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Grain size composition and transport of sedimentary organic carbon in the Changjiang River (Yangtze River) Estuary and Hangzhou Bay and their adjacent waters

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Abstract

Surface sediments from the Changjiang River (Yangtze River) Estuary, Hangzhou Bay, and their adjacent waters were analyzed for their grain size distribution, organic carbon (OC) concentration, and stable carbon isotope composition ($\delta^{13}C$). Based on this analysis, about 36 surface sediment samples were selected from various environments and separated into sand (>0.250 mm, 0.125–0.250 mm, 0.063–0.125 mm) and silt (0.025–0.063 mm) fractions by wet-sieving fractionation methods, and further into silt- (0.004–0.025 mm) and clay-sized (<0.004 mm) fractions by centrifugal fractionation. Sediments of six grain size categories were analyzed for their OC and $\delta^{13}C$ contents to explore the grain size composition and transport paths of sedimentary OC in the study area. From fine to coarse fractions, the OC content was 1.18%, 0.51%, 0.46%, 0.42%, 0.99%, and 0.48%, respectively, while the $\delta^{13}C$ was –21.64‰, –22.03‰, –22.52‰, –22.46‰, –22.36‰, and –22.28‰, respectively. In each size category, the OC contribution was 42.96%, 26.06%, 9.82%, 5.75%, 7.09%, and 8.33%, respectively. The OC content in clay and fine silt fractions (<0.025 mm) was about 69.02%. High OC concentrations were mainly found in offshore modern sediments in the northeast of the Changjiang River Estuary, in modern sediments in the lower estuary of the Changjiang River and Hangzhou Bay, and in Cyclonic Eddy modern sediments to the southwest of the Cheju Island. Integrating the distribution of terrestrial OC content of each grain size category with the $\delta^{13}C$ of the bulk sediment indicated that the terrestrial organic material in the Changjiang River Estuary was transported seaward and dispersed to the Cyclonic Eddy modern sediments to the southwest of the Cheju Island via two pathways: one was a result of the Changjiang River Diluted Water (CDW) northeastward extending branch driven by the North Jiangsu Coastal Current and the Yellow Sea Coastal Current, while the other one was the result of the CDW southward extending branch driven by the Taiwan Warm Current.

Key words: Changjiang River (Yangtze River) Estuary, Hangzhou Bay, grain size composition, organic carbon, material transport

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1 Introduction

The quantity and composition of sedimentary organic matter (OM) varies greatly in different marine environments. Identifying the distribution of OM and the factors that control its distribution in the various marine sedimentary environments is the key to understanding the global carbon cycle. The adsorption capacity of OM is significantly influenced by particle size and sediment composition. Sediment grain size distributions are controlled by factors such as hydrodynamic sorting conditions, circulation patterns, and material sources. Hydrodynamic sorting is one of the most active processes in the ocean, and has a very important role in sediment transport and deposition. Moreover, it directly controls sediment sorting, distribution, erosion, and deposition, and indirectly affects the distribution of sedimentary OM (Bergamaschi et al., 1997; Volkman et al., 2000; Song, 1997). The spatial range of hydrodynamic processes in the Changjiang River (Yangtze River) Estuary, Hangzhou Bay, and the adjacent waters extends to the Changjiang River Diluted Water (CDW), runoff from the Qiantang River, the reversing tidal current, the East China Sea Coastal Current, the Taiwan Warm Current (TWC), the North Jiangsu Coastal Current (NJCC), the Yellow Sea Coastal Current (YSCC), and the Yellow Sea Warm Current (Fig. 1). With a large amount of terrestrial matter and nutrient input, the burial efficiencies of riverine and marine OM in this area are

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38% and 5.5%, respectively, far higher than the world averages of 20% and 0.8% (Deng et al., 2006). It is therefore the ideal region for studying the transportation and grain size composition of sedimentary organic carbon (OC). A number of recent studies have focused on the sedimentary OC distribution of different sedimentary environments in the Changjiang River Estuary (Gao et al., 2007; Zhang et al., 2007; Wang and Xian, 2011; Zhu et al., 2011; Hu et al., 2012), including a preliminary study of the sedimentary OC distribution of various size fractions (Zhang et al., 2009). However, these studies mainly focused on the Changjiang River Estuary and its adjacent continental shelf, and excluded the Hangzhou Bay waters. Further research is needed on the OC distribution and its influencing factors on the entire sedimentary environment in this area, including the Changjiang River Estuary, Hangzhou Bay, and the adjacent continental shelf. In particular, there are some regions of interaction where the CDW is conveyed northward and encounters the NJCC as it moves southward, where the CDW is transported southward and encounters the Zhoushan Islands' barrier, and where the CDW flows eastward and is impeded by water masses of the YSCC and TWC. The hydrodynamic changes and water mass transport patterns in these regions are essential for predicting changes in OC in the ecological environment of the Changjiang River Estuary and its adjacent waters. Combining the distribution of the sedimentary grain size composition in the Pejrup M triangle with the regional circulation mode, the sedimentary environment of the Changjiang River Estuary, Hangzhou Bay, and the adjacent waters has been divided into eight grain size divisions (Fig. 1), and terrestrial materials in each division have been discussed in the context of the platinum element ratio and the rare earth element differential method (Zhang et al., 2013). In this study, which is based on previous studies, OC distribution in different sedimentary environments will be discussed, through further analysis of grain size, OC content, and the isotope composition of surface sediments in the study area. This study, which will provide basic data on water mass transport and exchange in the Changjiang River, the Huanghe River (Yellow River), or the East China Sea, will enhance our understanding of the sources, transport media, and depositional conditions of organic carbon, and how they influence the regional carbon cycle in coastal-shelf deposition systems; this information will also improve our understanding of the marine sedimentary OC cycle.

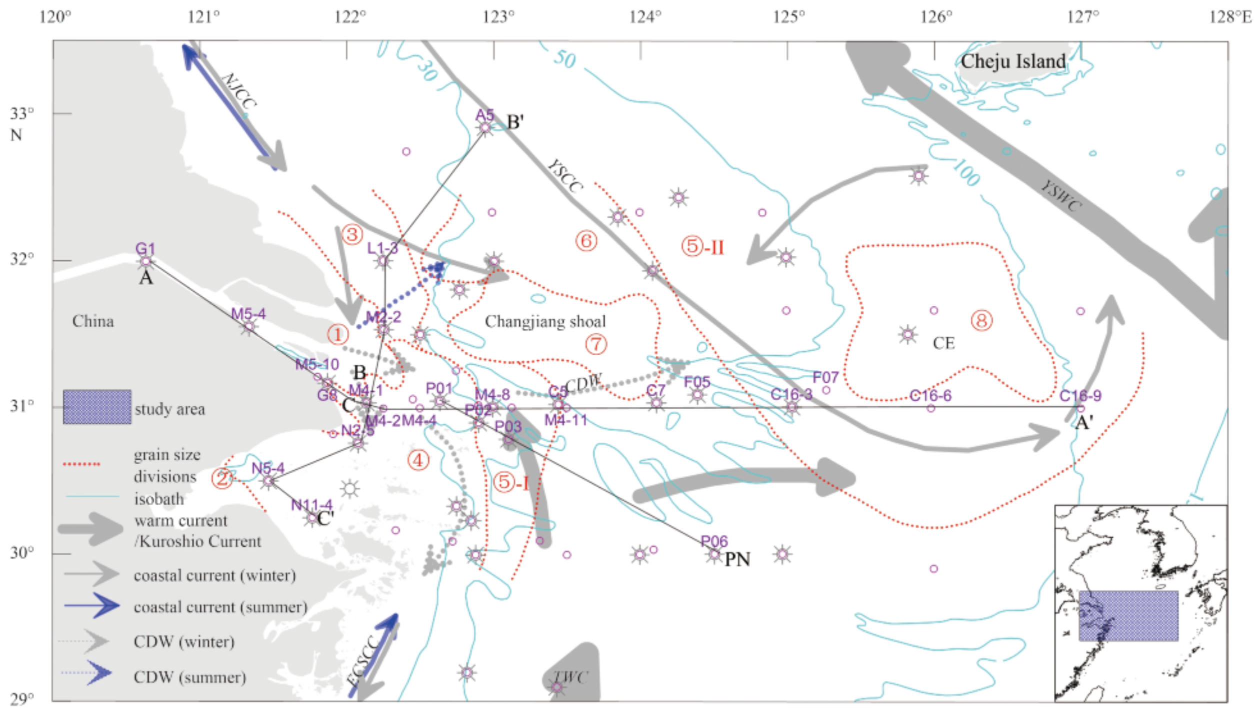

Fig. 1. Sampling stations of surface sediments in the study area and the regional circulation mode (winter and summer). Station information: ○ samples of bulk sediment analysis and ☼ samples of grain size fractionation. Circulation mode (Liu et al., 2003; Liu et al., 2010; Chen et al., 2000; Su, 2001; Yuan et al., 2008): CDW represents Changjiang River Diluted Water; NJCC North Jiangsu Coastal Current, ECSCC East China Sea Coastal Current, YSCC Yellow Sea Coastal Current, TWC Taiwan Warm Current, YSWC Yellow Sea Warm Current, KC Kuroshio Current, and CE Cyclonic Eddy. Grain size divisions in sedimentary environment (Zhang et al., 2013): ① Changjiang River upper estuary modern sediments, ② Qiantang River mouth modern sediments, ③ northeast of Changjiang River Estuary offshore modern sediments, ④ Changjiang River lower estuary-Hangzhou Bay offshore modern sediments, ⑤-I mixed sediments of the offshore modern sediments and continental shelf relict sediments, ⑤-II mixed sediments of modern sediments with relict sediments in continental shelf, ⑥ continental shelf relict sediments, ⑦ Changjiang shoal relict sediments, and ⑧ Cyclonic Eddy modern sediments to the Southwest of the Cheju Island.

2 Sample collection and analysis

The sampling stations were selected according to the topography and the hydrodynamic conditions in the study area. There were more sampling stations in the coastal zone and on the inner continental shelf where the terrain was more complex, and coastal water masses interacted strongly with continental shelf fronts. However, there were fewer sampling stations in the flatter terrain of the outer continental shelf. To understand the transport characteristics of OM derived from the Changjiang River, four sections were investigated: one cross-section in Hangzhou Bay, and the other three sections from the Changjiang River Estuary to the northeast, east, or southeast (PN) seaward, respectively. A total of 61 surface sediment samples were collected in 2006–2011, and were kept frozen (–20°C) until analysis in 2011 (Fig. 1).

Grain size was measured by a laser particle size analyzer in the Second Institute of Oceanography of the State Oceanic Administration, using a particle size interval of 0.5 φ. The analysis method was based on those specified by Specification for Oceanographic Survey, Part 8: Marine Geology and Geophysical Survey GB/T 12763.8–2007 (General Administration of Quality Supervision, Inspection and Quarantine of PRC and Standard Administration of PRC, 2007).

To understand the effect of coastal-shelf water masses on the transport of material in the Changjiang River, a total of 36 samples were selected in the interaction zone of the coastal-shelf water masses in the Changjiang River Estuary and Hangzhou Bay. Before analysis, the 36 samples were thawed, homogenized, and separated into the >0.250 mm, 0.125–0.250 mm, 0.063–0.125 mm, and 0.025–0.063 mm fractions using stainless steel sieves with deionized water (Zhang et al., 2009), and further into the 0.004–0.025 mm and <0.004 mm fractions by the centrifugation method (Poppe et al., 1988; Barbanti and Bothner, 1993; Ogrinc et al., 2005).

The bulk sediment and sediments from each grain size category were prepared for determination of OC and stable carbon isotope composition (δ^{13}C) (Fig. 1). The samples were acidified with aqueous 1 mol/L HCl to remove inorganic carbon. The samples were freeze dried after acidification, and converted to gas by combustion in an element analyzer (Thermo NE1112) that was connected to a mass spectrometer (Thermo Finnigan Delta plus AD) by a ConFlo interface to measure the amount of OC and δ^{13}C. The temperatures of the oxidizing and reducing reactors were 1 020°C and 650°C, respectively. The temperature of the filling column was 40°C. The reference gases were pure CO_2 from cylinders calibrated against Peedee Belemnite standard (PDB) carbonate. Reference materials, USGS-24 and GBW4408, were used to calibrate the pure CO_2 from the laboratory tank. The analytical precisions of the δ^{13}C and the OC measurements were about 0.2‰ and 0.1% respectively. Samples were analyzed by the Second Institute of Oceanography of the State Oceanic Administration.

3 Results

3.1 *Grain size analysis of the surface sediments*

The grain size distribution in the study area determined by laser particle analyzer showed that the content of sand, silt, and clay from 61 bulk surface sediments were, on average, 40.04%, 40.81%, and 19.15%, respectively, while the content of sand, silt, and clay from the bulk sediments of 36 typical fractionated samples were 43.26%, 38.78%, and 17.96%, respectively (Fig. 2). There was little difference between the two sets of samples, which suggests that the set of 36 samples was representative of the sediment grain size distribution in the study area. Using the sieve and centrifugal extraction methods, the content of the <0.004 mm, 0.004–0.025 mm, 0.025–0.063 mm, 0.063–0.125 mm, 0.125–0.250 mm, and >0.250 mm fractions was 16.07%, 27.09%, 5.73%, 9.52%, 30.96%, and 10.62%, respectively.

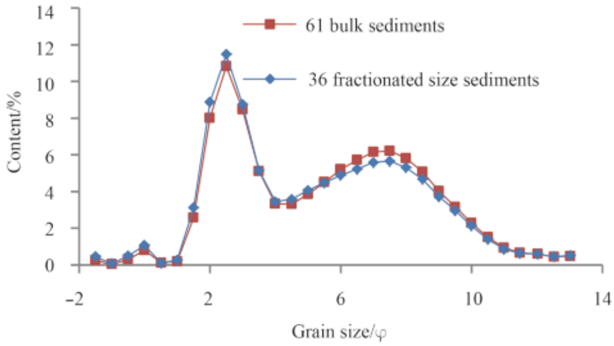

Fig. 2. Average content of each grain size interval for bulk surface sediments and typical fractionated size sediments in the study area.

3.2 *Organic carbon and stable isotopes* (δ^{13}C) *in the bulk samples*

The OC content of the sediment is the amount of OM preserved in the sediment that has not been mineralized, or influenced by the amount of primary productivity or by degradation processes. The OC content may represent the OM source, transport route, deposition process, and preservation (Meyers, 2003). The percentage of OC by weight (wt% OC) in the bulk surface sediments in the study area varied from 0.08% to 1.74%, with a mean of 0.45%. The stable carbon isotope composition (δ^{13}C) varied from –19.60‰ to –24.55‰, with a mean of –21.39‰. The high OC value was found in the offshore sediments in the study area (Fig. 3a). There are three OC-rich zones: (1) outside of the northern branch of the Changjiang River Estuary, (2) where the southern branch enters Hangzhou Bay, and (3) outside of the Zhoushan Islands' waters, which are affected by the northward branch of the Taiwan Warm Current and form a northeast/east branch, and have a trend of spreading to the Cyclonic Eddy area to the southwest of Cheju Island. In addition to the above three offshore OC-rich regions, the OC content in the Cyclonic Eddy modern sediments to the southwest of the Cheju Island is much higher than that in sediments in offshore OC-rich zones. The OC content of the eight grain size divisions were divided into three types by statistical analysis (Table 1). The first type (high) had an OC content >0.4%; this division included offshore modern sediments distributed in the northeast of the Changjiang River Estuary and the lower estuary of the Changjiang River and Hangzhou Bay (Subsets ③ and ④), and a mixture of modern sediments and continental shelf relict sediments (Subset ⑤-II) and the Cyclonic Eddy modern sediment to the southwest of the Cheju Island (Subset ⑧). The second type (medium) had an OC content of 0.3%–0.4%, and included modern sediments from the upper estuary of the Changjiang River, and a mixture of offshore modern sediments and continental shelf relict sediments (Subsets ⑤-I and ①). The third type (low) had an OC content of <0.3%, and included modern sediments from the mouth of the Qiantang River (Subset ②), and relict sediments on the continental shelf and in the Changjiang shoal (Subsets ⑥ and ⑦).

The OC origin contributes to the OC distribution in surface sediments. δ^{13}C has been used frequently to identify the material source (Gao et al., 2007; Jin et al., 2010; Cai et al., 2014). The δ^{13}C value of typical terrestrial OM is from –26‰ to –28‰, while the δ^{13}C of marine OM is relative heavy, and varies from –19‰ to –22‰. The δ^{13}C in the study area was lighter in the estuary than in the outer seas (Fig. 3b). This difference can be used to distinguish the transport characteristics between OC of terrestrial and marine origin. The δ^{13}C contour in the Changjiang River Estuary was divided into two branches, north and south, in the estuary. The north branch, extending northeastward, encountered the North Jiangsu Coastal Current, turned to the northeast at 32°N, and formed a tongue diffusing to Cheju Island; the transport trend of this branch was across the Yellow Sea Coastal Current into the Cyclonic Eddy. The branch that extended southward partly spread to the Hangzhou Bay, and other part flowed around the Zhoushan Islands to the south following the East China Sea Coastal Current; on the way it turned to the northeast, driven by the northward branch of Taiwan Warm Current.

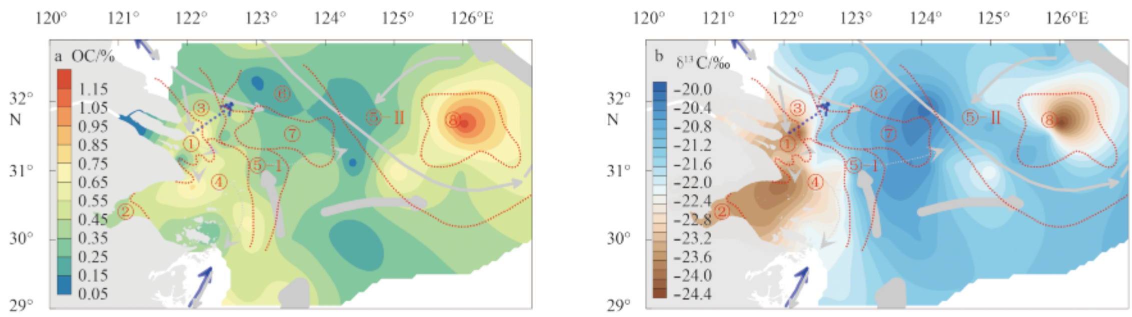

Fig. 3. Distribution of OC content and δ^{13}C of the surface sediment in the study area.

Table 1. Grain size divisions in sedimentary environment and OC content and its δ^{13}C in the study area

Grain size divisions in sedimentary environment	Hydrodynamic factors	OC/%	δ^{13}C/‰	Contribution of terrestrial OC/%
① Changjiang River upper estuary modern sediments	Changjiang runoff and reversing tidal current	0.37	–22.91	45
② Qiantang River mouth modern sediments	Qiantang runoff and reversing tidal current	0.25		
③ Northeast of Changjiang River Estuary offshore modern sediments	CDW and NJCC	0.45	–23.17	48
④ Changjiang River lower estuary-Hangzhou Bay offshore modern sediments	Reversing tidal current, Qiantang runoff, CDW	0.55	–22.46	40
⑤-I Mixed sediments of the offshore modern sediments and continental shelf relict sediments	CDW, TWC, NJCC	0.34	–21.28	23
⑤-II Mixed sediments of modern sediments with relict sediments in the continental shelf	YSCC, TWC, YSWC, KC, CDW	0.46	–21.17	22
⑥ Continental shelf relict sediments	NJCC, YSCC, TWC, CDW	0.26	–20.68	16
⑦ Changjiang shoal relict sediments	NJCC, CDW	0.21	–20.35	11
⑧ Cyclonic Eddy modern sediments to the southwest of the Cheju Island	YSCC, YSWC, KC, TWC	1.03	–21.81	30

3.3 *Organic carbon and stable isotopes (δ^{13}C) in each grain size category*

Coarse and fine grained particles have different transport paths; coarse-grained particles are transported in the bottom current, while fine-grained particles are transported as suspended load by upper water (Eisma, 1993). To understand OC lateral transport, the OC and the isotopic composition of each grain size category were analyzed (Fig. 4, Table 2). The OC content of the <0.004 mm, and 0.004–0.025 mm, 0.025–0.063 mm, 0.063–0.125 mm, 0.125–0.250 mm, and >0.250 mm fractions was 1.18%, 0.51%, 0.46%, 0.42%, 0.99%, and 0.48%, respectively. The δ^{13}C of each grain size category was –21.64‰, –22.03‰, –22.52‰, –22.46‰, –22.36‰, and –22.28‰, respectively. The OC content and δ^{13}C showed a U-shaped distribution as particle size became coarser or finer, indicating that the OC content was higher in coarse or fine particles than in the medium-sized categories. The OC content in the 0.063–0.125 mm fraction was the lowest (0.42%). This is consistent with results from other studies of similarly fractionated continental shelf, slope, pond, and lake samples (Keli et al., 1998; Cai et al., 2007). The δ^{13}C was heavier in the coarse or fine fractions than the medium-sized ones. The δ^{13}C value was the lightest (–22.52‰) in the 0.025–0.063 mm fraction, and was the heaviest (–21.64‰) in the <0.004 mm clay.

As can be seen, the OC of each grain size category formed different depocenters. OC in the >0.125 mm coarse-grained fraction was mainly enriched in offshore interactions between the Changjiang River Diluted Water, the Taiwan Warm Current, and the North Jiangsu Coastal Current (Figs 4m and p). OC in the 0.025–0.125 mm grain size category was mainly concentrated in river-coast areas, and in the Cyclonic Eddy area to the southwest of the Cheju Island, and its adjacent areas to the east of the Yellow Sea Coastal Current (Figs 4g and j). OC in the <0.025 mm fine grain category was mainly enriched in relict sediments on the continental shelf (Figs 4a and d).

The OC contribution of each grain size category can be calculated as the product of the OC content and the weight of each grain size category. Going from fine to coarse, the OC contribution in the <0.004 mm, 0.004–0.025 mm, 0.025–0.063 mm, 0.063–0.125 mm, 0.125–0.250 mm, and >0.250 mm fractions was 42.96%, 26.06%, 9.82%, 5.75%, 7.09%, and 8.33%, respectively, and showed a U-shaped distribution. The contribution in the 0.063–0.125 mm fraction was the smallest (5.75%), followed by the 0.125–0.250 mm fraction (7.09%). Approximately 69.02% of the OC was stored in approximately 43.16% of the <0.025 mm clay and fine silt, while 42.96% of the OC that had a heavy δ^{13}C value (–22‰) was stored in approximately 16.07% of the <0.004 mm clay. Figure 4 showed that there was a large OC contribution in (1) the <0.004 mm clay fractions in hypoxic areas of the

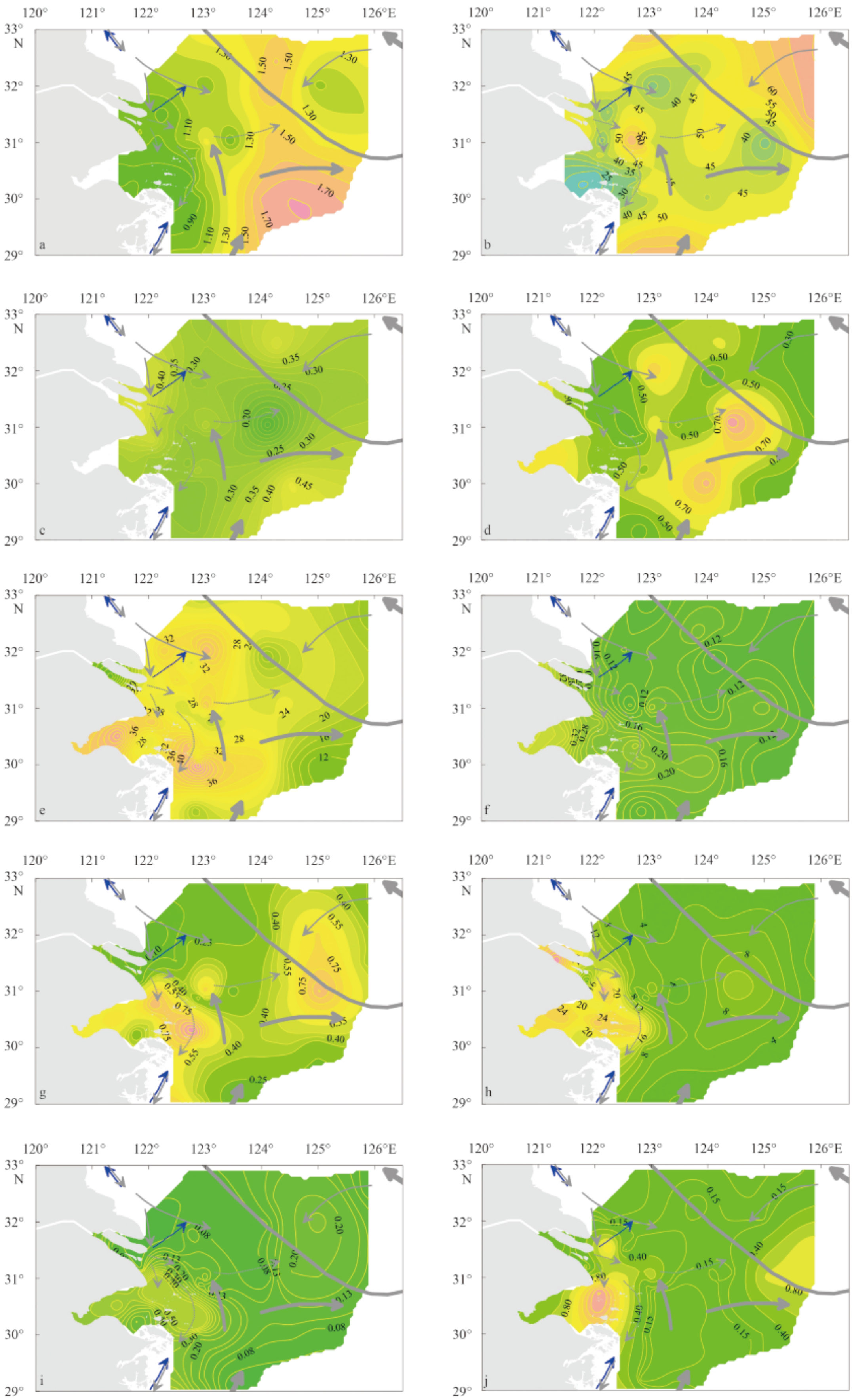

Fig. 4.

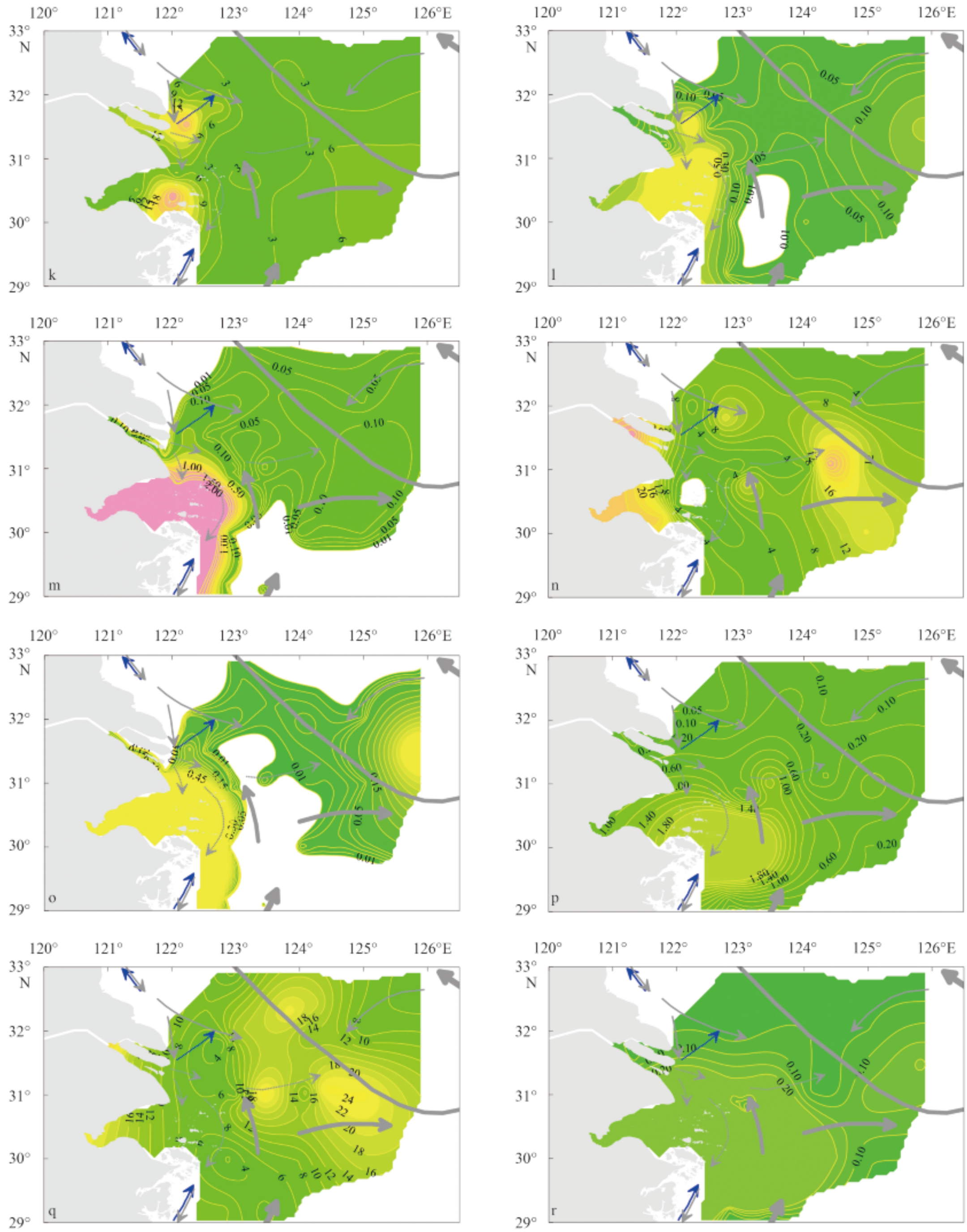

Fig. 4. Distribution of OC content, terrestrial OC and OC contribution in each size category. a. OC% for the grain size <0.004 mm fraction, b. OC contribution (%) for the grain size <0.004 mm fraction, c. terrestrial OC% for the grain size <0.004 mm fraction, d. OC% for the grain size 0.004–0.025 mm fraction, e. OC contribution (%) for the grain size 0.004–0.025 mm fraction, f. terrestrial OC% for the grain size 0.004–0.025 mm fraction, g. OC% for the grain size 0.025–0.063 mm fraction, h. OC contribution (%) for the grain size 0.025–0.063 mm fraction, i. terrestrial OC% for the grain size 0.025–0.063 mm fraction, j. OC% for the grain size 0.063–0.125 mm fraction, k. OC contribution (%) for the grain size 0.063–0.125 mm fraction, l. terrestrial OC% for the grain size 0.063-0.125 mm fraction, m. OC% for the grain size 0.125–0.250 mm fraction, n. OC contribution (%) for the grain size 0.125–0.250 mm fraction, o. terrestrial OC% for the grain size 0.125–0.250 mm fraction, p. OC% for the grain size >0.250 mm fraction, q. OC contribution (%) for the grain size >0.250 mm fraction, and r. terrestrial OC% for the grain size >0.250 mm fraction.

Changjiang River Estuary and in continental shelf relict sediments to the east of Yellow Sea Coastal Current (Fig. 4b), (2) 0.004–0.025 mm fine silt in the waters to the west of the Yellow Sea Coastal Current (Fig. 4e), and (3) in 0.025–0.063 mm coarse silt and 0.063–0.125 mm fine sand in the Changjiang River Estuary and Hangzhou Bay (Figs 4h and k). It is worth noting that there was a considerable OC contribution in the >0.125 mm sand components in continental shelf relict sediments (Fig. 4q).

Table 2. OC content, terrestrial OC and OC contribution in each size category

Grain size	<0.004 mm	0.004–0.025 mm	0.025–0.063 mm	0.063–0.125 mm	0.125–0.250 mm	>0.250 mm
OC/%	1.18	0.51	0.46	0.42	0.99	0.48
δ^{13}C/‰	–21.64	–22.03	–22.52	–22.46	–22.36	–22.28
Terrestrial OC/%	0.32	0.17	0.20	0.22	0.72	0.21
OC contribution/%	42.96	26.06	9.82	5.75	7.09	8.33
Sample numbers	33	35	36	35	32	23

4 Discussion

4.1 *Source of sedimentary organic carbon*

δ^{13}C has been widely used, not only to trace the migration of terrestrial OM in the sea, but also to estimate the quantity of various OC sources. Based on the isotopic mass balance equations of a two end-member mixing model for marine or terrestrial sources, the following is an empirical formula that can be used to estimate the contribution of terrestrial OM (*f*) (Minoura et al., 1997):

$$f = \frac{\delta^{13}C_{\text{marine}} - \delta^{13}C_{\text{sediment}}}{\delta^{13}C_{\text{marine}} - \delta^{13}C_{\text{terrestrial}}}.$$

In general, the δ^{13}C value of the terrestrial OC end-member is –26‰, and that of the marine source end-member is –20‰. The focus of this paper is the Changjiang River Estuary. The typical δ^{13}C value in sediments and suspended matter in the Changjiang River waters was around –26‰ for plants, –27.5‰ for forest soil, and –28‰ for paddy soil (Wu et al., 2007). The typical δ^{13}C value of terrestrial OM in the Changjiang River waters was –27.1‰ (Wu et al., 2002), and that of the OC end-member components of marine phytoplankton in the Changjiang River Estuary waters was –19.5‰ (Cai et al., 1992). In this study, the δ^{13}C values of the terrestrial and marine end-members in the Changjiang River Estuary were –27.1‰ and –19.5‰. The data measured in this study can be substituted into the above formula with the δ^{13}C values of two end-members, from which the relative contribution of terrestrial OM in sedimentary OM in the study area can probably be estimated.

The terrestrial OC content of the eight grain-size divisions in the sedimentary environment in the study area was divided into three types (Table 1). Sediments with a high contribution of terrestrial OC (>30%) were mainly in the coastal estuarine-bay area (Subsets ①, ③, and ④) and in the Cyclonic Eddy area to the southwest of Cheju Island (Subset ⑧). Sediments with a medium contribution of terrestrial OC (20%–30%) were in the mixed sediments in the modern sediments and relict sediments on the continental shelf (Subsets ⑤-I and ⑤-II). Sediments with a low contribution of terrestrial OC (<20%) were mainly in the relict sediments on the continental shelf and in the Changjiang shoal (Subsets ⑥ and ⑦). Previous studies have shown that using a two end-member mixing model will underestimate the terrestrial OC content in marine sediments, especially where there are fewer terrestrial, soil OM source inputs (Goñi et al., 2005; Gordon and Goñi, 2003). A large proportion of the OM inputs in the East China Sea were terrestrial; however, other studies in this area have shown that the branched-GDGT of soil OM had degraded by approximately 95% in the estuary (Zhu et al., 2011). The terrestrial OM contribution calculated by the two end-member model in the study area may therefore be underestimated in this paper.

At present, the terrestrial OC provenance cannot be identified by the δ^{13}C, even though it is a key to understanding the carbon cycle. In this paper, the terrestrial OC provenance will be discussed by integrating the OC and δ^{13}C data with other previous data. Cluster analysis of rare earth element and platinum family element data divided the terrestrial matter in the study area into three types as follows (Zhang et al., 2013) (Fig. 5): Type I mainly derived from the Changjiang River (Subsets ①, ③, ④, and ⑧), Type II mainly derived from the Huanghe River (Subsets ⑥ and ⑦), and Type III mainly derived from a mixture of Changjiang River and Huanghe River sediments (Subsets ⑤-I and ⑤-II). It was thought that δ^{13}C was depleted in surface sediments near the old Huanghe River Estuary in North Jiangsu, owing to the shallow water depth and strong hydrodynamic effects that caused sediment re-suspension, and resulted in high sediment concentrations, with one part spreading seaward on a concentration gradient, and the other part moving southward along the coast to the Changjiang River Estuary (Subset ③) (Cai et al., 2003). Large amounts of terrestrial plant debris in the 0.125–0.250 mm size category near the Zhoushan Islands (Subset ④) were identified when examined under the microscope (Zhang et al., 2009); the isotopic compositions of these sediments (N4-8 and N11-4) were –25.53% and –25.91%, respectively, and therefore were basically close to the plant end-member value of –26‰. Therefore, the source of the terrestrial OM in the Changjiang River Estuary (①, ③, ④) was mainly the Changjiang River, but also the old Huanghe River Estuary. Of course, some terrestrial OM from nearby islands was also found in this area.

The source of terrestrial matter in the Cyclonic Eddy area to the southwest of Cheju Island is contentious. Some researchers argue that, in winter, the Yellow Sea Coastal Current transports sediment from the Old Huanghe Delta to the East China Sea shelf, and forms a mud patch to the southwest of Cheju Island (Milliman et al., 1985; Yuan et al., 2008). The results of a comparative analysis of element ratios (Sc/Al, Zr/Ti, Cr/Th, Nb/Co) and Sr isotopic ratios (^{87}Sr/^{86}Sr) suggest that the southwest mud of Cheju Island (Subset ⑧) originated from the Changjiang River (Youn and Kim, 2011). Results of the regional ocean circulation model show that fine-grained sediment from the Changjiang River in winter and spring was partly carried to the southwest of Cheju Island (Subset ⑧) by the Yellow Sea Coastal Current and deposited there (Bain et al., 2013). As a result, some terrestrial OM derived from the Changjiang River Estuary was deposited in the Cyclonic Eddy area to the southwest of the Cheju Island. As shown in Fig. 3b, this shows that the terrestrial OM in the Changjiang River Estuary area was not only transported across the North Jiangsu Coastal Current, but also over the Yellow Sea Coastal Current, and transported into the CE area, influenced by the counter clockwise movement in the Cyclonic Eddy area, located to the southwest of the Cheju Island.

4.2 *Sedimentary organic carbon lateral transport*

Stable and radioactive isotope analyses of different grain size fractions in surface sediments in the estuary or bay shows that

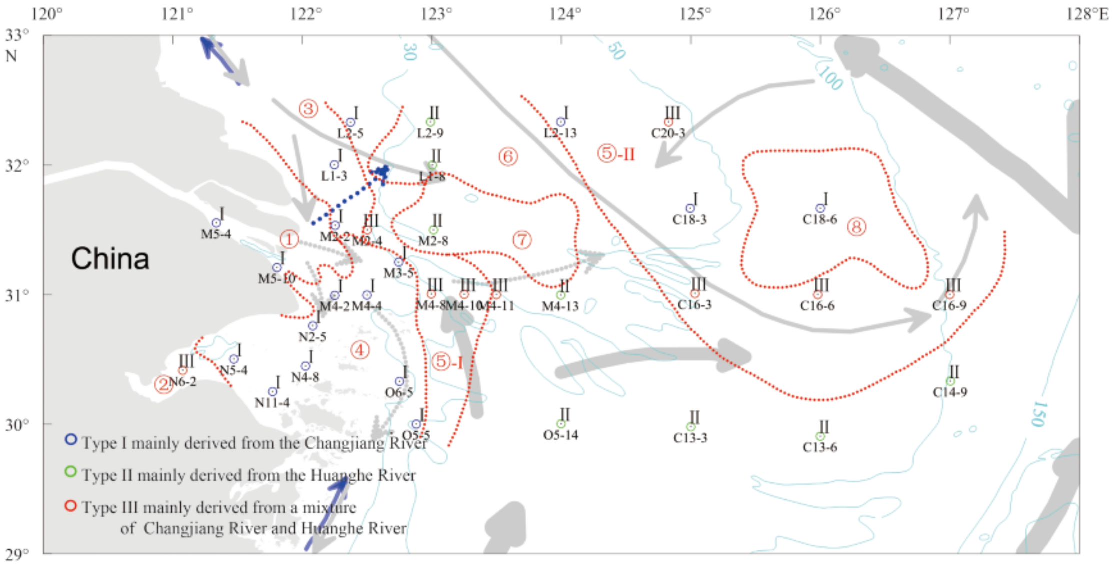

Fig. 5. Distribution on terrestrial provenance in the investigated area (Data from Zhang et al., 2013).

the OC content and the isotopic composition of each particle size category varies widely. Fine or coarse particles have markedly different material sources (Keli et al., 1998; Megens et al., 2002). The product of the terrestrial OC contribution and the OC content is the terrestrial OC content, which can be used to analyze the ratio of terrestrial OM in marine sediment. The terrestrial OC contents of the <0.004 mm, 0.004–0.025 mm, 0.025–0.063 mm, 0.063–0.125 mm, 0.125–0.250 mm, and >0.250 mm fractions were 0.32%, 0.17%, 0.20%, 0.22%, 0.72%, and 0.21%, respectively. The value was highest (0.72%) in the 0.125–0.250 mm fraction of samples from stations N4-8 and N11-4 near the Zhoushan Islands, and was the result of large amounts of terrestrial plant debris from the islands (Zhang et al., 2009). If the effect of large amounts of plant detritus from these two stations is ignored, the terrestrial OC content in the 0.125–0.250 mm fraction was 0.14%; the highest terrestrial OC content was in the <0.004 mm clay fraction (0.32%). Figure 4 shows the terrestrial OC content in each size category, which indicates the seaward transport path of terrestrial OC. The terrestrial OC from the Changjiang River and offshore islands into the sea formed the northeastward branch and spread to the CE area to the southwest of the Cheju Island at 30°N, driven by the northern branch of the Taiwan Warm Current outside the Zhoushan Islands' waters (Figs 4f, i and r). This result is different from those of previous studies of sediment transport induced by the Taiwan Warm Current that mainly focused on coastal areas where seaward sediment transport could have been blocked by the onshore bottom Ekman flows. The result is similar to those from the regional ocean circulation model that were based on observations from two cruises in July 2006 and January 2007 in the East China Sea; this model demonstrated that the Changjiang River sediment was mainly transported to the mud patch off the coast of Zhejiang and Fujian Provinces in winter and spring, after which the Taiwan Warm Current transported this sediment to the outer continental shelf of the East China Sea, rather than, as is the traditional view, blocking the seaward sediment transport (Bian et al., 2010). Of course, Figs 4h, k and n show that the OC contribution of the coarse-size fractions was mainly focused on the coastal areas. This means that only a small amount of terrestrial OC was transported across the Taiwan Warm Current.

Figures 4c, l and o show that terrestrial OC in the Changjiang River Estuary area moved with the northward branch seaward and turned to the northeast, driven by the North Jiangsu Coastal Current; it was obstructed by the Yellow Sea Coastal Current on the way north, and then turned to the southeast into the Cyclonic Eddy area. This is consistent with studies that showed that, in summer, the CDW moved to the southeast and then turned to the northeast direction of Cheju Island at 122°10′–122°30′E (Hu et al., 1995). This is similar to the latest result, which showed that the CDW moved northward and formed the northeast water tongue in summer, while one of the branches turned to the southeast and moved around the CE area during investigations in 2006–2007 (Wang et al., 2012; Bain et al., 2013). It also agrees with the results of a cluster analysis of rare earth elements and platinum element recognition, where the transport path of terrestrial material from the Changjiang River seaward was shown to be M2-2→L1-3→L2-5→L2-13→C18-3→C18-6 (Fig. 5).

The OC of each grain-size category showed that the OC contribution of the <0.025 mm fraction accounted for up to 69.02%. Cluster analysis of the OC and δ^{13}C and 1/2 φ interval particle size of 61 bulk sediments in the study area also confirmed that OC was closely correlated to the >6 φ (<0.022 mm) fine silt and clay fractions (Fig. 6). To determine the influence of hydrodynamic conditions on the contributions of the OC source and transport to OC distribution, the OC content and the δ^{13}C value and the proportions of the <0.022 mm fractions in surface sediment were determined for the different directions (Fig. 7): in addition to the classic section PN, northeast, east, and southeastern cross-sections were set along the terrestrial OC transport path from the Changjiang River Estuary to the continental shelf, and profile locations are shown in Fig. 1.

Section A-A′ begins to the west of the Changjiang River Estuary near Xuliujing, east of the shelf at a depth of 100 m. Overall, the content of the <0.022 mm fraction changed synchronously with the OC content, but, at Stas M4-4, P01 and M4-8 in the lower estuary of the Changjiang River, the content of fine grains increased and the OC content decreased. Previous research has indicated that strong tidal forces could result in re-suspension,

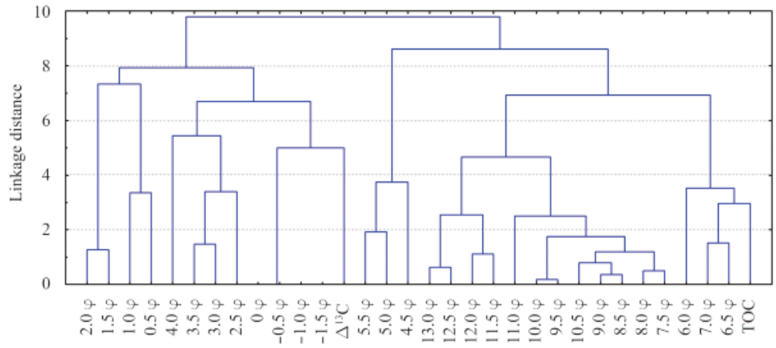

Fig. 6. Cluster analysis tree diagram for grain size content, OC content and δ^{13}C.

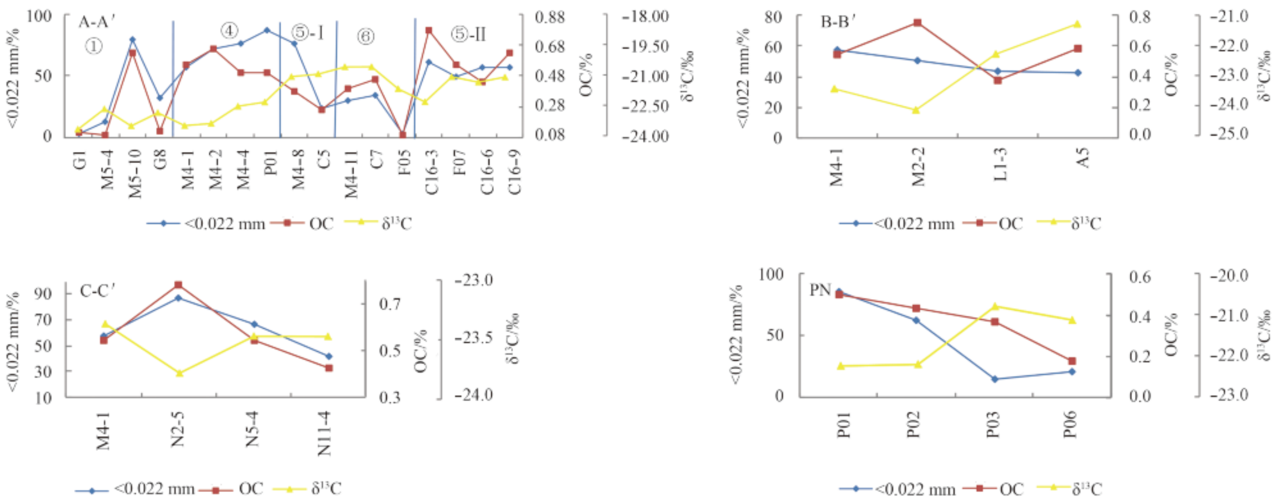

Fig. 7. Plot of the proportions of the grain size <0.022 mm fractions, OC content and δ^{13}C in surface sediments along the estuary to the continental shelf.

while winnowing, waves, and tides could transport terrestrial OM away from the fine particles, thereby preventing the accumulation of OM by the fine particles, resulting in degradation and loss of OM (Zhu et al., 2011; Keil et al., 1997). Research on modern marine sediments on the Washington coast has confirmed the hypothesis that a small portion of the OM can be clearly categorized as organic debris isolated in specific size classes (Keli et al., 1998). OC was reducing at three stations located in the shallow water (<50 m deep) on the inner continental shelf, which agrees with the distribution of a "cold pool" center (31°N, 123°E) outside of the Changjiang River Estuary (Liu, 2011). This may be affected by strong coastal storms that caused the OC content to decline in the lower estuary of the Changjiang River. In addition, the δ^{13}C values gradually increased from west to east, and reached a maximum value in continental shelf relict sediments. On both sides of the Yellow Sea Coastal Current, the δ^{13}C values (F05 and C16-3) were lighter, and were perhaps influenced by terrestrial material inputs. Moreover, the OC content at C16-3 was higher because of terrestrial material inputs from the Changjiang River Estuary driven by the east branch of the Taiwan Warm Current.

The OC content of the <0.022 mm fraction at Stas M2-2 and A5 in Profile B-B' in the northeast of the Changjiang River Estuary appeared abnormal. The OC content increased, while δ^{13}C became lighter at Sta. M2-2, indicating that there were inputs of terrestrial OC. Station M2-2 was influenced by terrestrial inputs from the southeast branch of the North Jiangsu Coastal Current (Zou et al., 1999; Cai et al., 2003), resulting in an increased contribution of terrestrial OC. The increasing OC and the δ^{13}C weighting at Sta. A5 indicate that the marine OC was overlain, because of OM inputs in this area, located in the southward path of the Yellow Sea Coastal Current.

In Profile C-C' from the Changjiang River lower estuary to Hangzhou Bay along the coast, the OC content was higher in the north than in the south, and coincided with the content of the <0.022 mm fractions. The contents were different between the north and the south because the stations in the south were further from the mainland than those in the north, where the terrestrial material inputs were higher. The δ^{13}C value was lighter at Sta. N2-5 in the north, indicating a contribution from terrestrial OM.

In Section PN that extended from the Changjiang River Estuary to the southeast outer shelf, with the exception of the OC content of station P03, there was a general gradual decrease in the OC content and the proportions of <0.022 mm fraction as distance from the river estuary increased. The station was located in an area where modern sediments were mixed with continental shelf relict sediments near the Changjiang River Diluted Water and the Taiwan Warm Current. Organic matter moving seaward from the Changjiang River Estuary was obstructed by the warm and salty northward branch of the Taiwan Warm Current, causing it to flocculate and sink to the seafloor. The plankton carried by the Taiwan Warm Current facilitated OC transfer to the seafloor (Liu, 2011; Cai et al., 2003). Results show that there was a significant change in the $\delta^{13}C$ value of the surface sediment.

The integration of the information on the distribution of terrestrial OC content of each grain size category, and $\delta^{13}C$ in bulk surface sediment has indicated that the terrestrial OM in the Changjiang River Estuary region was transported seaward and distributed to the Cyclonic Eddy area to the Southwest Cheju Island via two pathways: one was a result of the Changjiang River Diluted Water (CDW) northeastward-extending branch driven by the North Jiangsu Coastal Current and Yellow Sea Coastal Current, while the other one was the result of the CDW southward-extending branch driven by the Taiwan Warm Current. During transport from the estuary to the outer seas, there was a degree of OM degradation and loss in shallow water less than <50 m deep off the inner continental shelf, where sediment met with the "cold pool" outside of the Changjiang River Estuary. However, the OC content increased, and the $\delta^{13}C$ value changed dramatically where the Changjiang River Diluted Water interacted with the coastal circulation and the Taiwan Warm Current, such as at Sta. M2-2, where the CDW northeastward-extending branch met with the southward North Jiangsu Coastal Current, at Sta. A5 (near to the northeastward junction with the Yellow Sea Coastal Current), and at Sta. P03 (from the southward interaction with the northern branch of the Taiwan Warm Current).

5 Conclusions

(1) In the study area, the percentage of OC by weight in the bulk surface sediments varied from 0.08% to 1.74%, with a mean of 0.45%. The stable carbon isotope composition ($\delta^{13}C$) varied from –19.60‰ to –24.55‰, with a mean of –21.39‰. The OC content of the <0.004 mm, 0.004–0.025 mm, 0.025–0.063 mm, 0.063–0.125 mm, 0.125–0.250 mm, and >0.250 mm fractions was 1.18%, 0.51%, 0.46%, 0.42%, 0.99%, and 0.48%, respectively, and the $\delta^{13}C$ value in each grain size category was –21.64‰, –22.03‰, –22.52‰, –22.46‰, –22.36‰, and –22.28‰. The OC contribution of each grain size was 42.96%, 26.06%, 9.82%, 5.75%, 7.09%, and 8.33%. The OC content and its contribution were higher for coarse or fine particle sizes than for the middle-size fractions. Approximately 69.02% of the OC content was stored in the <0.025 mm clay and fine silt fractions.

(2) The OC content was high in modern sediments in the estuarine/bay offshore area and the Cyclonic Eddy area to the southwest of the Cheju Island, where the high contribution of terrestrial OC (>30%) was maybe mainly from inputs from the Changjiang River Estuary. The mixture of offshore modern sediments and continental shelf relict sediments had a medium OC content; contribution of terrestrial OC (20%–30%) in these sediments mainly derived from the Changjiang River Estuary and mixed inputs from the Huanghe River. The OC content in relict sediments of the continental shelf and Changjiang shoal was low, with the low contribution of terrestrial OC (<20%) mainly derived from Huanghe River sources inputs.

(3) Information on the distribution of the OC content integrated with $\delta^{13}C$ of the bulk sediment and each grain size category indicated that the terrestrial OC moving seaward from the Changjiang River Estuary was divided into three branches. One was northeastward, and was driven southward by the North Jiangsu Coastal Current then spread to the Cheju Islands; the load was conveyed to the CE area by the Yellow Sea Coastal Current, where it was deposited. The second branch spread partly to Hangzhou Bay. The third branch flowed around the Zhoushan Islands and then southward along the East China Sea Coastal Current, with a little part of it turning to the northeast on the way; this branch also had a tendency to transport material to the CE area driven by the northern branch of the Taiwan Warm Current.

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