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Distribution and source of organic matter in surface sediment from the muddy deposit along the Zhejiang coast, East China Sea

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ABSTRACT

To constrain organic matter compositions and origins, elemental (TOC, TN, C/N) and stable carbon ($\delta^{13}\text{C}$) and nitrogen isotope ($\delta^{15}\text{N}$) compositions are measured for surface sediments collected from muddy deposit along the Zhejiang coast, East China Sea. The results showed that the TOC, TN, C/N, $\delta^{13}\text{C}$, and $\delta^{15}\text{N}$ were 0.19–0.67%, 0.03–0.09%, 6.76–9.22, -23.43 to -20.26‰ , and 3.93–5.27‰, respectively. The $\delta^{13}\text{C}$ values showed that the mixing inputs of terrigenous and marine organic matter generally dominated sedimentary organic matter in the west part, and the sedimentary organic matters were mainly influenced by the marine organic matter in the east part of the study area. A stable carbon isotope two end member mixing model estimates $\sim 38\%$ terrestrial-derived and $\sim 62\%$ marine-derived inputs to sedimentary organic matter. Microbial mineralization strongly controls $\delta^{15}\text{N}$ values, and therefore cannot be used to identify the provenance of organic matter for the Zhejiang coast.

Estuarine and coastal areas are the direct channel between the land and ocean, trapping significant quantities of terrigenous and marine organic matter that is influenced by a complex series of physical, chemical, and biological processes during transport, deposition, and burial (Kennedy, 1984; Thornton and McManus, 1994). Preservation of organic matter in estuarine and coastal sediment is an important process in the global carbon cycle as more than 90% of the global marine organic carbon buried in continental margin sediment (Emerson and Hedges, 1988; de Haas et al., 2002).

In the coastal region, organic matter can be supplied by the marine organisms and terrestrial sources (Tesi et al., 2007). Identifying the provenance of organic matter in sediment is essential to understand the mechanisms of carbon cycling in the marine environment (Usui et al., 2006). Stable carbon and nitrogen isotope ratios and the ratio of total organic carbon to total nitrogen have been widely used to elucidate the source and fate of organic matter in the marine environment (Thornton and McManus, 1994; Naidu et al., 2000; Cloern et al., 2002; Wu et al., 2007; Zhang et al., 2007; Ramaswamy et al., 2008; Gao et al., 2012).

Previous reports have been published on carbon and nitrogen stable isotopes in sediment from the Yangtze estuary and adjacent area (Shi, 1993; Wu et al., 2000, 2002, 2003; Liu et al., 2006a; Zhou et al., 2006). However, the sampling stations of the surface sediment from the mud

deposit along the Zhejiang coast were very sparse. Therefore, the geochemical characteristics and provenance of surface sediments deposited farther south of the Yangtze River along the Zhejiang coast are not as well constrained. This study aims to quantify carbon and nitrogen isotopic compositions from muddy surface sediments along the Zhejiang coast to understand the spatial distribution of organic carbon and nitrogen stored in marine sediment on eastern China shelf. Geochemical signatures presented in this study will further constrain sediment provenance and be used to assess the relative contribution of terrigenous and marine sources to sedimentary organic matter found on the Zhejiang coast.

The study area is located in Zhejiang coast of the East China Sea, immediately south of the Yangtze River estuary (Fig. 1a). Yangtze River-derived sediments are transported onto the shelf predominantly by across-shelf dispersion and along-shelf advection (Liu et al., 2006b). During the summer, the Yangtze River discharges large amounts of water and sediment, with $\sim 32\%$ of the riverine sediment load estimated to be deposited at the river mouth sites during this time (Shen, 2001; Xu et al., 2015a). During the winter, intensified East Asian Monsoon activity and wave action re-suspend fine-grained river-derived sediments (Liu et al., 2006b, 2007a; Zheng et al., 2010; Xu et al., 2012). Initiation of the winter monsoon drives the formation of

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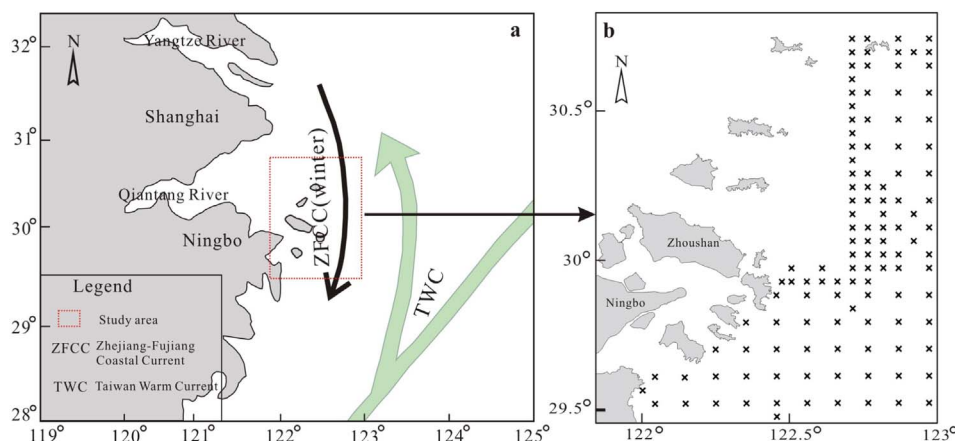


Fig. 1. Location of study area and related currents in the East China Sea (a), and sampling stations of surface sediment in the nearshore area of the Zhejiang coast (b).

upwelling circulation offshore associated with the Taiwan Warm Current (TWC), and downwelling in the near-shore region associated with the Zhejiang–Fujian Coast Current (ZFCC). The circulation of these currents generally dictates the distribution of fine-grained sediments within the inner shelf and form what is referred to as the “inner shelf mud area of the East China Sea” (Gu et al., 1997; Hu and Yang, 2001; Liu et al., 2006b, 2007a). This localized circulation prevents transportation of finer sediments to deeper waters in the East China Sea (Xu et al., 2013), which is consistent with observations indicating a lack of fine-grained muddy sediments in the middle and outer shelves which are covered by the relict sand (Liu et al., 2007a; Xu et al., 2009).

In the summer of 2012, 120 surface sediment samples were collected by box core near the coastal area of Zhejiang Province (Fig. 1b). Samples for geochemical analysis were taken from the upper 2 cm of each box core. All samples for organic matter analysis were frozen and stored in order to preserve organic signatures.

Freeze-dried and homogenized samples were analyzed for total organic carbon (TOC) and total nitrogen (TN) concentrations, as well as $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ compositions. To prepare each sample, 500 mg sediment was decalcified with 4 N HCl at room temperature for 12 h and then rinsed with deionized water four times. Decarbonated samples were then freeze-dried and transferred into tin capsules. Analysis of TOC and TN was conducted with a EuroEA3000 Elemental Analyzer with a standard deviation of $\pm 0.02\%$ dry weight ($n = 6$) for TOC, and $\pm 0.002\%$ dry weight ($n = 6$) for TN. Carbon and nitrogen isotopic compositions were determined separately, in duplicate, on decarbonated sediments by elemental analysis-isotope ratio mass spectrometry (EA-IRMS) using a Thermo Scientific Flash EA® 1112 Elemental Analyzer interfaced to a MAT253® Stable Gas Isotope Ratio Mass Spectrometer equipped with a ConFlo IV universal continuous flow interface. Standard deviation for isotopic ratios were $\pm 0.1\%$ ($n = 6$) for $\delta^{13}\text{C}$ and $\pm 0.3\%$ for $\delta^{15}\text{N}$ values ($n = 6$).

The concentrations in surface sediments from the study area range from 0.19% to 0.67% (mean $0.44 \pm 0.10\%$) for TOC, and 0.03% to 0.09% (mean $0.06 \pm 0.01\%$) for TN, respectively. Spatial variations in TOC and TN across the Zhejiang coast are similar and are shown in Fig. 2. Higher concentrations in both TOC and TN are located off the coast where clay-sized sediments are more prevalent (Xu et al., 2015b, 2016).

C/N values range from 6.76 to 9.22 (mean 7.71 ± 0.41). Higher values are documented in the southwest of the study area and lower values are found in the east (Fig. 3). The C/N ratio is an effective tool for tracing the predominant sources of organic matter in aquatic ecosystems (Matson and Brinson, 1990; Thornton and McManus, 1994; Andrews et al., 1998; Graham et al., 2001). Previous reports have shown that C/N ratios range from 15 to 20 for the terrestrial materials from Yangtze River and in the range of 5 to 10 for the adjacent coastal waters (Wu et al., 2003). The C/N values presented here ($\text{C/N} \approx 7.7$)

probably suggest that marine organic matter is the main source of organic matter to marine surface sediments along the Zhejiang coast. Given that the C/N ratios reflect the provenance of organic matter, it is expected that there exists a significant negative correlation between $\delta^{13}\text{C}$ and C/N ratio (Wu et al., 2003). However, it was shown that there are not contrary spatial distributions between $\delta^{13}\text{C}$ and C/N ratio (Figs. 3 and 4a). Therefore, the spatial distribution of C/N ratio in the study area should be caused by the decomposition process of organic matter (Wu et al., 2003) rather than the sources of organic matter. The C/N ratio modified by decomposition process could superimpose on the estimation of different sources of organic matter (Thornton and McManus, 1994).

The values in the surface sediment samples varied from -23.43% to -20.26% (average $-22.00 \pm 0.60\%$) for $\delta^{13}\text{C}$. The range of $\delta^{13}\text{C}$ of organic matter is broad in aquatic ecosystems, varying from -30% to -26% in runoff from terrigenous organic carbon (C-3 plants) and -22% to -18% for organic carbon from marine phytoplankton production (Cifuentes et al., 1996). The $\delta^{13}\text{C}$ gradient in sedimentary organic matter in the study area was characterized by a progressive seaward increase (Fig. 4a). The higher $\delta^{13}\text{C}$ values in the east of the study area could reflect sedimentation of a large quantity of marine organic matter. Previous studies have shown that the terrestrial organic matter in the East China Sea was mostly supplied by the Yangtze River (Deng et al., 2006; Kao et al., 2003). However, the $\delta^{13}\text{C}$ values in sedimentary organic matter from the study area were all larger than the suspended matter (-26.4% to -24.5% ; -25.5% to -23.1%) in the lower reach of the Yangtze River (Wu et al., 2015; Wang et al., 2012, respectively) and the suspended matter (-26.2% to -24.4%) in the middle and lower zone of the Yangtze River (Bao et al., 2014). The marine organic matters from the in situ planktons mixed with the terrigenous organic matters from the Yangtze River in the nearshore area and generally enlarge the $\delta^{13}\text{C}$ values, and then caused the $\delta^{13}\text{C}$ values different from the Yangtze River estuary. Other studies also found that there is a progressive change in the $\delta^{13}\text{C}$ value from the Yangtze river mainstream, through the Yangtze estuary to the East China Sea shelf (Hu et al., 2012; Li et al., 2012; Liu et al., 2006a; Wu et al., 2003, 2007; Zhang et al., 2007; Zhou et al., 2006).

The $\delta^{15}\text{N}$ values in the surface sediment samples varied from 3.93% to 5.27% (average $4.54 \pm 0.28\%$). It has been reported that the terrigenous detrital organic matter is generally characterized by a low $\delta^{15}\text{N}$ signature while the marine component has a relatively higher $\delta^{15}\text{N}$ value (Mariotti et al., 1984; Peterson et al., 1985; Thornton and McManus, 1994). However, the spatial distribution of $\delta^{15}\text{N}$ in the study area showed a contrary trend with higher values of $\delta^{15}\text{N}$ mainly distributed in the nearshore area locally similar with the C/N ratio (Figs. 3 and 4b). Higher $\delta^{15}\text{N}$ values corresponded with high C/N ratios, which suggested the higher $\delta^{15}\text{N}$ values are principally caused by the organic matter diagenesis (Thornton and McManus, 1994; Cifuentes et al., 1996).

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