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- Stable isotope ratio of C and N along Changjiang transport pathway
- Seasonal variation in stable isotope ratio of C and N in Changjiang Estuary
- Vertical profile of stable isotope ratio of C and N in Changjiang Estuary

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Stable isotope ratios of carbon and nitrogen in suspended organic matter: Seasonal and spatial dynamics along the Changjiang (Yangtze River) transport pathway

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Abstract Seven cruises were conducted in the Changjiang (Yangtze River) Estuary and the adjacent western East China Sea (ECS) from 2010 to 2012 to study the seasonal variations of $\delta^{13}C$ and $\delta^{15}N$ in suspended organic matter. In addition, two cruises in the northeastern ECS in July 2011 and in Tsushima Strait in July 2012 were conducted to evaluate the distribution patterns of these isotopes over the entire Changjiang transport pathway. In summer, the surface $\delta^{13}C$ was lowest in the Changjiang Channel, increasing from land to sea, reaching highest values in the central ECS, and then decreasing and remaining relatively constant. In winter, the surface $\delta^{13}C$ in the western ECS showed lower values with less variation in general. At most stations, $\delta^{13}C$ increased from the sea surface to the seabed, reflecting the degradation of sinking organic matter; however, these trends could be changed in the summer by surface phytoplankton accumulation. Combining data from all the Changjiang Estuary and western ECS cruises revealed that when the suspended particulate matter (SPM) was > 135 mg/L, the $\delta^{13}C$ values were fairly constant ($-24.5‰$ to $-20.5‰$); when the SPM was < 135 mg/L, the $\delta^{13}C$ values showed much greater variability ($-28.4‰$ to $-16.6‰$). The surface $\delta^{15}N$ also showed generally higher values in the central ECS in summer and lower values in winter. The seasonal variations of $\delta^{13}C$ and $\delta^{15}N$ were largely attributed to the SPM composition change, i.e., more phytoplankton cells in the summer whereas more resuspended sediment particles were present in winter.

1. Introduction

The behavior of particulate organic carbon (POC) and particulate nitrogen (PN) in estuaries and the adjacent shelf is complex. This is partly because the POC in these areas consists of several components with different origins, including higher plant detritus transported by rivers, in situ phytoplankton production, and resuspended organic sediments [*Wang et al.*, 2004; *Middelburg and Herman*, 2007]. Understanding POC behavior, budgets, and composition change in estuarine and shelf areas is essential to describe carbon cycle patterns on the global scale [*Hedges et al.*, 1997; *Chen and Wang*, 1999; *Schneider et al.*, 2003; *Deng et al.*, 2006].

The signatures of $\delta^{13}C$ and $\delta^{15}N$ have important scientific implications. Analyses of $\delta^{13}C$ and $\delta^{15}N$ are used in biogeochemical studies worldwide [e.g., *Barth et al.*, 1998; *Sheu et al.*, 1999; *Guo et al.*, 2004; *Sato et al.*, 2006] because fractionation of $^{13}C/^{12}C$ and $^{15}N/^{14}N$ occurs during an extensive variety of biogeochemical processes of organic matter [e.g., *Fernandez et al.*, 2003]. In estuarine and the adjacent shelf areas, $\delta^{13}C$ and $\delta^{15}N$ can be used to differentiate between terrestrial origin and in situ produced marine sources of POC [*Barth et al.*, 1998; *Alling et al.*, 2008; *Zhu et al.*, 2008]. Thus, knowledge of the distribution and variation of $\delta^{13}C$ and $\delta^{15}N$ in these areas can greatly improve our understanding of the biogeochemistry of carbon and nitrogen, e.g., the geographic extent of the high discharge of these biogenic elements from large rivers and the degree to which the river-discharged elements influence, and/or are modified by, the pelagic ocean.

The Changjiang (Yangtze River) is one of the largest rivers in the world, and the adjacent East China Sea (ECS) is the largest marginal sea in the northwestern Pacific that includes a large area of shallow continental shelf. The Changjiang transports large amounts of POC to the oceans [*Milliman et al.*, 1984], and most of this terrestrial-origin POC is trapped on the ECS shelf [*Deng et al.*, 2006; *Liu et al.*, 2007]. The Changjiang Estuary and the adjacent ECS shelf provide an ideal area to study the biogeochemical behavior of terrestrial-derived POC in oceans. The large quantities of organic substances that originate from the Changjiang may be completely modified before being ultimately deposited onto the ECS shelf or being transported further offshore to the deep ocean, on time scales of at least 1 to 2 months and on the spatial scale of several hundred kilometers [*Katoh et al.*, 2000; *Lie et al.*, 2003; *Chen et al.*, 2008; *Isobe and Matsuno*, 2008].

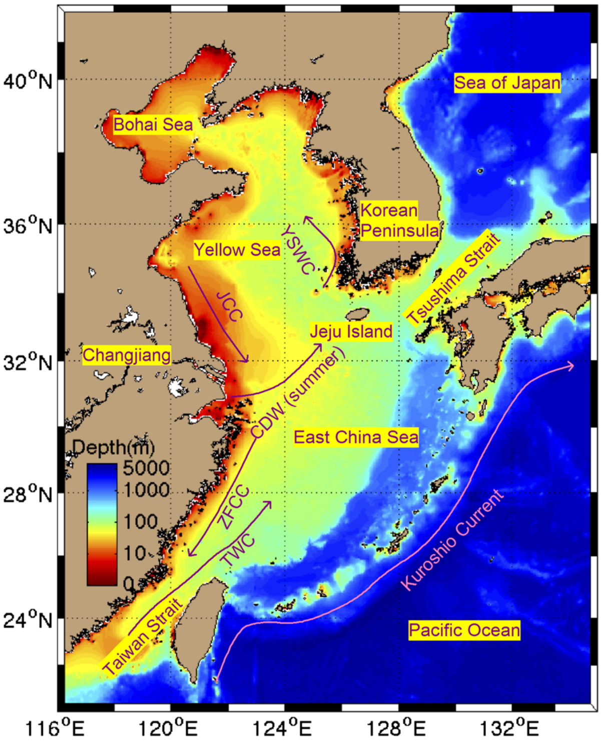

Figure 1. Bathymetry and main circulation currents of the study area. CDW: Changjiang Diluted Water, YSWC: Yellow Sea Warm Current, TWC: Taiwan Warm Current, JCC: Jiangsu Coastal Current, ZFCC: Zhejiang-Fujian Coastal Current.

Although previous studies have focused on variations in δ^{13}C and δ^{15}N in the POC of suspended particulate matter (SPM) in the Changjiang Estuary and the adjacent ECS shelf [*Tan et al.*, 1991; *Shi*, 1993; *Wu et al.*, 2003; *Zhang et al.*, 2007], most of them consisted of sampling during only one to two cruises with only a single transect or a relatively small survey area. Considering that the physical circulation in these areas is highly dynamic (influenced by the Changjiang discharge, the monsoon, and tidal pumping) on inter-annual, seasonal, and tidal time scales [*Ichikawa and Beardsley*, 2002; *Gao et al.*, 2009; *Li et al.*, 2011], it is necessary to conduct systematic studies of SPM δ^{13}C and δ^{15}N that include a number of cruises in different seasons and over multiple years. In addition, the Changjiang Diluted Water (CDW) can be pushed seaward in the summer as far as the area around Jeju Island and to the Tsushima Strait before it enters the Sea of Japan [*Isobe et al.*, 2002; *Chang and Isobe*, 2003; *Senjyu et al.*, 2006]. CDW can travel over the Changjiang transport pathway for more than 900 km. Therefore, it is also important to sample intensively over the entire area from the Changjiang Channel to the Tsushima Strait. To date, this type of work has rarely been reported in the literature.

The main aims of this study are as follows: (1) use data from three cruises conducted in the Changjiang Estuary and the adjacent western ECS in July 2012, in the northeastern ECS near Jeju Island in July 2011, and in the Tsushima Strait in July 2012, to describe the patterns of variation of δ^{13}C and δ^{15}N in suspended organic matters in the summer along the entire Changjiang transport pathway. (2) Use the data collected during the seven cruises in the Changjiang Estuary and western ECS between 2010 and 2012 to elucidate the seasonal patterns of variation of δ^{13}C and δ^{15}N in this area and to try to understand the main controlling factors. (3) Compare the results from this study with those in the literature to determine if the SPM composition and property (using the indices of δ^{13}C and the C/N molecular ratio) have changed greatly over time, as the amount of Changjiang-transported SPM has declined rapidly in recent decades due to the construction of reservoirs and dams within the Changjiang drainage basin [*Yang et al.*, 2005, 2006, 2007, 2011].

2. Materials and Methods

2.1. Site Description

The Changjiang Estuary is a mesotidal, partially mixed estuary characterized by semidiurnal tides with a mean tidal amplitude of 2.8 m [*Shi*, 2004]. The warm and saline Kuroshio Current flows northward along the ECS shelf break (Figure 1). As a result of this flow, the Taiwan Warm Current (TWC) and the Yellow Sea Warm Current (YSWC) contribute to a counterclockwise circulation in the ECS and in the Yellow Sea, respectively. These two currents induce a relatively cold and saline counter current, which includes the Jiangsu Coastal Current (JCC) in the north and the Zhejiang-Fujian Coastal Current (ZFCC) in the south (Figure 1). In winter, the ZFCC intensifies and the CDW flows southward along ZFCC in a narrow band along the inner shelf because of the low

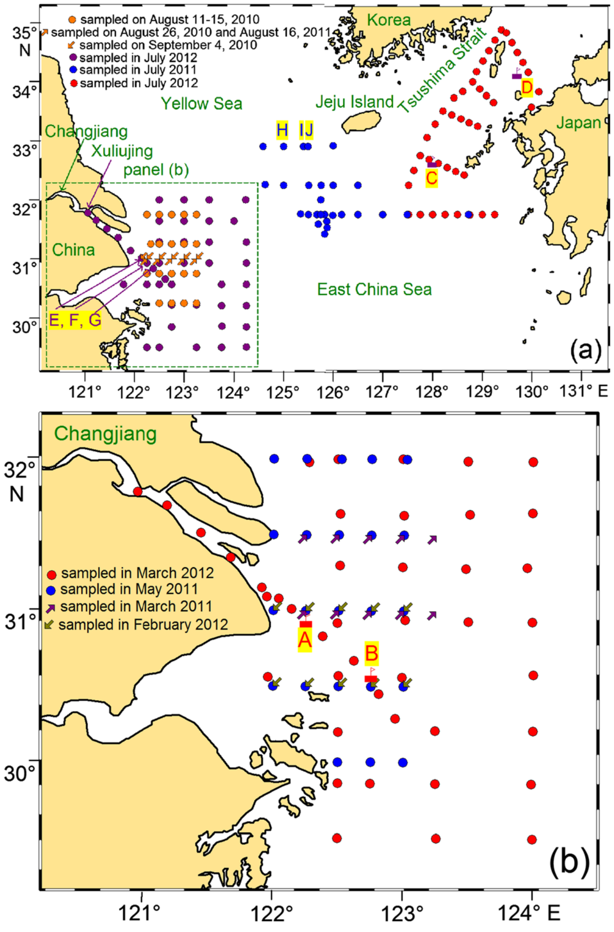

Figure 2. Map of the study area, sampling transects, and stations during the nine cruises.

Changjiang discharge and the prevailing northerly winds. Offshore, the TWC flows northward, carrying warm and saline waters. In summer, the northward TWC intensities and the southward ZFCC weakens. The CDW propagates northeastward across the shelf because of the combined effects of high river discharge, the prevailing southerly winds, and the intensified northward TWC [*Beardsley et al.*, 1985; *Ichikawa and Beardsley*, 2002; *Lee and Chao*, 2003]. During this period, the CDW covers most of the northern ECS and can be pushed to areas around the Jeju Island, through the Tsushima Strait, and finally into the Sea of Japan (Figure 1) [*Isobe et al.*, 2002; *Chang and Isobe*, 2003; *Senjyu et al.*, 2006].

2.2. Sampling

Figure 2 shows the sampling points along the latitudinal transects investigated during all cruises. Of the nine cruises in this study, seven were carried out in the Changjiang Estuary and the adjacent western ECS, in which one to nine transects were surveyed. During the August–September 2010 cruise in the western ECS, four transects were first surveyed during 11–15 August. In addition, to study the influence of a passing typhoon (#201007, named Kompasu, its tracking route can be found at http://typhoon.weather.com.cn/), one transect at latitude 31°N (Figure 2a) was surveyed once on 26 August (before the typhoon) and once on 4 September (after the typhoon). During the Changjiang Estuary and western ECS cruises in March 2012 and July 2012, much larger areas that included eight transects were investigated in the western ECS. In addition, samples from the transect along the Changjiang Channel direction, which covered the Turbidity Maximum Zone (TMZ) [*Jiang et al.*, 2013] and included more spatial-intensive sampling stations, were also collected (Figures 2a and 2b). To study the variations of δ^{13}C and δ^{15}N along the entire Changjiang transport pathway in summer, two cruises were conducted in July of 2011 and 2012, in the northeastern ECS and in the Tsushima Strait, respectively (Figure 2a).

During the March 2012 cruise in the western ECS, two anchor stations (A and B; Figure 2b) were investigated consecutively for 24 h, from 02:00 7 March to 02:00 8 March, and from 18:00 6 March to 18:00 7 March, respectively. The two stations were surveyed using two ships. Samples for δ^{13}C and δ^{15}N analysis were collected at both the surface and the bottom every 3 h at Station A and in the surface, middle, and bottom layers every 6 h at Station B. During the July 2012 cruise in the Tsushima Strait, two buoys (C and D; Figure 2a) were traced consecutively for 10 h in the Subsurface Chlorophyll Maximum (SCM) layer. The observation time was 06:00–16:00 on 25 July for Buoy Station C, and 06:00–16:00 on 22 July for Buoy Station D, and water samples from the SCM layer were collected every 2 h (due to a technical problem, samples were not collected at 14:00 at Buoy Station C).

During all cruises, samples were collected at the surface of all surveyed stations (except for sampling at Buoy Stations C and D), and samples from the middle (50% depth) and bottom (1–5 m above the sea floor) layers were collected at selected stations. During the cruises in the northeastern ECS and in the Tsushima Strait, water samples were collected from the SCM layer, instead of the middle layer. Once the water samples were brought on board the vessel, they were immediately filtered through pre-combusted Whatman GF/F filters with a pore size of 0.7 μm. At most 6 L of water was filtered for measurements of $\delta^{13}C$ and $\delta^{15}N$, although sometimes the volumes of the filtered water were restricted by the limited sample volumes. After filtration, the filters were rinsed with Milli-Q water to remove salts and then frozen at −20 °C until further analysis in the laboratory.

2.3. Measurements

In the laboratory, to measure concentrations of POC and PN, and the corresponding $\delta^{13}C$ (relative to V-PDB) and $\delta^{15}N$ (relative to air N_2), SPM filters collected from all cruises were oven-dried (50 °C for 48 h) and weighed. The weight differences between the dried filters and their counterparts before filtration were used to calculate SPM. The fractions of filters for POC and $\delta^{13}C$ analysis were exposed to concentrated HCl vapor for at least 48 h and then dried again. All filters for POC/$\delta^{13}C$ or PN/$\delta^{15}N$ analysis were packed tightly into tin cans before being measured with a Thermo Finnigan isotope ratio mass spectrometer (model: Delta plus XP, Thermo Electron Corporation, Bremen, Germany). The POC and PN concentrations were calculated by integrating the respective peaks (in units of Vs, i.e., Volt multiplied by second) produced during the measurements of $\delta^{13}C$ and $\delta^{15}N$. The accuracy of the stable isotope ratio results was guaranteed by frequent measurements of the standard materials of Caffeine (IAEA-600, −27.771‰ ± 0.043‰ for $\delta^{13}C$ and 1.0‰ ± 0.2‰ for $\delta^{15}N$), Cellulose (IAEA-CH-3, −24.724‰ ± 0.041‰ for $\delta^{13}C$), Potassium Nitrate (IAEA-NO-3, 4.7‰ ± 0.2‰ for $\delta^{15}N$), and Black Carbon (the Chinese Standard Material, GBW04407, −22.43‰ ± 0.07‰ for $\delta^{13}C$). The results of replicate samples showed that the precision for $\delta^{13}C$ and $\delta^{15}N$ was better than 0.1‰ and 0.2‰, respectively, if the integrated peak areas were higher than 50 Vs for $\delta^{13}C$ and higher than 25 Vs for $\delta^{15}N$. Thus, in this study, only $\delta^{13}C$ and $\delta^{15}N$ results with peak areas higher than 50 Vs and 25 Vs, respectively, were reported; the other $\delta^{13}C$ and $\delta^{15}N$ values were regarded as invalid results and discarded.

During each cruise, a Conductivity-Temperature-Depth (CTD) sensor (model: SBE-25, Sea-Bird Electronics; Bellevue, WA, USA) was used to record the salinity and temperature measurements. An Optical Backscatter Sensor (OBS) (model: OBS-3A or OBS-5A, Campbell Scientific; North Logan, UT, USA) was used to record the SPM turbidity (in units of NTU) measurements. The outputs of the CTD and OBS sensors were calibrated before each cruise. During the three western ECS cruises in August–September 2010, in May 2011, and in February 2012, the Laser In Situ Scattering and Transmissometry (LISST) instrument (Model: LISST-100X Type C, Sequoia Scientific Inc., Bellevue, WA, USA) was used to study the profiles of the total floc concentration (μL/L) and the mean grain size (μm) of field SPM [*Gartner et al.,* 2001; *Xia et al.,* 2004; *Mikkelsen et al.,* 2005, 2006; *Yuan et al.,* 2008; *Reynolds et al.,* 2010]. The LISST can measure the SPM concentrations in each of the 32 logarithmically arrayed size classes with mid-points ranging from 2.72 to 460 μm. Considering the generally high SPM in the western ECS, the LISST optical path length was shortened from 50 to 5 mm (for the February 2012 cruise) or to 10 mm (for August–September 2010 and May 2011 cruises), and the reported floc concentration results have therefore been multiplied by 10 and 5, respectively. Data were discarded if the corresponding optical transmission values over the path window were less than 15% [*Williams et al.,* 2007; *Gao et al.,* 2011]. After the CTD, OBS, and LISST data were retrieved, erroneous signals were removed and the remaining data were sub-sampled at every 1 m of depth and averaged. Therefore, the final data for temperature, salinity, turbidity, the total floc concentrations of 32 grain size classes, and the floc mean size used in this study were grouped into vertical bins of 1 m.

3. Results

3.1. Seasonal Variation of Surface $\delta^{13}C$

The results of the three cruises in the Changjiang Estuary and the adjacent western ECS in July 2012, in the northeastern ECS in July 2011, and in the Tsushima Strait in July 2012 were combined to obtain the POC $\delta^{13}C$ distribution in the surface SPM in summer over 900 km, i.e., from the station at Xuliujing (31°47′N and 120°56′E, Figure 2a) in the Changjiang Channel to the eastern Tsushima Strait (Figure 3). The $\delta^{13}C$ values in the surface SPM showed a clear pattern of variation along the entire Changjiang transport pathway: $\delta^{13}C$ had the lowest values (−27.3‰ to −25.9‰) in the Changjiang Channel (salinity < 0.4) then increased markedly in the TMZ and in the western and central ECS shelf, until the highest value of −16.6‰ was reached on the central ECS. At longitude 125°20′E, the values decreased sharply (to values < -20.3‰) and then remained relatively constant throughout the remaining transport areas in the northeastern ECS and in the Tsushima Strait. In the Tsushima Strait, the $\delta^{13}C$ may have been slightly elevated due to the proximity of the Korean and Japanese coasts (Figure 3).

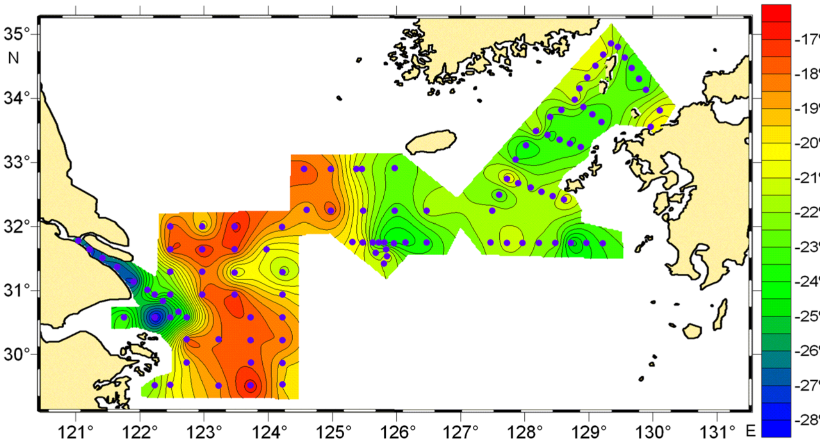

Figure 3. Distribution of particulate organic carbon (POC) δ^{13}C in the surface suspended particulate matter (SPM) along the Changjiang transport pathway in summer. The data are from the Changjiang Estuary and western East China Sea (ECS) cruise in July 2012, from the northeastern ECS cruise in July 2011, and from the Tsushima Strait cruise in July 2012.

Under the influence of the northerly monsoon and reduced Changjiang discharge in winter, the CDW flows southward in the narrow areas adjacent to the Chinese coast, instead of northeastward during the summer [*Beardsley et al.*, 1985; *Chang and Isobe*, 2003]. In March 2012, the δ^{13}C values for the surface SPM in the Changjiang Estuary and western ECS were fairly constant, with values ranging from −25.6‰ to −21.6‰ (Figure 4). The elevation of δ^{13}C in the central ECS shelf was not observed in Figure 4; thus, the pattern differs from that shown in Figure 3. Similar to the values observed in July 2012, the δ^{13}C values in the Changjiang Channel in March 2012 were also slightly lower than those in the ECS shelf during the same period (Figure 4).

In addition to those cruise results shown in Figures 3 and 4, the δ^{13}C distributions at the surface from the two other cruises (including four investigations) in summer and from the two other cruises in winter are shown in Figure 5. The results from a spring cruise (May 2011) are also displayed (Figure 5e). Compared to those two Changjiang Estuary and western ECS cruises in Figures 3 and 4, the cruises shown in Figure 5 were with smaller survey areas. Elevation of POC δ^{13}C values in the surface SPM at the seaward stations was observed in all four summer investigations (Figures 5a–5c, and 5f). In particular, the δ^{13}C values increased to levels higher than −18.3‰ at the three eastern stations along the sampling transect investigated on 26 August 2010 (Figure 5b). The two investigations shown in Figures 5b and 5c occurred within a time range of nine days, during which a typhoon passed through the ECS shelf and had a great influence on the study area. After the typhoon (on 4 September), the surface δ^{13}C values were greatly decreased compared to the five values for the same station sampled before the typhoon. As observed in March 2012 (Figure 4), during the two winter cruises in March 2011 and February 2012 (Figures 5d and 5g) the δ^{13}C values were overall lower compared to those in summer. During each of these two cruises, the lowest surface values (< −26.0‰) occurred at the most seaward stations (near 31°00′N and 123°00′–123°15′E) (Figures 5d and 5g).

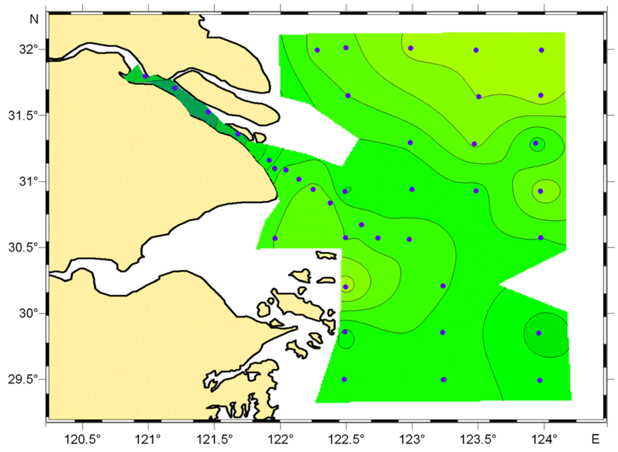

Figure 4. Distribution of POC δ^{13}C in the surface SPM during the Changjiang Estuary and western ECS cruise in March 2012. The color scale is the same as in Figure 3.

The results in Figures 3–5 show that the surface δ^{13}C distribution in SPM differed greatly between the summer and the winter cruises. Although the lowest values were observed in the

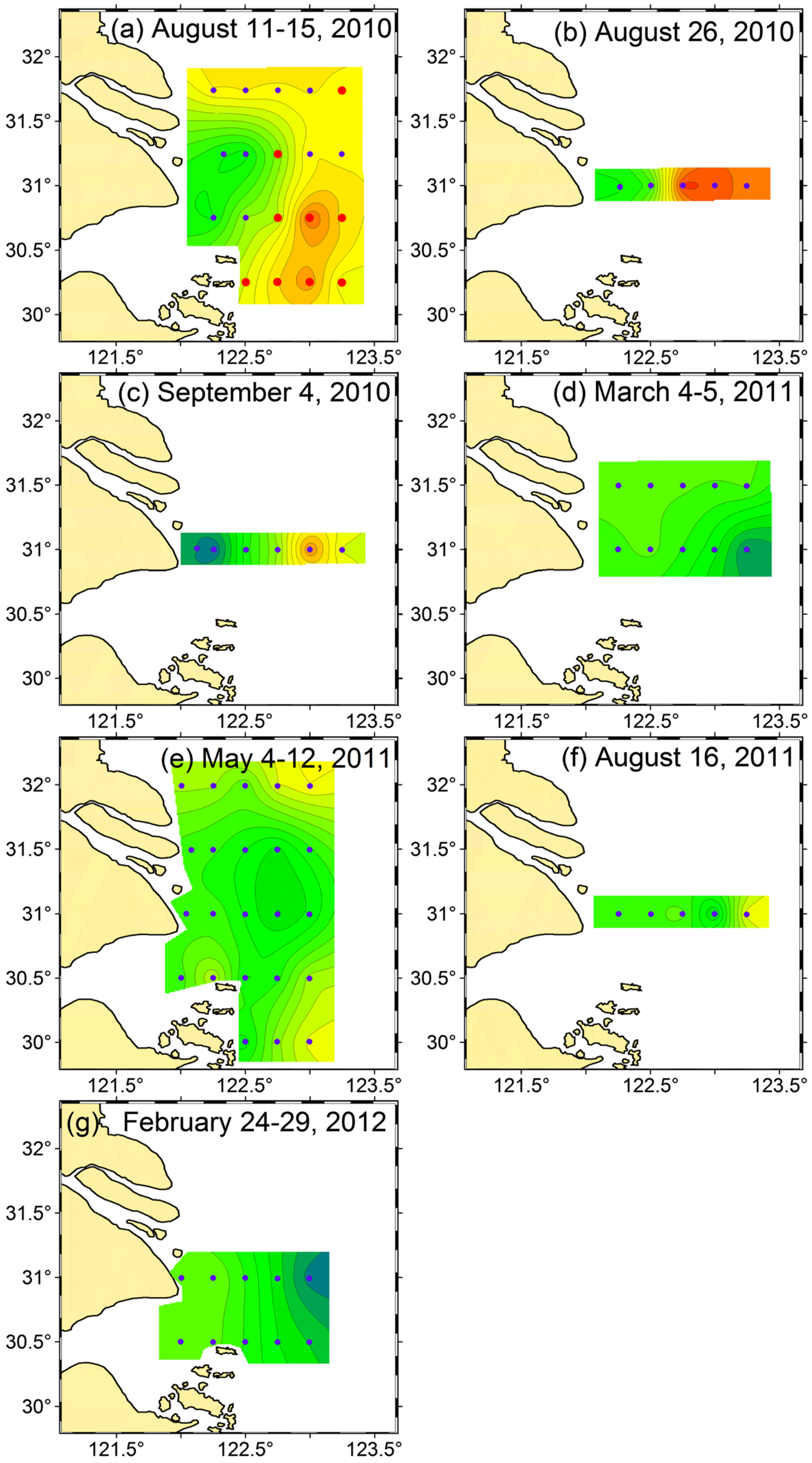

Figure 5. Distribution of POC $\delta^{13}C$ in the surface SPM during the remaining western ECS cruises that are not shown in Figures 3 and 4. The color scale is the same as in Figure 3. The red dots in panel (a) show the stations with the surface diatom accumulation.

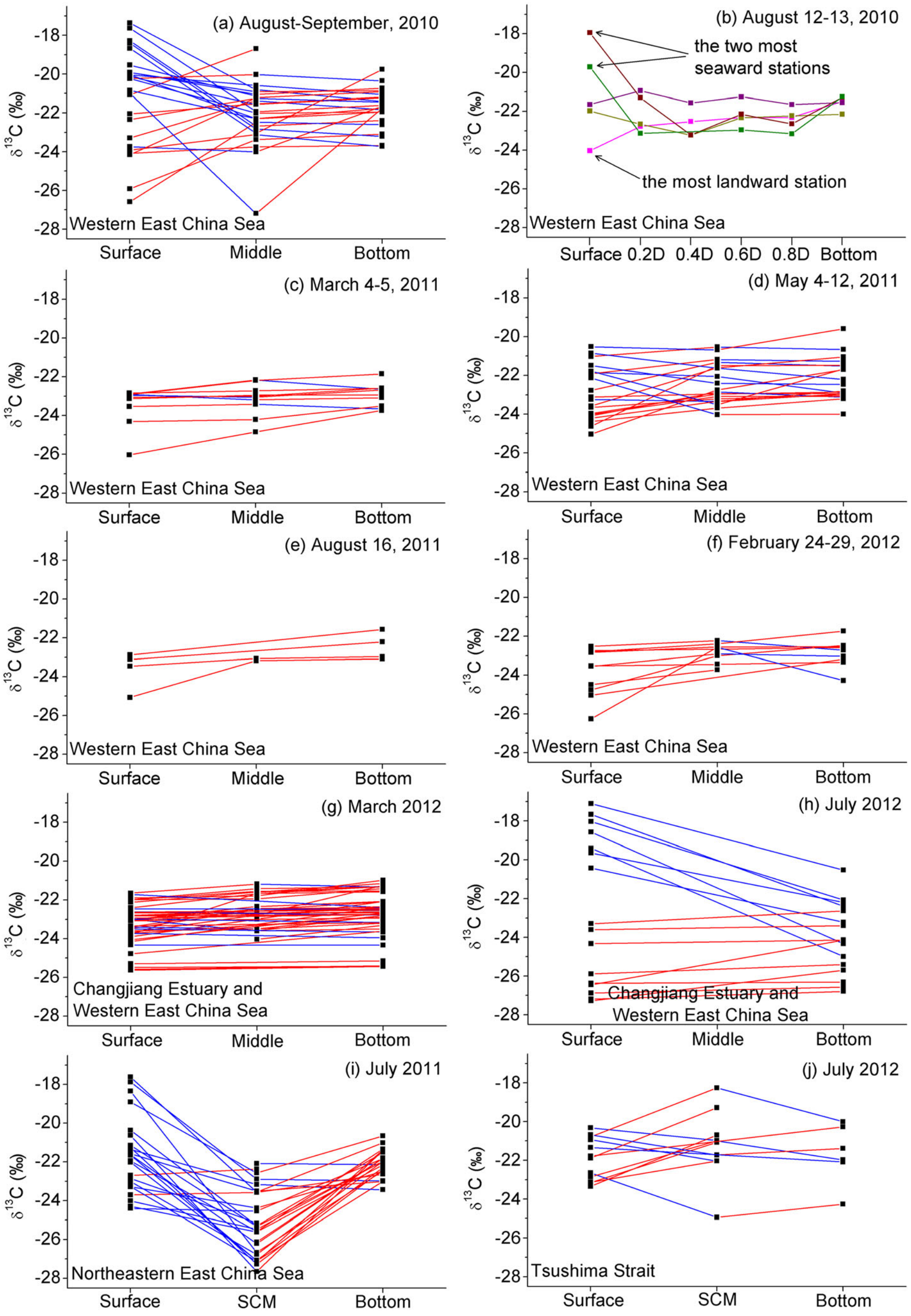

Figure 6. Vertical profiles of POC δ^{13}C in SPM from surface to middle layer (or SCM), and to the bottom in the Changjiang Estuary and western ECS cruises in different seasons, in the northeastern ECS cruise of July 2011, and in the Tsushima Strait cruise of July 2012. The red lines indicate the increasing downward trend while the blue lines indicate the decreasing trend.

Changjiang Channel during both seasons, in the western ECS an increasing trend of δ^{13}C from land to sea was generally observed in summer whereas a decreasing trend was found in winter. In the spring cruise in May 2011, the summer pattern of increasing surface δ^{13}C values from land to sea seemed to be observed (Figure 5e).

3.2. Vertical Variation of δ^{13}C

During all nine cruises, only surface samples were collected at some stations because of time constraints and insufficient sample volumes in the field. However, two to six samples from the sea surface to the bottom were collected at the other stations to obtain a vertical profile of δ^{13}C (Figure 6). In winter when the water column was generally vertically mixed in the western ECS, in most cases, the δ^{13}C values showed a

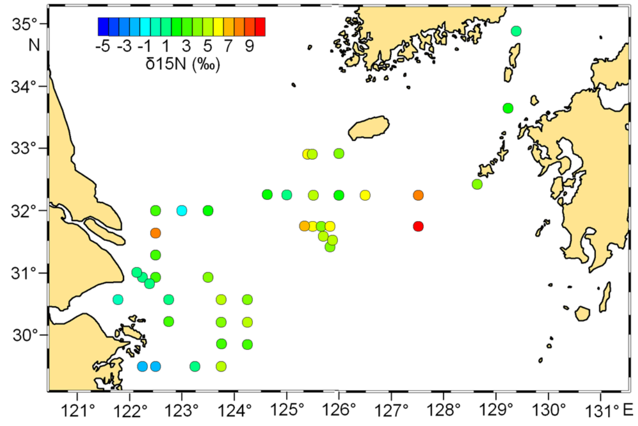

Figure 7. Distribution of SPM δ^{15}N at the surface along the Changjiang transport pathway in summer. The data are from the Changjiang Estuary and western ECS cruise in July 2012, from the northeastern ECS cruise in July 2011, and from the Tsushima Strait cruise in July 2012.

consistently increasing trend from the surface to the middle layer and to the bottom (Figures 6c, 6f, and 6g). In spring of May 2011, this increasing trend also was observed at most stations (Figure 6d). During the summer cruises when the water column was stratified, we expected to observe the same increasing trend as in the August 2011 cruise (Figure 6e). However, anomalous elevated δ^{13}C values were found at the surface of the offshore stations (Figures 6a, 6b, and 6h). Along the transect at latitude 30°45′N in August 2010 where more intensive samples from six vertical layers (surface, 0.2D (Depth), 0.4D, 0.6D, 0.8D, and bottom) were collected, the greatest vertical changes through the water depth occurred in the surface layers (Figure 6b). The pattern of increasing δ^{13}C values with depth commonly found in the western ECS suggests that when organic matter decomposition occurs during the SPM deposition process, the lighter carbon atoms (i.e., ^{12}C) are more easily removed from the organic matter substrate. However, this increasing trend could be transformed in summer by the sharp δ^{13}C increases at the surface (Figures 6a and 6h), which may be related to the accumulation of phytoplankton here (discussed later).

During the July 2011 cruise in the northeastern ECS and the July 2012 cruise in the Tsushima Strait (Figures 6i and 6j), samples were collected from the SCM layer, instead of the middle layer. During the northeastern ECS cruise, the most noticeable phenomenon was that POC from the SCM layer contained δ^{13}C values that were considerably lower than those from the surface and the bottom layers (Figure 6i). However, this pattern was not found during the July 2012 cruise in the Tsushima Strait, likely because it was a different study area and because fewer data were collected (Figure 6j). *Wang et al.* [2014] found that in ECS the surface water had mixed populations of micro-, nano-, and pico-phytoplankton, whereas in the SCM layer the water was dominated by the nano-phytoplankton. In the Tsushima Strait, the pico-phytoplankton was the dominant phytoplankton at the surface, while the micro-phytoplankton was the most important phytoplankton community in the SCM layer. The different phytoplankton communities between the surface and SCM in ECS and in the Tsushima Strait may at least partly explain the different vertical trends of δ^{13}C in POC between the two areas (Figures 6i vs. 6j).

3.3. Seasonal and Vertical Variation of δ^{15}N

Compared with δ^{13}C measurements, precise δ^{15}N measurements require the collection of much larger volumes of water. For this season, sufficient quantities of particulate matter were not always available after the field filtrations, and only a small number of valid δ^{15}N results were obtained in this study (Figure 7). The δ^{15}N values from three cruises (Changjiang Estuary and western ECS in July 2012, northeastern ECS in July 2011, and Tsushima Strait in July 2012) were combined, in a manner similar to δ^{13}C (Figure 3). Similarly, the limited data show that the δ^{15}N values increased slightly from the near-shore stations along the Chinese coast to the more seaward ECS shelf stations and then appeared to decrease from the northeastern ECS to the Tsushima Strait (Figure 7).

When comparing the δ^{15}N results from the sea surface in the western ECS between the three seasons, the δ^{15}N surface values were higher in the summer (average: 2.8‰ ± 2.2‰, $n = 42$; including the 19 values shown in Figure 8a and 23 values from the July 2012 cruise in western ECS shown in Figure 7) than in the spring of May 2011 (2.4‰ ± 1.9‰, $n = 10$; Figure 8b), and the lowest values were found in winter (−0.3‰ ± 2.9‰, $n = 9$; Figure 8c). The different stations where the data were obtained (Figures 7, 8a–8c) may also have contributed considerably to this seasonal difference in the average values.

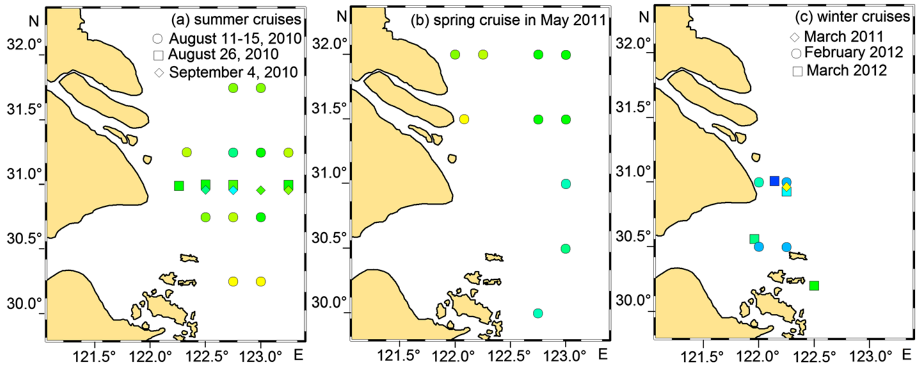

Figure 8. Distribution of SPM δ^{15}N in the surface of the Changjiang Estuary and western ECS cruises during different seasons. The results from the July 2012 cruise (that have been shown in Figure 7) are not included in panel (a). The color scale is the same as in Figure 7.

In contrast to the increasing δ^{13}C generally found in vertical profiles from the sea surface to the bottom in the Changjiang Estuary and western ECS (Figures 6a–6h), δ^{15}N showed a decreasing trend in most cases for all seasons (Figures 9a–9c). However, during the July 2011 cruise in the northeastern ECS (note the greatly reduced δ^{13}C values in the SCM during this cruise, as shown in Figure 6i), a consistent increasing trend of δ^{15}N with increasing depth was found in most cases (Figure 9d). These differing δ^{13}C and δ^{15}N profiles (Figure 6 vs. Figure 9) may suggest that during the mineralization of organic matter that accompanies SPM deposition, carbon and nitrogen may be incorporated via different biogeochemical processes, and they may behave differently in different areas.

3.4. δ^{13}C and δ^{15}N at the Anchor and Buoy Stations

Temporal variations of δ^{13}C and δ^{15}N were measured at the four anchor or buoy stations (Stations A, B, C, and D; Figures 2a and 2b). The results from these stations can help to elucidate the ranges of variation of δ^{13}C and δ^{15}N at the anchor stations under the influence of tidal currents in the Changjiang Estuary and western ECS (A and B), and at the buoy stations affected by the movement of the water masses through the Tsushima Strait (C and D). Stations A and B were investigated in March 2012. At Station A, δ^{13}C had an average value of −22.7‰ ± 0.1‰ at the surface ($n = 9$) and −22.6‰ ± 0.2‰ at the bottom ($n = 9$). The average values of δ^{15}N

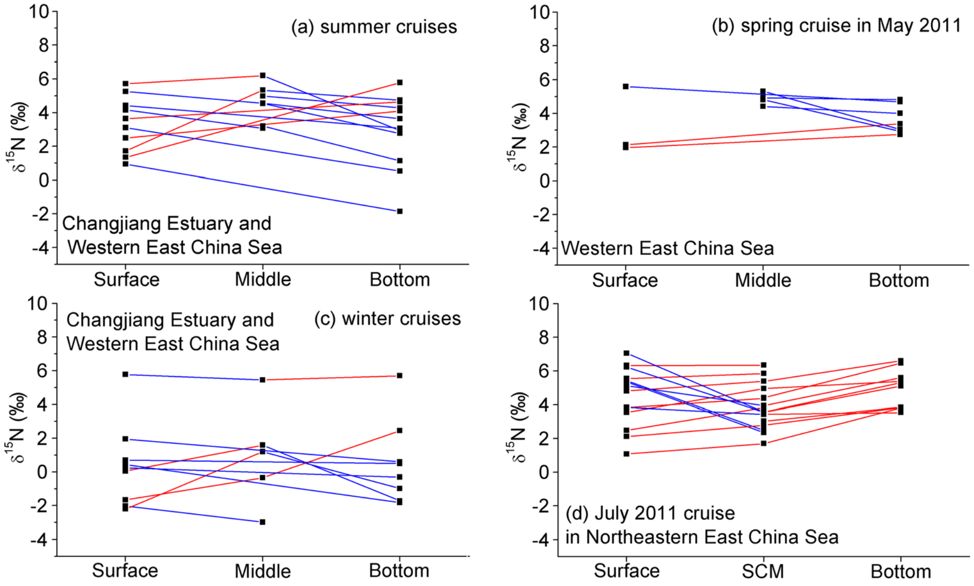

Figure 9. Vertical profiles of SPM δ^{15}N from the surface to the middle layer (or Subsurface Chlorophyll Maximum (SCM)), and to the bottom in the Changjiang Estuary and western ECS cruises during different seasons, and in the northeastern ECS cruise of July 2011. The red lines indicate the increasing downward trend while the blue lines indicate the decreasing trend.

at the surface and the bottom were −1.3‰ ± 2.8‰ ($n = 3$) and −2.5‰ ± 1.4‰ ($n = 6$), respectively. At Station B, the average $\delta^{13}C$ in the surface, middle, and bottom layers were −23.4‰ ± 0.3‰ ($n = 3$), −23.6‰ ± 0.1‰ ($n = 5$), and −23.1‰ ± 0.3‰ ($n = 4$), respectively. Due to insufficient nitrogen content accumulated on the filters, no valid $\delta^{15}N$ values were obtained at Station B. The $\delta^{13}C$ values at Station A were clearly higher than those at Station B throughout the vertical profile, and this trend of lower $\delta^{13}C$ at the more seaward station was consistent with the data trends often found from the winter cruises in the western ECS (Figures 5d and 5g).

Data for Stations C and D were obtained during the July 2012 cruise in the Tsushima Strait. During this period, both buoys moved approximately northeastward during the 10 h tracking period; the final tracking distances were 12.4 km for Buoy C and 15.6 km for Buoy D. To accumulate as much organic matter as possible on the filters, samples were collected from the SCM layer instead of the surface layer. During this cruise, a consistent trend for $\delta^{13}C$ had not been found along the vertical depth profiles from the transect survey (Figure 6j). The average $\delta^{13}C$ value for the SCM layer at Buoy C was −21.0‰ ± 0.5‰ ($n = 6$), which was significantly higher than that at Buoy D (−23.6‰ ± 1.2‰, $n = 4$) ($P < 0.01$). All $\delta^{15}N$ values at Buoy C were valid, and the average was 2.6‰ ± 1.2‰ ($n = 6$); however, all $\delta^{15}N$ values at Buoy D were invalid.

3.5. $\delta^{13}C$ and $\delta^{15}N$ in Phytoplankton Blooms

During the July 2012 cruise in the Changjiang Estuary and western ECS, a phytoplankton bloom was encountered at Station F (Figure 2a). Samples from two water patches that exhibited different colors (dark green and light green, separated by a distance of several meters) were collected. These two surface samples had much higher C (%) values (18.1% for dark green and 11.6% for light green water) than the bottom water (0.6%) or the surface waters at the two neighboring stations (Figure 2a, 1.0% for Station E and 1.8% for Station G). The $\delta^{13}C$ values of the two bloom samples were −26.5‰ (dark green) and −24.3‰ (light green). These values were somewhat lower than those at the bottom (−24.1‰) or in the surface waters from the two neighboring stations (−23.6‰ and −24.8‰ at Stations E and G, respectively). The $\delta^{15}N$ values at Station F were 1.3‰ for the dark green and 1.2‰ for the light green samples. Both were higher than the value of the surface sample from Station E (0.9‰). The $\delta^{15}N$ values at the bottom of Station F and at the surface of Station G were invalid.

Large-scale green macroalgae (*Ulva prolifera*) blooms broke out during the July 2011 cruise in the northeastern ECS, as had been observed in June 2008 [*Shi and Wang*, 2009; *Hu et al.,* 2010], and were often observed floating on the sea surface. During this cruise, two *U. prolifera* samples (at 31°39′N and 125°59′E on July 21, and at 31°48′N and 125°54′E on July 22) were collected, dried, thoroughly powdered and mixed in the laboratory, and then measured three times for both $\delta^{13}C$ and $\delta^{15}N$. The average $\delta^{13}C$ values for these two samples were −21.28‰ ± 0.06‰ ($n = 3$) and −21.94‰ ± 0.04‰ ($n = 3$), and the average $\delta^{15}N$ values were 5.8‰ ± 0.5‰ ($n = 3$) and 5.3‰ ± 0.4‰ ($n = 3$), respectively. These values were within the ranges of $\delta^{13}C$ and $\delta^{15}N$ measured for SPM collected during the same cruise (Figures 3 and 7).

Still during the northeastern ECS cruise in July 2011, a small-scale *Noctiluca scintillans* bloom was encountered at Station I when the ship transferred from Station H to J (Figure 2a), and the bloom sample was collected and filtered. The C (%) value of the bloom sample was 2.8%, whereas the values were 1.9% and 1.9%, respectively, for the two surface samples collected at Stations H and J. The $\delta^{13}C$ value of this sample (−21.5‰) was close to the surface value (−21.7‰) at the neighboring Station J, which was located several kilometers away. The $\delta^{13}C$ value of the surface SPM at Station H was −18.3‰, which was even higher than the bloom sample at Station I. The $\delta^{15}N$ value of the bloom sample at Station I (6.1‰) was higher than that at the surface of Station J (5.4‰). The surface $\delta^{15}N$ value at Station H was invalid.

3.6. Correlation Between $\delta^{13}C/\delta^{15}N$ and Other Parameters

$\delta^{13}C$ and C (%) showed a significant negative correlation during the February 2012 cruise in the western ECS for the surface samples and when results from all depth layers were combined, and a positive correlation during the July 2011 cruise in the northeastern ECS for the surface samples only (Table 1). However, neither a positive nor a negative correlation was found between these two parameters for any of the other seven cruises ($P > 0.05$). This is not expected because if the value of $\delta^{13}C$ is closely connected with the accumulation of phytoplankton, a positive correlation between $\delta^{13}C$ and C (%) should have been found. For $\delta^{13}C$ and C/N ratio (mol/mol), these two parameters showed a positive correlation during the July 2012 cruise in the

Table 1. Correlation Coefficients (*R*) Between $\delta^{13}C$, $\delta^{15}N$, and Other Parameters for Each of the Nine Cruises

R	$\delta^{13}C$ vs. C (%)	$\delta^{13}C$ vs. C/N	$\delta^{13}C$ vs. SPM	$\delta^{13}C$ vs. $\delta^{15}N$	$\delta^{15}N$ vs. C(%)	$\delta^{15}N$ vs. N(%)	$\delta^{15}N$ vs. C/N	$\delta^{15}N$ vs. SPM
August–September 2010 in western East China Sea (ECS)		$0.379^{*,a}$						
March 2011 in western ECS								
May 2011 in western ECS					$-0.714^{*,a}$ $-0.779^{**,b}$	$-0.731^{**,b}$		$0.689^{*,a}$
August 2011 in western ECS		$0.688^{*,b}$						
February 2012 in western ECS	$-0.776^{**,a}$ $-0.678^{**,b}$	$-0.651^{*,a}$ $-0.548^{**,b}$	$0.552^{**,b}$					
March 2012 in Changjiang Estuary and western ECS								
July 2012 in Changjiang Estuary and western ECS		$0.458^{**,a}$ $0.398^{**,b}$	$-0.269^{*,b}$	$0.586^{**,a}$			$0.410^{*,a}$	
July 2011 in northeastern ECS	$0.592^{**,a}$	$0.316^{**,b}$	$0.287^{*,b}$				$0.316^{*,b}$	
July 2012 in Tsushima Strait			$0.250^{*,b}$					

The two-star symbol (**) denotes the significant lever of $P < 0.01$ and the one-star symbol (*) denotes $P < 0.05$.
[a] means the correlation is for surface samples.
[b] means the correlation is for samples from all depths.

Changjiang Estuary and western ECS, and during the August–September 2010 cruise in the western ECS in the surface samples only. For the two cruises in the northeastern ECS in July 2011 and in the western ECS in August 2011, positive correlations were found only when results from all depth layers were combined. In contrast, during the February 2012 cruise in the western ECS, $\delta^{13}C$ and C/N ratio (mol/mol) showed a negative correlation. For $\delta^{13}C$ and SPM concentration (mg/L), no significant correlation was found if only surface samples were studied. However, when results from all depths were combined, significant positive correlation was found for the three cruises in the western ECS in February 2012, in the northeastern ECS in July 2011, and in the Tsushima Strait in July 2012. During the July 2012 cruise in the Changjiang Estuary and western ECS, a significant negative correlation between $\delta^{13}C$ and SPM concentration was found for samples from all depth layers.

In Table 1, the significant correlation between $\delta^{13}C$ and $\delta^{15}N$ was found only during the July 2012 cruise in the Changjiang Estuary and western ECS for surface samples. Significant negative correlation between $\delta^{15}N$ and C (%) was found during the May 2011 cruise in the western ECS. During this cruise, significant correlation was also found between $\delta^{15}N$ and N (%) when samples from all depths were combined and between $\delta^{15}N$ and SPM concentration (mg/L) when only surface samples were used. Positive correlation between $\delta^{15}N$ and C/N ratio (mol/mol) was found during the northeastern ECS cruise in July 2011 only when samples from all depths were combined, and during the Changjiang Estuary and western ECS cruise in July 2012 only when surface samples were used. Correlations for the other cruises were not studied because of the small amount of available $\delta^{15}N$ data.

We conclude that for most cruises no significant correlation between $\delta^{13}C$ and other parameters was found (Table 1). There seemed to be even less correlation for $\delta^{15}N$. For the two parameters of $\delta^{13}C$ and C/N ratio (mol/mol) that did show significant correlations during the five cruises in total, no consistently positive or negative correlation was found. This indicates that both $\delta^{13}C$ and $\delta^{15}N$ have unique and complex variation patterns over the Changjiang transport pathway.

Among the three cruises conducted in the Changjiang Estuary and the adjacent western ECS in July 2012, in the northeastern ECS in July 2011, and in the Tsushima Strait in July 2012, a significant correlation between $\delta^{13}C$ and C (%) for the surface samples was found only during the northeastern ECS cruise, and a significant correlation between $\delta^{13}C$ and C/N ratio (mol/mol) for surface samples was found only during the Changjiang Estuary and western ECS cruise (Table 1). In fact, along the Changjiang transport pathway from the Changjiang Channel to the Tsushima Strait, the variation patterns of C (%) and C/N ratio (mol/mol) were more complex than and different from that of $\delta^{13}C$ (Figure 10 vs. Figure 3). Both the Changjiang Channel and the northeastern ECS had low C (%) values, whereas high values occurred in the Tsushima Strait (Figure 10a). High C/N ratios (mol/mol) were found in the central ECS and the Tsushima Strait, whereas low values were recorded in the northeastern ECS (Figure 10b). The above results most likely suggest that $\delta^{13}C$ is a more suitable index than C (%) and C/N ratio (mol/mol) to trace the transport and dilution of the Changjiang terrestrial outputs of organic matter, as can partly be seen from the quite clear land-sea variation pattern in the Changjiang Estuary and the adjacent western ECS for $\delta^{13}C$ (Figures 3–5).

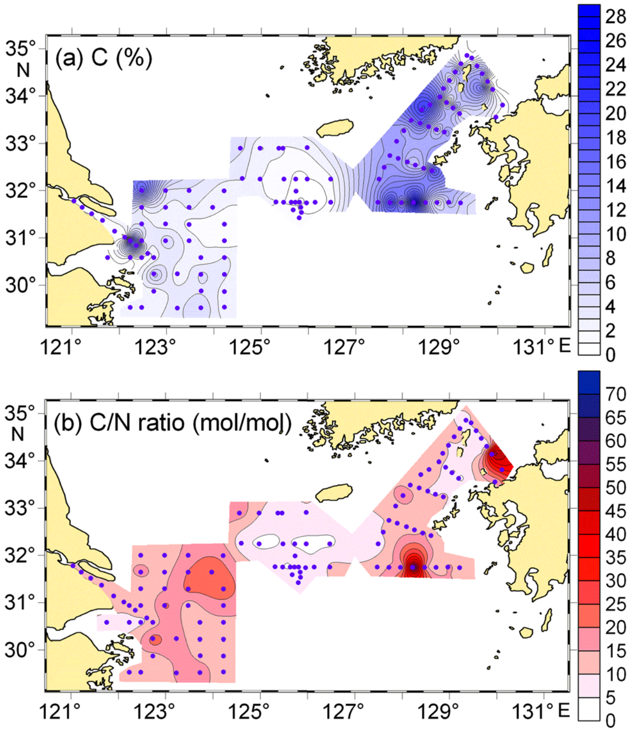

Figure 10. Distribution of SPM C (%) (a) and C/N molecular ratio (b) at the surface along the Changjiang transport pathway in summer. The data are from the Changjiang Estuary and western ECS cruise in July 2012, from the northeastern ECS cruise in July 2011, and from the Tsushima Strait cruise in July 2012.

4. Discussion

4.1. Large Variation of $\delta^{13}C$ With Low SPM

When all the $\delta^{13}C$ values from the Changjiang Estuary and western ECS for all depths, cruises, and seasons ($n = 417$) were plotted against the corresponding SPM value, an interesting phenomenon was detected (Figure 11). When the SPM amount was higher than 135 mg/L, the $\delta^{13}C$ values were fairly constant (−24.5‰ to −20.5‰). However, when SPM was lower than 135 mg/L, the $\delta^{13}C$ values showed a much greater range of variation (Figure 11).

Tan et al. [1991] found that $\delta^{13}C$ values of sediments from the ECS shelf fell within a narrow range from −22.9‰ to −20.8‰. *Kao et al.* [2003] reported that the $\delta^{13}C$ values of surface sediments over the entire ECS shelf (including the inner, middle, and outer shelf) ranged from −22.4‰ to −19.6‰. *Li et al.* [2012] reported that the $\delta^{13}C$ values in surface sediments from the inner ECS ranged between −23.1‰ and −20.9‰. All the above values were within [for *Tan et al.*, 1991; *Li et al.*, 2012] or similar to [for *Kao et al.*, 2003, although their study area was different from the present study] the $\delta^{13}C$ value range for the samples with higher SPM concentration in this study (−24.5‰ to −20.5‰, with SPM > 135 mg/L), suggesting that sediment resuspension may have largely contributed to these higher-SPM samples.

In Figure 11, 43 samples had $\delta^{13}C$ values higher than −20.5‰, and 35 samples had $\delta^{13}C$ values lower than −24.5‰. Among the 43 samples with high $\delta^{13}C$ and low SPM concentration (<135 mg/L), 38 were collected from the surface water during the summer. In addition, the salinity of all 43 samples was higher than 20.7 and the seawater temperature was higher than 23.1 °C for all the 38 surface samples in summer. Among the 35 samples that had low $\delta^{13}C$ and low SPM concentration (<135 mg/L), 18 samples were collected from the Changjiang Channel (eight from March 2012 and 10 from July 2012, salinity < 0.4) and six were from areas close to the Changjiang mouth in summer (i.e., still under the influence of the terrestrial materials, salinity of 11.5–22.2). The other 11 samples were from the more seaward stations in the western

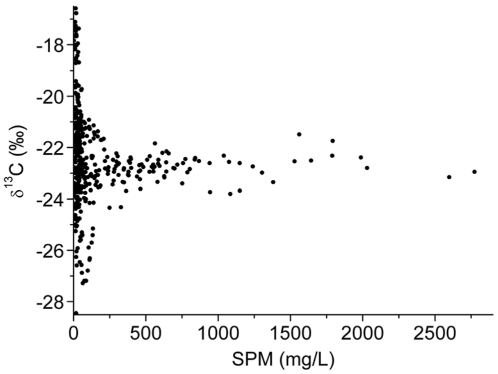

Figure 11. Variation of POC $\delta^{13}C$ against SPM. The data are all available $\delta^{13}C$ values from all depths and all seven Changjiang Estuary and western ECS cruises.

ECS during all seasons (29°51′–31°30′N, 122°30′–124°15′E; salinity $>$ 25.0), in which six were from surface and middle layers at the most seaward stations during each winter cruises (salinity $>$ 33.6) and likely under the influence of the intruding Kuroshio Subsurface Water with low δ^{13}C from -31‰ to -27‰ [*Wu et al.*, 2003].

Among the 18 samples collected in the Changjiang Channel (with low δ^{13}C and low SPM concentration, salinity $<$ 0.4, mentioned above), although no significant correlation between δ^{13}C and seaward distance was found for the samples from the March 2012 cruise ($n = 8$, $P > 0.05$; collected at the surface and bottom of four stations); a significant negative correlation between the two parameters was found for the samples from the July 2012 cruise ($R^2 = 0.47$, $n = 10$, $P = 0.03$; collected at the surface and bottom of five stations), contrary to the seaward increasing trend of δ^{13}C during this season when considering all samples over the much larger area (the whole western ECS) and over the much greater salinity range.

Based on the results from Figures 3–5 and 11, the basic patterns of variation of δ^{13}C along the Changjiang transport pathway can be described as follows: in summer, the Changjiang water with the lowest δ^{13}C in POC is discharged into the estuarine TMZ, where both the SPM concentration (up to values $>$ 700 mg/L, according to *Jiang et al.* [2013]) and salinity begin to increase. During transport over the TMZ, the POC δ^{13}C values in SPM increase. When the surface salinity is higher than 20 in ECS, the surface CDW often has phytoplankton accumulation in summer. At this point in time and space, the POC δ^{13}C values reach their highest levels within the entire Changjiang transport pathway. These values then decline and remain relatively constant throughout the remaining part of pathway until to the Tsushima Strait.

In winter, when the CDW encounters the estuarine TMZ, the POC δ^{13}C values in SPM increase and then remain relatively constant over the TMZ, even over the pelagic area where the SPM concentrations decrease sharply. In the western ECS, unlike those in summer, the δ^{13}C values do not increase because phytoplankton blooms do not occur frequently in this season. Instead, the CDW changes direction, flows southward, and then encounters the water masses influenced by the Kuroshio Subsurface Water [*Wu et al.*, 2003], when the δ^{13}C values often decrease to levels similar to or even lower than those in the Changjiang Channel.

4.2. Influence of Phytoplankton on δ^{13}C

During the western ECS cruise in 11–15 August in 2010, phytoplankton samples were collected by trawls with a pore size of 22 μm, and the concentrated samples were fixed with neutralized formalin. Phytoplankton species were identified and counted in the laboratory using an inverted microscope at 200× or 400× magnifications (L. Gao, East China Normal University, unpublished data, 2010). Considerable accumulation of the diatom *Chaetoceros* spp. was found at several stations (indicated by the red dots in Figure 5a), and these stations had higher surface salinity in general than those without the bloom (26.9 ± 2.9 vs. 21.9 ± 5.9; $P = 0.007$). However, the expected higher C (%) average value at the surface of the stations with a bloom compared with those without bloom was not found (2.2% ± 1.0% for the former vs. 2.3% ± 1.4% for the latter). Although the higher average δ^{13}C value at surface was recorded at the stations with a bloom (-20.2‰ ± 1.4‰ vs. -21.4‰ ± 1.9‰), the difference was not statistically significant ($P > 0.05$).

Our LISST data support the hypothesis that the increase in δ^{13}C at seaward stations during the summer cruises (Figures 3, 5a–5c, and 5f) may have resulted from the phytoplankton accumulation in the surface waters. During the western ECS cruises in summer (August–September 2010), spring (May 2011), and winter (February 2012), the profiles of total floc concentrations (μL/L) and mean size (μm) were recorded by LISST, together with turbidity profiles collected by OBS. During the summer and the spring cruises when the water columns were stratified in the western ECS, OBS-derived turbidity increased considerably from the surface to the bottom, which reflects the SPM deposition and bottom resuspension (Figures 12a–12d). During the winter cruise in February 2012, however, the water was vertically mixed and turbidity was relatively constant throughout the water column (Figure 12e). The index of total floc concentration from LISST increased consistently downward in May 2011 (Figure 12i) and remained relatively constant in February 2012 (Figure 12j), and during these two cruises, the floc mean size (also from LISST) generally exhibited slightly decreasing or constant profiles, respectively (Figures 12n and 12o). In contrast, during the August–September cruise in 2010 the total floc concentration profiles (especially those at the seaward stations) were no longer consistent with those for turbidity because the maximum floc concentration

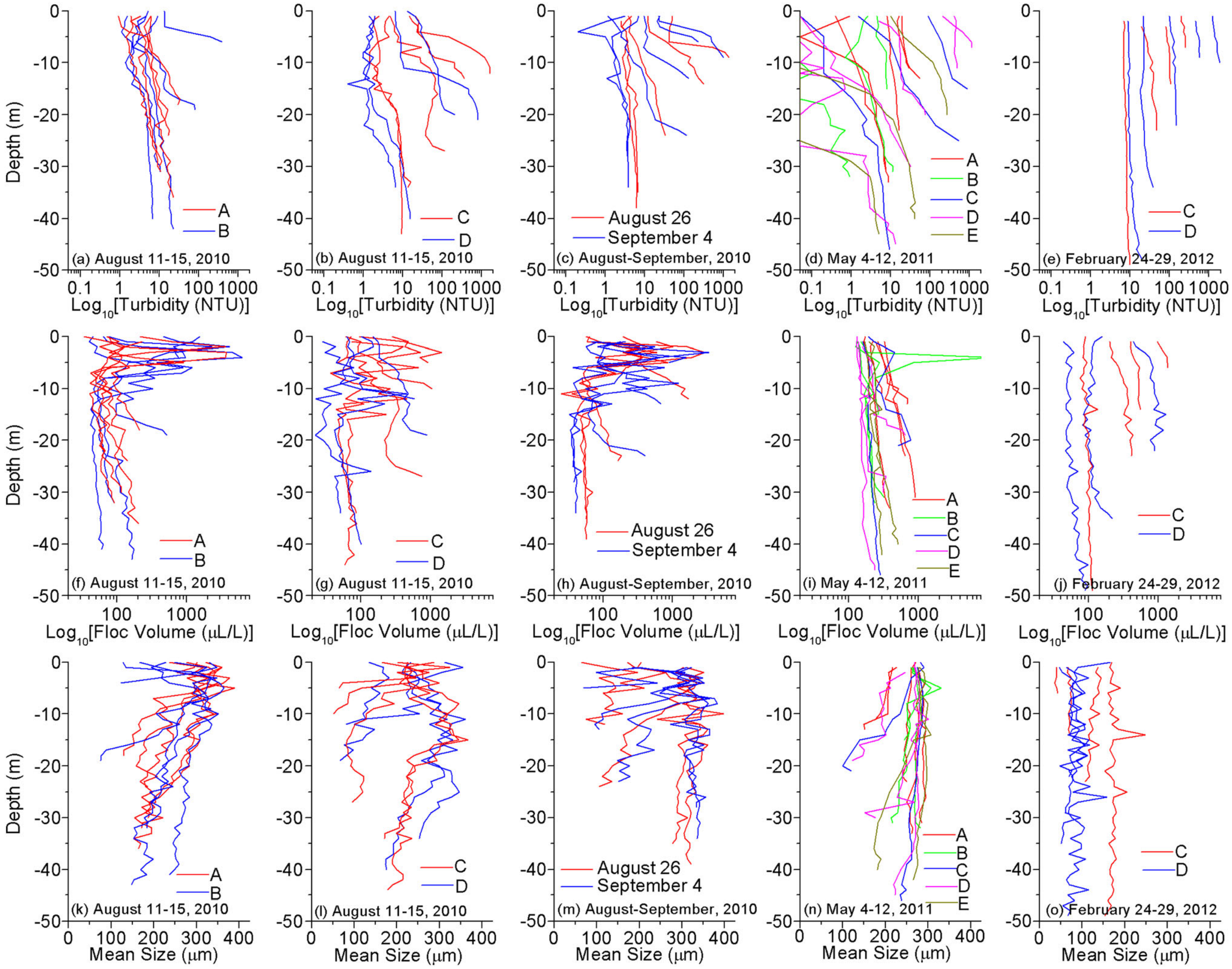

Figure 12. Vertical profiles of seawater turbidity (from Optical Backscatter Sensor (OBS)), and total floc concentration and mean grain size (from Laser In Situ Scattering and Transmissometry (LISST)) in the three cruises when both OBS and LISST data are available. Note the $\log_{10}$ scales for turbidity and total floc concentration. From north to south, the four latitudinal transects in the August–September 2010 cruise are denoted by Transects A, B, C, and D; the five transects in the May 2011 cruise are denoted by Transects A, B, C, D, and E; and the two transects in February 2012 cruise are denoted by Transects C and D. These latitudinal transects are indicated in Figure 2.

appeared in the near-surface layers (Figures 12f–12h). At the same time, the mean size also was highest in these near-surface layers (Figures 12k–12m). These results suggest that the peak floc concentration and the maximum size in the profiles from the summer cruise may have resulted from the near-surface accumulation of phytoplankton, although this accumulation was not recorded by the OBS turbidity signals (Figures 12a–12c).

Two issues remain unresolved. First, visually identified phytoplankton blooms were encountered during the cruises in the Changjiang Estuary and western ECS in July 2012 (Station F in Figure 2a) and in the northeastern ECS in July 2011 (the *Ulva prolifera* bloom, as well as the *N. scintillans* bloom found at Station I in Figure 2a). The collected *Ulva prolifera* samples and the filtered SPM samples collected from Stations F and I should consist of purer phytoplankton cells, yet the δ^{13}C values of these samples were all less than −21‰. Thus, the presence of the bloom cannot explain the increase of δ^{13}C observed at the seaward stations during the summer cruises (Figures 3, 5a–5c, and 5f). Second, during the northeastern ECS cruise in July 2011, considerably lower δ^{13}C values were found in the SPM samples from the SCM layer (Figure 6i), and these results appear to be inconsistent with the field observations. Because the chlorophyll content is the index used to quantify phytoplankton biomass, the higher chlorophyll content in the SCM layers should have led to higher POC δ^{13}C values in SPM.

Harmelin-Vivien et al. [2008] reported that in the Gulf of Lions in the northwestern Mediterranean, the surface POC had a systematically lower δ^{13}C than the corresponding micro-phytoplankton collected from the fluorescence maximum layer at the same station. This seems to differ from what we found for the SCM in the northeastern ECS (Figure 6i). However, *Zhuang et al.* [2007] extracted pure chlorophyll molecules from SPM collected in the Changjiang Estuary and the adjacent western ECS and found that the δ^{13}C values of these pure molecules ranged from −32.29‰ to −28.17‰ (average: −29.75‰), which were considerably lower than those of the original POC substrates (range: −23.19‰ to −17.92‰, average: −20.67‰). These above findings illustrate the importance of differentiating between the carbon present in the chlorophyll molecules, in the phytoplankton cells, and in other POC molecules within SPM [*Chang et al.*, 2003], because there is most likely a large difference in δ^{13}C among these different organic carbon pools within POC [*Qian et al.*, 1996; *Sachs et al.*, 1999; *Sachs and Repeta*, 2000]. Furthermore, the δ^{13}C values of bulk POC may also be greatly influenced by the δ^{13}C signatures of the local dissolved inorganic carbon (DIC) [*Sheu et al.*, 1996; *Barth et al.*, 1998] and dissolved organic carbon (DOC) molecules [*Wang et al.*, 2004]; this issue in the Changjiang Estuary and ECS requires further study.

4.3. Comparison With Previous Studies

Tan et al. [1991] measured the POC δ^{13}C of surface SPM samples in the Changjiang Estuary and the adjacent ECS in June 1980 and found that the values varied between −25.4‰ and −19.7‰. In winter (November 1981), however, the values ranged between −26.6‰ and −23.7‰. During the two cruises in a similar area, *Shi* [1993] found that the surface SPM in winter (January 1986) had POC δ^{13}C values ranging from −25.64‰ to −22.64‰. However, in summer (July 1986) when salinity was less than 25, the δ^{13}C values ranged between −26.39‰ and −22.03‰. When salinity was higher than 25, the values ranged between −21.79‰ and −18.64‰ [*Shi*, 1993]. The results from *Tan et al.* [1991] and *Shi* [1993] are in agreement with the results obtained in this study during both summer and winter. *Wu et al.* [2003] studied the δ^{13}C and δ^{15}N values of SPM in autumn (October–November 2000), along the transect that began near the Changjiang mouth and ended at the Ryukyu Island. They found that the water near the Kuroshio Subsurface Water had a δ^{13}C value below −30‰, which was lower than those in the Changjiang Channel reported in this study (Figures 3 and 4) and was consistent with our data showing that lower δ^{13}C values were often found in the eastern stations of the western ECS in all seasons and at all depths (e.g., those surface samples at the most seaward stations in the winter cruises shown in Figures 5d and 5g). *Zhang et al.* [2007] studied the δ^{13}C values in the Changjiang Estuary in July 2001 and did not find the elevated δ^{13}C values at the seaward stations, which was most likely because their study area was too close to the Changjiang mouth.

In recent decades, the Changjiang sediment discharge fluxes have declined sharply due to the construction of a large number of dams and reservoirs within the Changjiang drainage basin [*Yang et al.*, 2005, 2006, 2007, 2011]. The chemical and biological environments have also been greatly modified [*Jiao et al.*, 2007; *Zhou et al.*, 2008; *Dai et al.*, 2011; *Yu et al.*, 2011; *Gao et al.*, 2012]. Therefore, it is useful to compare the δ^{13}C and C/N ratio (mol/mol) in SPM of the Changjiang Estuary and the adjacent ECS from this study with those in the literature (Figures 13 and 14). Because the POC δ^{13}C values in the Changjiang Estuary and western ECS display considerable seasonal variations (described above), the δ^{13}C and C/N molecular ratio in Figures 13 and 14 were plotted against salinity specific for each sampling month (if available). The δ^{13}C values in the high-salinity water were lower in the winter months (Figures 13a, 13b, and 13i) and greatly elevated in the summer months (Figures 13e–13g). When comparing the results between different years (e.g., for July and August; Figures 13e and 13f), no clear inter-year differences were observed. Similarly, in Figure 14 the C/N molecular ratio versus salinity plots did not differ greatly between recent years (2010–2012; Figures 14a–14c, 14e–14g) and past years (1980–1981; Figures 14d and 14h).

4.4. Seasonal Variation of δ^{13}C Along the CDW Transport Pathway

The δ^{13}C results measured in the Changjiang Channel (−25.6‰ to −25.1‰ in March 2012, and −27.3‰ to −25.4‰ in July 2012, salinity < 0.4) suggest that the Changjiang basin is dominated by plants using the C3-pathway of CO_2 uptake, rather than those employing the C4-pathway [*Wu et al.*, 2007], because these two categories of plants have distinct δ^{13}C ranges, −34‰ to −24‰ for the former, and −16‰ to −9‰ for the latter [*Smith and Epstein*, 1971; *Onstad et al.*, 2000]. During the summer cruises, when the Changjiang terrestrial materials are transported out of the Changjiang mouth, the CDW encounters the intensified TWC

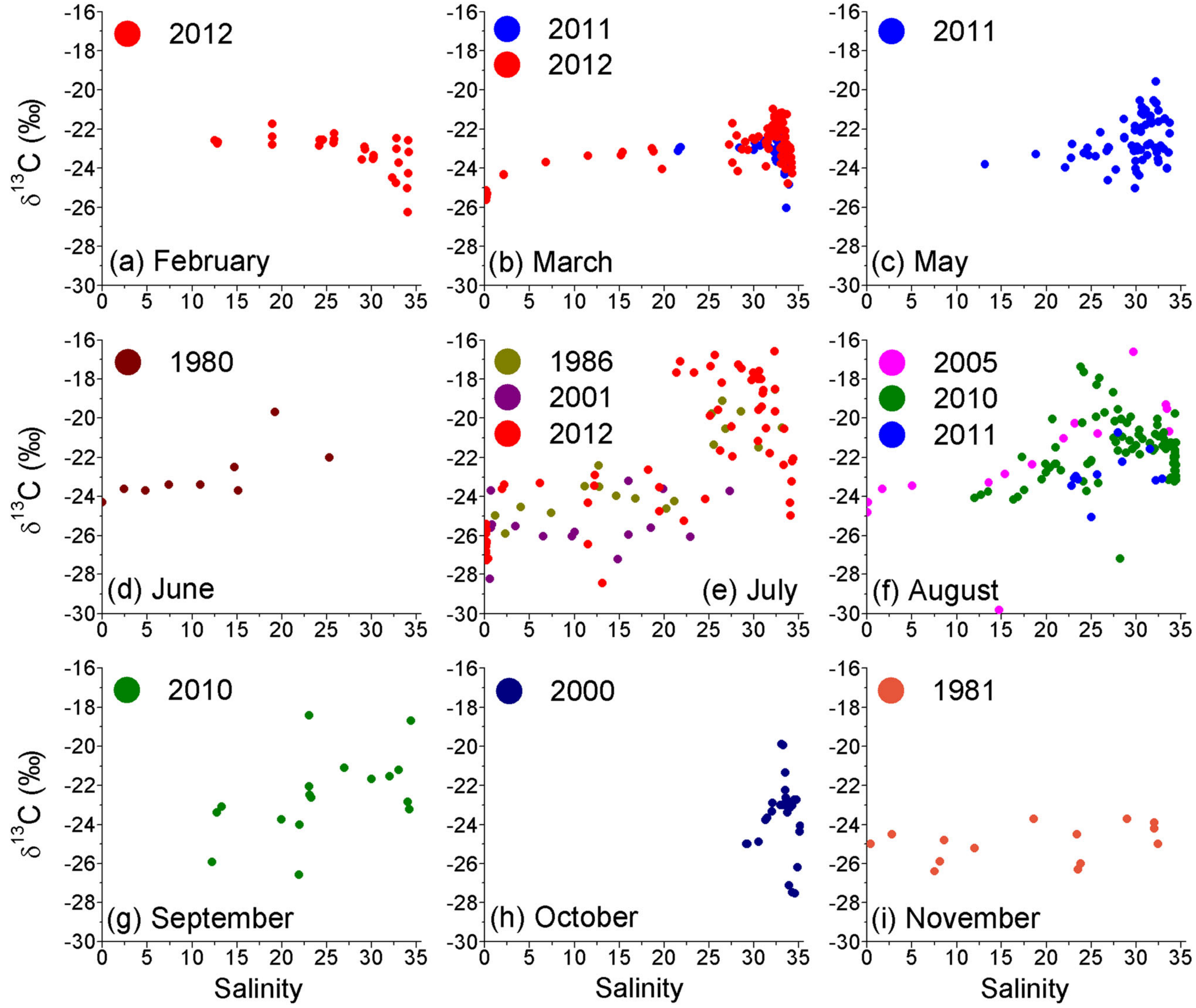

Figure 13. Variation of POC δ^{13}C in SPM of all depths against salinity during each month available for the Changjiang Estuary and the adjacent ECS. The data are from the present study as well as digitized from literature, including *Tan et al.* [1991] for June 1980 and November 1981, *Lin* [2007] for August 2005, *Shi* [1993] for July 1986, *Wu et al.* [2003] for October 2000, and *Zhang et al.* [2007] for July 2001.

that flows northeastward parallel to the 50-m isobath until approximately 30°N latitude [*Zhu et al.*, 2004; *Chen and Sheu*, 2006; *Moon et al.*, 2010]. According to *Gong et al.* [1996], the TWC is defined by water with a salinity of 31–34.2 and temperatures higher than 23 °C. During the summer cruise in July 2012 in the Changjiang Estuary and western ECS, with a relatively large study area, the TWC signals were found at the surface of the three eastern-most stations along the southern-most 29.5°N latitude. The δ^{13}C values at the surface and bottom of the three stations ranged from −22.2‰ to −16.6‰, showing a large variation range but in agreement with the range of variability at the other stations (−28.4‰ to −16.8‰). Therefore, the δ^{13}C values at these stations appear to depend more on the local phytoplankton biomass, and the potential change caused by the TWC intrusion appears to be minor. For Kuroshio water, *Wu et al.* [2003] found a depleted δ^{13}C signature (−31‰ to −27‰) for the Kuroshio Subsurface Water. However, during all summer cruises in the western ECS in this study, no Kuroshio signal (salinity > 34.2, according to *Gong et al.* [1996]) was recorded. During the northeastern ECS cruise in July 2011, the Kuroshio signal was detected at > 40 m depth at two stations; however, no depleted δ^{13}C signature was found (all the three δ^{13}C values in Kuroshio were invalid). During the Tsushima Strait cruise in July 2012 with a large input of Kuroshio water, no δ^{13}C values less than −25.5‰ were found, which is inconsistent with the low δ^{13}C signatures found by *Wu et al.* [2003] in Kuroshio Subsurface Water in a different

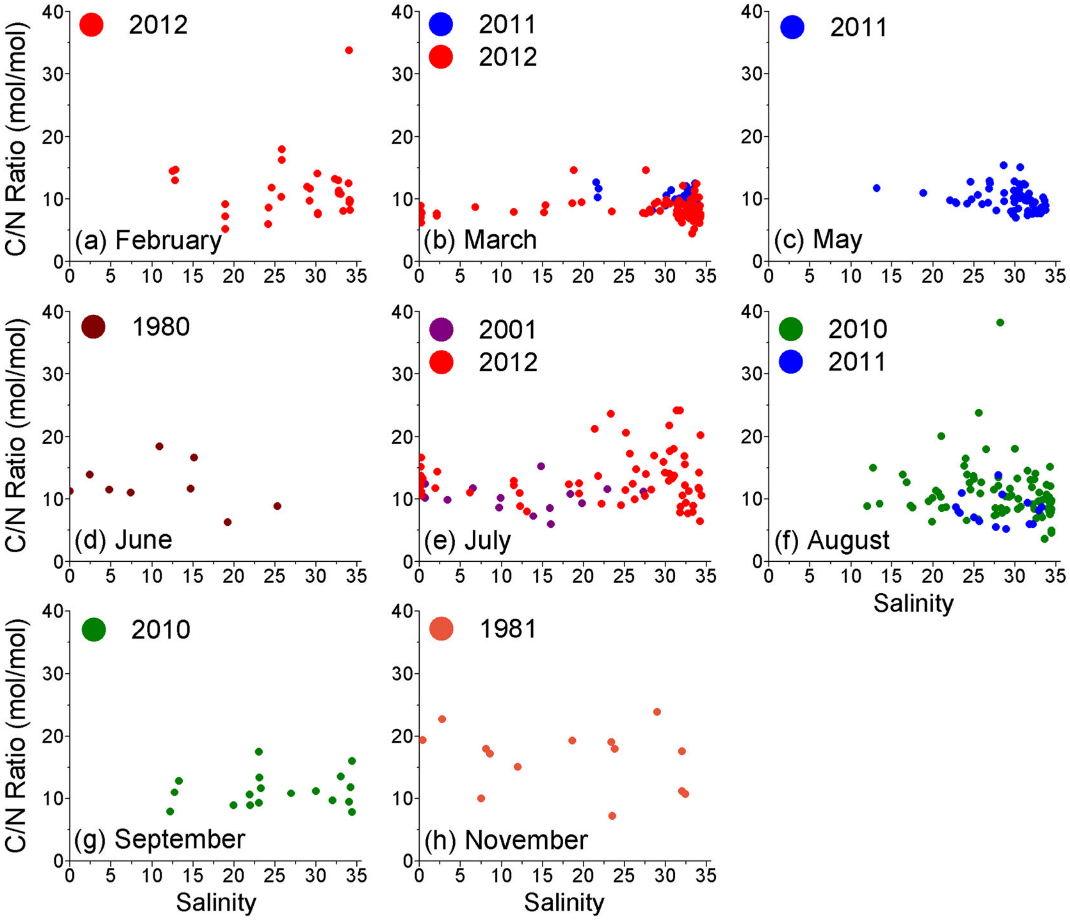

Figure 14. Variation of SPM C/N molecular ratio of all depths against salinity during each month available for the Changjiang Estuary and the adjacent ECS. The data are from the present study as well as digitized from literature, including *Tan et al.* [1991] for June 1980 and November 1981, and *Zhang et al.* [2007] for July 2001.

area (approximately 28°–29°N and 126°–127°E). These results again suggest that the POC δ^{13}C values are determined more by the local microbial and biological processes, than by the origin of the water mass.

In the Tsushima Strait, the surface POC δ^{13}C values were relatively constant with a small range of variability of −24.0‰ to −20.2‰, which was lower than those from the central ECS (Figure 3). *Wang et al.* [2014] found mixed populations of micro-, nano-, and pico-phytoplankton at the surface in ECS whereas pico-phytoplankton dominated at the surface in the Tsushima Strait. This different phytoplankton size and community may have contributed greatly to the different POC δ^{13}C values measured in these two areas, as well as the generally higher C (%) and higher C/N ratio (mol/mol) at the surface in the Tsushima Strait than in the northeastern ECS (shown in Figures 10a and 10b, respectively).

In winter, the TWC is not as strong as in summer [*Zhu et al.*, 2004; *Chen and Sheu*, 2006], and the CDW flows southward along the Chinese coasts. Conversely, the YSWC becomes stronger in winter, which may greatly influence the POC δ^{13}C values in the northeastern ECS near the Jeju Island [*Ma et al.*, 2006; *Xu et al.*, 2009]. However, based on the results of the March 2012 cruise in the western ECS, the δ^{13}C values (−22.0‰, −21.6‰, and −21.5‰ in the surface, middle, and bottom layers, respectively) at the most northeastern stations (32°N and 124°E) still fell into the variation ranges of those at the inner stations (−25.6‰ to −21.0‰, the surface values can be found in Figure 4). According to *Gong et al.* [1996], no TWC signal was found in all our winter cruises in the western ECS (water with salinity of 31–34.2 and temperatures > 23 °C). During the two winter cruises in March 2011 and in February 2012, the depleted δ^{13}C signatures at the surface of the most seaward stations (Figures 5d and 5g) may still reflect the potential influence of the intruding Kuroshio water that is characterized by low δ^{13}C values (−31‰ to −27‰, according to *Wu et al.* [2003]).

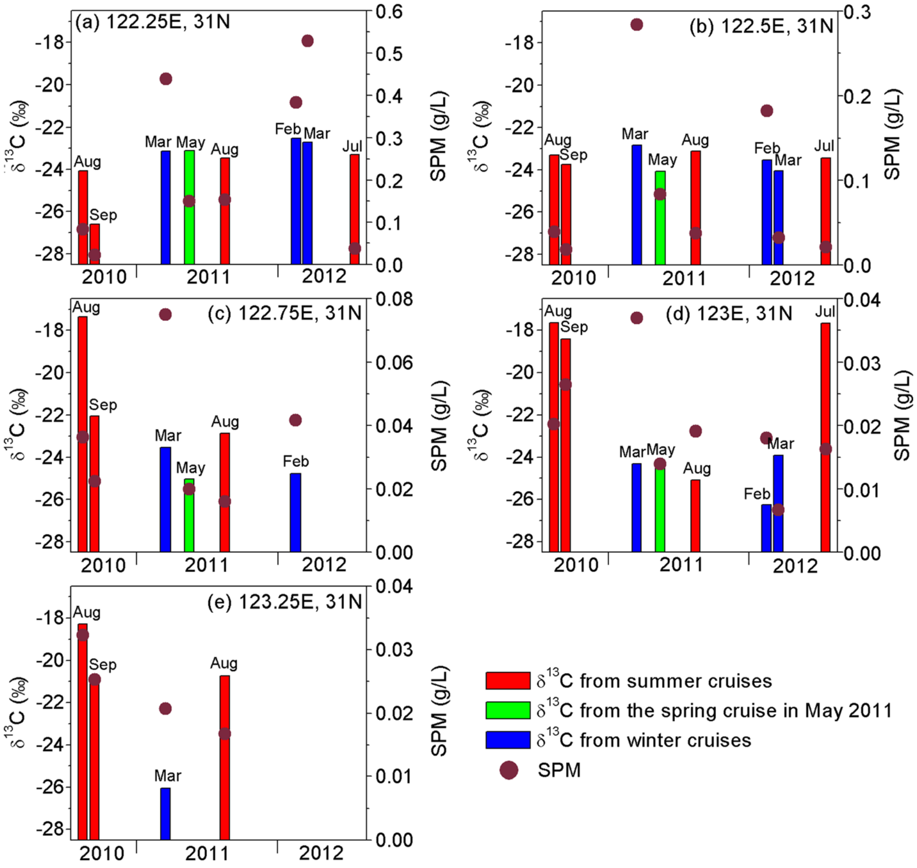

Figure 15. Temporal variation of POC δ^{13}C in suspended particulate matter (SPM) and the SPM amount at the surface of the five stations along the transect at 31°N, which has been surveyed repeatedly during the study period from 2010 to 2012.

In the western ECS in general, the POC δ^{13}C values appear to be heavier in summer than in winter. To show this seasonal variation pattern more clearly, the variations of surface δ^{13}C at the stations along 31°N are plotted in Figure 15. As shown in Figures 3–5, during the three years from 2010 to 2012 this line has been investigated eight times. At the two inner stations where the SPM at surface was relatively higher, no distinct difference of δ^{13}C between summer and winter cruises was observed (Figures 15a and 15b). However, at the three seaward stations where the SPM at surface was generally low, high δ^{13}C values (> -22‰) were recorded for the summer cruises (Figures 15c–15e). At each station in Figure 15, the higher surface SPM values appeared during the winter cruises, especially for the inner stations. *Yamaguchi et al.* [2013] showed that in spring and summer when the blooms appeared, the SPM in ECS was dominated by phytoplankton cells and phytoplankton-related organic particles. However, in winter, the SPM in ECS was dominated by resuspended sediment particles. This seasonal SPM composition change may largely explain the different distribution and variation of δ^{13}C between the summer and winter cruises, as well as why the higher δ^{13}C values in POC (> -20.9‰) can only be found in summer along the Changjiang transport pathway (Figures 3–5, 13, and 15).

5. Conclusion

Understanding the distribution and variation of δ^{13}C and δ^{15}N in POC can provide information on the biogeochemistry of carbon and nitrogen in the highly dynamic land-sea interaction area like the Changjiang Estuary and the adjacent ECS shelf. It can also help to trace how the properties and composition of the Changjiang-derived POC change during its long-distance transport process over ECS. In addition, the stable isotope ratios of carbon and nitrogen are useful in differentiating the POC composition, and potentially act as an efficient tool to study environmental change in the coastal areas on decadal time-scales because the SPM δ^{13}C and δ^{15}N may respond to the changed phytoplankton productivity in oceans, as well as to changed terrestrial fluxes from land. In this study, the most distinctive variation of δ^{13}C is its seasonal change over ECS.

In summer, after the ^{13}C-depleted POC was transported out of the Changjiang mouth, the δ^{13}C values were greatly elevated in response to the enhanced contribution by phytoplankton cells and the related organic material. In winter, however, the surface δ^{13}C values over the ECS shelf were relatively low compared with those in summer and remained relatively constant over this large study area. The δ^{15}N values in POC also appear to be lower in winter than in summer. The lower δ^{13}C and δ^{15}N values over ECS in winter were primarily attributed to the elevated amount of resuspended sediment in the SPM during this season. Our results also show that in most cases of the western ECS cruises the δ^{13}C values increased from surface to bottom, most likely caused by the degradation processes ongoing in the settling organic particles. No clear decadal change in δ^{13}C and C/N molecular ratio in SPM over the ECS was found compared with the Changjiang transported SPM fluxes that have decreased sharply in recent decades. More detailed studies are needed to answer questions, such as how the SPM δ^{13}C and δ^{15}N variations respond to the different bloom species with different sizes, how the δ^{13}C and δ^{15}N values in the bulk POC interact with those for the DOC and DIC, and how these δ^{13}C and δ^{15}N values respond to the changing contribution of each specific biogenic molecule to the seawater POC.

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