

Spatial variability in the abundance, composition, and age of organic matter in surficial sediments of the East China Sea

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[1] Understanding the sources and fate of organic matter (OM) sequestered in continental margin sediments is of importance because the mode and efficiency of OM burial impact the carbon cycle and the regulation of atmospheric CO₂ over long time scales. We carried out molecular (lignin-derived phenols from CuO oxidation), elemental, isotopic ($\delta^{13}\text{C}$, $\Delta^{14}\text{C}$), and sedimentological (grain size and mineral surface area) analyses in order to examine spatial variability in the abundance, source, age of surface sediments of the East China Sea. Higher terrigenous organic matter values were found in the main accumulating areas of fluvial sediments, including the Changjiang (Yangtze) Estuary and Zhejiang-Fujian coastal zone. Isotopic and biomarker data suggest that the sedimentary OM in the inner shelf region was dominated by aged ($\Delta^{14}\text{C} = -423 \pm 42\%$) but relatively lignin-rich OM ($\Lambda = 0.94 \pm 0.57$ mg/100 mg OC) associated with fine-grained sediments, suggesting important contributions from soils. In contrast, samples from the outer shelf, while of similar age ($\Delta^{14}\text{C} = -450 \pm 99\%$), are lignin poor ($\Lambda = 0.25 \pm 0.14$ mg/100 mg OC) and associated with coarse-grained material. Regional variation of lignin phenols and OM ages indicates that OM content is fundamentally controlled by hydrodynamic sorting (especially, sediment redistribution and winnowing) and in situ primary production. Selective sorption of acid to aldehyde in clay fraction also modified the ratios of lignin phenols. The burial of lignin in East China Sea is estimated to be relatively efficient, possibly as a consequence of terrigenous OM recalcitrance and/or relatively high sedimentation rates in the Changjiang Estuary and the adjacent Zhejiang-Fujian mud belt.

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1. Introduction

[2] In the modern ocean, river-dominated continental shelves represent a major sink of terrestrial organic carbon (OC) and are globally important regions for OC burial in the marine environment [Burdige, 2005; Hedges and Keil, 1995]. Large fluxes of land-derived OC, enhanced marine productivity, and high sedimentation rates are contributing factors leading to the significance of these regions in terms of carbon cycling and sequestration [Aller and Blair, 2004; Gordon and Goni, 2004; Tesi et al., 2007]. However, carbon cycling

in these spatially heterogeneous and dynamic environments, including the relative importance of organic matter (OM) remineralization, burial, and across-margin export is not fully understood [Zhu et al., 2011a]. In particular, variation of OC composition (e.g., ancient OC and recent soil C, marine fresh materials, etc.) and modes of particle association may lead to different extents of oxidation in surface sediment [Blair and Aller, 2012].

[3] A variety of geochemical approaches have been employed to elucidate the composition and fate of terrestrial and marine OC in continental shelves sediments, including OC/N ratios, $\delta^{13}\text{C}$ compositions, and the abundance and distribution of specific biomarker compounds (including lignin phenols, plant wax *n*-alkanes, phytoplankton sterols, etc.). Studies using multiple biogeochemical indicators, primarily biomarkers in combination with isotopic compositions, have proven useful in elucidating the spatial variability of OC composition and sources of organic matter in complex environment of coastal shelves [Drenzek et al., 2007; Goni et al., 2005; Zhu et al., 2011a; 2013]. Radiocarbon (^{14}C) can provide an important constraint on the predepositional history of terrestrial and marine OC [Eglinton et al., 1997; Griffith

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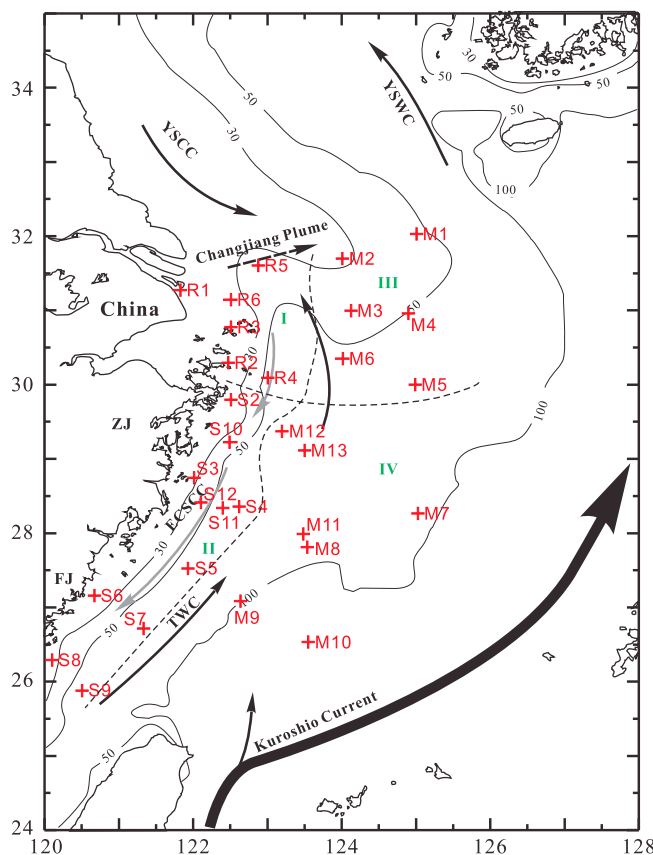


Figure 1. Map of the spatial distribution of the sampling stations in the ECS. The dashed lines divide the study region into four subregions described as I, II, III, and IV. ZJ and FJ are the abbreviations of Zhejiang and Fujian.

et al., 2010], and the recognition of fossil OC in marine environment [Drenzek *et al.*, 2007; Galy *et al.*, 2007].

[4] Continental shelves are sites of extensive reworking and associated organic carbon remineralization [Aller and Blair, 2004]. Recent studies suggest that the burial of OC in river-dominated shelves is initially controlled by particle delivery and deposition, such as riverine export and dispersal [Allison *et al.*, 2000; Blair and Aller, 2012]. Physical reworking of shelves sediments driven by bottom currents and tides can increase the oxygen exposure of sediments and also add relatively fresh, planktonic organic matter to the sediment [Aller and Blair, 2004; Hedges and Keil, 1995]. For example, on the northern Louisiana shelf, periodic resuspension and redistribution of surface sediments by storms result in accelerated the degradation of organic matter [Goñi *et al.*, 2006].

[5] Approximately one third of organic matter buried in marine sediments may be of terrestrial origin [Burdige, 2005]. Pre-aged soil-derived terrestrial OC or petrogenic OC derived from erosion on sedimentary rocks may comprise a significant proportion of the OC in river-dominated margins [Griffith *et al.*, 2010; Hilton *et al.*, 2011]. The heterogeneous composition of terrigenous organic matter (OM_{terr}) (TOM) may lead to errors in assessments of its contribution to OM buried in marine sediments when traditional two-end-member mixing models are applied [Gordon and Goni, 2004].

[6] The East China Sea (ECS) represents an important sea system with shallow (< 130 m in average) but broad (> 500 km) shelf, providing a unique setting to examine the fate of OM_{terr}. In the context of the supply and fate of OM_{terr} [Deng *et al.*, 2006], the Changjiang is the most important feature. The inner continental shelf (up to ~60 m in depth) receives a large amount material from the Changjiang during each flood season [Qin *et al.*, 1996]. Much of this material is resuspended during winter storms and is transported southward toward the Taiwan Strait [DeMaster *et al.*, 1985; Liu *et al.*, 2006]. The outer continental shelves of ECS (water depth, 60–200 m) are minimally influenced by terrestrial input. Most of this area retains the morphological features and sedimentary characteristics of the last stage of the Late Pleistocene, and has been classed as a “relict deposit” [Emery, 1968]. The fate of Changjiang-derived material in ECS shelves has been the subject of intensive investigation [DeMaster *et al.*, 1985; Liu *et al.*, 2006]. These studies have revealed that the sediment mainly disperses alongshore in a southward direction, and is largely confined to the inner shelf [Huh and Su, 1999; Lin *et al.*, 2002]. Limited amounts of fine-grained material could, however, be transported laterally across the shelf [Hung *et al.*, 1999; Oguri *et al.*, 2003]. However, these interpretations are mostly based on bulk geochemical analyses or sedimentological investigations. Detailed geochemical studies utilizing source-specific biomarkers (e.g., lignin phenols) and radiocarbon characteristics of organic materials are presently rare [Zhu *et al.*, 2011a; Li *et al.*, 2012].

[7] A recent study indicates the Changjiang subaqueous delta front (122°10′–30′E, 31°–31°20′N) will continue to erode as long as the annual discharge from the river remains below ~270 Mt/yr of sediment [Yang *et al.*, 2007]. Sediment discharge of the Changjiang, as measured at Datong, fell from 490 Mt/yr in the 1950s and 1960s to ~150 Mt/yr in recent years because of dam construction in the basin [Yang *et al.*, 2011]. The impact of these changes in sediment supply on carbon cycling in the ECS remains uncertain.

[8] The goal of this study is to investigate in detail the source and distribution of organic matter in surface sediments of East China Sea using both the stable carbon isotopic compositions and radiocarbon contents of bulk organic carbon as well as the abundance and composition of molecular markers of vascular plant input (lignin-derived phenols), and to place this information within a sedimentological context. In this way, we seek information on OM_{terr} supply and dispersal in this important shelf sea system.

2. Materials and Methods

2.1. Study Site and Samples

[9] Surface sediment samples were collected from the inner and outer shelf of the East China Sea during 2004–2006 (Figure 1 and Table 1). Sediments were collected using a 50 × 50 cm box corer, which was subsampled into 0.5 or 1 cm intervals aboard ship and then frozen (−20°C) until they were freeze-dried for subsequent analysis in the laboratory.

2.2. Analytical Methods

[10] Grain size characteristics were measured directly on aliquots of the surface sediment samples using a Coulter LS 100Q (Coulter Company, USA), after treatment with 5% H₂O₂ and 0.2 M HCl to dissolve organic matter and

Table 1. Sample Locations, Elemental Analysis, and Isotopic Composition of Surface Sediments From the ECS^a

| Location | Sample Location | Date | Longitude | Latitude | Water Depth (m) | Region | Mean Grain Size (μm) | %clay | %silt | %sand | SA m^2/g | %OC | %TN | N/C | $\delta^{13}\text{C}$ | Delta ¹⁴ C | ¹⁴ Cage | f _{ANC} | f _{MOD} |
|-------------|-----------------|-----------------|-----------|----------|-----------------|--------|-----------------------------------|-------|-------|-------|--------------------------|------|-------|--------|-----------------------|-----------------------|--------------------|------------------|------------------|
| Inner shelf | S2 | November, 2006 | 122.51 | 29.80 | 23 | II | 5 | 40 | 60 | 0 | 21 | 0.47 | 0.05 | 0.097 | -21.66 | -437.1 | 4560±40 | 0.44±0.04 | 0.50±0.04 |
| | S3 | November, 2006 | 122.01 | 28.74 | 18 | II | 5 | 44 | 56 | 0 | 19 | 0.69 | 0.09 | 0.107 | -23.18 | -477.2 | 5150±35 | 0.48±0.04 | 0.52±0.04 |
| | S4 | November, 2006 | 122.62 | 28.36 | 73 | II | 5 | 42 | 58 | 0 | | 0.53 | | | -21.56 | -437.3 | 4560±30 | 0.44±0.04 | 0.50±0.04 |
| | S5 | November, 2006 | 121.93 | 27.52 | 80 | II | 4 | 46 | 54 | 0 | | 0.62 | 0.10 | 0.139 | -22.89 | -422.6 | 4360±30 | 0.42±0.04 | 0.58±0.04 |
| | S6 | November, 2006 | 120.68 | 27.15 | 20 | II | | | | | 27 | 0.61 | 0.09 | 0.123 | | | | | |
| | S7 | November, 2006 | 121.34 | 26.71 | 73 | II | 6 | 40 | 60 | 0 | 10 | 0.59 | 0.07 | 0.106 | -22.25 | -450.9 | 4760±35 | 0.45±0.04 | 0.55±0.04 |
| | S8 | November, 2006 | 120.10 | 26.29 | 32 | II | 4 | 48 | 52 | 0 | 28 | 0.69 | 0.09 | 0.109 | -22.48 | -385.7 | 3860±30 | 0.38±0.04 | 0.62±0.04 |
| | S9 | November, 2006 | 120.51 | 25.87 | 66 | II | 6 | 39 | 61 | 0 | 14 | 0.67 | 0.10 | 0.124 | -19.14 | -408.8 | 4170±35 | 0.40±0.04 | 0.60±0.04 |
| | S10 | June, 2006 | 122.50 | 29.23 | 40 | II | 10 | 35 | 65 | 0 | | 0.57 | 0.09 | 0.129 | -21.62 | | | | |
| | S11 | June, 2006 | 122.39 | 28.34 | 63 | II | 8 | 41 | 59 | 0 | | 0.61 | 0.09 | 0.134 | -21.47 | | | | |
| | S12 | June, 2006 | 122.10 | 28.41 | 38 | II | 10 | 37 | 64 | 0 | 17 | 0.47 | 0.07 | 0.135 | -21.75 | | | | |
| | R1 | July, 2005 | 121.82 | 31.28 | | I | 13 | 34 | 63 | 3 | 18 | 0.53 | 0.07 | 0.118 | -23.26 | | | | |
| | R2 | July, 2005 | 122.48 | 30.30 | | I | 16 | 26 | 71 | 4 | 15 | 0.36 | 0.05 | 0.120 | -23.90 | -486.7 | 5300±35 | 0.49±0.04 | 0.51±0.04 |
| R3 | July, 2005 | 122.50 | 30.78 | | I | 15 | 29 | 68 | 3 | 15 | 0.47 | 0.06 | 0.104 | -23.89 | -395.1 | 3980±35 | 0.39±0.04 | 0.61±0.04 | |
| R4 | July, 2005 | 123.00 | 30.10 | | I | 9 | 41 | 58 | 1 | | 0.30 | 0.05 | 0.132 | -20.39 | | | | | |
| R5 | July, 2005 | 122.88 | 31.61 | | I | 8 | 41 | 59 | 0 | 10 | 0.41 | 0.05 | 0.111 | -21.90 | -328.3 | 3140±40 | 0.33±0.05 | 0.67±0.05 | |
| R6 | November, 2006 | 122.50 | 31.15 | 17 | I | 6 | 38 | 61 | 0 | 16 | 0.56 | 0.08 | 0.116 | -22.75 | -417.2 | 4280±35 | 0.41±0.04 | 0.59±0.04 | |
| Outer shelf | M1 | June, 2006 | 125.00 | 32.03 | 47 | III | 143 | 20 | 31 | 49 | 13 | 0.36 | 0.06 | 0.13 | -21.90 | -311.7 | 2940±35 | 0.31±0.05 | 0.69±0.05 |
| | M2 | September, 2004 | 124.00 | 31.70 | | III | 39 | 27 | 50 | 23 | 4 | 0.26 | 0.05 | 0.172 | -22.34 | -421.3 | 4340±35 | 0.41±0.04 | 0.59±0.04 |
| | M3 | June, 2006 | 124.12 | 31.01 | 42 | III | 239 | 5 | 7 | 88 | 5 | 0.08 | 0.02 | 0.218 | -19.86 | | | | |
| | M4 | June, 2006 | 124.89 | 30.96 | 57 | III | 12 | 36 | 61 | 4 | | 0.37 | 0.06 | 0.131 | -21.78 | -340.5 | 3290±30 | 0.34±0.04 | 0.66±0.04 |
| | M5 | June, 2006 | 124.98 | 30.00 | 57 | III | 199 | 5 | 7 | 88 | 11 | 0.20 | 0.04 | 0.184 | -21.30 | -326.1 | 3110±30 | 0.32±0.05 | 0.68±0.05 |
| | M6 | September, 2004 | 124.00 | 30.36 | | III | 179 | 14 | 19 | 67 | | 0.30 | 0.04 | 0.129 | -23.57 | -584 | 6990±40 | 0.58±0.03 | 0.42±0.03 |
| | M7 | November, 2006 | 125.01 | 28.27 | 100 | IV | 156 | 7 | 7 | 86 | 13 | 0.28 | 0.03 | 0.097 | -22.31 | -494.2 | 5420±45 | 0.49±0.04 | 0.51±0.04 |
| | M8 | November, 2006 | 123.53 | 27.81 | 91 | IV | 157 | 6 | 7 | 87 | 5 | 0.21 | 0.04 | 0.164 | -25.00 | | | | |
| | M9 | November, 2006 | 122.63 | 27.08 | 106 | IV | 84 | 14 | 11 | 76 | 5 | 0.29 | 0.04 | 0.146 | -22.13 | -476.8 | 5150±35 | 0.48±0.04 | 0.48±0.04 |
| | M10 | November, 2006 | 123.54 | 26.53 | 151 | IV | 86 | 13 | 15 | 72 | 5 | 0.21 | 0.04 | 0.147 | -22.80 | -423 | 4360±45 | 0.42±0.04 | 0.58±0.04 |
| | M11 | June, 2006 | 123.47 | 27.99 | 89 | IV | 175 | 12 | 15 | 72 | 8 | 0.26 | 0.04 | 0.147 | -20.54 | | | | |
| | M12 | November, 2006 | 123.19 | 29.37 | 67 | IV | 34 | 24 | 20 | 57 | 15 | 0.72 | | | -23.55 | -634.1 | 8020±40 | 0.63±0.03 | 0.37±0.03 |
| | M13 | June, 2006 | 123.50 | 29.12 | 68 | IV | 10 | 45 | 54 | 1 | 10 | 0.32 | 0.06 | 0.170 | -22.14 | -392.1 | 3940±30 | 0.39±0.04 | 0.61±0.04 |
| Average | | | | | | 116 | 18 | 23 | 59 | 8 | 0.30 | 0.05 | 0.15 | -22.25 | -440 | | | | |

^aSA: surface area; N/C is based on the atomic molar ratio, f_{MOD} and f_{ANC} are estimated for the fraction of modern carbon and ancient carbon based on radiocarbon C-14 data.

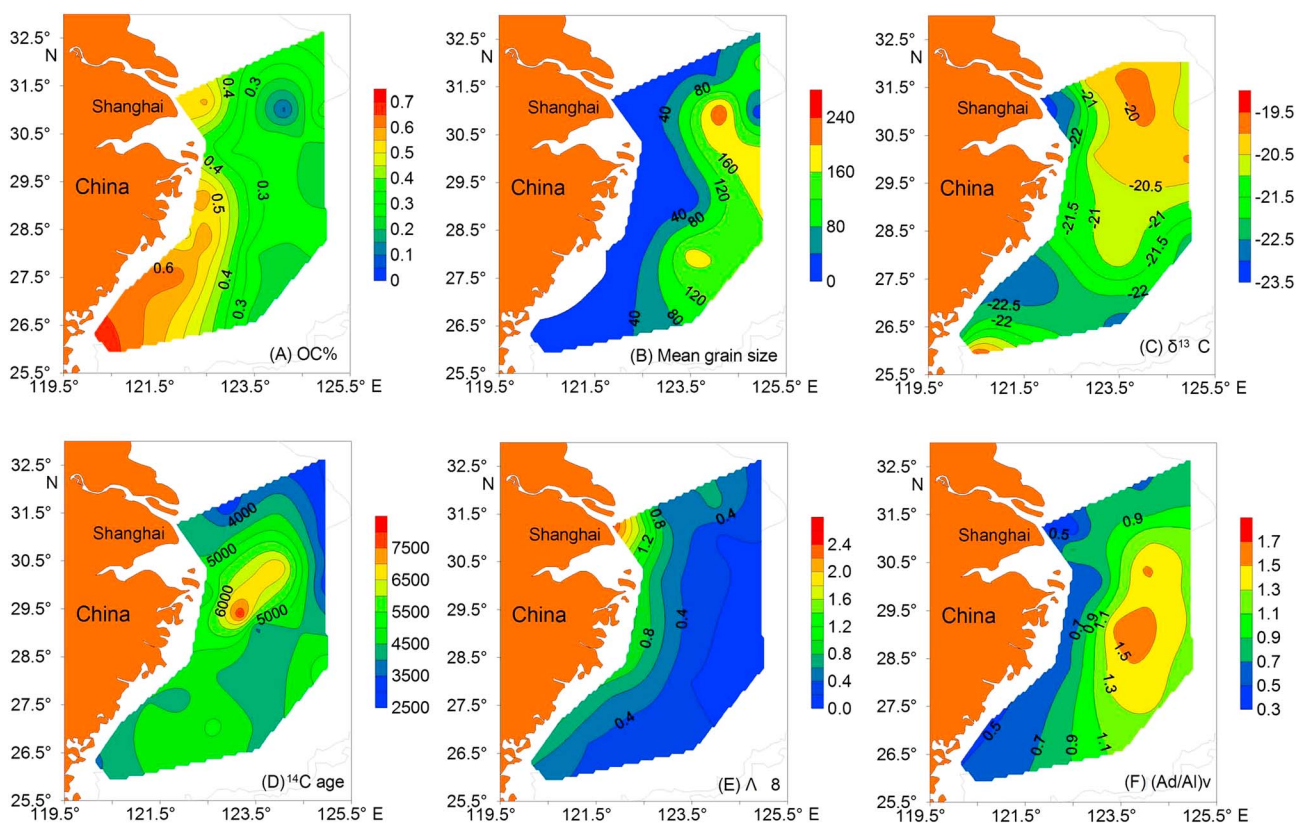


Figure 2. Spatial contours of geochemical parameters of surface sediments in the ECS. A: the percentage of organic carbon; B: mean grain size; C: stable carbon isotopes; D: ^{14}C ages; E: $\Delta^8 = \text{C} + \text{V} + \text{S}$; F: $(\text{Ad}/\text{Al})_v$ is the acid-to-aldehyde ratio of V phenols.

biogenic carbonate, with an analytical precision within $\pm 5\%$. The remaining sediments were ground to pass an 80-mesh ($187.5\ \mu\text{m}$) sieve for elemental, isotopic and lignin analysis.

[11] The concentrations of organic carbon and total nitrogen (relative precision $\pm 5\%$) were determined using a CHNOS Elemental Analyzer (Model: Vario EL III). Weight percentages of organic carbon were analyzed after removing the carbonate fraction by vapor phase acidification. Weight percentages of total nitrogen were analyzed similar but without acidification. Stable carbon isotopic compositions of decarbonated sediments were determined using a Flash EA1112 Elemental Analyzer connected to an Isotope Ratio Mass Spectrometer produced by the Finnigan MAT Co. (Model: DELTA plus/XP). $^{13}\text{C}/^{12}\text{C}$ ratios are expressed relative to the PDB standard by the conventional δ notation. Analytical precision, determined by replicate analysis of the same sample, was $\pm 0.2\%$.

[12] The mineral-specific surface area (SA) of unground sediment samples was determined using a BEL-MAX apparatus equipped with highly precise gas sensor for low pressure measurement. The specific surface area was evaluated using the Brunauer-Emmett-Teller (BET) equation. Prior to the measurement, the samples were first outgassed at 300°C under vacuum for 10 h.

[13] Lignin phenols were measured according to the methods of Hedges and Ertel [1982] and Yu *et al.* [2011]. Briefly, dried and ground sediments (0.5–1.0 g) were oxidized by CuO in oxygen-free bombs. The bombs were heated at 160°C for 3 h. Ethylvanillin was then added as

the recovery standard before samples were acidified to $\text{pH} < 2$, extracted into 99:1 (volume ratio) ethyl acetate/petroleum ether, dried, and stored (-20°C) until further analysis. Samples were redissolved in acetonitrile and derivatized using BSTFA + 1%TMCS for measurement using a HP 6890 Gas Chromatography (DB-1 column, FID). The oven temperature increased from 100°C to 270°C by $4^\circ\text{C}\ \text{min}^{-1}$ and held for 10 min finally. Lignin phenols were quantified with calibration curves of commercial standards and corrected by the recovery of ethylvanillin. The mean analytical precision of lignin parameters used in this study was better than $\pm 10\%$.

[14] Radiocarbon analyses were performed at the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) facility at Woods Hole Oceanographic Institution following. After removal of inorganic carbon with 10% HCl at 60°C and drying, sedimentary OC was combusted to CO_2 and then converted to graphite via Fe/ H_2 catalytic reduction. The graphite was pressed into AMS targets and analyzed by AMS. Radiocarbon values are reported as $\Delta^{14}\text{C}$ (‰) and Fraction modern (Fm) [Stuiver *et al.*, 1986].

2.3. Statistical Analysis

[15] Pearson's correlation was used to test the correlation between different parameters (water depth, grain size, carbon isotopes, and lignin phenols, etc.) (p values were presented as two-tailed probability). All analyses were performed using SPSS 13.0 for Windows (SPSS Inc., US).

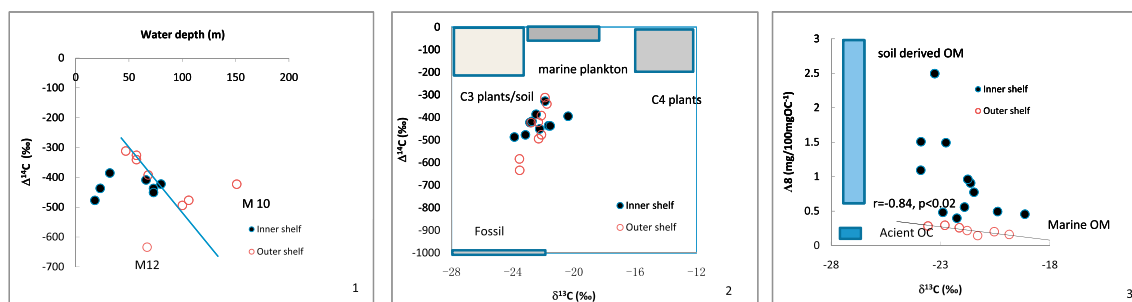


Figure 3. (1) Water depth versus $\Delta^{14}\text{C}$ distribution, (2) $\Delta^{14}\text{C}$, and (3) $\Lambda 8$ versus $\delta^{13}\text{C}$ distribution in the surface sediment samples in the ECS.

3. Results

3.1. General Characteristics of Sedimentary Organic Matter

[16] The composition of bulk sediments from the inner ($< \sim 60$ m) and outer ($> \sim 60$ m) shelf of ECS is presented in Table 1 and Figure 2. Mean grain sizes of outer shelf samples (10–239 μm , average 116 μm) were much higher than those of the inner shelf (4–16 μm , ave. 8 μm). With the exception of few outer shelf samples characterized by a higher clay-silt content, most coarse-grained samples displayed a significant negative relationship between grain size and water depth ($R = -0.77$, $p < 0.05$). Similar results were also observed for inner shelf samples. Inner shelf samples were dominated by clay-silt fraction while most outer shelf samples were dominated by %sand (Table 1). There was also considerable variability in mineral-specific surface area (SA) values. The average SA value for inner shelf samples was $17.5 \text{ m}^2 \text{ g}^{-1}$ (max. $28.2 \text{ m}^2 \text{ g}^{-1}$), while the average value for outer shelf samples was $8.39 \text{ m}^2 \text{ g}^{-1}$. Average values for %OC and %TN values were 0.54% and 0.07% for inner shelf samples, and 0.30% and 0.05% for outer shelf samples, respectively. All samples yielded SA-normalized OC loadings (OC:SA) between 0.2 and 0.7 mg C m^{-2} (ave., 0.36 mg C m^{-2}). These loadings are lower than those observed in many river and marine sediments (0.5–1.0) but higher than OC:SA values reported for river-dominated margins (0.25 ± 0.08) [Hedges and Keil, 1995; Keil et al., 1997; Burdige, 2005]. Similar to the OC loadings, measured TN:SA ratios (0.2–0.8) are low compared to typical values of 0.7–0.8 reported for river suspended sediments and marine depocenter sediments [Mayer et al., 1998]. There is no significant relationship between mean grain size and %OC for inner shelf samples, while with the exception of two fine-grained sediment samples, OC loadings of outer shelf sediments are inversely correlated with mean grain size ($R = -0.63$, $p < 0.05$).

3.2. Bulk Organic Matter Composition

[17] There is a slight increase of TN/OC ratio from inner shelf to outer shelf samples (Table 1). A linear relationship exists between OC and TN ($R = 0.93$, $p < 0.001$) with a positive intercept of average inorganic nitrogen content at 0.005% [Goñi et al., 1998]. The modified organic nitrogen (ON)/OC atomic ratios (corrected for this percentage of inorganic nitrogen) ranged from 0.09 to 0.13 for inner shelf samples and 0.08 to 0.16 for outer shelf samples. Sediments from

the inner and outer shelf regions exhibit distinct differences in average stable carbon isotopic composition of bulk organic carbon ($\delta^{13}\text{C}_{\text{OC}}$) in (–19.1‰ to –23.9‰) with more depleted values observed in the shallow regions of the inner shelf. In the northern region of the outer shelf, enriched $\delta^{13}\text{C}_{\text{OC}}$ values (~ -21 ‰) were observed compared to the other sandy region (~ -23 ‰). This contrast was also observed in a previous study [Zhu et al., 2011a] (Figure 2).

[18] The radiocarbon contents (expressed as $\Delta^{14}\text{C}$ values) of surface sediments varied between –328.3‰ and –634.1‰, corresponding to conventional ^{14}C ages of 3140 to 8020 years before present (years BP) (Table 1 and Figure 2). The oldest ^{14}C age was returned for a station on the outer shelf (M12) characterized by finer-grained sediment with a higher SA value. With the exception of samples M12 and M10, we observed contrasting behaviors between water depth and $\Delta^{14}\text{C}$ among samples shallow region and over 50 m isobath, with deeper samples yielding older ages and shallower samples yielding fresher ages among over 50 m isobath region while OM ages getting fresher with increasing depth in shallow region (Figure 3.1). No strong relationship appears to exist between $\Delta^{14}\text{C}_{\text{OC}}$ and $\delta^{13}\text{C}_{\text{OC}}$ in inner shelf sediments, whereas these parameters appear correlated in outer shelf sediments ($R = 0.89$, $p < 0.001$) (Figure 3.2).

[19] Based on %OC, N/C ratios, and mean grain size, our study area could be divided into four subregions: Reg. I, River & Delta (average values of %OC, N/C, and grain size at 0.41%, 0.12, and 12 μm , respectively), dominated by % silt-clay but minor %sand; Reg. II, ZJ-FJ coast (0.59%, 0.12, and 6 μm , respectively, homogenized in %silt-clay); Reg. III, Paleo-Changjiang Delta (the Yangtze shoal) (0.26%, 0.16, and 135 μm , respectively, big variability in %sand (4–88)); and Reg. IV, Outer shelf (average values of 0.33%, 0.14, and 100 μm , respectively, less %clay-silt than Reg. III) (Figure 1), which was also suggested in Qin et al. [1996]. Liu et al. [2010] described similar depositional settings based on grain size composition and magnetic distribution.

3.2.1. CuO Product-Based Parameters

[20] The abundance of vanillyl, syringyl, and cinnamyl (V + S + C) lignin-derived phenols ($\Lambda 8$) (mg/100 mg OC) decreased seaward from the river mouth to the shelf, but exhibited no discernable decrease southward along the Zhejiang (ZJ) – Fujian (FJ) Coast (Table 2 and Figure 2). The highest lignin content was detected at the river mouth station (R1) with a $\Lambda 8$ value of 2.50 mg/100 mg OC. Lignin contents differed significantly between inner and outer shelf sediments with average

Table 2. Concentrations and Ratios of the CuO Oxidation Products of Surface Sediments From the ECS^a

| | $\Delta 8$ | P | V | S | C | | | | | | | |
|-------------|------------|-----------|------|------|------|------|------|--------|----------------------|-------|------------------|--------------------|
| | Yields, | mg/100 mg | OC | | | S/V | C/V | LPVI | (Ad/Al) _v | P/S+V | OC _{BM} | OC _{SOIL} |
| Inner shelf | | | | | | | | | | | | |
| S5 | 0.48 | 0.39 | 0.26 | 0.20 | 0.02 | 0.80 | 0.08 | 143.52 | 0.66 | 0.85 | | |
| S6 | 0.83 | 0.27 | 0.48 | 0.32 | 0.04 | 0.67 | 0.08 | 137.89 | 0.47 | 0.35 | | |
| S7 | 0.40 | 0.23 | 0.22 | 0.16 | 0.02 | 0.69 | 0.08 | 138.89 | 0.59 | 0.61 | | |
| S8 | 0.67 | 0.29 | 0.38 | 0.26 | 0.03 | 0.68 | 0.07 | 140.51 | 0.45 | 0.45 | | |
| S9 | 0.45 | 0.26 | 0.25 | 0.17 | 0.03 | 0.69 | 0.11 | 139.73 | 0.59 | 0.60 | | |
| S10 | 0.91 | 0.48 | 0.45 | 0.38 | 0.07 | 0.85 | 0.15 | 147.87 | 0.59 | 0.57 | | |
| S11 | 0.77 | | 0.37 | 0.34 | 0.06 | 0.93 | 0.17 | 151.95 | 0.66 | | | |
| S12 | 0.96 | 0.79 | 0.51 | 0.39 | 0.06 | 0.75 | 0.12 | 142.66 | 0.56 | 0.87 | | |
| R1 | 2.50 | 0.94 | 1.25 | 1.08 | 0.16 | 0.86 | 0.13 | 147.79 | 0.49 | 0.40 | | |
| R2 | 1.09 | | 0.62 | 0.40 | 0.07 | 0.65 | 0.11 | 137.86 | 0.55 | | | |
| R3 | 1.51 | 1.11 | 0.83 | 0.59 | 0.08 | 0.71 | 0.10 | 140.26 | 0.93 | 0.78 | | |
| R4 | 0.49 | | 0.35 | 0.12 | 0.02 | 0.36 | 0.06 | 121.84 | 0.57 | | | |
| R5 | 0.56 | | 0.40 | 0.14 | 0.02 | 0.36 | 0.04 | 121.14 | 0.78 | | | |
| R6 | 1.49 | 0.40 | 0.79 | 0.63 | 0.08 | 0.80 | 0.10 | 144.13 | 0.37 | 0.29 | | |
| Average | 0.92 | 0.53 | 0.50 | 0.37 | 0.06 | 0.72 | 0.11 | 140.90 | 0.59 | 0.61 | | |
| Outer shelf | | | | | | | | | | | | |
| M1 | 0.32 | | 0.18 | 0.12 | 0.02 | 0.64 | 0.11 | 137.31 | 0.78 | | | |
| M2 | 0.56 | | 0.29 | 0.25 | 0.03 | 0.86 | 0.09 | 146.44 | 0.72 | | 0.21 | 0.79 |
| M3 | 0.16 | | 0.05 | 0.10 | 0.01 | 1.88 | 0.28 | 162.94 | 1.26 | | 0.52 | 0.48 |
| M4 | 0.22 | | 0.14 | 0.06 | 0.01 | 0.44 | 0.09 | 126.86 | 1.20 | | 0.28 | 0.72 |
| M5 | 0.15 | | 0.06 | 0.08 | 0.01 | 1.29 | 0.19 | 144.98 | 1.20 | | 0.34 | 0.66 |
| M6 | 0.28 | | 0.13 | 0.11 | 0.04 | 0.88 | 0.34 | 155.15 | 1.53 | | 0.05 | 0.95 |
| M7 | 0.10 | | 0.05 | 0.04 | 0.01 | 0.71 | 0.13 | 141.21 | 1.11 | | 0.21 | 0.79 |
| M10 | 0.29 | 0.39 | 0.13 | 0.16 | 0.01 | 1.26 | 0.07 | 161.74 | 1.17 | 1.35 | 0.15 | 0.85 |
| M11 | 0.20 | 0.34 | 0.11 | 0.08 | 0.01 | 0.69 | 0.10 | 139.20 | 1.45 | 1.79 | 0.43 | 0.57 |
| M13 | 0.26 | 0.47 | 0.13 | 0.11 | 0.02 | 0.84 | 0.16 | 147.96 | 1.68 | 1.99 | 0.23 | 0.77 |
| Average | 0.25 | 0.40 | 0.13 | 0.11 | 0.02 | 0.95 | 0.16 | 146.38 | 1.21 | 1.71 | | |

^a $\Delta 8 = C + V + S$, normalized to 10 mg of organic carbon; C = ferulic acid + cinnamic acid; V = vanillin + acetovanillone + vanillic acid; S = syringaldehyde + acetosyringone + syringic acid; P = p-hydroxybenzaldehyde + p-hydroxybenzoic acid + p-hydroxyacetophenol; (Ad/Al)_v is the acid to aldehyde ratio of V phenols; LPVI: lignin phenol vegetation index; OC_{BM} and OC_{SOIL} are the estimation of the contribution from benthic algae OC and soil OC based on stable isotope composition.

$\Delta 8$ values of 0.95 and 0.29, respectively. These values are comparable to those in the Washington margin, the Gulf of Mexico, and Peru margin [Prah et al., 1994; Sampere et al., 2008; Bergamaschi et al., 1997].

[21] $\Delta 8$ values decreased with increasing of water depth and mean grain size. They also showed moderate positive correlation with SA for inner shelf samples but a weak negative correlation among outer shelf samples. There is an apparent correlation between $\delta^{13}C_{OC}$ versus $\Delta 8$ for outer shelf samples (Figure 3.3), whereas inner shelf samples do not exhibit a clear relationship.

[22] Among the CuO oxidation products, vanillyl and syringyl phenols were present in approximately equal proportions and together accounted for 50–90% and 30–90% of V + S + C phenols in outer and inner shelf sediments, respectively. The p-hydroxybenzenes were single most abundant compound class among the CuO reaction products, accounting for as high as 50–60% of total phenols in outer shelf sediments. Their proportional abundance in inner shelf sediments was lower (20–40%). The data collected from Gulf of Mexico displayed similar CuO product signatures from the river to the shelf [Gordon and Goni, 2003].

[23] Average values of syringyl to vanillyl (S/V) and cinnamyl to vanillyl (C/V) ratios were 0.70 and 0.10, respectively, for inner shelf sediments (Figure 4.1). These values are close those observed from the lower reaches of the Changjiang [Yu et al., 2007], 0.95 and 0.16 for outer shelf (Table 2), similar to values for the Gulf of Mexico [Gordon

and Goni, 2003], higher S/V ratios than the values for the Washington margin [Prah et al., 1994], but lower than those reported by Zhu et al. [2011a] from the same region. A linear relationship between S/V and C/V values for inner shelf samples indicates that samples are mainly composed of lignin derived from angiosperm/gymnosperm woody tissue admixed with nonwoody angiosperm tissue. Increasing S/V and C/V ratios seaward implied increasing proportions of angiosperm leaf and/or grass-derived lignin in outer shelf region, which is also observed in the Louisiana shelf [Goñi et al., 1998]. The lignin phenol vegetation index (LPVI) was calculated according to Tareq et al. [2004]: $LPVI = \left\{ \frac{S(S+1)}{(V+1)+1} \right\} \times \left\{ \frac{C(C+1)}{(V+1)+1} \right\}$; where V, S, and C are expressed in % of the sum of the eight vanillyl, syringyl, and cinnamyl phenols. The LPVI values of inner shelf and outer shelf samples were quite similar. High LPVI values from 120 to 160 likely are consistent with a higher contribution of angiosperms woods. The correlation between LPVI and $\Delta 8$ values in Reg. I suggested that decreasing $\Delta 8$ values covaried with decreasing LPVI (Figure 4.2).

[24] Values for (Ad/Al)_v differ between inner shelf and outer shelf samples (Table 2), with much higher values for outer shelf sediments (average at 1.21, relative to 0.59 for the inner shelf). This observation is contrasted to the results from Washington coast by Keil et al. [1998]. However, the (Ad/Al)_v values exhibit the correlation with %clay among samples collected from Reg. I and IV: finer samples tend to have higher (Ad/Al)_v values (Figure 4.3).

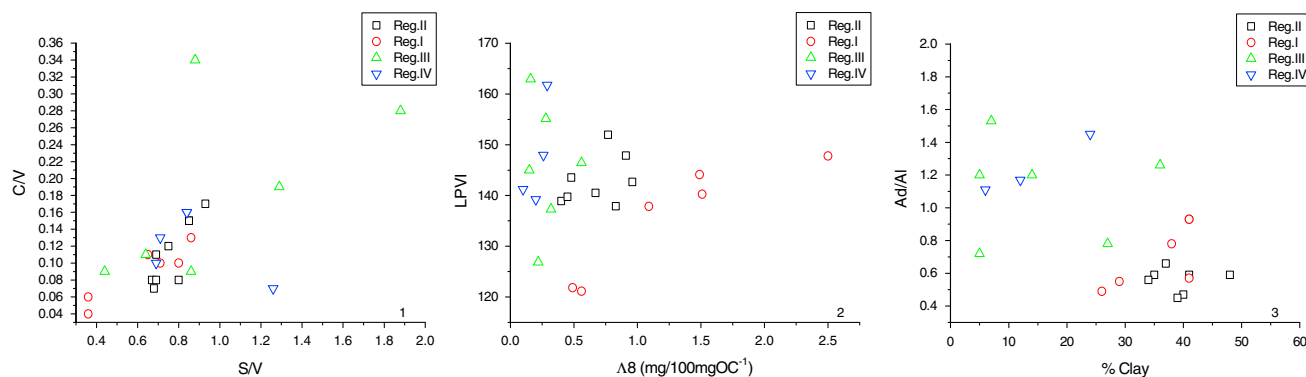


Figure 4. 1 S/V versus C/V, (2) LPVI versus $\Delta 8$ (normalized to 10 mg of organic carbon), (3) (Ad/Al)_v versus %clay distribution in the surface sediment samples in the ECS.

4. Discussion

4.1. Burial of Terrigenous Organic Matter (TOM) in the ECS

[25] The Changjiang is known to be the dominant present-day source of OM_{terr} to the ECS [Deng *et al.*, 2006]. Recent studies have established that the terrestrial signature is mostly confined to a region within 200–250 km off the Changjiang Estuary [Zhang, 1999; Wu *et al.*, 2003], and a band of muddy sediment skirting the inner shelf along the ZJ-FJ Coast [Liu *et al.*, 2006; Zhu *et al.*, 2011a, 2011b]. Terrestrial particles are also transported across the shelf with low-density Changjiang diluted water in the surface layer. Material that escapes the inner shelf may be deposited in the relict sandy region [Xu *et al.*, 2009], or transported to the outer shelf, especially in winter [Guo *et al.*, 2002].

[26] ¹⁴C ages of sediments from the four regions exhibit the following trend of increasing age: IV > II > I > III. The freshest ages found in Reg. III suggest the modification of bulk ages of organic matter in this shallow region with high primary production [Zhu, 2007]. The average radiocarbon contents of OC between inner shelf and outer shelf samples were comparable. This differs from prior observations in the Mexico Gulf where sandier sediment were characterized by lower $\Delta^{14}\text{C}$ values (i.e., older ¹⁴C ages) [Gordon and Goni, 2003]. The explanation for such kind of distributions is the original terrestrial OM have old ages due to the dominated soil OM derived material [Wu *et al.*, 2007]. Although we did detect the elder ages of outer shelf samples, the modification of local primary production or other sources (e.g., benthic OM) makes the bulk ages look younger than we expect.

[27] The lignin phenols parameters also illustrated some differences among the four regions, especially for lignin concentrations (I > II > IV > III). A previous investigation into the composition of organic matter carried by the Changjiang suggested that soil-organic material was a dominant component [Wu *et al.*, 2007]. Most Changjiang material is distributed close to the Changjiang Delta although some material can be dispersed as far south as 26°N by the winter-intensified China Coastal Current [Xu *et al.*, 2009]. As a consequence of long-distance transport involving multiple sedimentation-remobilization episodes, coupled with degradation processes and mixing with other sources, there are no distinct relationships between molecular indicators of vascular plant input (e.g., $\Delta 8$) with bulk isotopic

properties (e.g., $\delta^{13}\text{C}_{\text{OC}}$), or with LPVI or Ad/Al ratios with %clay for Reg. II samples (Figure 4).

[28] Atmospheric deposition of lignin phenols over East China Sea might be significant especially considering increasing dust storm aerosol from northwest China can be transported over the coastal regions to the open oceans [Zhang *et al.*, 2010]. Organic aerosols may contain plant materials (fragments; pollens), biomass burning, road, soil, and mineral dust, etc., which will be potential sources for lignin phenols. Previous study showed that the compositions of lignin phenols in Houston aerosols likely are derived from woody tissues of flowering plants, with minor influence from soft tissues and pollens [Shakya *et al.*, 2011]. From our data, there is only minor shifting in C/V and LPVI ratios from inner to outer shelf. It is thus difficult to elucidate the potential contribution of organic aerosols derived lignin in two contrasting environment.

[29] The bulk properties (e.g., %OC, grain size, $\Delta^{14}\text{C}_{\text{OC}}$) reveal that the outer shelf sediments are heterogeneous compared with inner shelf samples, especially Reg. III (Figures 2 and 3). Reg. III is shallow water and strong dynamic region, which is characterized as highly heterogeneous and lowest lignin phenols and fresher ages compared to Reg. IV (typical relict region). It has previously been reported that relict sandy sediments can efficiently trap fine particulate matter [Bacon *et al.*, 1994]. Organic matter inputs from benthic macroalgae, as observed for Mid-Atlantic Bight shelf relict sediments, represent a possible additional source of organic matter to these sediments [Rusch *et al.*, 2003]. The observed relationship between $\Delta 8$ and stable carbon isotopes (Figure 3) suggests a simple two-end-member mixing, involving a ¹³C-enriched end-member ($\delta^{13}\text{C} -16.0\text{‰}$) with an isotopic composition close to that reported for benthic macroalgae [-17.0‰ ; France, 1995]. Using a $\delta^{13}\text{C}$ value (-24‰) for paleosol-derived organic matter for the other end-member, we made the estimation of the contribution from these two sources (Table 2). Higher contribution of benthic macroalgae was observed in Reg. III than Reg. IV except M6 station. Such kind of distribution is consistent with the Chl *a* concentrations in the sediments [Zhu, 2007].

[30] As illustrated by the variation of S/V and C/V ratios between the two major regions (inner and outer shelves) (Table 2), S/V and C/V ratios increased from the coast to the shelf, which might suggest increasing proportions of angiosperm leaf- and/or grass-derived lignin in offshore sediments as other continental shelf systems [e.g., Louisiana shelf

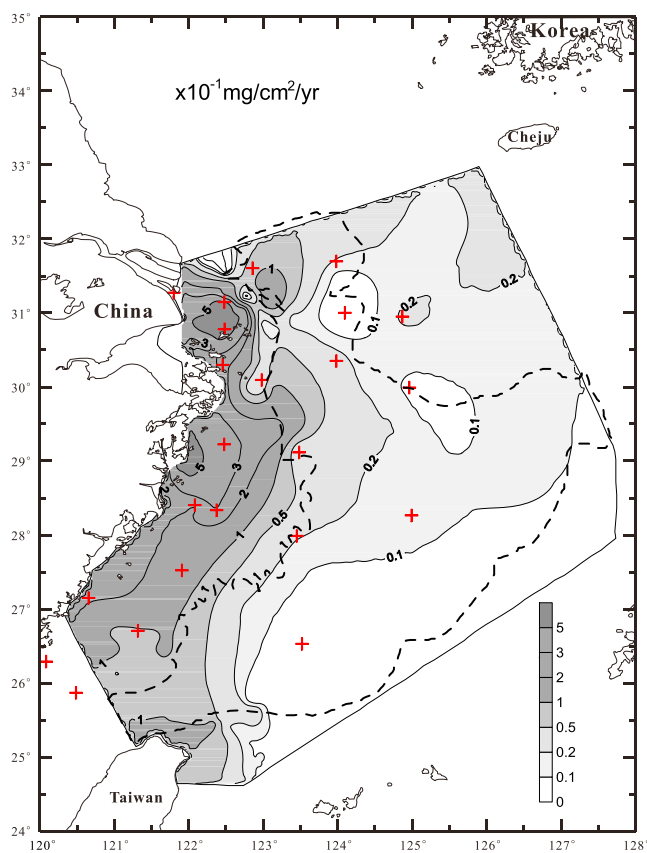


Figure 5. Spatial distribution of lignin flux over the ECS.

- Goñi *et al.*, 1998; NW Iberian margin, Schmidt *et al.*, 2010]. The preferential offshore transport of lignin derived from grass and leaf tissue may reflect the impact hydrodynamic sorting processes. However, it was reported that sorption and desorption also can modify the ratio of S/N and C/N, especially sorption with variable minerals [Hernes *et al.*, 2007].

[31] Based on sedimentary lignin phenol concentrations and sediment mass accumulation rates over the study region, we estimate a total burial flux of lignin phenols in the ECS of 4.2×10^4 t/yr [Yang *et al.*, 2008; Figure 5]. Lignin accumulation rates (10^{-1} mg lignin $\text{cm}^{-2} \text{yr}^{-1}$) (estimated by mass accumulated rate \times lignin concentration) were highest in nearshore stations and decreased dramatically in outer shelf. The highest lignin accumulation rates were observed out of the Changjiang Estuary, overlapped with muddy area, which is regarded as the first sink of Changjiang sediment [Liu *et al.*, 2011]. Reg. II's material also mainly derived from same source due to retransportation and dispersal. There is a good declined gradient of lignin accumulation rate from Reg. II to Reg. IV. But the distribution in Reg. III is more heterogeneous. Tesi *et al.* [2008] also pointed out winnowing and resuspension of fine particle during strong wave energy substantially modified the deposit, especially in shallow stations (such as Reg. I and III).

[32] Calculated terrestrial organic matter (OM_{TERR}) burial efficiencies are compared with published data in Table 3. Burial efficiency is estimated as the OC:SA value of the marine sediment sample multiplied by the proportion of OM_{TERR} relative to total OC pool divided by the OC:SA of riverine suspended particles (0.5 ± 0.1 mg C m^{-2}). The

resulting estimation of OM_{TERR} burial efficiency from inner shelf (24.7%) is slightly higher than the global value for river-dominated margins (22%) estimated by Burdige [2005] or for margins in general by reported by Schlunz and Schneider [2000; 20%], but lower than that estimated by Deng *et al.* [2006; 38%]. The estimation of Deng and coworkers derived from a stable carbon isotope mixing model, which may carry greater uncertainty due to the variability in organic matter sources and heterogeneity in organic matter composition over the entire ECS. The greater burial efficiency observed in ECS is consistent with level of terrestrial input and high sediment deposition rates. Nevertheless, the estimated efficiency is lower than that calculated for the Amazon shelf [30%, Aller, 1998], and much lower than that of the Bay of Bengal Bay [$\sim 100\%$, Galy *et al.*, 2007], which are characterized by even higher sediment accumulation rates. This result is in agreement with the finding in Peru Margin that postdepositional winnowing lowered the inventory of OC in the sediment [Bergamaschi *et al.*, 1997].

4.2. Factors Influencing the Accumulation of Organic Matter in the Shelf

[33] Identifying the environmental factors which control organic matter content and composition in dynamic coastal systems is challenging, but essential for interpreting the sedimentary record of organic carbon supply and burial [Lange *et al.*, 2010], and for predicting how these systems may respond, and contribute, to future carbon cycle perturbations. Various factors have been implicated in relation to organic matter preservation in marine environment, including primary productivity in the overlying column, bottom water oxygen concentration, local sedimentation rate, hydrodynamic sorting, winnowing, and mineral surface area [e.g., Bergamaschi *et al.*, 1997; Keil *et al.*, 1997; Schulte *et al.*, 2000]. We explore some of these factors in the context of the ECS.

[34] The marked difference of sediment rate between inner and outer shelves of the ECS [Deng *et al.*, 2006] could be at least partly responsible for the different organic geochemical characteristics (e.g., lignin phenol compositions) in the two regions. In the shallow inner shelf region, biogeochemical characteristics may more easily be controlled by physical (i.e., high turbulence during storm events) or biological (e.g., bioturbation) mixing processes [Zonneveld *et al.*, 2010]. Also, periodic hypoxia events have been reported to occur in shallow bottom waters (water depth < 50 m) between 28°N and 32°N [Zhang *et al.*, 2010]; however, because of the ephemeral nature of this phenomenon there is no corresponding organic carbon maximum in surface sediments underlying the hypoxic zone. Primary production is recognized as impor-

Table 3. Estimation of Terrestrial Organic Matter Burial Rates in the ECS

| Sediment Type | OC:SA (mg C m^{-2}) | TOM/ Σ OM (%) | TOM Burial Efficiency (%) |
|-------------------------------------|----------------------------------|-------------------------|------------------------------|
| Inner shelf sediment | 0.33 ± 0.11 | 37 ± 19 | 24.7 ± 4.5 |
| Outer shelf sediment | 0.36 ± 0.16 | 14 ± 8 | 10.7 ± 2.6 |
| River-dominated margin ^a | 0.25 ± 0.08 | 67 ± 24 | 22 ± 5 |
| Nondeltaic margin ^a | 0.86 ± 0.11 | 16 ± 4 | 17 ± 4 |

^aData from Burdige [2005].

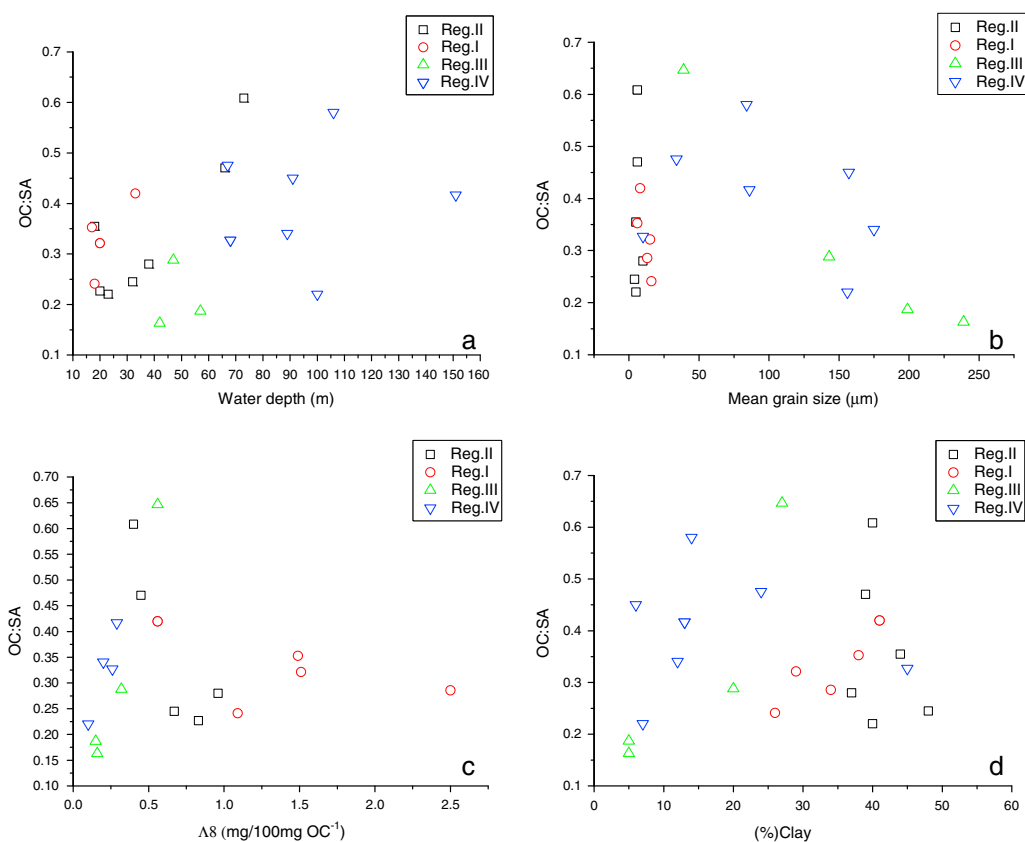


Figure 6. (a) Water depth, (b) mean grain size, (c) $\Delta 8$ (normalized to 100 mg of organic carbon), and (d) %clay versus the ratio of OC: surface area distribution in the surface sediment samples in the ECS.

tant factor to controlling the quantity of OM buried in marine sediment [Schulte *et al.*, 2000]. Higher primary productivity is generally observed near to the Changjiang Estuary as a consequence of the huge riverine supply of nutrients [Zhang, 1999]; however, this pattern does not fit the spatial distribution of organic matter in inner shelf sediments. More broadly, annual primary production is reported to be higher in the northern than the southern ECS [Tan and Shi, 2006]; however, there is no significant north-to-south gradient in organic carbon contents of outer shelf sediments. But we did observe fresher ages of organic matter in Reg. III. Resuspension and redistribution of fresh organic carbon, including lateral transport to the outer shelf, could be partly responsible for this discrepancy [Zhu *et al.*, 2011a]. Overall, however, the burial efficiency of marine OC is low, and similar in magnitude to the global average of 0.8% reported by Berger *et al.* [1989].

[35] A recent radioisotope tracer study revealed that fine-grained riverine suspended particles can be dispersed both along the coastlines from north to south and transported from nearshore to offshore regions adjacent to the Changjiang Estuary via the influence of local current systems [Du *et al.*, 2010]. Such differential transport processes have been shown to be an important mechanism for dispersal of terrestrially derived organic matter over river-dominated margins [Keil *et al.*, 1997; Bianchi *et al.*, 2002]. However, modification of lignin phenols along the dispersal was suggested from the variation of S/V, C/V, and Ad/Al ratio in Reg. II (Figure 4).

[36] Thus, hydrodynamic characteristics such as grain size distribution exert a strong influence on OC concentration and

composition of ECS sediments. Specific mineral surface area does not show an increasing from offshore to the shelf as the Mexico Bay [Gordon and Goni, 2004]. However, OC:SA ratios showed increase with water depth, especially in Reg. I and II (Figure 6a). In particular, for outer shelf samples, coarser-grained sediments generally exhibit lower OC:SA ratios (Figure 6b). This correlation between OC and mineral surface area suggests a role for mineral surfaces as a control on sedimentary OC content in this region [Mayer, 1994]. We also notice that OC:SA has correlations with %clay in Reg. I and III but scattered in other two regions (Figure 6c). This may be linked to the dispersal of Changjiang diluted water especially in summer period. The contrasting relationships between lignin phenol content and mineral surface area for Inner and Outer shelf sediments suggest that different processes influence OC composition in these two regions (Figure 6d). For Outer shelf sediments, the significant correlation between these parameters is consistent with an association of lignin residues with fine-grained minerals, whereas inner shelf sediments showed negative correlation except few higher lignin phenol samples from the estuary region. Besides hydrodynamic sorting, sorption is also one of important factors to control organic matter preservation in marine sediment [Keil *et al.*, 1994; Mayer, 1994], especially for lignin phenols. In Figure 4, the coherent variation of Ad/Al ratio with %clay in Reg. I and IV may suggest the selective sorption of acid than aldehyde in clay fraction. The big shift of Ad/Al ratio from inner shelf to outer shelf may result from combination of selective sorption and diagenetic process,

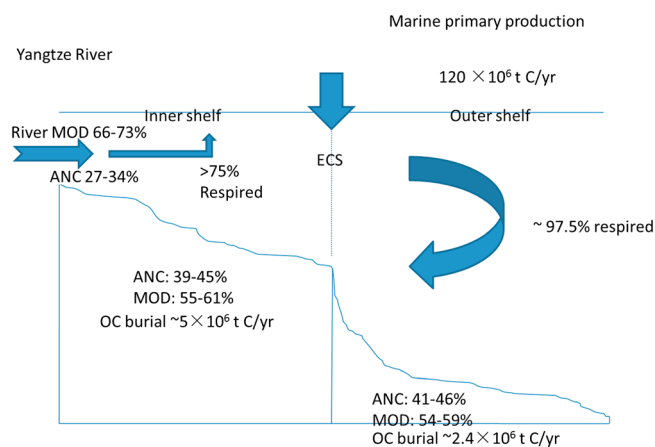


Figure 7. The estimated budget of modern and ancient carbon in inner and outer shelf of the ECS. Data derived from Deng *et al.* [2006], Wu *et al.* [2007], Liu *et al.* [2011], and this study.

which might explain the contrasting result of Ad/AI with grain size compared to Washington coast [Keil *et al.*, 1998].

4.3. ^{14}C Contents of OC in Shelf Sediment and its Implications

[37] The ^{14}C content of OC in surface sediments can provide important constraints on the sources of OM and on sedimentological processes influencing organic carbon burial preserved on continental margins [Griffith *et al.*, 2010]. Plankton in modern-day surface seawater displays enriched $\Delta^{14}\text{C}$ values (55–200‰). $\Delta^{14}\text{C}$ values of suspended or settling particulate organic matter in the surface waters of the northwestern Pacific Ocean have been reported to range from -59 to $+37$ ‰ [Honda *et al.*, 2000]. Assuming that surface ocean primary productivity represents the dominant source of OC to the sediments, and that sediment accumulation rates are rapid and dominate over sediment mixing processes, the ^{14}C content of surface sedimentary OC should reflect that of this sinking particulate organic matter. In this study, OC in surface sediments is consistently depleted in ^{14}C ($\Delta^{14}\text{C} \leq -300$ ‰), implying significant contributions pre-aged organic matter. Contributions of aged carbon of continental origin to surface shelf sediments have been observed in the other river-dominated margins [Gordon and Goni, 2003; Goni *et al.*, 2005]. On the inner shelf of the ECS, where sediments are strongly influenced by the Changjiang, continentally derived (terrestrial/fossil/anthropogenic) inputs of aged carbon (e.g., soils, petrogenic C from rock erosion, black carbon, and other products of fossil fuel utilization) likely represent the major source of pre-aged organic matter [Griffith *et al.*, 2010]. The $\Delta^{14}\text{C}$ values from inner shelf (survey during 2004–2006) were more negative than the data of other study (survey during 2009 and 2010) from same region which might be due to spatial and temporal variability [Li *et al.*, 2012], or might be related to decreasing sediment load from Changjiang. Indeed, $\Delta^{14}\text{C}$ values of Inner shelf were similar to those of particles and sediments recovered from mainstem of the Changjiang (in the range of -350 – -250 ‰). On the Outer shelf, $\Delta^{14}\text{C}$ values are also low despite the lack of strong terrigenous influence (as exemplified by low abundances of lignin marker

compounds; Table 2 and Figure 2). For these locations, the older ^{14}C ages may reflect winnowing, redistribution, and current-driven transport of marine and terrestrial OC via bottom currents [Griffith *et al.*, 2010; Zhu *et al.*, 2011a]. Bottom nepheloid layers, which have been observed to extend from off the Changjiang Estuary to the shelf break [Peng and Hu, 1999], can be significant in sediment transfer across the shelf, especially in winter.

[38] In addition to sediment redistribution phenomena, small contributions of fossil fuel-derived black carbon can significantly affect bulk OC ^{14}C values [Dickens *et al.*, 2004]. Available data indicate that black carbon accumulates on the inner shelf and sandy sediments of the outer shelf as high as 20–50%, which is considerably higher compared to other shelf system ($< 10\%$) [Huang *et al.*, 2012; Li *et al.*, 2012]. Graphitic black carbon dominated in outer shelf sediments with content increasing with grain size. At present it is not possible to distinguish the respective influence of sediment redistribution versus fossil carbon inputs on OC ^{14}C ages. Compound-specific radiocarbon measurements may be of use in this regard. However, in the absence of this information, we can derive minimum estimates for the portion of aged carbon that is accumulating in ECS sediments based on simple isotopic mass balance considerations [Goni *et al.*, 2005; Drenzek *et al.*, 2007]. The fraction of ancient OC (f_{ANC}) and modern OC ($f_{\text{MOD}} = 1 - f_{\text{ANC}}$) of each sample can be estimated from the $\Delta^{14}\text{C}$ values according to:

$$\Delta^{14}\text{C} = [f_{\text{ANC}} \times \Delta^{14}\text{C}_{\text{ANC}}] + [f_{\text{MOD}} \times \Delta^{14}\text{C}_{\text{MOD}}] \quad (1)$$

where $\Delta^{14}\text{C}_{\text{ANC}}$ and $\Delta^{14}\text{C}_{\text{MOD}}$ represent the ^{14}C values of modern and ancient end-members. Assuming $\Delta^{14}\text{C}_{\text{ANC}} = -1000$ ‰ and $\Delta^{14}\text{C}_{\text{MOD}} = 0 \pm 50$ ‰ for the isotopic mass calculations [Goni *et al.*, 2005; Drenzek *et al.*, 2007] yields results indicating that ancient organic matter constitutes an important component of the OC in all samples (Table 1). With the exception of M6 and M12, inner shelf and outer shelf samples contain comparable amounts of OC_{ANC} (35–45 % of OC). The low $\delta^{13}\text{C}$ values of the two stations exhibiting anomalously low ^{14}C contents may indicate contributions from pre-aged soil-derived OM. The fraction of OC_{ANC} observed in ECS is significantly lower than Canadian Beaufort Shelf but higher than the northern Gulf of Mexico [Goni *et al.*, 2005].

[39] Using a carbon budget for the ECS developed by Deng *et al.* [2006], combined with $\Delta^{14}\text{C}$ data stemming from this study and from Li *et al.* [2012], we derive a budget for modern and ancient organic carbon in inner shelf and outer shelf sediments of the ECS (Figure 7). The load of OC from the Changjiang and supplied via marine primary production was derived from previous studies [Wu *et al.*, 2007]. Modern organic carbon preservation in the ECS estimated by f_{MOD} and OC burial rates indicates substantial loss of modern organic matter ($\sim 97.5\%$ respired) prior to burial, similar to the findings of Goni *et al.* [2005] for the Beaufort Shelf. This modern organic carbon accounts for over 55% of the total carbon burial in ECS. For terrestrial OM, we calculate a burial rate based on the terrestrial burial efficiency ($\sim 25\%$ in this study) plus terrestrial load supplied by the Changjiang (4.5×10^6 t/yr, long-term average). The resulting estimate of terrestrial OC burial rate, 1.13×10^6 t/yr, is very close to that

estimated for the inner shelf by *Deng et al.* [2006]. However, the burial of OC_{ANC} in the inner shelf ($\sim 2 \times 10^6$ t/yr) is higher than annual input from the Changjiang ($\sim 1.5 \times 10^6$ t/yr). This may suggest either an overestimation of OC_{ANC} in the inner shelf (e.g., organic matter aging during sediment resuspension and reworking), or additional potential sources of ancient OM, such as from atmospheric deposition, the small rivers in ZJ and FJ region, the Taiwan Islands [e.g., *Hilton et al.*, 2011], from the abandoned Yellow River Delta to the north. This is the first result about the estimation of the burial of ancient OM versus modern OM in ECS. Due to global change (e.g., decreasing in riverine sediment load and frequent red tides events, etc.), the composition of riverine OM may increase labile fraction or higher primary production is observed in the shelf, at the same time, erosion in the delta will accelerate to release older materials from sediments, which will influence regional/global biogeochemical cycles and the shelf's ability to sequester atmospheric CO₂. Therefore, there remains considerable uncertainty in our ability to adequately quantify carbon exchange from land to the coastal ocean.

5. Conclusions

[40] Spatial variation in the supply, dispersal, and decomposition of terrestrial and marine materials in the contemporary ECS manifests itself as a complex mosaic of organic matter contents and compositions in underlying surface sediments that are expressed at both the bulk and molecular level. Terrestrial residues constitute the dominant component of OM in inner shelf sediments whereas marine OM is important in outer shelf deposits. The net loss of sedimentary OM during along-shore and across-shelf transport is dominated by the oxidation of terrestrial OM, including loss of pre-aged organic materials. Distinguished higher BC content may modify the original carbon ages significantly.

[41] Climate change and anthropogenic disturbances in the Changjiang basin have potential to dramatically alter the sources, fluxes, and composition of organic carbon delivered to the ECS. Indeed, although terrestrial material from the Changjiang dominates OC in inner shelf, sediment supply has significantly decreased recently as a consequence of dam emplacement on the river main stem [*Yang et al.*, 2007], with major potential implications for regional carbon and sediment budgets. Correspondingly, small river systems along ZJ-FJ coastline and sediment supply from Taiwan Island may thus exert an increasing influence on OC delivery and fate in the ECS. Moreover, records indicate the ZJ-FJ coast has experienced more frequent storms during past decade [*Shao*, 2005], which may further increase sediment loads from small mountainous river systems in the coastal range [*Liu et al.*, 2008]. Overall, the balance of depositional and erosional processes in the ECS, and its impact on carbon cycling within this major continental shelf sea system, remains uncertain and warrants further and ongoing study.

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