**Diverse origins and pre-depositional histories of organic matter in contemporary Chinese
marginal sea sediments**

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ABSTRACT

Marginal seas are estimated to account for up to 90% of organic carbon (OC) burial in marine sediments, and thus play an important role in global carbon cycle. However, comprehensive assessments of carbon budgets for marginal sea systems are challenging due to their inherent complexity, with spatial and temporal variability in carbon inputs and dispersal processes. We

examine the Bohai Sea and Yellow Sea (BS-YS) in order to further our understanding of sedimentary OC delivery, translocation and accumulation in a shallow marginal sea system. Bulk properties and the content and isotopic compositions ($\Delta^{14}\text{C}$, $\delta^{13}\text{C}$) of source-specific plant wax *n*-alkyl lipid biomarkers were determined for a suite of surficial sediment samples. Variable $\delta^{13}\text{C}$ values (-25.1 to -28.5‰) and contemporary radiocarbon ages of short-chain *n*-fatty acids (FAs; C_{16} , C_{18}) reflect modern autochthonous marine and/or fresh terrestrial plant input. In contrast, extremely depleted $\Delta^{14}\text{C}$ values (-932 to -979‰) of short-chain *n*-alkanes (C_{16} , C_{18}) suggest a predominant input from sedimentary rocks (petrogenic OC) or petroleum. Abundance-weighted average $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values of long-chain leaf wax lipids ($\text{C}_{26+28+30}$ *n*-FAs, $\text{C}_{24+26+28}$ *n*-alkanols, $\text{C}_{27+29+31}$ *n*-alkanes) are -29.1 ± 1.1 to $-30.2 \pm 0.3\text{‰}$, and -286 ± 150 to $-442 \pm 119\text{‰}$, respectively, illustrating that terrestrial OC delivery is dominated by pre-aged (~ 3000 – 5000 ^{14}C yrs) C_3 vegetation sources.

A coupled carbon-isotopic mixing model, based on the bulk and compound-specific biomarker $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values, is used to partition the BS-YS sedimentary OC into three components that reflect both origins and transport processes. For all sampling sites, 31–64% is modern/contemporary OC, 24–49% is pre-aged terrestrial OC, and 7–26% is fossil OC, the latter likely derived from both physical erosion of ancient sedimentary rocks and fossil fuel sources. Pre-aged soil OC is most prominent in front of the modern and old Huanghe (Yellow River) delta (48% and 49%), and fossil OC is most significant north of the old Huanghe mouth (26%). Significant pre-aged soil contributions (33%) are also evident for sites further offshore, where transport and deposition of eolian dust supply may be important. For the three major deposition areas of the BS-YS system (Bohai Basin, sub-aqueous Huanghe delta and central south YS basin), we estimate that about 3.02 Mt/yr of refractory, plant-derived pre-aged soil OC and 0.98 Mt/yr of ^{14}C -depleted fossil OC accumulates in surface sediments, corresponding to 35% and 11% of sediment TOC, respectively. Compared with estimates for fluxes from corresponding sources, the burial efficiency is close to 100% for pre-aged soil OC and 70% for fossil OC, implying efficient OC burial in delta and shelf environments. Re-burial of these two pools of terrigenous OC only affects carbon cycling on millennial and longer timescales respectively, and exerts little influence on the modern carbon cycle (<100 yr). Carbon isotopic compositions of source specific biomarkers

are a useful tool not only for constraining OC sources and transport vectors, but also for delineating their impact on the contemporary carbon cycling in marginal sea systems.

1. INTRODUCTION

OC burial in marine and terrestrial sedimentary basins comprises the second largest long-term sink for atmospheric CO₂ on geological time-scales (Burdige, 2005; Galy et al., 2007). Due to high biological productivity, supply of allochthonous materials and sedimentation rates, marginal seas are regionally and globally significant sites of both carbon sequestration and remineralization. Despite occupying less than 10% of the global ocean area, continental shelves, including marginal seas, have been proposed to account for up to 90% of organic matter burial in the modern ocean (Hedges and Keil, 1995) and 20% of the net ocean CO₂ uptake (2.0±0.6 Gt C) (Battle et al., 2000). However, considerable uncertainties persist regarding the nature and magnitude of carbon burial because marginal seas reside at the land-ocean continuum, and are inherently dynamic and spatially heterogeneous systems. OC supplied from adjacent continents via fluvial and eolian processes is augmented by marine OC produced in overlying waters, (Bianchi, 2011; Blair and Aller, 2012), resulting in diverse fluxes, reactivities and ages of organic matter deposited in marginal sea sediments. The extent to which OC burial constitutes a net carbon source or sink, strongly depends upon the origin of sedimentary OC.

OC in marginal sea sediments can be considered to comprise of three components: marine OC, terrestrial biospheric OC, and petrogenic OC from erosion of sedimentary rocks or from anthropogenic activities (Blair and Aller, 2012; Drenzek et al., 2007; Galy et al., 2008; Goñi et al., 2005; Kusch et al., 2010b). These different OC pools can influence the carbon cycle over varying timescales. Recent marine and terrestrial OC can be reactive and hence fully or partially degraded during transport and sedimentation processes, resulting in recycling of carbon to the atmosphere (Aller and Blair, 2004; Bianchi, 2011). On the other hand, burial of this recently synthesized “biospheric” OC in sediments reflects sequestration of modern C. Temporary storage or protracted translocation of biospheric OC within and between intermediate reservoirs (e.g., soils, floodplain deposits, mobile mudbelts) can influence remineralization/burial timescales. In contrast, recycling and reburial of “fossil OC” without significant alteration exerts a minimal net influence on

atmospheric CO₂ (Galy et al., 2008; Galy and Eglinton, 2011). Anthropogenic activity has also contributed to the burial of fossil OC in marine sediments (Eglinton et al., 1997; Hedges et al., 1997; Pearson and Eglinton, 2000). Overall, the difference between oxidation and burial of the biospheric OC governs the net sink of atmospheric CO₂ over shorter timescales (decades to thousands years) whereas exhumation and re-burial of fossil OC has no net effect on atmospheric CO₂ (Galy et al., 2008; Galy and Eglinton, 2011).

Terrestrial OC discharged by large rivers (e.g., Brahmaputra-Ganga, Huanghe) is usually thought to be relatively refractory as a consequence of long-term storage and exposure to biological degradation in continental reservoirs (Eglinton et al., 1996; Feng et al., 2013a; Galy and Eglinton, 2011; Kusch et al., 2010b). In contrast, extensive loss of terrestrial OC in some estuarine and deltaic environments (Aller et al., 2008; Bianchi, 2011; Hedges et al., 1997; Keil et al., 1997) suggests that terrestrial OC can be reactive. In addition, dynamics associated with wind-borne and fluvial transport of terrestrial OC are quite distinct, with significant implications for the physicochemical characteristics of terrestrial OC supplied to the adjacent marginal seas. Shallow marginal seas are characterized by extensive horizontal material transport by coastal currents, storm and tidal activity, each affecting the fate of terrestrial and marine OC.

Association with mineral surfaces exerts strong influence not only on the reactivity of OC (Blair and Aller, 2012; Bouchez et al., 2014; Keil et al., 1997; Meyers, 1994), but also on its susceptibility to redistribution (Goñi et al., 1998). OC content generally positively correlates with mineral-specific surface area (SA), and is inferred to reflect organic matter association with, and protection by, fine-grained minerals (Blair and Aller, 2012; Goni et al., 2008; Keil et al., 1997). Moreover, hydrodynamic particle sorting processes in the benthic boundary layer promote resuspension, lateral transport and redistribution of OC (Kusch et al., 2010a; Xu et al., 2012). These complex patterns of OC supply and dispersal render it challenging to assess controls on the nature and burial efficiency of organic matter in marginal sea sediments (Kusch et al., 2010b; Ohkouchi et al., 1997). Nevertheless, constraining the various sources and ages of sedimentary OC components is essential for improved understanding of the various sedimentological and biogeochemical processes.

The BS-YS is a large river-influenced marginal sea system and is considered an important

sink for atmospheric CO₂ (Chen and Borges, 2009; Oh et al., 2000). Only ~5% of the sediment derives from marine production or from atmospheric deposition (Li et al., 2012; Zhou et al., 2014). In contrast, roughly 95% of sediment supply to this region is by the modern (ave., 930 Mt/yr for the period 1900–2007) Huanghe and via re-distribution of old Huanghe delta material (ave., 792 Mt/yr) over the past 100 years (Peng et al., 2010; Zhou et al., 2014). However, sediment export of the modern Huanghe to the ocean has dramatically decreased since the 1960s due to human activities (i.e., mainly dam construction and soil conservation) and enhanced drought, with an amount of 151 Mt/yr from 2000 to 2007 (Peng et al., 2010). The abundance of old/refractory terrestrial OC (Zhan et al., 2013) and high clastic sediment loads are considered to be responsible for enhanced carbon burial efficiencies in the BS-YS (Keil et al., 1997), representing a potentially important sink for terrestrial and fossil OC (Tao et al., 2015).

Several approaches have been used to examine the spatial distribution of marine and terrestrial OC in BS-YS sediments. These indicate that terrestrial OC burial predominantly occurs in the modern/old Huanghe delta and subaqueous delta systems whereas marine OC burial predominates in the central basin depocenters (Cai et al., 2003; Xing et al., 2014). The detection of combustion-derived (pyrogenic) PAHs also suggests that eolian OC is another source for sedimentary OC in the center basin and more distal regions (Wu et al., 2001). Sedimentological and hydrodynamic processes also influence OC spatial distribution, with enhanced accumulation of OC occurring in fine-grained depocenters in the BS-YS basin (Xing et al., 2011; Yang and Youn, 2007), and an inverse correlation has also been observed between OC content and sediment grain size in Bohai Sea sediments (Hu et al., 2009). Compound-specific carbon isotopic compositions of source-specific biomarkers can provide direct constraints on end-member values of source materials in order to assess OC inputs (Eglinton et al., 1997; Freeman et al., 1990), and distinguish between pre-aged terrestrial and fossil OC. This approach has proven useful in identifying diverse carbon sources in marine sediments (Drenzek et al., 2007; Eglinton et al., 1997; Feng et al., 2013a). However, it has not yet been applied in the Chinese marginal seas, even though Huanghe suspended sediment studies reveal that aged OC is the dominant component of river OC supplied to the BS-YS system (Tao et al., 2015; Wang et al., 2012).

In order to develop a more detailed assessment of the composition, source and age of organic

matter delivered to the BS-YS, we examine compound-specific ^{13}C and ^{14}C characteristics of source-specific biomarkers in a suite of surface sediment samples. Spatial variations in the isotopic characteristics of these biomarkers are used to evaluate the influence of both transport and diagenetic processes as controlling factors on OC burial. A coupled isotopic mass balance model based on biomarker ^{13}C and ^{14}C is applied to quantify the relative proportion of modern, pre-aged soil and fossil OC in surface sediments of this marginal sea system.

2. MATERIAL AND METHODS

2.1. Study area

The BS-YS is bordered by China and the Korean peninsula, with a maximum depth of 103 m and covers an area of approximately $457 \times 10^3 \text{ km}^2$ (Fig. 1). The sedimentology, mineralogy and geochemistry of BS-YS sediments have been intensively studied (Alexander et al., 1991; Shi et al., 2003; Yang and Youn, 2007; Yang and Liu, 2007). Sedimentation rates (Alexander et al., 1991) and mineral grain size distributions (Hu et al., 2009; Shi et al., 2003) have distinct spatial patterns reflecting both the source of materials and hydrodynamic regimes controlling settling and erosion. Overall, the content of fine-grained material (clay, $<4 \mu\text{m}$) and sedimentation rate show opposite spatial patterns. Lower clay contents and higher sedimentation rates near the coasts reflect direct inputs of coarse particles from the adjacent landmasses, and winnowing by the highly energetic coastal currents. In contrast, the YS is a deposition center under the control of cyclonic circulation and a cold water gyre that focus sediments from surrounding areas (Shi et al., 2002). Higher clay contents and lower sedimentation rates in the basin center are caused by the general counterclockwise circulation in the YS which limits the supply of coarse materials to the basin center, as well as by quiescent conditions that favor deposition of fine-grained sediments. Over the past 100 years, the primary sources of sediment to the YS-BS are the modern Huanghe and the old Huanghe delta (Li et al., 2012; Milliman and Syvitski, 1992; Zhou et al., 2014). The modern Huanghe-derived clayey silt sediment is directly discharged into the western Bohai Sea with an accumulation rate of 557 Mt/yr. Coastal currents transport a portion of the fine-grained sediment from the Huanghe is transported eastward via the Bohai Strait to the YS, building the Shandong subaqueous delta at an accumulation rate averaging $\sim 70 \text{ Mt/yr}$ (Liu et al., 2004; Yang and Liu,

2007). Between 1128 and 1855 CE, the Huanghe emptied into the south YS and formed a huge delta along the Jiangsu coast, whereas the modern, anthropogenically-influenced Huanghe river mouth is at (or near) the location of the mouth prior to 1128 CE, and for most of the Holocene. The old Huanghe delta was abandoned after the course of the river was changed to its modern-day point of discharge in the BS in 1855 CE. Subsequently, wave and tidal activity and coastal currents have continued to erode the subaerial and subaqueous old Huanghe delta. Roughly 50% of the sediment deposited at the center of the south YS and south of Cheju Island derives from the old Huanghe delta (Zhou et al., 2014), with the rest mainly from the modern Huanghe and wind-derived dust materials.

2.2. Sampling and bulk analysis

Surface (0–5 cm) and near-surface (0–30 cm) sediments from 9 locations in the BS-YS region were collected respectively using a box corer (during the 973 Cruise by the Ocean University of China, OUC, in April of 2012), and a grab sampler (during a cruise by the Qingdao Institute of Marine Geology in December of 2008). In addition, a sandy sediment sample (OYR0) was obtained from the old Huanghe subaerial delta (Fig.1, Table S1). All samples were stored frozen in aluminum foil and subsequently freeze-dried.

An aliquot of bulk, homogenized sediment sample was processed for elemental and bulk isotopic analysis (^{13}C and ^{14}C). After removal of inorganic carbon with 12 N HCl fumigation (Komada et al., 2008), the TOC, TN and bulk $\delta^{13}\text{C}$ values ($\delta^{13}\text{C}_{\text{TOC}}$) were determined at the Stable Isotope Facility within the Climate Geology group at ETH with a standard deviation of 0.002 wt % ($n = 6$) and 0.1‰ ($n = 6$), respectively; whereas bulk $\Delta^{14}\text{C}$ values ($\Delta^{14}\text{C}_{\text{TOC}}$) were measured on a gas-ion source MICADAS accelerator mass spectrometry (AMS) system at the Laboratory for Ion Beam Physics, ETH Zürich. Another aliquot was used for sedimentological characterization including mineral-specific surface area (SA) and grain size analysis. After removing organic matter (300 °C, 12 h), SA and mean grain size were measured by a 5-point BET method on a NOVA 4000 surface area analyzer (Quantachrome) and on a Malvern laser diffraction grain sizer, respectively (Keil et al., 1997; Mayer, 1994).

2.3. Contents and carbon isotope analysis of *n*-alkyl lipid biomarkers

The detailed methods for extraction, purification and isolation of target biomarkers have been provided in Tao et al. (2015). About 40–150 g freeze-dried and homogenized sediments were extracted and separated to obtain three classes of target *n*-alkyl lipid biomarkers including *n*-fatty acid methyl esters (*n*-FAMES), *n*-alkanes and *n*-alkanol acetates. To check purity and for quantification, a small aliquot (typically 1/500) of each fractions was injected on a HP 7890 series gas chromatograph (GC) equipped with FID and a VF-1ms capillary column (30 m × 0.25 mm i.d., 0.25 µm film thickness). An aliquot (<5%) of each fraction was kept for stable carbon isotopic analysis. The remaining individual compounds, or groups of compounds (*n*-alkanes, *n*-alkanol acetates and *n*-FAMES) were further purified and isolated by preparative capillary gas chromatography (PCGC) prior to compound-specific ¹⁴C analysis. Using repeated injections, 20 to 100 µg of carbon of individual lipid target compounds was collected using a Gerstel preparative fraction collector (Eglinton et al., 1996).

Aliquots of the *n*-FAME and *n*-alkane fractions obtained from each sample were measured in duplicate by GC-isotope ratio mass spectrometry (GC-IRMS), a HP 6890 GC coupled with a Thermo Delta-V system (Tao et al., 2015). The δ¹³C values of *n*-FAMES were subsequently corrected for the contribution of the added methyl carbon (−42.56±0.41‰) in order to derive *n*-FA δ¹³C values and their respective errors. Uncertainty of δ¹³C values ranged from 0.1‰ to 1.9‰, averaging 0.3‰ for *n*-alkanes and 0.2‰ for *n*-FAs. Results are given in ‰ relative to VPDB.

Decarbonated sediment samples, as well as corresponding *n*-alkyl lipids and processed procedural blanks were analyzed for radiocarbon content on a gas-ion source MICADAS accelerator mass spectrometry (AMS) system at the Laboratory for Ion Beam Physics, ETH Zürich. All radiocarbon data were expressed as Δ¹⁴C and corresponding radiocarbon year (before 1950 AD). All the Δ¹⁴C of *n*-FAMES and *n*-alkanol acetates were corrected for the derivative methyl and acetyl carbon, respectively, to derive inherent Δ¹⁴C values and associated errors for underivatized compounds. In our study, the analytical uncertainty for ¹⁴C analysis of *n*-alkyl lipid compounds isolated with the PCGC procedure ranges from 6 to 40‰ (ave., 12‰) after propagation of error. This offset is comparable to that reported by Hou et al. (2010) and Feng et al. (2013a) (details provided in Tao et al., 2015).

3. RESULTS

3.1. Bulk characteristics

TOC and TN content, TOC isotopic compositions ($\delta^{13}\text{C}_{\text{TOC}}$, $\Delta^{14}\text{C}_{\text{TOC}}$) and mineralogical parameters for BS-YS surface sediments are presented in Table 1. TOC and TN contents vary from 0.04 to 1.28% and 0.04 to 0.19%, respectively, and generally increase with distance offshore and with the highest value (1.28% and 0.19%) at the center of the south YS (site H07). TOC and TN contents display a statistically significant correlation and a positive intercept (Fig. S1, $R^2 = 0.98$, $n = 8$, $P < 0.001$), suggesting contributions from inorganic N sources at some locations (Schubert and Calvert, 2001). Molar ratios of TOC and TON (TOC/TON) for all the sediments range from 6.7 to 9.4, with the lowest value near the coast northwest of the old Huanghe delta (SYS600) and lower values (7.8–8.4) also occurring in the area of the delta and central south YS basin. SA and mean grain size values range from 4.1 to 36.5 m^2/g and 8 to 175 μm , respectively. The highest SA and lowest mean grain size values both occur in the center mud area of south YS basin (H07) whereas near delta-coastal samples (e.g., SYS600 and B67) generally show higher mean grain size and lower SA (Table 1).

Generally, TOC show gradual enrichment in ^{13}C and ^{14}C from river delta to offshore sites (Fig. 2). $\delta^{13}\text{C}_{\text{TOC}}$ values range from -21.7 ± 0.1 to $-23.1 \pm 0.1\text{‰}$ with the lowest value at old Huanghe delta area (SYS454), and lower values east of the modern Huanghe delta area (B67) and the highest value at the central south YS area (N05). $\Delta^{14}\text{C}_{\text{TOC}}$ values range from $-435 \pm 7\text{‰}$ to -191 ± 9 with the lowest value near the old Huanghe delta, and highest values in the central south YS area (H07).

3.2. Lipid biomarker abundances

The TOC-normalized contents of the long-chain ($\geq n\text{-C}_{24}$) *n*-alkyl lipid biomarkers generally decrease from the river delta to offshore, with the lowest values at the center of south YS (H07 and N05), whereas short-chain homologues show no systematic spatial variation (Fig. S2).

n-FAs are the most abundant among the three *n*-alkyl lipid biomarker classes, with the total contents ranging from 933 to 2731 $\mu\text{g/g}$ TOC for *n*-C₁₄ to C₃₂. The total *n*-FAs contents are 2 to 12

times higher than those of *n*-alkanes (78 to 1222 $\mu\text{g/g}$ TOC for *n*-C₁₅ to C₃₃) and *n*-alkanols (167 to 635 $\mu\text{g/g}$ TOC for *n*-C₁₈ to C₃₂). These ranges are similar to those previously reported for the BS-YS shelf (Bigot et al., 1989; Xing et al., 2014). Embedded within this overall trend are distinct molecular-level patterns. Specifically, *n*-FAs exhibit a bimodal distribution with an even-carbon number predominance, with pronounced carbon number maxima (C_{max}) at *n*-C₁₆ and at *n*-C₂₄, *n*-C₂₆ or *n*-C₂₈ (Fig. S2A). The carbon preference index ($\text{CPI}_{\Sigma 24-32\text{FA}}$) of long-chain *n*-FAs ranges between 3.4 to 4.7, significantly lower than values (5.2 to 6.0) of suspended particles carried by the Huanghe (Tao et al., 2015), with the highest values at the modern Huanghe and old Huanghe delta (B67 and SYS454) and the lowest value in the center mud area of the south YS (H07) (Fig. S2A). *n*-Alkanols are consistently dominated by long-chain C₂₂–C₃₂ homologues with a strong even-carbon-number predominance ($\text{CPI}_{\Sigma 24-32\text{Alkanol}} = 7.0\text{--}10.6$), and a C_{max} at *n*-C₂₂ or *n*-C₂₈, the latter being site-dependent. Generally, samples near the river delta have higher CPI values with a maximum at *n*-C₂₈, whereas samples offshore have lower CPI and less pronounced carbon number maxima (Fig. S2B). *n*-Alkanes also exhibit a bimodal distribution with maxima at *n*-C₁₆, *n*-C₁₇ or *n*-C₁₈ and at *n*-C₂₉ or *n*-C₃₁, and with a distinct even-over-odd carbon number predominance for shorter chain ($\leq C_{24}$) homologues, and odd-over-even carbon number predominance for long-chain ($> C_{24}$) homologues,. The $\text{CPI}_{\Sigma 25-33\text{Alkane}}$ ranges between 3.5 to 5.8, except for Station SYS600, which exhibits a noticeably lower CPI value of 2.1 (Fig. S2C).

3.3. Molecular isotopic characteristics of *n*-alkyl lipid biomarkers

The $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values of individual compounds, or groups of compounds are shown in Figure 3 and Figure 4. In order to obtain purified saturated *n*-FA fractions, unsaturated lipids were removed prior to carbon isotopic analysis. Therefore, molecular information discussed in this study only refers to saturated lipids. Short-chain *n*-FAs (C₁₄–C₂₀) tend to be enriched in ^{13}C (-20.6 ± 0.2 to $-28.6\pm 0.1\text{‰}$) relative to the longer-chain homologues (C₂₄–C₃₂, -27.0 ± 0.1 to $-31.0\pm 0.1\text{‰}$). Longer-chain *n*-FAs are generally 1 to 8‰ depleted in ^{13}C relative to corresponding TOC values, whereas short-chain odd *n*-FAs (*n*-C₁₅ and *n*-C₁₇) are more enriched in ^{13}C than TOC by *ca.* 1 to 2‰ (Fig. 3A). For radiocarbon, a trend of decreasing ^{14}C content of *n*-FAs with increasing chain-length is observed. Short-chain FAs (*n*-C₁₆ and *n*-C₁₈) displayed a relatively

narrow range of $\Delta^{14}\text{C}$ values (-140 ± 19 to $+37\pm 10\text{‰}$) and corresponding radiocarbon ages ($1,150\pm 178$ yr to modern) are either greater than modern or are within or close to the estimate of the minimum deposition time (Table S1). Long-chain FA ($\geq n\text{-C}_{24}$) homologues displayed a larger range and generally lower $\Delta^{14}\text{C}$ values (-168 ± 8 to $-510\pm 12\text{‰}$; ^{14}C ages between $1,420\pm 80$ and $5,660\pm 190$ yr). In comparison to TOC, short-chain *n*-FAs are systematically ^{14}C -enriched (younger) whereas long-chain homologues exhibit no systematic enrichment or depletion (Fig. 3A and Fig. 4).

$\delta^{13}\text{C}$ values were not determined for *n*-alkanols but their $\Delta^{14}\text{C}$ values for even-carbon-numbered homologues ($n\text{-C}_{18} - \text{C}_{28}$) were found to vary markedly, ranging from $-518\pm 13\text{‰}$ to $+1\pm 27\text{‰}$, corresponding to ^{14}C ages of $5,800\pm 220$ yr to modern. Similar to *n*-FAs, $\Delta^{14}\text{C}$ values generally tend to decrease with increasing chain length from C_{18} to C_{28} , with highest $\Delta^{14}\text{C}$ values (-59 to -140‰) for short-chain C_{18} *n*-alkanols and lower values (-183 to -518‰) for long-chain C_{26} or C_{28} *n*-alkanols. However, at the modern Huanghe delta site (B67), the long chain homologue C_{28} is significantly enriched in ^{14}C relative to the C_{26} homologue (Fig. 3B), suggesting a shorter residence time and/or greater fresh plant contribution to the *n*- C_{28} homologue.

The $\delta^{13}\text{C}$ values of C_{16} to C_{35} *n*-alkanes (ranging from -25.5 ± 0.3 to $-31.4\pm 0.1\text{‰}$) are similar to those of the C_{16} to C_{31} *n*-fatty acids. Specifically, $\delta^{13}\text{C}$ systematically decreases from C_{16} to C_{31} by approximately 3 to 5‰, and then gradually increases from C_{33} to C_{35} by 2 to 5‰. Among all the homologues, *n*- C_{29} or C_{31} exhibits the most ^{13}C -depleted value (-29.8 ± 0.1 to $-31.4\pm 0.1\text{‰}$) and is more depleted in ^{13}C than TOC by *ca.* 4 to 9‰ (Fig. 3C). This is a typical depletion of lipids versus bulk biomass (e.g., Schouten et al., 1998; Wiesenberg et al., 2004) or sedimentary OC (Naraoka and Ishiwatari, 1999). The $\Delta^{14}\text{C}$ values of all measured *n*-alkanes are generally lower than TOC, by as much as 710‰. $\Delta^{14}\text{C}$ values of *n*- $\text{C}_{27+29+31}$ alkanes were highly variable ($-236\pm 12\text{‰}$ to $-615\pm 13\text{‰}$), corresponding to differences in conventional ^{14}C age of $2,100\pm 130$ to $7,610\pm 270$ yr, respectively. Even-carbon-numbered long-chain *n*-alkanes (*n*- $\text{C}_{24+26+28+30}$) exhibited lower $\Delta^{14}\text{C}$ values ($-471\pm 30\text{‰}$ to $-921\pm 25\text{‰}$; corresponding to $5,050\pm 460$ to $20,300\pm 2550$ ^{14}C yr) than their odd-carbon-numbered counterparts (*n*- $\text{C}_{27+29+31}$), while the short-chain homologues (*n*- C_{16+18}) were most strongly ^{14}C -depleted, with $\Delta^{14}\text{C}$ values ranging from -932 ± 31 to $-979\pm 18\text{‰}$ ($21,480\pm 3620$ to $30,880\pm 6620$ yr) (Fig. 3C and Fig. 4).

4. DISCUSSION

4.1. Sedimentological and mineralogical controls on OC loading and distribution

Association with mineral surfaces has been proposed as a primary control on OC stability in marine sediments (Keil et al., 1997; Mayer, 1994). Based on our limited data, TOC contents of the BS-YS sediments have a positive correlation with SA and a negative correlation with mean grain size (Fig. S3A, B), revealing that fine-grained sediments contain higher TOC. Measured OC:SA ratios of 0.10 to 0.35 mg C m⁻² (Fig. S3A) are significantly lower than those reported for many rivers and shelf sediments (OC:SA = 0.4 to 1.0 mg C m⁻²) but similar to previous data reported for high-sedimentation-rate deltaic and low-sedimentation-rate deep-sea environments (<0.4 mg C m⁻²; Blair and Aller, 2012). However, low OC:SA values (0.1~0.2 mg C m⁻²) were also observed from modern Huanghe suspended particles, and from Fly River relict floodplain sediments (Goñi et al., 2014; Keil et al., 1997; Tao et al., 2015). Thus, the relatively low OC loading in BS-YS sediments, especially samples near the Huanghe and old Huanghe delta, likely reflects the supply of extensively weathered relict terraces or floodplain sediments derived from the Chinese loess plateau that are low in OC (SA= 21.0±2.4 m²/g, OC% = 0.3±0.2%, OC:SA = 0.14±0.10 mg C m⁻²; Liu et al., 2003; Liu et al., 2008).

Alternatively, such low OC:SA ratios could be attributed to a high content of high-surface-area smectite in the clay fraction of the Yellow Sea, especially on its western flank (Yang et al., 2003 and references therein). Furthermore, correlations of OC:SA ratio versus C/N ratio and δ¹³C_{TOC} values (Fig. S4A and B) suggest that low carbon loadings are associated with lower C/N ratios and relict terrestrial OC, consistent with strong weathering and degradation (Huanghe, OYR0 and SYS600); in contrast high carbon loadings suggest addition of higher C/N ratio and ¹³C-enriched marine OC. A positive linear relationship between Δ¹⁴C_{TOC} and OC:SA ratio also supports the above interpretation (Fig. S4C), suggesting substantial contributions of old and highly weathering OC from the Huanghe particles (Tao et al., 2015; Wang et al., 2012) into near deltaic samples and higher contribution of fresh (¹⁴C modern) marine OC into offshore samples characterized by higher OC loadings.

4.2. OC provenance in BS-YS sediments

4.2.1. Significance of aged OC

In our study, samples exhibit a relatively narrow range of C/N ratios (6.7–9.4) that are consistent with that of marine planktonic biomass (4 to 10; Meyers, 1994), but C/N ratios of Huanghe suspended particulate samples are also low (Fig. S4A; Liu et al., 2003; Tao et al., 2015; Wang et al., 2012). The lower C/N ratios, which are likely caused by nitrogen associated with mineral phases (Schubert and Calvert, 2001) and low OC loading on input (Keil et al., 1997; Tao et al., 2015) as seen in Huanghe drainage basin sediment, limit the use of this property as a source indicator at multiple (more than two end-member) source-influenced sites. $\delta^{13}\text{C}_{\text{TOC}}$ values increase slightly with distance offshore and are higher than those of POC from the modern Huanghe (ave. $-24.0 \pm 0.2\text{‰}$; Fig. 2A), consistent with a greater proportion of marine OC ($\delta^{13}\text{C}_{\text{planktonic}} = -21\text{‰}$; Fry and Sherr, 1989; Hedges et al., 1997 and references therein) as reported by other studies of the BS-YS (Cai et al., 2003; Xing et al., 2011; Xing et al., 2014). However, the relatively high $\delta^{13}\text{C}_{\text{TOC}}$ value ($-21.7 \pm 0.1\text{‰}$) and old ^{14}C age (6330 ± 110 yr) for the old Huanghe subaerial delta sample (OYR0) may reflect strong weathering and degradation processes, again rendering $\delta^{13}\text{C}_{\text{TOC}}$ values ambiguous as a source indicator. At this site, C/N and $\delta^{13}\text{C}_{\text{TOC}}$ values here don't encode source exclusively.

For all marine sediments, the ^{14}C ages for TOC ($1,640 \pm 90$ to $4,520 \pm 90$ yr) are significantly older than corresponding minimum depositional ages based on average ^{210}Pb -based age models (see Table S1) for the box cores (approximately over 1 to 230 yr), implying significant pre-aged OC contributions to the sediment. Old OC in the BS-YS sediments may derive from pre-aged higher plant OC or fossil OC emanating from modern and abandoned rivers, but also may be a consequence of aging during lateral transport. High terrigenous input including supply of pre-aged soil or ancient fossil OC is the likely cause of lower $\Delta^{14}\text{C}$ values at the river-proximal locations (Wang et al. 2012; Tao et al., 2015). Samples near the old Huanghe delta (Fig. 2B; -428 ± 7 to $-549 \pm 6\text{‰}$) are older than that of modern Huanghe POC (ave. $-417 \pm 17\text{‰}$; Tao et al., 2015), and grain size is larger than suspended particulates in the modern Huanghe (Table 1; Tao et al., 2015). These differences likely reflect re-oxidation and redistribution of old Huanghe delta sedimentary OC, which promotes removal of labile (younger) OC, leaving older OC behind. However, other

sediments are more enriched in ^{14}C content than the modern Huanghe POC, indicating addition of younger marine OC. Resolving the causes and quantifying the amount of aged OC require molecular-level ^{14}C information.

4.2.2. Molecular $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ constraints on the origin of OC

Abundance and isotopic compositions of *n*-alkyl lipid biomarkers were examined in order to further constrain the source and composition of sedimentary OC from the BS-YS system.

Short-chain FA homologues - indicators of modern/contemporary OC

Short-chain *n*-FAs (C_{14} – C_{18}) are generally as abundant, or more abundant, than their longer-chain homologues (C_{24} – C_{32}) (Fig. S2A), suggesting significant marine (algal or bacterial) contribution to the sedimentary OC (Drenzek et al., 2007; Feng et al., 2013a). However, the relative abundance for short-chain *n*-FAs shows no clear increase in the central south YS despite this area being a site of enhanced marine OC accumulation (Cai et al., 2003; Xing et al., 2011; Xing et al., 2014). Lower sedimentation rates in this region may promote selective turnover of more labile organic compounds (e.g., short-chain FAs). In contrast, high relative abundances of short-chain FAs in samples near the modern Huanghe delta include high proportions of bacterial *n*-fatty acids (*n*- C_{15} and *n*- C_{17} , Fig. S2A; Zou et al., 2006), suggesting supply of labile terrestrial biospheric organic matter (e.g., plant debris and recent detritus from surface soil horizons) fueling strong microbial activity in the coastal deltaic region (Bianchi, 2011). Thus, the proportion of different FAs appears to be a function not only of OC source, but also the chemical and physical environment in which they are deposited. Further in-depth lipid biomarker investigations including bacterially-derived lipids (e.g., iso- and anteiso branched FAs, short-chain unsaturated FAs) would aid in distinguishing contributions from different biomass sources (e.g., bacteria, phytoplanktons or fresh terrestrial) in these environments.

A decrease in carbon isotope ratios ($\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$) of sedimentary *n*-FAs and *n*-alkanols with increasing chain length (Fig. 3A, 3B), implies a variety of sources from old to modern for different chain-lengths. Variable $\delta^{13}\text{C}$ values and younger ^{14}C ages in short-chain (C_{14} – C_{18}) *n*-FAs (Fig. 3A and Fig. 4) also indicate significant contributions from contemporary marine and/or fresh terrestrial plant sources. The most abundant homologue in all samples, *n*- C_{16} FA, displays the

highest $\Delta^{14}\text{C}$ values, ranging from -106 ± 11 to $+12\pm 10\text{‰}$ (^{14}C ages from 840 ± 100 yr to modern), and generally exhibits a spatial pattern where coastal or estuary sites are older than offshore sites, with the lowest $\Delta^{14}\text{C}$ value near the old Huanghe delta (Fig. 5A). Depleted ^{14}C signals in short-chain FAs near estuaries may reflect the dynamic sedimentary environment (Li and Gao, 2002), or potentially consumption of aged fluvially-derived OC by heterotrophic bacteria. However, corresponding $\delta^{13}\text{C}$ values for $n\text{-C}_{16}$ FA in the BS-YS sediments are up to 2.8‰ higher than the same FA in suspended particulate matter in the modern Huanghe ($-27.9\pm 0.5\text{‰}$; Fig. 6A). For all sites, the range in $n\text{-C}_{16}$ FA $\delta^{13}\text{C}$ values (-25.1 ± 0.1 to $-27.3\pm 0.1\text{‰}$) is 4 to 6‰ lower than typical values for marine or C_4 plant biomass (Aller and Blair, 2006; Aller et al., 2008; Fry and Sherr, 1989; Hedges et al., 1997), an offset that is similar to that reported between FAs and bulk biomass of marine algae/bacteria and C_4 vascular plants (Schouten et al., 1998; Wiesenberg et al., 2004). Intensive agricultural activities and extensive wetlands along the lower Yellow River may be responsible for local inputs of ^{13}C -enriched terrestrial OC. Taken together, $n\text{-C}_{16}$ FA carbon isotopic compositions imply a mixed origin from contemporary marine and terrestrial sources.

In contrast, the most abundant short-chain (C_{16} , C_{18}) n -alkanes exhibit very low $\Delta^{14}\text{C}$ values (Fig. 4), ranging from -932 ± 31 to $-979\pm 18\text{‰}$ (^{14}C ages of $21,480\pm 3620$ to $30,880\pm 6620$ yr), implying substantial fossil (^{14}C -dead) contributions to $n\text{-C}_{16+18}$ alkanes.

Long-chain homologues - indicators of pre-aged terrestrial biospheric OC

Long-chain ($\geq \text{C}_{24}$) n -FAs, n -alkanols and odd-numbered n -alkanes are generally considered to derive from vascular plant waxes. TOC-normalized contents for long-chain n -alkyl lipid biomarkers (C_{24+} compounds) reveal a decreasing trend from the delta to offshore sites, implying diminishing terrigenous OC abundance with distance from the coast. The $\delta^{13}\text{C}$ values of long-chain n -FAs ($\text{C}_{24}\text{--}\text{C}_{32}$) and odd-numbered n -alkanes ($\text{C}_{25}\text{--}\text{C}_{31}$) suggest C_3 vascular plant sources predominate, and their corresponding $\Delta^{14}\text{C}$ values are highly variable but display a pre-aged signal (Fig. 3 and Fig. 4). These observations suggest that terrestrial plant-derived lipids can experience protracted storage on land prior to delivery to the BS-YS, as well as during subsequent dispersal within this marginal sea system. High $\Delta^{14}\text{C}$ variability of these wax lipids may reflect differences in the type and proximity of different terrestrial plant sources (i.e. agricultural crops, trees, grasses, bryophytes, wetland emergent vegetation). Alternatively, differing

propensities for storage and mobilization could influence the abundance and carbon isotopic compositions of specific plant wax lipids (Goñi et al., 1998). We thus report the abundance-weighted average $\Delta^{14}\text{C}$ values of long-chain *n*-FAs ($\text{C}_{26+28+30}$), *n*-alkanols ($\text{C}_{24+26+28}$) and *n*-alkanes ($\text{C}_{27+29+31}$) as a reflection of corresponding average “residence times” prior to burial due to their relatively coherent $\delta^{13}\text{C}$ values. $\Delta^{14}\text{C}$ values of *n*- $\text{C}_{26+28+30}$ FAs, *n*- $\text{C}_{24+26+28}$ alkanols and *n*- $\text{C}_{27+29+31}$ alkanes display a similar spatial pattern (Fig. 5B, C, D), with generally lower values near the Huanghe extended subaqueous delta and the terminus of the old Huanghe materials along the coastal region (-401 ± 8 to $-510\pm 12\text{‰}$, -329 ± 5 to $-518\pm 13\text{‰}$ and -473 ± 14 to $-615\pm 15\text{‰}$, respectively), and somewhat higher values in the center of the basin (-217 ± 8 to $-265\pm 8\text{‰}$, -246 ± 9 to $+1\pm 27\text{‰}$ and -236 ± 12 to $-361\pm 13\text{‰}$, respectively).

Except for the H07 site, *n*- $\text{C}_{26+28+30}$ FAs (ave., -358‰ , 3,580 yr) and *n*- $\text{C}_{24+26+28}$ alkanols (ave., -327‰ , 3,210 yr) show a similar range and are generally more enriched in ^{14}C content than corresponding *n*- $\text{C}_{27+29+31}$ alkanes (ave., -442‰ , 4790 yr; Fig. 4), suggesting either that the former two groups have faster turnover (or transport) times, or that there are contributions of fossil *n*-alkanes to the latter. In terms of source heterogeneity, carbon isotopic variations among certain plant wax components could be potentially caused by different supply and transport pathway, such as fluvial discharge versus eolian transport (Eglinton et al., 2002; Kusch et al., 2010b), or by surface versus bottom currents (Bian et al., 2013; Guo et al., 2006; Guo et al., 2003). Alternatively, there are reports of long-chain (C_{24} – C_{30}) saturated *n*-alkyl lipids in marine biomass (Rommerskirchen et al., 2006; Schouten et al., 1998; Volkman et al., 1980b), potentially complicating the interpretations of OC sources in terms of terrestrial plant wax input. In our study, sediment samples far away from modern and old Huanghe delta (BF01) and the two central south YS basin sites (H07, N05) have quite different abundance patterns of the long-chain ($\geq \text{C}_{22}$) homologues in which the C_{24} or C_{26} for *n*-FAs and C_{22} for *n*-alkanols are most abundant (Fig. S2A, B). The FA distributions are partially influenced by those found in Chinese eolian dust samples which exhibit carbon number maxima (C_{max}) at C_{24} (Guo et al., 2003), as well as marine biomass with pronounced carbon number maxima at C_{22} or C_{24} for *n*-FAs and C_{22} for *n*-alkanols (Volkman et al., 1998; Volkman et al., 1980a; Volkman et al., 2008). In contrast, samples along the coast tend to maximize at C_{28} , consistent with their distributions in Huanghe suspended and bed sediments

(Bigot et al., 1989; Tao et al., 2015). The differing chain length distributions of the long-chain ($\geq C_{22}$) *n*-FAs and *n*-alkanols again suggest these two groups of compounds in the BS-YS sediments carry varying uncertainty in terms of source exclusivity. Corresponding stable carbon isotopic values also support this interpretation. The abundance-weighted averaged $\delta^{13}C$ compositions for *n*- $C_{26+28+30}$ FAs vary from -27.4 ± 0.3 to -30.5 ± 0.1 ‰ and exhibit $\delta^{13}C$ values in samples from the center of the south YS that are over 3‰ higher than those (-31.5 ± 0.1 ‰) in front of modern Huanghe (Fig. 6B). This trend could be consistent with a partial marine source for *n*- $C_{26+28+30}$ FAs, or it could be explained by ^{13}C -enriched eolian contribution such as from C_4 plant-derived OC. Reported $\delta^{13}C$ values for long-chain *n*-alkanes (-26.8 to -33.0 ‰) in aerosols and dust samples from the northeastern China coastal region suggest a predominant C_3 plant origin mixed with some C_4 plant contribution (Guo et al., 2006). Compared with relatively constant and lower $\delta^{13}C$ values of plant wax lipids in the modern Huanghe (-31.4 to -32.1 ‰, Tao et al., 2015), the spatially variable and higher $\delta^{13}C$ values of FAs in the BS-YS system imply non-Huanghe source inputs. Meanwhile, the relative abundance of long-chain *n*-FAs ($C_{26+28+30}$) versus long-chain *n*-alkanes ($C_{27+29+31}$) tends to increase from the delta to offshore sites (1.2 to 8.7), with highest values in the central south YS (N05). This trend, combined with lower corresponding ratios within or near the river (1.1 to 2.4), suggests a marine contribution for long-chain *n*-FAs. We therefore suspect that *n*- $C_{26+28+30}$ FAs in the BS-YS sediment samples reflect predominantly pre-aged terrestrial C_3 plant source (riverine and eolian) (1900–5670 yr) admixed with FAs of marine origin in the south YS basin. Confirmation of this interpretation will require further constraints, such as biomarker δD measurements (Sauer et al., 2001).

Compared with $C_{26+28+30}$ *n*-FAs and $C_{24+26+28}$ *n*-alkanols, *n*- $C_{27+29+31}$ alkanes display a more coherent ^{13}C trend offshore (-29.6 ± 0.1 to -30.9 ± 0.1 ‰) and the lowest $\Delta^{14}C$ values. This suggests that long-chain *n*-alkanes may be partially derived from a fossil source (s) characterized by more uniform $\delta^{13}C$ values and lower CPI values (Fig. S2C and Fig. 6C). We have previously inferred that long-chain odd-carbon-numbered *n*-alkanes carried by Huanghe suspended particles are influenced by thermogenic hydrocarbons from a fossil source (Tao et al. 2015). Overall, coherent ^{13}C and ^{14}C -depleted characteristics of *n*- $C_{27+29+31}$ alkanes in all the BS-YS sediment samples suggest a dominant pre-aged C_3 plant source (riverine and eolian) (2100–7610 yr), admixed with a

fossil component near the coastal sites, as indicated by lower CPI values (e.g. CPI=2.1 at SYS600 in Fig. S2C).

4.3. Quantification of different OC contributions using coupled isotopic model

4.3.1. A coupled carbon isotopic mixing model

Discussions of the contents and carbon isotopic compositions of source specific biomarkers (Section 4.2.2 based) reveal a complex picture in terms of sources and transport vectors for terrestrial and marine OC. Despite this complexity, this information provides the foundation for an approach to understand carbon supply and accumulation processes in the Chinese marginal seas. A three end-member mixing model based on the $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values of bulk and source-specific biomarkers is used here to estimate the relative fractional proportions of modern/contemporary (f_M), pre-aged soil (f_S), and fossil (f_F) OC in BS-YS surface sediments. All three components are distinguished by ^{14}C age, and are not necessarily derived from a unique source. The calculation and notation follows similar equations to those used by Tao et al. (2015):

$$\Delta^{14}\text{C}_{\text{TOC}} = f_M \times \Delta^{14}\text{C}_M + f_S \times \Delta^{14}\text{C}_S + f_F \times \Delta^{14}\text{C}_F$$

$$\delta^{13}\text{C}_{\text{TOC}} = f_M \times \delta^{13}\text{C}_M + f_S \times \delta^{13}\text{C}_S + f_F \times \delta^{13}\text{C}_F$$

$$1 = f_M + f_S + f_F$$

We constrain individual source end-member values based on site-specific $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values of diagnostic biomarkers (Table S2). In this model, 'Modern OC' source indicates a mixture of marine and fresh/labile terrestrial OC characterized by short turn-over and delivery times (young ^{14}C ages), rather than the 'classic' marine vs. terrestrial. We use $n\text{-C}_{16}$ FA, a ubiquitous and labile compound in natural environment that shows a high abundance in biomass and in the surface/organic layer of soils (Otto and Simpson, 2005; Schouten et al., 1998), as a representative biomarker of modern/contemporary biospheric OC. In the BS-YS system, $\Delta^{14}\text{C}$ values of $n\text{-C}_{16}$ FA in low-energy depositional sites (+6 to +12‰) approach that of surface-ocean dissolved inorganic carbon (>0‰; Levin and Hesshaimer, 2000; Pearson et al., 2001). Although old ^{14}C ages for $n\text{-C}_{16}$ FA (13 ± 78 to 840 ± 100) have been observed in the high-energy depositional sites (Fig. 5A), these approach corresponding minimum depositional ages based on average ^{210}Pb -based age models (see Table S1), and thus imply a modern/contemporary OC source. Hence, modern/contemporary

OC is therefore assumed to carry $\delta^{13}\text{C}_{16\text{FA}}$ and $\Delta^{14}\text{C}_{16\text{FA}}$ equivalent to $\delta^{13}\text{C}_{\text{M}}$ and $\Delta^{14}\text{C}_{\text{M}}$ for each site in the model. A pre-aged soil OC end-member is defined as plant-derived OC which has been stored in intermediate reservoirs such as soils, floodplains, lakes etc., and then subsequently exported to the marine environments via fluvial and/or eolian transport. Long-chain functionalized *n*-alkyl lipids (*n*-C_{26, 28, 30} FAs, *n*-C_{24, 26, 28} alkanols, *n*-C_{27, 29, 31} alkanes) show high relative abundances in soils, where they are closely associated with mineral particles (Feng et al., 2013b; Xu et al., 2009) and may experience long residence times ($\geq 10^3$ ¹⁴C yrs), but exhibit lower relative abundances in fresh materials where short-chain homologues predominate. With respect to the pre-aged soil end member ($\delta^{13}\text{C}_{\text{S}}$ and $\Delta^{14}\text{C}_{\text{S}}$), long-chain *n*-C₂₆₊₂₈₊₃₀ FAs are regarded as most appropriate candidates. Their corresponding $\Delta^{14}\text{C}$ values (–217 to –510‰) are unaffected by fossil OC inputs, whereas those of *n*-C₂₇₊₂₉₊₃₁ alkanes (–236 to –615‰) are artificially too low due to fossil contributions from the latter (Kusch et al., 2010b; Pearson and Eglinton, 2000; Tao et al., 2015). However, marine biomass may produce *n*-C₂₆ and *n*-C₂₈ FAs and long-chain (C_{24, 26, 28}) *n*-alkanols (Rommerskirchen et al., 2006; Volkman et al., 1998; Volkman et al., 1980a; Volkman et al., 2008) that would likely be most significant at distal sites. In our study, large isotopic variations among *n*-C₂₆, *n*-C₂₈ and *n*-C₃₀ FAs (110–151‰) at distal/offshore sites (B25 and N05) reflect uncertain and variable amounts of other OC sources (e.g., marine contributions), yielding higher $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ values for *n*-C₂₆ and *n*-C₂₈ FAs (Fig. 3A). Hence, values for $\delta^{13}\text{C}_{\text{S}}$ and $\Delta^{14}\text{C}_{\text{S}}$ at offshore sites (B25 and N05) were therefore based on $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values of *n*-C₃₀ FA.

Because of petrogenic OC contributions from the Huanghe drainage basin and intensive anthropogenic activity (including offshore petroleum exploitation in the Bohai Basin), fossil OC is defined as a mixture of a sedimentary rock (petrogenic) source and petroleum-derived OC. *n*-C_{16,18} alkanes in our samples display extremely low $\Delta^{14}\text{C}$ values (–932 to –979‰; Fig. 4), indicating a predominantly fossil origin ($\Delta^{14}\text{C}_{\text{F}} = -1000\text{‰}$). Given AMS measurement uncertainty for small samples (15–30 μgC), we define values $\leq -920\text{‰}$ as “¹⁴C-dead”. Hence, fossil OC in the mixing model assumes a similar $\delta^{13}\text{C}_{\text{F}}$ value as those of *n*-C_{16,18} alkanes from each site and $\Delta^{14}\text{C}_{\text{F}}$ as –1000‰.

Prior studies have shown that significant differences exist between $\delta^{13}\text{C}$ values of specific lipids and bulk carbon in biomass, with lipid components generally exhibiting greater ¹³C depletion

as a consequence of biosynthetic fractionation and related effects. The reported magnitude of these offsets ($\delta^{13}\text{C}_{\text{bulk}} - \delta^{13}\text{C}_{\text{compound}}$) is 5–10‰ for algae and 7–10‰ for vascular plants (Ballentine et al., 1996; Collister et al., 1994; Schouten et al., 1998; Wiesenberg et al., 2004). Isotopic offsets between *n*-alkanes and bulk organic materials are not uniform as they may reflect both biosynthetic processes and the effects of diagenesis and thermal alteration. In some cases *n*-alkanes are isotopically similar to bulk OC in the host sedimentary rock (Eglinton, 1994), whereas others have observed up to a 7‰ offset in $\delta^{13}\text{C}$ between *n*-alkanes and petrogenic materials (e.g., immature bitumens, coals etc.; Drenzek et al., 2007; Goñi et al., 2005; Wiesenberg et al., 2004). Since we presently have no data on $\delta^{13}\text{C}_{\text{bulk}} - \delta^{13}\text{C}_{\text{compound}}$ offsets of typical source materials within and surrounding the Chinese marginal seas, in this study, we therefore apply a moderate (4–7‰) offset to the measured $\delta^{13}\text{C}$ values for each of the biomarker classes in defining the three end-members in order to approximate corresponding $\delta^{13}\text{C}_{\text{bulk}}$ values. After applying this correction, calculated values of $\delta^{13}\text{C}_{\text{M}}$ (–18.1 to –23.3‰) are consistent with a mixed signal of marine biomass and fresh terrestrial C_3 -dominated vegetation in northern China and in the modern Huanghe. Similarly, calculated $\delta^{13}\text{C}_{\text{S}}$ values (–20.6 to –26.5‰) encompass that of C_3 vegetation, loess/paleosols from the Chinese Loess Plateau and OC in fluvial material from the modern Huanghe. Lastly, $\delta^{13}\text{C}_{\text{F}}$ values (–20.1 to –24.2‰) are similar to bulk $\delta^{13}\text{C}$ of OC in source rocks in the upper Huanghe which are predominantly low-to-medium grade metamorphic rocks and coal series (Jia et al., 2003; Liu et al., 2007). We sought to improve our three end-member mixing model using matrix of calculations by first fixing FA-constrained sources ($\delta^{13}\text{C}_{\text{M}}$, $\delta^{13}\text{C}_{\text{S}}$) with offsets from 4 to 7‰, and then by varying alkane-constrained $\delta^{13}\text{C}_{\text{F}}$ also with offset from 4 to 7‰ (Fig. S5). Offsets less than 4‰ or more than 7‰ were found to return implausible values of f_{S} or f_{M} (i.e., <0), implying that the chosen range (4–7‰) is appropriate in the absence of values for isotopic differences between bulk OC and specific compounds specific to the Chinese marginal seas. Model outputs are relatively sensitive to change with variations of the $\delta^{13}\text{C}_{\text{bulk}} - \delta^{13}\text{C}_{\text{compound}}$ offset, showing largest influence on f_{S} but minor influence on f_{F} , consistent with previous studies (Tao et al., 2015). For the convenience of discussion, the following reported values for f_{M} , f_{S} and f_{F} reflect model results integrated across prescribed $\delta^{13}\text{C}_{\text{bulk}} - \delta^{13}\text{C}_{\text{compound}}$ offsets between 4 and 7‰, with associated model-derived uncertainties given as standard deviations (Table 2).

4.3.2. Fractional contributions of different source OC components

The proportion of modern/contemporary OC in BS-YS surface sediments ranges from 31–64%, with values generally higher for sites offshore (central south YS, N05; and offshore subaqueous Huanghe delta, B25; $f_{M, avg} = 63\%$, 64% , respectively) compared to sites near the modern and old Huanghe delta ($f_{M, avg} = 32\%$; 31% , respectively) (Fig. 7, Table 2). This spatial pattern is consistent with previous estimates derived from other molecular proxies which showed an increasing contribution from marine production and a decreasing supply of fluvial-supplied soil material for sites toward the center of the basin (Hu et al., 2009; Wang et al., 2014; Xing et al., 2014).

Our model yields pre-aged soil OC contribution varying from 24–49%, with the lowest contribution in the modern Huanghe distal delta (B25, BF01; $f_{S, avg} = 24\text{--}28\%$) and the highest contribution near the modern and old Huanghe inner delta ($f_{S, avg} = 48\%$; 49% , respectively). These results are consistent with the general pathway of fluvial sediment transport within the BS-YS system (Yang and Liu, 2007; Zhou et al., 2014). Surprisingly, $f_{S, avg}$ is also estimated to account for up to 33% of TOC in one of the offshore sites (H07) (Fig. 7, Table 2), implying significant contributions of pre-aged terrestrial OC to the central south YS. However, $\Delta^{14}C_S$ at this site is relatively high, which means younger ^{14}C age, compared to coastal sites (Fig. 5B, C, D). Wind-borne pre-aged (soil) OC could be a potential source of this terrestrial OC. Large dust/aerosol plumes from China travelling to the east/northeast may entrain biological materials from vegetation and upper soil layers, carrying a relatively ^{14}C -enriched terrestrial OC offshore (Kusch et al., 2010b). However, as dust mostly originates from the loess terrain and deserts in northwest China, it is not straightforward to distinguish between the eolian derived- and Huanghe-borne pre-aged OC contributions.

Fossil OC is a significant component of OC in the BS-YS, varying from 7–26%. High values are returned for samples most strongly influenced by Huanghe sediments (B67, $f_{F, avg} = 17\%$), subaqueous Huanghe delta mud area (BF01, $f_{F, avg} = 16\%$), old Huanghe delta mud area (SYS454, $f_{F, avg} = 19\%$), and Haizhou Bay (SYS600, $f_{F, avg} = 26\%$) (Fig. 7, Table 2). This pattern implies that fossil OC in BS-YS surface sediments was supplied by both the modern and old Huanghe. The

high proportion of fossil OC in the central south YS basin (11%) most likely originates from the old Huanghe delta, consistent with the conclusion of a separate study (Zhou et al., 2014). Moreover, based on *n*-alkane abundance and low $CPI_{\Sigma 25-33Alkane}$ (2.1, Fig. S2C), Haizhou Bay (SYS600) may be contaminated with fossil fuels. Fossil OC exported from mountainous rivers draining active margin settings has been reported to account for up to, and in some cases or more than, half of OC burial in adjacent marine sediments (Blair et al., 2003; Kao et al., 2014; Raymond and Bauer, 2001). In comparison, contribution of fossil OC to the northern Chinese marginal seas from the Huanghe – a river system developed on a passive margin is substantially lower. This also appears to be the case for the Changjiang (Yangtze river; Li et al., 2015). In addition to potential differences in the abundance and nature of fossil OC in source rocks, less extensive fossil OC export and sedimentary accumulation may reflect oxidation, sequestration or dilution of fossil OC during long-distance transport and protracted storage in intermediate reservoirs (Tao et al., 2015), including those stemming from anthropogenic activity (e.g., dam construction; Li et al., 2015).

4.4. Implications for carbon cycling & budgets

Modern, pre-aged and fossil OC pools contribute to global and regional budgets, influence the carbon cycle over varying timescales and are likely to vary with changes in climate and anthropogenic activities. Although there are inherent uncertainties in the mass balance approach described here, it provides an important means to distinguish and assess the fate of these modern, pre-aged and fossil OC components in continental margin sediments. Combining prior estimates of sediment flux in the BS-YS system (Fig. 1 and Table 3) and our dual isotopic mass balance estimates, we calculate a supply and accumulation budget for the three different OC components (modern, pre-aged, fossil) for the Chinese marginal seas, averaged over ~ 100 years (Fig. 8 and Table 3). This time interval is selected because our sediment sampling interval (0~10 cm) and physical mixing of surface sediments permit only time-integrated estimates. For each site, total POC and sedimentary OC annual fluxes are estimated by average OC% $\times$ average annual sediment flux (Q_S ; Mt/yr), whereas different sourced OC flux is calculated by average OC% $\times$ average annual sediment flux (Q_S ; Mt/yr) $\times$ average fraction of individual OC component (f_M, f_S or f_F). Over the past 100 years, modern Huanghe sediment flux has changed markedly due to human

activities and drought, decreasing from ~1100–1200 Mt/yr prior to the 1960s, to 754 Mt/yr from 1960–1999, and most recently to 151 Mt/yr (2000–2007). We therefore use a time-integrated average sediment flux of 930 Mt/yr over the past 100 years in our calculation (Fig. 1 and Table 3). Tao et al. (2015) estimated the average proportions of different OC components in modern Huanghe suspended sediments using a approach and found that modern/contemporary, pre-aged (soil) and fossil OC accounted for $16\pm9\%$, $53\pm12\%$ and $31\pm3\%$ of POC, respectively ($\pm$ standard deviation here refers to uncertainty of the calculation method depending on the variations of the specific $\delta^{13}\text{C}_{\text{bulk}}-\delta^{13}\text{C}_{\text{compound}}$ offset; Table 3). We assume that variations in water discharge and total suspended sediment (TSS) concentration of the modern Huanghe resulting from human activity (e.g., dam building or water and sediment regulation by human activities) do not significantly affect OC composition, and specifically the proportions of the different OC components, but will strongly affect the overall flux of OC exported to the ocean. Temporal fluctuations in the proportions of different OC components stemming from seasonal changes in terrestrial primary production, precipitation etcetera (Tao, 2014) are taken into account through determination of annual average values. Thus, modern values of average fraction of different OC components (f_M , f_S and f_F) are assumed to be relatively constant within the past 100 years.

Estimates of sedimentary OC flux vary widely between studies (Alexander et al., 1991; Li et al., 2012; Saito, 1998; Yang et al., 2003; Zhou et al., 2014). Here, we apply a recent sediment budget model of the YS-BS basin using 100-year time scale that is modified from Li et al. (2012) and Zhou et al. (2014). Collected data reveal that the sediment supply from the modern Huanghe (~930 Mt/yr) results in sediment accumulation of 557 Mt/yr in the Bohai Sea basin and 70 Mt/yr in the subaqueous Huanghe delta (area M1 and M2 of Li et al. 2012). The remaining ~300 Mt/yr emanating from the modern Huanghe has been suggested to be transported to the YS (Liu et al., 2009). This estimate is concordant with our calculated sediment budget for the YS, but is at the upper limit of other studies, which range from about 6.8 Mt/yr (Martin et al., 1993) to 300 Mt/yr (Liu et al., 2009). Sediment supply from the old Huanghe delta area (792 Mt/yr) and modern Huanghe (300 Mt/yr) to the YS roughly equals estimates of sediment accumulation in this marginal sea region that includes the southern Jiangsu coast (113 Mt/yr) and its offshore slope (200 Mt/yr), the Changjiang delta (38 Mt/yr), the central south YS (542 Mt/yr, including south of

Cheju Island mud area), the Min-Zhe mud belt (35 Mt/yr) and the southeast YS mud areas (40~150 Mt/yr; SEYSM of Lim et al., 2007) along the Korean peninsula (Fig. 1). The Changjiang sediment contribution has been stable, without significant shifts in river's path at least the past few thousand years. Most of the Changjiang suspended sediments have been trapped in the estuary and nearby coastal areas towards the south (Li et al., 2001; Yang et al., 2003; Yang et al., 2002).

Based on our calculations (Fig. 8 and Table 3), a total of 5.62 Mt/yr of OC has been supplied by the modern Huanghe (3.44 Mt/yr) and the old Huanghe (2.18 Mt/yr). For pre-aged soil OC, the modern Huanghe supplied 1.82 Mt/yr and the old Huanghe supplied 0.88 Mt/yr (total, 2.70 Mt/yr). With respect to fossil OC, the modern Huanghe supplied 1.07 Mt/yr and the old Huanghe supplied 0.49 Mt/yr, for a total of 1.56 Mt/yr. In BS-YS basin, 2.77, 0.38 and 5.48 Mt/yr sedimentary OC accumulate in the BS basin, Shandong Peninsula subaqueous delta, and central south YS basin, respectively, for a total of 8.63 Mt/yr. In the three depositional sub-regions, this corresponds to 1.27, 0.10 and 1.65 Mt/yr for pre-aged soil OC (total, 3.02 Mt/yr), respectively, and 0.36, 0.05 and 0.57 Mt/yr (total, 0.98 Mt/yr) for fossil OC, respectively. The calculated total accumulation of pre-aged soil OC (3.02 Mt/yr) is larger than the supply (2.70 Mt/yr) from the two major sources. Uncertainties in our estimates resulting from the limited number of core-top samples analyzed, and unknown $\delta^{13}\text{C}_{\text{bulk}}-\delta^{13}\text{C}_{\text{compound}}$ offsets in the model calculation may be the cause of this discrepancy (Table 3 and Fig. S5). However, inaccurate fluvial flux values, other sources of terrestrial OC (e.g., dust), and different transport processes may also be responsible.

Another approach (see Fig. 8) takes into consideration these last two factors (transport processes and dust sources). Therein, the BS and subaqueous Huanghe delta in the north YS have been commonly regarded as the depocenters of modern Huanghe OC, respectively accumulating 1.27 and 0.10 Mt/yr of pre-aged soil OC, 0.36 and 0.05 Mt/yr of fossil OC. The remainder of the pre-aged soil OC (0.46 Mt/yr) and fossil OC (0.65 Mt/yr) from the modern Huanghe is inferred to be transported to, and to accumulate in, the central south YS. Additionally, net sediment erosion rate of the old Huanghe delta is estimated at 792 Mt/yr over the past 100 years, but only about half of this amount (424 Mt/yr sediments) could be delivered to the central south YS basin (Zhou et al., 2014). Based on these estimates, the old Huanghe contributes 0.47 Mt and 0.26 Mt of pre-aged soil OC and fossil OC to the central south YS annually, whereas, the amount of pre-aged soil and

fossil OC accumulation is calculated to be 1.65 and 0.57 Mt/yr, respectively. In order to balance estimates of pre-aged soil OC accumulation rates in the central south YS and assuming no degradation of terrigenous OC during transportation, an unidentified contribution of 0.72 Mt/yr pre-aged soil OC is required, such as via atmospheric deposition or the Yellow Sea Warm Current (YSWC). Previous estimates of eolian dust supply also indicate that this could affect the sedimentary OC budget of the YS surface sediments (Huang et al., 2014; Zhang et al., 1993; Zhang and Friedlander, 2000). Irrespective of additional sources and measurement uncertainties, our estimates imply very high burial efficiency for pre-aged soil OC in the central south YS basin. Similar calculations for fossil OC indicate a missing sink of 0.34 Mt of fossil OC (Fig. 8), which may reflect oxidation and/or restricted transmission for sedimentary rock detritus during transport and sedimentation (Galy et al., 2008). Meanwhile, anthropogenic activities can impact the transport and burial of both pre-aged soil OC and fossil OC fractions in the China marginal seas, which need to be considered in future studies for better estimates of the system's carbon budgets.

Overall, in the three sediment deposition areas (Bohai Basin, subaqueous Huanghe delta and central south YS), reburial of allochthonous is estimated to account for 46% of OC accumulation (a total of 4.00 Mt/yr). For comparison, a total of 4.63 Mt/yr of modern OC is buried in the three main sediment accumulation regimes, the latter reflecting the capacity for short-term (< 100 yr scale) transfer and subsequent sequestration of C recently fixed by the biosphere in this marginal sea system.

5. CONCLUSIONS

Spatial variations in the content and carbon isotopic compositions of TOC and source specific lipid biomarkers in the BS-YS surficial sediments reflect source heterogeneity as well as transport and transformation processes. A coupled isotopic mixing model based on compound-specific and bulk $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ data was applied to derive estimates for accumulation of different OC types in the BS-YS system. Based on this approach we find that OC in BS-YS surface sediments is, on average, comprised of ca. 31–64% modern OC, 24–49% pre-aged terrestrial (soil) OC, and 7–26% fossil OC. The greatest proportions of modern OC accumulate at the center of the south YS and offshore subaqueous Huanghe delta (63%, 64%, respectively), whereas maximum proportions of

pre-aged soil OC are observed at modern and old Huanghe delta (48% and 49%, respectively), and the highest proportion of fossil OC is found north of the old Huanghe mouth (26%). We also find significant contributions of pre-aged soil OC (up to 33%) at more distal sites, potentially reflecting augmented terrestrial OC supply via atmospheric (dust) transport and deposition.

The supply and the fate of OC derived from different sources has significant implications in terms of regional and global-scale carbon cycle processes. On one hand, ~46% of OC accumulation (4.00 Mt/yr) in the BS-YS surface sediments is derived from pre-aged (i.e., several thousand-year-old) plant-derived OC (likely from soils) and ancient fossil OC (>30,000 yr), with burial efficiencies close to 100% and 70%, respectively. These two pools of terrigenous OC influence carbon cycling over longer (thousand-to-million-year) time scales. On the other hand, accumulation of modern OC, estimated at 4.63 Mt/yr, contributes directly to the sequestration of atmospheric CO₂, and influences carbon cycle on shorter time scales (<100 yr) within this marginal sea system.

The findings reported here also have implications for biomarker-based reconstructions from marginal sea sediment records. Protracted storage and aging of vascular plant OC prior to export and accumulation in marine basins may induce substantial lags in climate and vegetation signals. Thus, the fidelity of organic biomarker records in marine sediments, both in a temporal and spatial context, must be carefully assessed for accurate paleoclimate reconstruction.

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Figure Captions

Fig. 1. A map of sampling sites (filled circles: marine sediments; open circle: the old Huanghe subaerial delta), surface circulation (arrows) and mud area (grey shaded area) of the Bohai Sea, North Yellow Sea, South Yellow Sea and north East China Sea. Current: BSCC = Bohai Sea Coastal Current (black arrows); YSCC = Yellow Sea Coastal Current (black arrows); NJCC = North Jiangsu Coastal Current (blue arrows); CDW = Changjiang Diluted Water (blue dotted arrows); YSWC = Yellow Sea Warm Current (red dotted arrow).

Fig. 2. Spatial variations of bulk OC isotopes: (A) $\delta^{13}\text{C}_{\text{TOC}}$; (B) $\Delta^{14}\text{C}_{\text{TOC}}$. Symbol size and color is proportional to values (more negative values correspond to smaller symbol sizes and colder colors; $\delta^{13}\text{C}$ values range from -20 to -30‰ , and $\Delta^{14}\text{C}$ values range from $+100$ to -900‰).

Fig. 3. Compound-specific $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ compositions of individual and/or groups of *n*-alkyl lipids from each site, including samples from the Huanghe delta (gray circle), the BS basin (magenta circle), the Subaqueous Huanghe delta (blue diamond), the south YS basin (red triangle) and the Older Huanghe delta (black square). (A) *n*-fatty acids (*n*-FAs); Some $\Delta^{14}\text{C}$ data are combinations of similar homologues: Site B43, BF01, H07, SYS454 and SYS600 *n*-C₂₆ (*n*-C₂₆₊₂₈₊₃₀); site B43, SYS600 *n*-C₁₆ (*n*-C₁₆₊₁₈). (B) *n*-alkanols; Some $\Delta^{14}\text{C}$ data are combinations of similar homologues: Site B25 and N05 *n*-C₁₈ (*n*-C₁₈₊₂₀); Site H07 *n*-C₂₈ (*n*-C₂₆₊₂₈); site SYS454, SYS600 *n*-C₂₈ (*n*-C₂₄₊₂₆₊₂₈). (C) *n*-alkanes from each site; Some $\Delta^{14}\text{C}$ data are combinations of similar homologues: All sites *n*-C₃₁ (*n*-C₂₇₊₂₉₊₃₁); All sites, *n*-C₂₈ (*n*-C₂₄₊₂₆₊₂₈₊₃₀); All sites, *n*-C₁₈ (*n*-C₁₆₊₁₈). Bulk ($\delta^{13}\text{C}_{\text{TOC}}$ and $\Delta^{14}\text{C}_{\text{TOC}}$) values are shown for comparison.

Fig. 4. A plot of $\delta^{13}\text{C}$ versus $\Delta^{14}\text{C}$ compositions of individual and/or groups (short-chain homologues, circle; middle-chain homologues, triangle; long-chain homologues, diamond) of *n*-alkyl lipids (fatty acids, green; alkanes, blue) and bulk OC (black square) from each marginal sea sites and the Huanghe SPM (from Tao et al., 2015). The filled symbols are for marine sediments, and the unfilled symbols are for the Huanghe SPM.

Fig. 5. Spatial variations in $\Delta^{14}\text{C}$ values of *n*-alkyl lipids in surface sediments: (A) *n*-C₁₆ FA, (B) *n*-C₂₆₊₂₈₊₃₀ FAs, (C) *n*-C₂₄₊₂₆₊₂₈ alkanols, (D) *n*-C₂₇₊₂₉₊₃₁ alkanes. Symbol size and color is proportional to values (more negative values correspond to smaller sizes and colder colors; $\Delta^{14}\text{C}$ values range from $+100$ to -900‰).

Fig. 6. Spatial variations in $\delta^{13}\text{C}$ values of *n*-alkyl lipids in surface sediments: (A) *n*-C₁₆ FA, (B) *n*-C₂₆₊₂₈₊₃₀ FAs, and (C) *n*-C₂₇₊₂₉₊₃₁ alkanes. The composite values for multiple homologues shown

in B and C represent abundance-weighted average values. Symbol size and color is proportional to values (more negative values correspond to smaller sizes and colder colors; $\delta^{13}\text{C}$ values range from -24 to -34‰).

Fig. 7. Coupled isotopic mass balance results for the fractional contributions of OC from modern (blue bar), pre-aged soil (green bar) and ancient fossil (black bar) sources in surface sediments of the BS-YS basin over a range of prescribed $\delta^{13}\text{C}_{\text{Bulk}} - \delta^{13}\text{C}_{\text{Compound}}$ offsets (4–7‰). The height of each bar is proportional to their percent contribution (bars shown at the bottom are set for 100%).

Fig. 8. Terrigenous (pre-aged soil and fossil) OC budget (Mt/yr) in the BS-YS basin. Positive values in the boxes indicate different sourced OC accumulation; and negative values above and below the arrowed lines indicate OC supply through dispersal or net erosion process of the modern and Older Huanghe delta in different sediment regimes. Dust or other source contributions to the Bohai Sea and the modern Huanghe subaqueous delta areas are not shown, as they are beyond the scope of this study. The minimum contribution from non-Huanghe OC source to the central SYS is given based on a source-sink mass balance calculation.

Table 1. Bulk characteristics of surficial sediments from the BS-YS and Old YR subaerial delta

Samples	Types	TOC%	TN%*	TOC/TON*	SA	Mean grain size	OC:SA	$\delta^{13}\text{C}$	$\Delta^{14}\text{C}$
				[mol]	[m ² /g]	[μm]	[mg C/m ²]	[‰]	[‰]
B67	Silt	0.41	0.07	8.3	16.1	23	0.26	-22.8	-337
B43	Clayey silt	0.58	0.09	9.0	21.7	25	0.27	-22.2	-242
B25	Silt	0.67	0.10	9.2	19.7	20	0.34	-22.8	-226
BF01	Silt	0.42	0.06	9.4	12.1	36	0.35	-22.5	-270
H07	Clayey Silt	1.28	0.19	8.4	36.5	8	0.35	-22.1	-191
N05**	Clayey Silt	0.74	0.12	7.8	29.5	12	0.25	-21.7	-235
SYS454	Clayey silt	0.41	0.07	8.2	19.4	21	0.21	-23.1	-428
SYS600	Silty sand	0.14	0.04	6.7	12.9	126	0.11	-22.2	-435
OYR0	Sand	0.04	n.d.	n.d.	4.1	175	0.10	-21.7	-549

*Molar ratio of TOC and TN content was corrected for mineral-associated inorganic N based on the method in Schubert and Calvert (2001).

The sediment sample at **N05 site is a subsurface sample (0–30 cm depth)

Table 2. Coupled isotope mass balance results for the average fractional contributions of modern (f_M), pre-aged soil (f_S) and fossil OC (f_F) in the Huanghe suspended matter and marine surface sediments over a range of prescribed $\delta^{13}\text{C}_{\text{Bulk}}-\delta^{13}\text{C}_{\text{Compound}}$ offsets (4–7‰). $\pm$ standard deviation here refers to uncertainty of the calculation method depending on the variations of the specific $\delta^{13}\text{C}_{\text{bulk}}-\delta^{13}\text{C}_{\text{compound}}$ offset.

sites	Location	f_M	f_S	f_F
B67	Huanghe delta	0.32 $\pm$ 0.13	0.48 $\pm$ 0.18	0.20 $\pm$ 0.05
B43	Bohai Sea basin	0.50 $\pm$ 0.09	0.43 $\pm$ 0.13	0.07 $\pm$ 0.04
B25	Subaqueous Huanghe delta	0.64 $\pm$ 0.06	0.24 $\pm$ 0.12	0.12 $\pm$ 0.06
BF01	Subaqueous Huanghe delta	0.56 $\pm$ 0.10	0.28 $\pm$ 0.18	0.16 $\pm$ 0.07
H07	South Yellow Sea basin	0.56 $\pm$ 0.15	0.33 $\pm$ 0.21	0.11 $\pm$ 0.06
N05	South Yellow Sea basin	0.63 $\pm$ 0.13	0.27 $\pm$ 0.17	0.10 $\pm$ 0.05
SYS454	Old Huanghe delta	0.31 $\pm$ 0.17	0.49 $\pm$ 0.25	0.20 $\pm$ 0.08
SYS600	Haizhou Bay	0.42 $\pm$ 0.11	0.32 $\pm$ 0.22	0.26 $\pm$ 0.11
River	Huanghe	0.16 $\pm$ 0.09*	0.53 $\pm$ 0.12*	0.31 $\pm$ 0.03*

* Data are from Tao et al., 2015

Table 3. A burial budget for three different OC components (modern, pre-aged, fossil) in the BS-YS basin. Also included is the estimated supply of these OC components by the modern and Old Huanghe dispersal systems

Location	Representative sites	Q_s^2	OC ³		Modern OC ³		Pre-aged soil OC ³		Fossil OC ³	
		[Mt/yr]	[%]	[Mt/yr]	$[f_M]$	[Mt/yr]	$[f_S]$	[Mt/yr]	$[f_F]$	[Mt/yr]
Huanghe	Kenli station ¹	−929	0.37	−3.44	0.16±0.09	−0.55±0.32	0.53±0.12	−1.82±0.40	0.31±0.03	−1.07±0.09
Old Huanghe delta	SYS454, SYS600	−792	0.28	−2.18	0.37±0.14	−0.80±0.31	0.41±0.24	−0.88±0.51	0.22±0.10	−0.49±0.21
Total supply		−1721		−5.62		−1.35±0.45		−2.70±0.65		−1.56±0.23
Bohai Sea	B67, B43	557	0.50	2.77	0.41±0.11	1.14±0.30	0.46±0.15	1.27±0.42	0.13±0.04	0.36±0.12
Subaqueous Huanghe delta	B25, BF01	70	0.55	0.38	0.60±0.08	0.23±0.03	0.26±0.15	0.10±0.06	0.14±0.07	0.05±0.03
South Yellow Sea basin	H07, N05	542	1.01	5.48	0.60±0.14	3.26±0.77	0.30±0.19	1.65±1.05	0.10±0.05	0.57±0.28
Total burial		1169		8.63		4.63±0.83		3.02±1.13		0.98±0.31

¹A previous study site at the Huanghe downstream (Tao et al., 2015)

² Q_s indicates sediment flux data for different BS-YS areas derived from Li et al., 2012, Peng et al., 2010 and Zhou et al., 2014. Positive values indicate deposition and negative values indicate dispersal or net erosion of the modern and old Huanghe material.

³Calculated bulk and three different source OC flux data for different BS-YS areas based on $Q_s \times OC\%$ and $f_i \times OC\% \times Q_s$ ($i = M, S$ and F), respectively. Positive values indicate OC burial and negative values indicate OC supply through dispersal or net erosion processes of the modern and old Huanghe. OC% and f_i in the individual location is the average value of several representative sites collected from the corresponding sediment regime.

Figure1

Figure2A

Figure2B

Figure3A

Figure3B

Figure3C

Figure4

Figure5A

Figure5B

Figure5C

Figure5D

Figure6A

Figure6B

Figure6C

Figure7

Figure8