



Sources and degradation of sedimentary organic matter in the mud belt of the East China Sea: Implications from the enantiomers of amino acids

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ABSTRACT

Total organic carbon (TOC), total N (TN) and amino acids (AAs, including the bacterial biomarker D-AAs) were determined in sediment cores from the mud belt of the East China Sea (ECS). The concentration of total hydrolyzable AAs (THAAs) ranged from 3.35 to 13.44 $\mu\text{mol/g}$ dry wt (dw) of the sedimentary organic matter (SOM), exhibiting a decreasing trend downcore. Major constituents of the THAAs were glycine (Gly), L-serine (L-Ser), L-alanine (L-Ala), L-glutamic acid (L-Glu) and L-aspartic acid (L-Asp), whereas the D-aspartic acid (D-Asp), D-glutamic acid (D-Glu), D-serine (D-Ser) and D-alanine (D-Ala), along with non-protein AAs (γ -aminobutyric acid and β -alanine) together accounted for ca. 7% of the THAA pool. Given the C/N ratio values, the OM in three cores was predominantly of marine origin. Based on D-Ala yield, bacterial OM represented on average 18% of TOC and ca. 30% of TN. C-normalized THAAs (THAA-C%) was a sensitive indicator of SOM diagenetic alteration in the upper 40 cm of sediment. However, the degradation index (DI) and reactivity index (RI) values for the cores did not exhibit any definite trend, which may indicate that these two indicators were not sensitive in the early stage of sediment degradation. Negative correlation between the D/L ratio of Ala and THAA-C% suggested a close coupling between the extent of degradation and the accumulation and/or selective preservation of bacterial material in the sediments. In addition, correlation between clay content and THAA-C% suggested that fine grained particles played an important role in affecting the quality of SOM.

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1. Introduction

Estuary and shelf seas represent a transition zone between terrestrial and marine environments. Owing to high terrestrial input and high marine primary productivity, > 90% of marine sedimentary organic matter (SOM) is buried along continental margins (Hedges and Keil, 1995). Recent studies have shown that marine OM and terrigenous OM in coastal sediments can exhibit contrasting reactivity and fate (Bourgoin and Tremblay, 2010). Traditionally, terrigenous OM has been considered recalcitrant, so sediments dominated by a terrestrial input would be expected to be less reactive than those dominated by phytoplankton material (Alkhatib et al., 2012). Sediment total organic carbon (TOC), total

nitrogen (TN) and TOC/TN (C/N) are useful proxies for studying the sources of SOM. Terrestrial vascular plants are rich in cellulose and therefore have higher C/N (20–400) than marine phytoplankton (C/N 4–6; Hedges et al., 1986). In addition to bulk elemental analysis, biomarkers (e.g. Hedges and Parker, 1976; Gordon and Goñi, 2003; Bianchi and Canuel, 2011; Li et al., 2012) have been commonly used to track the sources, behavior and degradation state of OM in coastal regions. Amino acids (AAs) are the primary form of N in terrestrial and marine organisms and represent important constituents of living and detrital OM (Cowie and Hedges, 1992). They act as the most labile fraction of bulk OM and can also provide insight into OM source and diagenetic alteration (Cowie and Hedges, 1992; Dauwe and Middelburg, 1998; Dauwe et al., 1999; Kaiser and Benner, 2008). Many studies have shown that AAs are good indicators of the degradation state of particulate OM (POM), dissolved OM (DOM) and SOM (Amon et al., 2001; Davis et al., 2009; Kaiser and Benner, 2009; Bourgoin and Tremblay, 2010; Fernandes et al., 2014; Chen et al., 2016). For

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example, several indicators of OM diagenetic state have been established, based on the yield of C and N as total hydrolyzable AAs (THAAs), reactivity index (RI) and degradation index (DI; Jennerjahn and Ittekkot, 1997; Dauwe and Middelburg, 1998; Dauwe et al., 1999; Davis et al., 2009). Heterotrophic bacteria have been recognized for their critical role in the transformation and mineralization of OC and N in the marine environment (Kawasaki and Benner, 2006). The presence of D-AAs in marine sediments may be linked to the peptidoglycan in the cell walls of bacteria (Moriarty and Hayward, 1982; Hagström et al., 2000). Peptidoglycan is a unique bacterial cell wall heteropolymer, containing D-alanine, D-aspartic acid, D-glutamic acid and D-serine (Kaiser and Benner, 2008). Since the D-enantiomers of AAs occur almost exclusively in peptidoglycan (Schleifer and Kandler, 1972; Benner and Kaiser, 2003), they can be used as biomarkers for bacterially derived OM in sediments (McCarthy et al., 1998; Kaiser and Benner, 2008; Bourgoin and Tremblay, 2010). In addition, non-protein AAs such as β -alanine and γ -aminobutyric acid may be formed from microbial decarboxylation of AAs (Lee and Cronin, 1982) and be used as an indicator for microbial reworking of OM in sediments.

Understanding of the source, composition and reactivity of SOM in the East China Sea (ECS) is still limited. A number of studies have been conducted to evaluate the source, transport and burial of OM in the Changjiang Estuary and its adjacent seas, using multiproxy approaches based on stable isotopes and a broad suite of biomarkers (e.g. Deng et al., 2006; Zhang et al., 2007; Gao et al., 2008; Zhu et al., 2008; Hu et al., 2012; Li et al., 2012, 2014). However, few studies have focused on the use of AAs as an indicator to investigate the source and fate of SOM in the ECS. The objectives of the present study were to (i) identify the source, nature and degradation state of SOM in the mud belt of the ECS, (ii) assess the influence of microbial reworking on the SOM in sediment cores based on the composition of D- and L-AAs and (iii) compare the effectiveness of the different degradation indicators for SOM degradation in the area.

2. Sampling and methods

2.1. Study area

The ECS is one of the largest shelf seas in the world, with an average water depth of ca. 72 m. Ca. 90% of terrestrial material entering the ECS shelf is derived from input from the Changjiang River (Zhou et al., 2010). The rich supply of nutrients from the river contributes to the high primary productivity (437 mg C/m²/d) in the ECS (Liu et al., 2015a). The mechanisms of OM transport in the region depend primarily on discharge from the river and coastal currents (Huh and Su, 1999; Chen et al., 2003). Studies have suggested that much of the terrestrial sediment in the ECS was transported southward along the Zhejiang-Fujian coast, forced by the Changjiang Diluted Water (CDW) and Jiangsu Coastal Current (JCC), where the sediment was deposited west of 123° E due to the barrier effect of the shear force from the Zhejiang-Fujian Coastal Current (ZFCC) and the Taiwan Warm Current (TWWC), forming the mobile mud belt on the inner shelf of the ECS (Liu et al., 2007; Fig. 1). Liu et al. (2006, 2007) indicated that this mobile mud belt exhibits a high sediment accumulation rate along the Zhejiang-Fujian coast, with an annual sedimentation rate of ca. 1.5 cm/yr.

2.2. Sample collection

Three cores were collected from the Zhejiang-Fujian mud belt using a stainless steel box corer onboard the R/V Ke Xue No. 1 in October 2014 (Fig. 1). They were collected at a water depth of

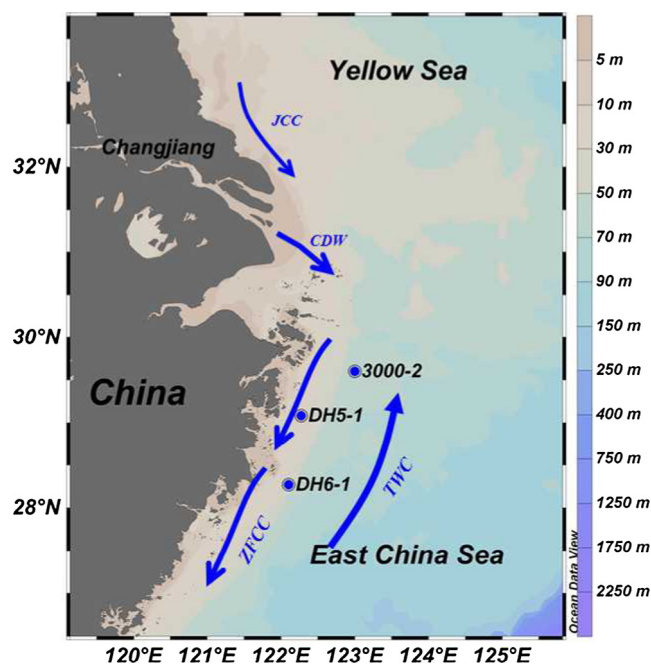


Fig. 1. Regional ocean circulation patterns and sampling stations in the East China Sea. JCC, Jiangsu Coastal Current; TWC, Taiwan Warm Current; CDW, Changjiang Diluted Water; ZFCC, Zhejiang-Fujian Coastal Current. Figure was generated using Ocean Data View software (Schlitzer, 2014. Ocean Data View. <http://odv.awi.de>).

48.7 m at station 3000-2 (123.000°E, 29.600°N), 36.8 m at station DH5-1 (122.276°E, 29.082°N) and 38.8 m at station DH6-1 (122.104°E, 28.270°N), respectively. Samples were retrieved using sub-corers (10 cm i.d.) to recover an undisturbed surface. Recovery was 36 cm at 3000-2, 32 cm at DH5-1 and 34 cm at DH6-1. Then, three sub-cores were cut into 2 cm intervals, transferred to clean glass vials and immediately frozen at -20°C onboard. After being brought to the laboratory on land, the samples were freeze-dried and homogenized by grinding in an agate mortar prior to analysis. The sampling stations represent the major transport and deposition pathway of Changjiang-derived sediments initially deposited and remobilized in the Zhejiang-Fujian coastal mud area.

2.3. Grain size analysis

Grain size composition was analyzed using a laser particle size analyzer (Mastersizer 2000; Malvern Instruments Ltd., UK). Three categories were distinguished: $< 4\ \mu\text{m}$ for clay, $4\text{--}63\ \mu\text{m}$ for silt and $> 63\ \mu\text{m}$ for sand. The samples were analyzed in duplicate and the precision was better than 3% ($n = 6$).

2.4. TOC and TN

The homogenized dry samples were acidified with 4 mol/l HCl at room temperature for 24 h to remove carbonate. Thereafter, they were rinsed with Milli-Q water and dried in an oven at 50°C . The sediments were then used for bulk TOC and TN analysis. These were performed using a Perkin-Elmer 2400 II CHN analyzer. The analytical precision for TOC and TN was $\pm 0.02\ \text{wt}\%$ and $\pm 0.01\ \text{wt}\%$, respectively ($n = 5$).

2.5. AAs

THAAs (L-AAs, D-AAs and two non-protein AAs - β -Ala and γ -Aba) were determined using the *o*-phthalaldehyde (OPA) and N-isobutyryl-L/D-cysteine (IBLC/IBDC) derivatization methods and separated using high performance liquid chromatography

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