 2002). Thus, defining the spatiotemporal and vertical distributions of iodine isotopes in seawater is of fundamental importance for their effective use as tracers.

The East China Sea (ECS), as a typical ocean marginal sea, contains oceanic input currents, seasonally variable coastal currents and large river inputs. It has been reported that the turnover time of the ECS shelf water was approximately 1.3 yrs and that those of coastal and estuarine waters were much lower, about 30 days (Tan et al., 2018; Wang et al., 2018); these times are significantly lower than the residence time of $^{129}$I in ocean water. Although the solubility of $^{129}$Iodine is still highly soluble in estuaries ($K_d < 200 \text{ L kg}^{-1}$, Takata et al., 2013) compared to that in the ocean, it is still highly soluble, and it can thus be used to trace the exchange of water masses in marginal seas, even with abundant sediment discharge from rivers. The vertical profiles associated with the estimated inventory and spatial distribution of $^{129}$I and $^{127}$I are relevant to the long-range transport of other conservative-behavior radionuclides (e.g., $^3$H, $^{90}$Sr, $^{137}$Cs and U isotopes) and oceanic currents. In recent years, increased numbers of Nuclear Power Plants (NPPs) have been constructed along the coast of the ECS, such as Qinshan in Haiyan City, Tianwan in Lianyungang City and Sanmen in Taizhou City. The potential releases of anthropogenic radionuclides, including iodine isotopes, from these NPPs to the environment have
aroused profound concern. Thus, it is necessary to determine the baseline of concentrations of $^{129}$I and $^{127}$I and their movement behaviors in the ECS, which can provide vital information for evaluating the possible impacts of these NPPs.

Some studies have reported the level of $^{129}$I and $^{127}$I in the seawater of marginal systems in the Northwestern Pacific (e.g., Cooper et al., 2001; Povinec et al., 2013; Casacuberta et al., 2017), however, the time-series and vertical profiles of two isotopes are still limited, especially in the ECS (Suzuki et al., 2010; Hou et al., 2013). In our previous work (Liu D. et al., 2016), by observing the spatial distribution of iodine isotopes (i.e., $^{129}$I and $^{127}$I) concentration and species in the seawater of the ECS in August, we found that $^{129}$I decreased from the coast to offshore and the species of $^{129}$I and $^{127}$I were dominated by iodate in the shelf water but existed mainly as iodide in the Changjiang Estuary. We also found that the influence of Fukushima nuclear accident in the $^{129}$I level of the ECS was not detectable until August 2013. In the present work, we extended our research to focus on the inventory and temporal changes of $^{129}$I and $^{127}$I; we also quantified the source of $^{129}$I in the ECS seawater by observing the spatial distribution and vertical profile of $^{129}$I. The temporal distribution pattern can be established by combining with the data from our previous study (Liu D. et al., 2016). These results are expected to promote our understanding of not only the movement of iodine isotopes in estuarine and continental shelf areas but also the long-term transport of other conservative radionuclides by oceanic current.

2. Materials and methods

2.1. Study area

The ECS, which is one of the broadest shelves in the world, is a typical river-dominated marginal sea located between the Asian continent and the Pacific
The north border of ECS is the ligature between Qidong and Cheju Island, south border of ECS is located in the southern part of Taiwan Strait and the east border is the Kyushu Island, the Ryukyu Islands and Taiwan Island. ECS is ~400 km long and has a maximum width of ~640 km, with a mean and maximum water depth of 349 m and 2719 m, respectively. The ECS shelf is relatively flat, slanting from the continental shelf toward the southeast, with a mean gradient of 0.04% (Wang et al., 2016). There are more than 40 rivers flowing into the ECS, including the largest river in China and the third-largest river in the world, Changjiang, which discharges abundant sediment and freshwater to the ECS. Due to the heavy influence of monsoons, there are two seasonally variable coastal currents, and the Changjiang Diluted Water (CDW) exists in the ECS. The Yellow Sea Coastal Current (YSCC) and the Zhejiang–Fujian Coastal Current (ZFCC) are southward-flowing in winter and spring but reverse or even disappear in summer. The CDW flows southward in winter and spring but northeastward in summer. In the northern region of the ECS, the YSCC intersects with the northward Yellow Sea Warm Current (YSWC) and forms a loop current. From the open sea side, there are two northward-flowing currents, i.e., the Taiwan Warm Current (TWC) and the Kuroshio Current (KC). The KC is one branch of the boundary current in the Western Pacific, which comprises surface, subsurface, intermediate and deep-water regions. The surface water of the KC ranges from 80 m to 200 m and the subsurface water is below the surface water, with a maximum depth of 450 m; it is characterized by high salinity (~35 psu) (Ichikawa and Beardsley, 2002).

2.2. Sampling and analysis

Seawater samples, including 26 surface seawater samples (<2 m in depth) and 18 vertical profiles, were collected during the R/V “Dongfanghong 2” Cruise between
Among these samples, there were three samples (i.e., M2, M3 and M5) collected in the Southern Yellow Sea. Then, samples were filtered (Φ 0.45 μm) in situ and stored in 2 L polyethylene plastic bottles in the dark under for subsequent laboratory analyses. Meanwhile, the temperature and salinity of the seawater samples were measured in situ during sampling. The iodine isotopes data collected from the surface seawater samples during August 2013 were reported in our previous study (Liu D. et al., 2016).

The method for iodine separation from seawater was modified from Hou et al. (2010). Briefly, 1000 ml of filtered seawater were placed in a beaker and spiked with 0.2 mg of $^{127}$I carrier (Woodward iodine, Woodward Iodine Corporation, Oklahoma, USA) and 200 Bq $^{125}$I; then, 0.50 ml of 2.0 mol L$^{-1}$ NaHSO$_3$ solution was added. Then, 6 mol L$^{-1}$ HNO$_3$ was added to adjust the pH to < 2 to convert the iodine species to iodide. A total of 28 ml of 0.01 mol L$^{-1}$ AgNO$_3$ solution was slowly added while stirring to coprecipitate iodide as AgI-AgCl. After the supernatant was discarded, AgI-AgCl precipitate was separated by centrifuge and sequentially rinsed with 3 mol L$^{-1}$ HNO$_3$, H$_2$O, and 7.5%, 2.5%, and 1% NH$_3$·H$_2$O to remove Ag$_2$SO$_3$ and excessive AgCl and AgBr until 1-3 mg of precipitate was obtained for AMS measurements. The chemical yield of iodine in the chemical separation was obtained by measuring the $^{125}$I in the precipitate using a NaI gamma detector (Model FJ-2021, Xi'an Nuclear Instrument factory, Xi'an, China), which was found to be 85-95%. Procedural blanks were prepared using the same procedures as the samples. These blanks contained deionized water instead of samples.

The separated AgI–AgCl coprecipitate was dried in an oven at 60–70 °C, homogeneously pulverized, and mixed with niobium powder (325-mesh, Alfa Aesar, Ward Hill, MA) in a 1:5 mass ratio. The mixture was pressed into a copper holder
using a pneumatic press (Zhenjiang Aode Presser Instruments Ltd.). The $^{129}\text{I}/^{127}\text{I}$ atomic ratios in the prepared targets were measured by AMS using a 3MV Tandem AMS system (HVEE) in the Xi’an AMS center. All $^{127}\text{I}^{5+}$ ions were measured as charges (current) using a Faraday cup, and $^{129}\text{I}^{5+}$ was measured using a gas ionization detector. All samples were measured for 6 cycles, with 5 min per sample in each cycle. The procedural background $^{129}\text{I}/^{127}\text{I}$ ratio was measured to be $1.0 \times 10^{-13}$, which is approximately two orders of magnitude lower than that measured in the samples. The measurement uncertainty for the samples was less than 3.5% (Liu D. et al., 2016). A detailed description of the AMS system and $^{129}\text{I}$ measurements was reported by Hou et al. (2010).

The concentration of $^{127}\text{I}$ in seawater was measured by ICP-MS (Thermo Scientific, X series II, USA) after undergoing a 10-time dilution using 1% ammonium solution. The samples were spiked with Cs+ (2 ng ml$^{-1}$), which was also used as an internal standard. The detection limit of $^{127}\text{I}$ was determined to be 0.02 ng ml$^{-1}$, which is more than 2 orders of magnitude lower than the measured value in the diluted seawater.

3. Results

3.1. The spatial distribution of iodine isotopes in the ECS

The iodine isotopes, temperature and salinity values in the surface water of the ECS are listed in the supplemental file (Table S1). The $^{127}\text{I}$ concentrations in surface seawater ranged from 15.6 to 54.7 μg L$^{-1}$ (mean of 33.2±1.7 μg L$^{-1}$, n=26) in October, which were similar to those observed in August (Figure 3). The lowest $^{127}\text{I}$ concentrations were observed near the Changjiang Estuary, ranging from 15.6 to 25.0 μg L$^{-1}$. This phenomenon are comparable to the values in the water as the same as that observed in August of the same year (Figure 3). The lower $^{127}\text{I}$ concentrations...
in the Changjiang Estuary should be attributed to freshwater input with the low $^{127}$I concentration from the Changjiang River. Even lower $^{127}$I concentration of 3.1-7.1 μg L$^{-1}$ were observed in the three locations of the inner part of the Changjiang Estuary. The maximum values of $^{127}$I were found in the southeastern region of the ECS, and this trend was the same as that in August. However, the concentrations of $^{127}$I in the northeastern region of the ECS were much lower in October (26.0-38.5 μg L$^{-1}$) than that in August (41.8-47.4 μg L$^{-1}$). The $^{129}$I concentrations in the surface seawater of the ECS are (0.61-1.94) × 10$^7$ atoms L$^{-1}$ (mean of (1.22±0.09) × 10$^7$ atoms L$^{-1}$, n=26) in October, which are slightly lower than those in August. However, this value is 1-2 orders of magnitude higher than the pre-nuclear level of 0.043 × 10$^7$ atoms L$^{-1}$ (Snyder et al., 2010). The spatial distribution pattern of $^{129}$I is the opposite of that of $^{127}$I, i.e., $^{129}$I is higher in the Changjiang Estuary but relatively lower in the offshore seawater. The measured $^{129}$I concentrations in the surface seawater in this study are comparable to the reported values in surface water of the North Pacific (20°N–45°N) ((0.59–3.7) × 10$^7$ atoms L$^{-1}$) (Guilderson et al., 2014), Japan Sea ((0.28–5.8) × 10$^7$ atoms L$^{-1}$) (Cooper et al., 2001), and southern Indian Ocean ((0.60–0.80) × 10$^7$ atoms L$^{-1}$) (Povinec et al., 2011), but higher than that in Antarctic region ((0.11–0.31) × 10$^7$ atoms L$^{-1}$) (Xing et al., 2017), lower than that in the Bering Sea ((1.8–131) × 10$^7$ atoms L$^{-1}$) (Cooper et al., 2001) and North Atlantic (31°N–50°N) ((4.0–127) × 10$^7$ atoms L$^{-1}$) (He et al., 2013), and much lower than that in the Baltic Sea ((0.25–17) × 10$^{10}$ atoms L$^{-1}$) (Yi et al., 2011), North Sea ((0.26–38) × 10$^{10}$ atoms L$^{-1}$) (Hou et al., 2007), Celtic Sea ((0.03–1.2) × 10$^{10}$ atoms L$^{-1}$) (He et al., 2014) and Irish Sea ((6.2–47) × 10$^{10}$ atoms L$^{-1}$) (Schnabel et al., 2007), where they were heavily contaminated by the discharge from the reprocessing plants at La Hague (France) and Sellafield (UK).
The $^{129}$I/$^{127}$I ratios in the surface seawater collected in October 2013 varied from $4.72 \times 10^{-11}$ to $14.7 \times 10^{-11}$ (mean of $(7.02 \pm 0.49) \times 10^{-11}$, n=26), which are significantly lower than those observed in August but more than one order of magnitude higher than the pre-nuclear level ($1.5 \times 10^{-12}$, Snyder et al., 2010). The distribution pattern of $^{129}$I/$^{127}$I is similar to that of $^{129}$I concentrations, which means that the $^{129}$I concentrations dominate the distribution pattern of $^{129}$I/$^{127}$I ratios. The maximum values of $^{129}$I/$^{127}$I ratios were found in the estuary area in both seasons (Figure 3). The second-highest $^{129}$I/$^{127}$I ratios (~1.6$\times 10^{-11}$) were found in the Jiangsu Coast in both seasons. In the northeastern region of the ECS, the $^{129}$I/$^{127}$I ratios were higher in August (>1.2$\times 10^{-11}$) than that in October (<0.9$\times 10^{-11}$). $^{129}$I/$^{127}$I ratios in the freshwater of the Changjiang were much higher (~100$\times 10^{-11}$) than those in the coastal areas of the ECS (Liu et al., 2016). This can be attributed to the fact that the $^{129}$I/$^{127}$I ratio of the Changjiang input is largely diluted by high $^{127}$I concentration seawater when the land-sourced $^{129}$I (with low $^{127}$I level) water is transported southward along the coast (Wu et al., 2013; Wang et al., 2016). It has also been reported that when the $^{129}$I/$^{127}$I ratios in the freshwater of the Mississippi River (113$\times 10^{-11}$-519$\times 10^{-11}$) (Oktay et al., 2001) were dramatically higher than those discharged into the coastal area of continental Louisiana, the $^{129}$I/$^{127}$I ratios were decreased from 113$\times 10^{-11}$-519$\times 10^{-11}$ (Oktay et al., 2001) to (16$\times 10^{-11}$-41$\times 10^{-11}$) (Schwehr et al., 2005).

### 3.2. The vertical profiles of iodine isotopes in the water column

The vertical profile of iodine isotopes in the sampling stations in the ECS are plotted in Figure 4 and Figure 5. In section N (Figure 4), narrow variations of $^{127}$I concentrations were observed at all three stations. The $^{129}$I concentrations showed a maximum value ($2.5 \times 10^7$ atoms L$^{-1}$) at a depth of 12 m at station N2; the maximum value of the $^{129}$I/$^{127}$I ratio was also found in the same layer of N2. Similar trends were
also found at stations A1 and A3, where the $^{127}\text{I}$ and $^{129}\text{I}$ concentrations decreased from surface to bottom water, but the $^{129}\text{I}/^{127}\text{I}$ ratios were relative constant. In section B, the $^{127}\text{I}$ concentrations first increased and then decreased with increasing depth at the two stations (i.e., B1 and B2) near the river mouth, but they were constant at station B3. The $^{129}\text{I}$ concentrations showed a narrow variation with depth at stations B2 and B3, but they decreased from the surface water to the bottom layer at station B1. The $^{129}\text{I}/^{127}\text{I}$ ratios decreased from the surface waters to the 25-m layers at three stations, and the maximum values decreased from B1 to B2.

In sections C, D and F (Figure 5), there are six stations located in coastal areas and two stations (F8 and C12) located in the pathway of the KC. In the coastal areas, the concentrations of $^{129}\text{I}$ and $^{127}\text{I}$ varied little at different depths, which suggests that the vertical profiles of iodine isotopes associated with the water mass were well-mixed in the water column in these areas. The uniform vertical distributions of temperature and salinity (Table 1) also support this statement. In addition, the $^{129}\text{I}/^{127}\text{I}$ ratios in the depth profile were constant at stations C4 and D1, but they showed higher values at the bottom layers at stations C3 ($9.2\times10^{-11}$) and D2 ($13\times10^{-11}$). Relatively higher values were also observed in the 17-m layers of stations F1 and F2. In an offshore station (i.e., D4) through which the TWC flows, the $^{129}\text{I}/^{127}\text{I}$ ratios were constant, and temperature and salinity also showed a very narrow range of variation. In the margin of the ECS shelf where the KC flows, the $^{127}\text{I}$ concentrations of seawater showed higher values ($40 \sim 60 \mu g \text{ L}^{-1}$) in the vertical profiles at stations F8 and C12, and the highest values were found in the 300-m and 400-m layers at stations F8 and C12, respectively. The $^{129}\text{I}$ concentrations at station F8 showed relatively minor variations, but those at station C12 showed a maximum value in the 100-m layer and then decreased with increasing depth. The $^{129}\text{I}/^{127}\text{I}$ atomic ratios were constant at the
surface layer and then reached their highest values at the 100-m layer at station C12 and the 180-m layer at station F8.

4. Discussions

4.1. The inventory of $^{129}$I in the ECS water column

The inventory of $^{129}$I (I, atoms m$^{-2}$) in the seawater column can be estimated using the following equation (modified from Suzuki et al., 2010):

$$ I_x = \sum_{i=1}^{n} \left[ \frac{1}{2} ^{129}C_i \right] (d_{i+1} - d_i) + ^{129}C_1 d_1 + ^{129}C_n (d_b - d_n) $$

where $n$ is the number of sampling depth, $^{129}C_i$ represents the concentrations (atoms m$^{-3}$) of $^{129}$I in seawater at depth $i$, $d_i$ and $d_b$ refer to the $i$th sampling depth of seawater (m) and the total depth to the bottom (m, total depth of the water column), respectively. The inventories of $^{129}$I were estimated to be (0.23-0.79) ×10$^{12}$ atoms m$^{-2}$ (mean of (0.46±0.15) ×10$^{12}$ atoms m$^{-2}$) in shallow waters (27-65 m in total depth) and (6.5-7.3) ×10$^{12}$ atoms m$^{-2}$ (mean of (6.9±0.6) ×10$^{12}$ atoms m$^{-2}$) in deep waters (440-500 m in total depth) (Table 1 and Figure 6). The inventories of $^{129}$I in the shallow waters generally increased from the coasts to the offshore region. The deeper stations hold a higher inventory because they contain more volume of water compared to shallow stations or shelf stations. In order to understand the sources and sinks of $^{129}$I, here we only compared the inventory in first 100 m depth of the water column with other regions in the world. Generally, the inventories of $^{129}$I in the upper 100 m water column of the ECS were comparable to those of the North Pacific ((1.7-3) ×10$^{12}$ atoms m$^{-2}$) (Povinec et al., 2010; Suzuki et al., 2010), the Southern Indian Ocean ((0.6-0.8) ×10$^{12}$ atoms m$^{-2}$) (Povinec et al., 2011) and the Gulf of Mexico (1.5×10$^{12}$ atoms m$^{-2}$) (Schink et al., 1995), which suggest a similar dominant source initially from the global fallout for these regions. But our results were clearly lower than those.
from the offshore of Fukushima ((8-24) ×10^{12} \text{ atoms m}^{-2}) (Hou et al., 2013), and this enhanced values observed in the offshore of Fukushima which should be attributed to the release from the Fukushima accident. The Arctic Ocean ((3.4-141) ×10^{12} \text{ atoms m}^{-2}) (Smith et al., 1998; Alfimov et al., 2004a) and Baltic Sea ((60-6834) ×10^{12} \text{ atoms m}^{-2}) (Alfimov et al., 2004b; Aldahan et al., 2007a) also had very high $^{129}$I inventories compared to the ECS. The $^{129}$I concentration in the upper 100 m water column of the North Atlantic Ocean (6×10^{7} \text{ atoms L}^{-1}-4.7 \times 10^{11} \text{ atoms L}^{-1}) (Edmon et al., 2001; Schnabel et al., 2007; Alfimov et al., 2004c, 2013) were also much higher than that in the ECS, and thus the $^{129}$I inventories are expected be much higher than those of the ECS. The enhanced $^{129}$I inventories in the Arctic and the North Atlantic ocean were attributed to the marine discharge of $^{129}$I from the nuclear reprocessing facilities at La Hague and Sellafield (Alfimov et al., 2004c). Overall, the inventory estimation could offer a good approach to check whether if there are extra sources than global fallout, or others.

4.2. Sources and sinks of $^{129}$I in the ECS

In the use of $^{129}$I as tracers for the assessment of water mass in marginal sea, its source terms should be first assessed. $^{129}$I in the ECS is initially from the atmospheric fallout which were mainly released from the weapon tests before 1970 and reprocessing plants Sellafield (UK) and La Hague (France) during 1970s and 1990s (Fan et al., 2016; Bautista et al., 2016; Zhang et al., 2018). Then it was finally reached to the ECS by means of river, direct atmospheric input and/or ocean currents (Liu D. et al., 2016). Our previous study suggested that the main sources of $^{129}$I in the water column of the ECS are riverine input, atmospheric fallout and oceanic input (including adjacent sea and open sea inputs), but their contributions were not quantified (Liu D. et al., 2016). The sinks of $^{129}$I are mainly storage in the water
Based on the inventory of $^{129}$I, we could estimate the total amount of $^{129}$I in the water column of the ECS. Due to the dramatic differences in the $^{129}$I inventory between shallow and deep waters, it is more reasonable to separately calculate the total $^{129}$I in these waters. The total surface area of the ECS corresponding to the $^{129}$I inventories was estimated by Google Earth to be $2.56 \times 10^{11}$ m$^2$, which is divided into two parts by the 100-m isobaths ($2.185 \times 10^{11}$ m$^2$ for the shallow waters and $4.65 \times 10^{10}$ m$^2$ for the deep waters) (Figure 6). Then, the total $^{129}$I in the water column of the ECS can be estimated to be $(4.2 \pm 0.4) \times 10^{23}$ atoms (88 g) by using the mean inventory in the two parts ($(0.46 \pm 0.15) \times 10^{12}$ atoms m$^{-2}$ for the shallow waters (<100 m) and $(6.9 \pm 0.6) \times 10^{12}$ atoms m$^{-2}$ for the deep waters (400-500 m)) with each the corresponding surface area. This value is far smaller than those in water column of the nuclear reprocessing facilities (La Hague, France and Sellafield, UK) contaminated seas, including the Baltic Sea (15 kg), North Sea (86 kg), Nordic Sea (1100 kg), Barnets Sea (366 kg) and Kara Sea (113 kg) (Aldahan et al., 2007a). As the turnover time of the ECS shelf water was ~1.3 yrs (Wang et al., 2018), the total flux of $^{129}$I stored in the water column was estimated to be $(3.2 \pm 0.3) \times 10^{23}$ atoms yr$^{-1}$ using the total $^{129}$I $(4.2 \pm 0.4) \times 10^{23}$ atoms) divided by the turnover time of the ECS.

### 4.2.1 Sources of $^{129}$I in the ECS

Based on the inventory of $^{129}$I, we could estimate the total amount of $^{129}$I in the water column of the ECS. Due to the dramatic differences in the $^{129}$I inventory between shallow and deep waters, it is more reasonable to separately calculate the total $^{129}$I in these waters. The total surface area of the ECS corresponding to the $^{129}$I inventories was estimated by Google Earth to be $2.56 \times 10^{11}$ m$^2$, which is divided into
two parts by the 100-m isobaths ($2.185 \times 10^{11}$ m$^2$ for the shallow waters and $4.65 \times 10^{10}$ m$^2$ for the deep waters) (Figure 6). Then, the total $^{129}$I in the water column of the ECS can be estimated to be $(4.2\pm0.4) \times 10^{23}$ atoms (88 g) by using the mean inventory in the two parts ($(0.46\pm0.15) \times 10^{12}$ atoms m$^{-2}$ for the shallow waters (<100 m) and $(6.9\pm0.6) \times 10^{12}$ atoms m$^{-2}$ for the deep waters (400-500 m)) with each—the corresponding surface area. This value is far smaller than those in water column of the nuclear reprocessing facilities (La Hague, France and Sellafield, UK) contaminated seas, including the Baltic Sea (15 kg), North Sea (86 kg), Nordic Sea (1100 kg), Barents Sea (366 kg) and Kara Sea (113 kg) (Aldahan et al., 2007a). As the turnover time of the ECS shelf water was ~1.3 yrs (Wang et al., 2018), the total flux of $^{129}$I stored in the water column was estimated to be $(3.2\pm0.3) \times 10^{23}$ atoms yr$^{-1}$, the total $^{129}$I ((4.2\pm0.4) \times 10^{23} \text{ atoms}) and the turnover time of the ECS.

The concentrations of the dissolved $^{129}$I in the inner region of the Changjiang Estuary were reported to be $(1.7-4.0) \times 10^7$ atoms L$^{-1}$ (mean of $(2.6\pm1.2) \times 10^7$ atoms L$^{-1}$, n=3) during August 2013 (Liu D. et al., 2016). Using this mean value and the annual water discharge of the Changjiang to the ECS ($9.05 \times 10^{11}$ m$^3$ yr$^{-1}$) (Yang et al., 2015), we can estimate that the mean annual $^{129}$I input derived from the Changjiang water is $(2.4\pm1.1) \times 10^{22}$ atoms, which is much lower than the inventory of $^{129}$I in the ECS. Since there are no any reported data about the particulate $^{129}$I in the Changjiang river water, here, we use a simple equation to roughly estimate the $^{129}$I input ($I_{sed}$, atmos yr$^{-1}$) by sediment discharge from the Changjiang:

$$I_{sed} = I_{wat} \times TSM \times K_d$$  \hspace{1cm} (2)$$

where $I_{wat}$ is the $^{129}$I input by water discharge from the Changjiang ($(2.4\pm1.1) \times 10^{22}$ atoms yr$^{-1}$), TSM is the mean concentration of total suspended matter discharged from
the Changjiang to the ECS \((1.4 \times 10^{-4} \text{ ton m}^{-3})\), which is estimated by dividing the annual sediment discharge \((1.3 \times 10^{8} \text{ ton yr}^{-1})\) (Yang et al., 2015) by the water discharge of the Changjiang. The \(K_d\) of iodine was reported to be 40-200 m \(^3\) ton \(^{-1}\) in the estuaries (Takata et al., 2013). Considering that the mean TSM of the Changjiang is very high, here, we use the maximum value (200 m \(^3\) ton \(^{-1}\)). Finally, the \(I_{\text{sed}}\) is estimated to be \((6.8 \pm 3.2) \times 10^{20} \text{ atoms yr}^{-1}\), which is very small (~3% of \(I_{\text{wat}}\)) compared to the total inventory of \(^{129}\text{I}\) (<0.2% of the total inventory of \(^{129}\text{I}\) in the ECS). Overall, the contribution of \(^{129}\text{I}\) from the Changjiang riverine input to the ECS is just a small fraction (7.5%) of the total flux of \(^{129}\text{I}\) in the water column \((3.2 \pm 0.3) \times 10^{23} \text{ atoms yr}^{-1}\) of the ECS, although the CDW significantly affects the seasonal variation of \(^{129}\text{I}\) in the water column near the Changjiang Estuary. Since the total discharge from other rivers only contributes 10% of freshwater to the ECS (Milliman and Farnsworth, 2013), the input of \(^{129}\text{I}\) from these rivers to the ECS is less than 0.75% of the total inventory, assuming that the \(^{129}\text{I}\) concentrations in these rivers are the same as those in the Changjiang.

Direct atmospheric fallout should also contribute \(^{129}\text{I}\) to the ECS. Although there are no reported data at the ECS, we can use other data from the nearest stations at the same latitude as the ECS, i.e., Tokyo to the north (35.5 °N) and Ishigaki Island to the south (24.0 °N). During the Fukushima accident, the westerly wind brought and deposited most of the released \(^{129}\text{I}\) in the northern North Pacific Ocean (Tumey et al., 2014; Casacuberta et al., 2017), causing negligible deposition in the ECS. The \(^{137}\text{Cs}\) in the seawater collected in the ECS after the Fukushima accident confirmed this assumption (Zhao et al., 2018). These investigations revealed that the \(^{129}\text{I}\) released from the Fukushima accident was mainly deposited in the land and offshore waters near the Fukushima NPP, which had a very small influence on the sea areas of
southwest Japan (Hou et al., 2013; Casacuberta et al., 2017). Thus, the pre-accident

\[ ^{129}\text{I} \] deposition data from Tokyo and Ishigaki Island can be used to roughly estimate

the contribution of direct atmospheric fallout to the ECS. Toyama et al. (2013) estimated that the atmospheric depositional \( ^{129}\text{I} \) in Tokyo was \( 6.2 \times 10^{10} \) atoms \( \text{m}^{-2} \text{yr}^{-1} \) during 2003 and that in Ishigaki Island was \( 0.69 \times 10^{10} \) atoms \( \text{m}^{-2} \text{yr}^{-1} \) at 2003. By multiplying the median of \( ^{129}\text{I} \) deposition (\( 3.4 \times 10^{10} \) atoms \( \text{m}^{-2} \text{yr}^{-1} \)) in the two stations with the surface area of the ECS (\( 2.56 \times 10^{11} \) \( \text{m}^{2} \)), we can estimate that the atmospheric fallout of \( ^{129}\text{I} \) to the ECS is \( 0.087 \times 10^{23} \) atoms \( \text{yr}^{-1} \). This value accounts for approximately 3% of the total \( ^{129}\text{I} \) flux in the water column of the ECS.

### 4.2.2 \( ^{129}\text{I} \) budget in the ECS

Due to the high solubility and long residence time of iodine in the ocean, reports of its concentrations in marine sediments are very limited, especially in the marginal sea of the NW Pacific (Fan et al., 2016), which makes the estimation of the sedimentary deposition of \( ^{129}\text{I} \) difficult in the ECS. However, considering the low TSM in the ECS (< 10 mg \( \text{L}^{-1} \)) and the lower \( K_d \) values (< 30 L kg\(^{-1} \)) of \( ^{129}\text{I} \) in seawater relative to those in estuaries (< 30 L kg\(^{-1} \)) (Huang et al., 2013; Takata et al., 2013), it is expected that the amount of \( ^{129}\text{I} \) buried in ECS sediment is small compared to that stored in the water column. The \( ^{129}\text{I} \) inventories in water column were reported to be ten times higher than those buried in the sediments from the North Sea, Nordic Sea and North Atlantic Ocean (Aldahan et al., 2007a). Without considering the sedimentary buried \( ^{129}\text{I} \) in the ECS, we can obtain a lower limit (\( (2.9 \pm 0.3) \times 10^{23} \) atoms \( \text{yr}^{-1} \)) of net input of \( ^{129}\text{I} \) from the Northwest Pacific via the KC and TWC and the Yellow Sea via the YWCC (Fig. 5), which represents approximately 90% of the flux of \( ^{129}\text{I} \) in the ECS. A previous study suggested that the oceanic input of Pu from the
Pacific Proving Ground (PPG) in the Northwest Pacific dominated the mass balance of Pu in the ECS sediment and that a small portion of Pu was likely derived from the Yellow Sea (Wang et al., 2017). It is reasonable that the very conservative $^{129}$I could be transported from the Northwest Pacific to the ECS and that some portion of $^{129}$I could be derived from the Yellow Sea via the YSCC and NJCC.

4.3. Implication of $^{129}$I/$^{127}$I for tracing water mass movement

4.3.1 The potential use of $^{129}$I/$^{127}$I as a tracer for identifying the water mass in the ECS

Multi-currents and biological activities (e.g., enrichment by phytoplankton) could affect the distribution of iodine isotopes in the ECS (Butler et al., 1981; Liu D. et al., 2016). The $^{129}$I concentrations decreased from the coast to offshore, whereas $^{127}$I is mainly oceanic, and its concentration distribution showed a reverse pattern relative to $^{129}$I and generally increased with salinity ($r$=0.53, $p<0.01$). The $^{129}$I/$^{127}$I ratios could help alleviate the biological effects and thus are more significant than single iodine isotopes in tracing the exchange of water masses in the ECS. The $^{129}$I/$^{127}$I ratios showed very weak linear correlation with the salinity ($r$=-0.46, $p<0.01$), which implies that the ECS waters are not resulting from the simply two-endmember (fresh and salt water) mixing (Fig. 6a). Instead, three main endmembers can be identified in October 2013: the CDW water, YSCC water and KC water, characterized by the salinity and $^{129}$I/$^{127}$I ratios of $<25$ and $12 \times 10^{-11}$, $32.5$ and $18 \times 10^{-11}$, $35.5$ and $6 \times 10^{-11}$, respectively. The highest $^{129}$I/$^{127}$I ratios were observed in the middle layer of the water column at stations A3 and N3. These values are similar to those of surface sediments ($((177-189) \times 10^{-12})$ at Jiaozhou Bay in the Yellow Sea (Fan et al., 2016). Relatively high levels of $^{129}$I were also observed in the surface water of the northernmost region of the ECS (stations M2 and M3). Therefore, the high $^{129}$I
contents in the middle layer at stations A3 and N3 may have originated from the Yellow Sea water, which flows south and downwards to the middle layer of the northern ECS (Lie et al., 2000). The $^{129}\text{I}/^{127}\text{I}$ ratios vs. salinity distribution in the KC waters showed a distinct pattern (with the highest salinities and relatively lower $^{129}\text{I}/^{127}\text{I}$ ratios) compared to other regions. It is noted that the water derived by the KC and TWC cannot be separated by $^{129}\text{I}/^{127}\text{I}$ ratios and salinity and thus they are considered as one source.

From the Figure 6a, the upper 10 m water at station D1 seems to be derived from the CDW waters, and those of surface water at station F1 showed the similar salinity and $^{129}\text{I}/^{127}\text{I}$ ratios, which suggests that CDW-derived water could transport southward to F1 via the ZFCC during October. While at the bottom layer of the station D1, the salinity and $^{129}\text{I}/^{127}\text{I}$ ratio lay in the middle range of CDW water and KC water values, which suggest a mixing between these two endmembers. At station D2, the 39-m layer water showed a mixing signal between YSCC water and KC water, which is similar to the a recent in-situ observation conducted nearby (28 °N, 122 °E) during February and March 2014 that they deduced the YS water could be transported to the ECS (Liu et al., 2018). Our result suggests that the YSCC water might also be transported to the inner shelf (29 °N, 122 °E) of ECS during October via the intermediate or near-bottom layer water. In the south offshore (east of 30 m isobath), the spatial and vertical distribution of $^{129}\text{I}/^{127}\text{I}$ ratios were dominated by the KC input, which bring abundant warm and salty water associated with lower $^{129}\text{I}/^{127}\text{I}$ ratios iodine isotopes to the coastal areas (Guo et al., 2006; Zhang et al., 2017). The $^{129}\text{I}/^{127}\text{I}$ ratios vs salinity distribution of surface water during August 2013 showed similar pattern with those during October that three sources can be observed. However, the $^{129}\text{I}/^{127}\text{I}$ ratios vs salinity both in the south coast and offshore of the ECS showed the
signal close to the KC water, reflecting a stronger intrusion of the TWC and KC to coastal areas of the ECS during summer relative to autumn. From Figure 9a, the KC waters could intrude into the inner shelf near the Zhe-Min Coast via the bottom layer and Changjiang Estuary (Ichikawa and Beardsley, 2002).

The source of the water mass in the north of the ECS is very complex due to the joint interaction of the multi-currents, tide, monsoon and topography. Wu et al. (2014) hypothesized that a small portion of the CDW plume could extend along the Jiangsu coast in winter, which is mainly driven by the tide-induced Stokes drift. The CDW waters signal were also observed in the surface water of the Jiangsu coast both in October (e.g., stations M2), suggests that such extension of the CDW plume might also exist in autumn. The signal of Changjiang River input was also found in other parameters of sediment in the Jiangsu coast, e.g., its magnetic minerals, trace elements and clay minerals (Zhang et al., 2012; Lu et al., 2015). These observations further suggest that Changjiang-derived iodine isotopes can be transported to the Jiangsu coast via the tide-induced coastal current (e.g., autumn and winter). From Figure 6, the iodine isotopes in the north of the ECS are derived from the mixing of the CDW, YSCC, and KC waters. Lian et al. (2016) used $\delta^{18}$O and $\delta$D as tracers to suggest that the subsurface waters of the KC could intrude near the Changjiang river mouth. In the present study, the $^{129}$I/$^{127}$I ratios vs. salinity distribution showed that the KC (and TWC) water could arrive at the north part (32 °N, 123 °E) of the Changjiang Estuary (e.g., station 48, 49 and N2). As mentioned above, the signal of YSCC water were observed in the intermediate layer of the Northern ECS (i.e., N2 and A3). But for the surface water, the $^{129}$I/$^{127}$I ratios vs. salinity distribution showed a mixing between CDW water and the KC/TWC water. It means that an upwelling which is originated from the KC/TWC water meeting the shallower topography in the
Changjiang Estuary (31°N, 122.5°E) mixed with the CDW plume and transported northward. Besides, the signal of the KC water was also observed in bottom layer of the station N3 and N4, which further suggests the complex structure of multi-currents in the northern ECS.

Overall, the spatial and vertical distribution of iodine isotopes is significantly affected by the complex current system in the ECS. The diagram of $^{129}$I/$^{127}$I ratios-traced water mass structure in the ECS is showed as Figure 7. Assessing the land-sea interactions of nutrients, especially the estimation of oceanic contributions via the KC, has consistently been one of the most difficult and important scientific problems in river-influenced marginal systems (Liu et al., 2016). Our study showed that the $^{129}$I/$^{127}$I ratio is sensitive to water mass exchange in the ECS and that most $^{129}$I is originated from oceanic input; thus, the $^{129}$I/$^{127}$I ratio could serve as a good indicator for better tracing the land-sea interactions of nutrients in marginal systems.

4.3.2 Implication of $^{129}$I/$^{127}$I for seawater vertical mixing process

The vertical profiles of both $^{129}$I/$^{127}$I ratios and $^{129}$I concentrations generally showed narrow variations in south offshore waters with depths between 30 m and 100 m (Figures 4 and 5), indicating the vertical mixing of seawater in the middle shelf of ECS. In a station offshore Fukushima (31), the significantly higher $^{129}$I concentrations that were released by the Fukushima accident were observed in the top 20-m waters during June 2011 (Hou et al., 2013); this depth likely represents a mixing layer during a 3-month timescale. However, in deep waters of the ECS (e.g., F8 and C12), the $^{129}$I/$^{127}$I ratios and $^{129}$I concentrations showed fluctuations in the vertical profiles. The maximum ratios were observed in the subsurface water (100-m depth) of the KC (e.g., C12). The maximum values of the $^{129}$I/$^{127}$I ratio in the water column were also found in the 25-m-deep water of the Middle Atlantic Bight (36-37°N) in 1993 (Santschi et
The mechanism of such maximum $^{129}\text{I}/^{127}\text{I}$ ratios in the subsurface water of the ocean has not been well established. The existence of maximum $^{90}\text{Sr}$ and $^{137}\text{Cs}$ concentrations in subsurface water was also observed in the central NW Pacific Ocean (Povinec et al., 2003; Hong et al., 2012). Buesseler and Livingston (1997) used the subsurface Cs maximum which corresponding to Chernobyl fallout to indicate the vertical mixing process near the Bosporus, Black Sea. Biological activity might affect the species components (iodate and iodide) of $^{129}\text{I}$, but it exerts less influence on the vertical distribution of $^{129}\text{I}/^{127}\text{I}$ ratios (Zhang and Hou, 2013). Instead, physical circulation mixing should play an important role in the existence of the subsurface maximum $^{129}\text{I}/^{127}\text{I}$ ratios. As mentioned above, the $^{129}\text{I}$ released by the La Hague (France) and Sellafield (UK) were much higher than the nuclear tests. Thus, the layer with maximum values might represent the year of 1989, which had the maximum fallout of $^{129}\text{I}$ initially from the reprocessing plants at La Hague and Sellafield (Hou et al., 2000). Besides, the sedimentary records of $^{129}\text{I}/^{127}\text{I}$ ratios also showed the peaks corresponded to this year in the Jiaozhou Bay and Taal Lake, Philippines (Bautista et al., 2016; Fan et al., 2016). If we assume that the layer with maximum $^{129}\text{I}/^{127}\text{I}$ ratios represent the year of 1989, we could roughly estimate that the mixing velocities of surface and subsurface waters were 4 and 6 m $\text{y}^{-1}$ at station C12 and the Middle Atlantic Bight, respectively. More reliable vertical mixing velocities could be better elucidated by using much higher-resolution $^{129}\text{I}$ profiles in the water column and more precise time-series (1950s-present) of $^{129}\text{I}$ fallout data in the relevant region.

5. Conclusions

Based on the results of surface waters at 26 stations and water column at 18 stations in the ECS, the following conclusions can be drawn:
The inventories of $^{129}$I were estimated to be $(0.23-0.79) \times 10^{12}$ atoms m$^{-2}$ (n=16) in shallow waters (<100 m) and $(6.5-7.3) \times 10^{12}$ atoms m$^{-2}$ (n=2) in deep waters (400-500 m). The $^{129}$I inventories of upper 100 m waters in the ECS were comparable to (significantly lower than) those of other region (without being) contaminated by the nuclear accidents or nuclear reprocessing facilities, which suggests that the $^{129}$I inventory is useful to check whether if there are extra sources than global fallout, or others.

The $^{129}$I/$^{127}$I atomic ratios varied with the water mass flowing in August and October, with higher values $(10 \times 10^{-11} - 20 \times 10^{-11})$ in the coastal regions of the ECS and lower values $(<8 \times 10^{-11})$ in the offshore. The total inventory of $^{129}$I in the water column of the ECS was $(4.2 \pm 0.4) \times 10^{23}$ atoms (~88 g). The contributions from the Changjiang river input (<7.5%) and direct atmospheric fallout (<2.7%) to the total $^{129}$I in the ECS are much less than the oceanic input from the Yellow Sea via the YSWC and the West Pacific via the KC (the contribution from the Fukushima accident is negligible). These observations suggest that the $^{129}$I/$^{127}$I atomic ratios are potentially used to trace the of the land-ocean interaction in the marginal seas.

The vertical distribution of the $^{129}$I/$^{127}$I atomic ratios and salinity are useful to trace the water mass exchange in the ECS, and the water masses in the ECS present stratification during their confluence; the Changjiang waters could be transported to the North Jiangsu coast both in August and October; and the TWC water could intrude to Northern part of the Changjiang mouth (32°N, 123°E). Besides, the water profiles of iodine isotopes are also helpful to trace the seawater mixing process in ocean
Acknowledgments

Original data on the salinity, temperature, $^{127}\text{I}$, $^{129}\text{I}$ concentrations and $^{129}\text{I}/^{127}\text{I}$ atomic ratios of the surface water and sampling locations were listed in the Table S1 of this paper. This research was supported by the Natural Science Foundation of China (41706089), Ministry of Science and Technology of China (No. 2015FY110800) and State key Laboratory of Loess and Quaternary Geology, China. Samples were collected under the support of the Ministry of Science and Technology of PR China (2011CB409801). We appreciate Dr. Qi Liu for his contribution for the measurement of $^{129}\text{I}$ in all samples in Xi’an AMS center.

References


Beam Interactions with Materials and Atoms, 294, 542-546.


Lian, E., Yang, S., Wu, H., Yang, C., Li, C., & Liu, J. T. (2016). Kuroshio subsurface water feeds the wintertime Taiwan Warm Current on the inner East China Sea shelf.


Povinec, P. P., Livingston, H. D., Shima, S., Aoyama, M., Gastaud, J., Goroncy, I., ...


Raisbeck, G. M., Yiou, F., Zhou, Z. Q., & Kilius, L. R. (1995). $^{129}$I from nuclear fuel reprocessing facilities at Sellafield (UK) and La Hague (France); potential as an oceanographic tracer. Journal of Marine Systems, 6(5-6), 561-570.


Figure captions:

Figure 1. Location of sampling stations in the ECS during the October cruise in 2013 (surface water in red and vertical profiles in blue). The regional surface current in the autumn is modified after Wu et al., 2014: Yellow Sea Coastal Current (YSCC); Yellow Sea Warm Current (YSWC); Changjiang Dilute Water (CDW); Zhejiang-Fujian Coast Current (ZFCC); Taiwan Warm Current (TWC); Kuroshio Current (KC). The bathymetry (m) of the ECS shelf is shown by grey lines.

Figure 2. Spatial distribution of $^{127}$I and $^{129}$I concentrations and $^{129}$I/$^{127}$I atoms ratios in the surface water in the ECS during August (replotted from Liu D. et al., 2016) and October of 2013. This figure was generated using the Surfer mapping software package using Kriging interpolation based on a digital elevation model (DEM) with $6 \times 6$ km grid resolution.

Figure 3. The vertical profiles of $^{127}$I and $^{129}$I concentrations and $^{129}$I/$^{127}$I atom ratios in the ECS water column during October of 2013. The iodine isotopes distributions are plotted as three group for different regions: Northern part of the Changjiang river mouth, inner shelf the ECS and offshore region.

Figure 4. Spatial distribution of $^{129}$I inventories in water column in the ECS during October of 2013. The shallow and deep waters are dived by the 100-m isobaths.

Figure 5. The mass balance of $^{129}$I in the seawater of the ECS.

Figure 6. Distribution of $^{129}$I/$^{127}$I atom ratios vs. salinity in seawater of the ECS during October (a) and August (b) 2013. CJE represents the Changjiang Estuary.

Figure 7. Diagram of the water mass structure in the ECS during October traced by the $^{129}$I/$^{127}$I ratios. S, I and B represent the surface waters, intermediate waters and bottom waters in the ECS, respectively.