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Widespread dispersal and aging of organic carbon in shallow marginal seas

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ABSTRACT

The occurrence of pre-aged organic carbon (OC) in continental margin surface sediments is a commonly observed phenomenon, yet the nature, sources, and causes of this aged OC remain largely undetermined for many continental shelf settings. Here we present the results of an extensive survey of the abundance and radiocarbon content of OC in surface sediments from the northern Chinese marginal seas. Pre-aged OC is associated with both coarser (>63 µm) and finer (<63 µm) sedimentary components; measurements on specific grain-size fractions reveal that it is especially prevalent within the 20–63 µm fraction of inner shelf sediments. We suggest that organic matter associated with this sortable silt fraction is subject to protracted entrainment in resuspension-deposition loops during which it ages, is modified, and is laterally dispersed, most likely via entrainment within benthic nepheloid layers. This finding highlights the complex dynamics and predepositional history of organic matter accumulating in continental shelf sediments, with implications for our understanding of carbon cycling on continental shelves, development of regional carbon budgets, and interpretation of sedimentary records.

INTRODUCTION

Continental shelves are the major loci of organic carbon (OC) burial in the oceans and the confluence of terrestrial and marine realms; it is crucial to understand the processes that lead to the efflux of CO_2 and sequestration of OC on continental shelves in order to predict potential changes to this important and dynamic component of the carbon cycle. Physicochemical interactions between organic matter (OM) and the mineral matrix are typically invoked as the primary mode of stabilization and sequestration of OM in sediments (Kennedy and Wagner, 2011). Hydrodynamic processes play a critical role in the dispersal and distribution of mineral-associated OM on continental margins and in the deep sea (McCave and Hall, 2006; Inthorn et al., 2006). Protracted entrainment within resuspension-deposition loops and exposure to oxic conditions may, in turn, influence the properties of OC that accumulates in underlying sediments. Accordingly, the distribution, composition, reactivity, and age of OM preserved in continental margin sediments may be controlled to a significant degree by OM-mineral interactions, with important implications for regional and global carbon budgets (Burdige, 2005; Deng et al., 2006). However, much remains to be understood concerning the spatial and temporal dimensions of OM-mineral interactions and associated transport processes, and their impacts on source, composition, and amount of OM accumulating on continental margins and in the deep sea.

The ^{14}C ages of OC in the surface mixed layer of shallow continental shelf sediments are, in many cases, older than expected if the OM had originated from modern-day biological production in the overlying water column or on the adjacent land mass (Tao et al., 2015). These less-than-modern ^{14}C ages (i.e., $\Delta^{14}C < 0‰$) imply that there is a contribution of OM from petrogenic sources, and/or that there is a time offset between production and deposition of marine and/or terrestrially derived biospheric OM.

Here we present a comprehensive assessment of the ^{14}C content of OC in both bulk samples and specific grain-size fractions of surface sediments from the northern Chinese marginal seas (CMS), including the Bohai Sea, Yellow Sea, and East China Sea (Fig. 1A). The CMS composes one of largest shallow marginal seas in the world where two major rivers, the Yellow and Yangtze Rivers, discharge vast quantities of sediment (1.08 and 0.5 × 10^9 t/yr, respectively; Yang et al., 2003, and references therein) into an extensive (~1000 km wide, ~3000 km long)

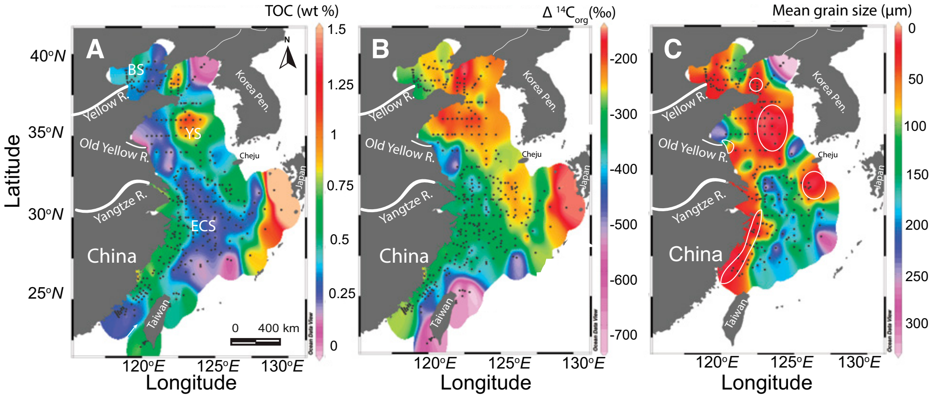

Figure 1. Geographical variations in bulk properties of surface sediments in the Chinese marginal seas (BS—Bohai Sea; YS— YS—Yellow Sea; ECS—East China Sea). A: Total organic carbon (TOC, %). B: $\Delta^{14}C_{org}$ (‰). C: Mean grain size (µm). Black dots represent sample locations. Areas of fine-grained sediment accumulation (red regions in C) match well with those delineated by Qiao et al. (2011). Pen.—peninsula.

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shallow shelf sea where seasonal currents and other hydrodynamic influence exert complex and dynamic controls on the sediment distribution (Chen, 2009) (Fig. 2B). An extensive suite of more than 300 new ^{14}C and ^{13}C measurements are combined with previously published data to yield a detailed picture of the spatial variability in OC characteristics for the CMS. When combined with sedimentological information, they shed new light on hydrodynamic controls on OC age and distribution in this extensive marginal sea system.

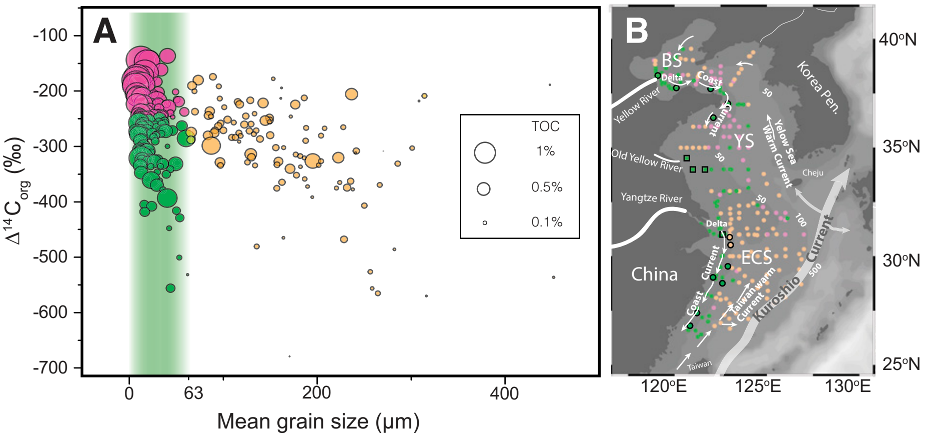

Figure 2. Sedimentological control on bulk total organic carbon (TOC, %) and $\Delta^{14}C_{org}$ (‰) in surface sediments from the Chinese marginal seas (BS—Bohai Sea; YS— YS—Yellow Sea; ECS—East China Sea). A: Relationship between $\Delta^{14}C_{org}$ and mean grain size for surface sediments. Circle size represents approximate TOC (%). Circles represent samples with mean grain size of <63 µm and $\Delta^{14}C_{org}$ <–250‰ (green), mean grain size <63 µm and $\Delta^{14}C_{org}$ >–250‰ (magenta), and mean grain size >63 µm (yellow). Vertical green bar highlights those samples with a mean grain size <63 µm, including those that exhibit depleted $\Delta^{14}C_{org}$ values. B: Locations of the three corresponding sample types. Inner shelf and highly energetic regime samples selected for geochemical analysis of grain-size fractions are highlighted by black circles (n = 12) and squares (n = 4), respectively. Regional circulation patterns are also shown; white arrows indicate the inferred current directions and bathymetric contours (modified from Chen, 2009; Liu et al., 2007). Pen.—peninsula.

METHODS

Surface sediments (0–1, 0–2, or 0–5 cm) were collected from the CMS during different cruises (Table DR1 in the GSA Data Repository[1]). Particle size analysis was performed on bulk sediments after freeze-drying and removal of OM (350 °C for 12 h) using a Mastersizer 2000 (Malvern Instruments Ltd.) laser-diffraction instrument (Geological Institute, ETH-Zürich). For carbon isotopic analyses on size fractions, wet sediment samples were separated into <20, 20–32, and 32–63 µm and coarser fractions using stainless-steel mesh sieves in <1 h (in order to minimize OM losses). Freeze-dried bulk sediment samples and corresponding grain-size fractions were analyzed for OC content and stable carbon isotopic composition at ETH Zürich. Prior to analysis, inorganic carbon was removed from dried samples by fumigation with concentrated HCl (37%, 72 h) and drying over NaOH pellets (72 h) in a desiccator at 60 °C. Radiocarbon analysis was performed at the Laboratory of Ion Beam Physics, ETH Zürich.

RESULTS

Bulk OC ^{14}C contents ($\Delta^{14}C_{org}$) of surface sediments exhibit marked spatial variability (average −305‰ ± 102‰, 1σ; n = 320), with ^{14}C-enriched, i.e., relatively young, OC (−174‰ to −280‰) in the central Yellow Sea and on the outer shelf of the East China Sea, and older OC (−274‰ to −682‰) along the inner edge of the East China Sea, in front of the old and modern Yellow River delta and the Yangtze River delta, and adjacent to the Island of Taiwan (Fig. 1; Table DR1). Total organic carbon (TOC) contents of surface sediments vary from <0.01% to 2.14% (average 0.5% ± 0.3, 1σ; n = 240). Mean grain size of bulk sediments (Fig. 1C) varies from 6.2 µm to 452.8 µm (average 83.7 µm; n = 190).

A subset of 16 samples spanning the nearshore regimes of the CMS was chosen for separation and geochemical characterization of specific grain-size fractions. The samples (black symbols in Fig. 2B) were selected from regions that are proximal to the mouths of the modern Yangtze and Yellow and old Yellow Rivers and also downcurrent from these major river systems. Marked age variability is evident among grain-size fractions derived from the same surface sediment, with $\Delta^{14}C_{org}$ values ranging from −777‰ to −218‰ (n = 48; Table DR2). We also measured $^{14}C_{org}$ of coarser fractions (63–125, 125–250, 250–500, >500 µm) in highly energetic regimes (at H20, H21, H23, P01) with a large range of values (–551‰ ± 148‰; n = 11). Stable carbon isotope compositions ($\delta^{13}C_{org}$ values) also exhibit marked variability among grain-size fractions (–24.6‰ to −20.4‰; n = 48). OC contents of grain-size fractions ranged from 0.07% to 1.22% (n = 48), with smaller size fractions generally characterized by higher TOC values.

DISCUSSION

Hydrodynamic processes are considered to exert strong influence on the type, amount, and dispersal of OC accumulating in sediments of the CMS (DeMaster et al., 1985). The marked heterogeneity in $\Delta^{14}C_{org}$ values of surface sediments does not, however, exhibit a straightforward relationship with grain size (Figs. 1B, 1C, and 2A). Hydrodynamic particle sorting would result in decreasing $\Delta^{14}C_{org}$ values with increasing grain size, and $\Delta^{14}C_{org}$ values are negatively correlated with mean grain size for offshore sediments (Fig. 2A, yellow and magenta; >20 m water depth, $r^2 = 0.53$). This relationship may reflect various factors: diminished preservation of fresh OM on coarser particles due to lower mineral surface area protection (Aller, 1998), sluggish transport of larger particles (Huettel et a., 2014), sediment winnowing processes (Hedges et al., 1999), enhanced OM remineralization as a consequence of greater permeability of coarser sediments (Huettel et al., 2014), or export of sedimentary rock–derived petrogenic OC eroded from Taiwan island via episodic storm events (Hilton et al., 2008). Overall, greater proportions of refractory OM from old carbon sources and protracted lateral transport may account for the greater proportion of pre-aged OC in coarser fractions in deeper waters.

In sharp contrast to this trend, samples dominated by finer grained sediments (mean grain size, <63 µm; Fig. 2A, green and magenta symbols) do not exhibit any correlation between grain size and $\Delta^{14}C_{org}$ ($r^2 = 0.1$), and include a population of samples characterized by significantly pre-aged OC (Fig. 2A, green; $\Delta^{14}C_{org}$ values <–250‰) with OC contents of up to 1.14%. The latter observations may be partly explained by fluvial supply of terrestrial materials containing fossil (^{14}C dead) or pre-aged biospheric OC to the marginal seas (Tao et al., 2015). Plumes of aged OC emanate from these point sources, dispersed by prevailing seasonal current systems in the region (Fig. 1B). However, this cannot be the sole cause of this distribution pattern. For example, suspended OM from the Yangtze River mouth is characterized by higher $\Delta^{14}C_{org}$ values ($\Delta^{14}C_{org}$: −103‰ to −129‰; Wang et al., 2012) relative to those of adjacent inner shelf sediments. Similarly, the average reported $\Delta^{14}C_{org}$ value of Yellow River particulate OM collected across all seasons is −417‰ ± 17‰ (Tao et al., 2015), while in the corresponding deltaic area,

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[1] GSA Data Repository item 2016259, supplementary figures and tables, is available online at www.geosociety.org/pubs/ft2016.htm, or on request from editing@geosociety.org.

we found that $\Delta^{14}C_{org}$ values of the 20–32 μm fraction are lower (–604‰ at B45). The contrast in OC ages between shallow (inner) and deeper (outer) shelves may reflect the influence of hydrodynamic processes where differential particle transport and inherent $\Delta^{14}C_{org}$ variations among grain-size fractions impart changes in bulk sediment characteristics.

Finer grained sediments characterized by older OC (Fig. 2A, green) were derived mostly from shallow inner shelf and subaqueous delta environments that are prone to wind- and tidally-driven sediment resuspension processes (Wang et al., 2011; Yang et al., 2011) (Fig. 2B). Local and regional currents mobilize, entrain, and redistribute sediments (Chen, 2009); satellite images (Fig. DR1) clearly show trajectories of large-scale sediment dispersal. In winter, Yangtze-derived fine-grained sediments are carried southward in the bottom layers by an intensified Chinese Coastal Current, transporting materials parallel to the coastline, forming the muddy regimes (Liu et al., 2007; Yang et al., 2011). These seasonal currents induce sediment sorting, and ^{14}C analyses of OC residing in different grain-size fractions were undertaken on representative samples (n = 16) from shallow regions (<50 m) in an effort to understand hydrodynamic controls on the scatter exhibited in mean grain and $\Delta^{14}C_{org}$ values in finer grained sediments (<63 μm; Fig. 2B, green). For several locations (n = 12) it is evident that the 20–32 μm fraction exhibits lower $\Delta^{14}C_{org}$ than corresponding smaller (<20 μm) and larger (>32 μm) fractions (Fig. 3B, *t*-test, $p < 0.05$). For example, at a Yellow River prodelta location (station B45), the $\Delta^{14}C_{org}$ values of this fraction are ~255‰ and 220‰ lower (~4000 ^{14}C yr older) than those of <20 μm and 32–63 μm fractions, respectively (Table DR2). In some highly energetic regimes local conditions may mobilize coarser materials. For example, in the Yangtze River prodelta (P01) and the region ~200 km northward (e.g., H20, H21, H23), where wave and tidal action promotes vigorous sediment resuspension (Wang et al., 2011), $\Delta^{14}C_{org}$ values of coarser fractions (e.g., 32–63 μm) are lower than corresponding 20–32 μm fractions (Fig. DR2B). The theoretical relationship between critical shear stress and grain size of spherical quartz implies that particle sizes centered around 20 μm have the greatest potential to be eroded and remobilized when compared with the more cohesive clay fractions and the coarser silt and sand fractions (Thomsen and Gust, 2000; McCave and Hall, 2006; Fig. 3A). This corresponds to the sortable silt fraction (10–63 μm) as defined by McCave and Hall (2006). For practical purposes (i.e., sieve mesh sizes), we adopt a slightly narrower range (20–63 μm) here. The relatively ^{14}C-depleted values of OM associated with this sortable silt fraction in subaqueous delta and inner shelf sediments of the CMS are consistent with the influence of particle resuspension processes. Systematic ^{14}C relationships between the sortable silt and other grain-size fractions are evident in 12 samples (*t*-test, $p < 0.05$), but they do not hold across the entire suite of samples investigated ($p > 0.05$). This is likely due to the wide diversity of depositional environments in which other factors such as particle density and shape, flow viscosity, and particle interactions (e.g., aggregation) may play a role (Thomsen and Gust, 2000).

As remobilized particles enter the bottom boundary layer (BBL), they may contribute to the formation of benthic nepheloid layers that can persist for extended periods of time and result in translocation of entrained particles over considerable distances prior to eventual sedimentation and burial. In the Yellow River delta and in the adjacent Bohai Sea, winter storm waves and tidal currents induce enhanced sediment resuspension (to 100 mg/L in the BBL; Yang et al., 2011). Enhanced BBL sediment transport on the East China Sea inner shelf has also been indicated from modeling studies (Bian et al., 2013) and observations (Li et al., 2013). The BBL is characterized by significant physical, chemical, and biological gradients that promote oxic degradation and transformation of labile OM (Keil et al., 2004; Thomsen and Gust, 2000). While it is not known if BBL processes preferentially act upon materials residing in the sortable silt fraction, residual OM within this fraction is likely to become more refractory and increase in ^{14}C age as a consequence of its protracted residence in the benthic nepheloid layer and participation in repeated sediment resuspension-deposition cycles (Aller, 1998; Aller and Blair, 2004). Notably, the pattern of $\delta^{13}C_{org}$ values among grain-size fractions echoes that of $\Delta^{14}C_{org}$, with relatively low values for the 20–32 μm fraction (Fig. 3C; $p < 0.01$). This suggests that during resuspension and lateral transport the sortable silt fraction loses OC (Fig. 3D) and retains a greater proportion of ^{13}C-depleted OC due to preferential mobilization of terrestrial material, selective loss of marine OC relative to terrestrial OC, or enhanced degradation of ^{13}C-enriched marine OM (e.g., hydrolysable amino acids, carbohydrates) relative to more refractory ^{13}C-depleted marine OM components (Hwang and Druffel, 2003). Prevailing and seasonally oscillating coastal currents transport and disperse entrained sediment both northward and southward in the CMS (Chen, 2009), with attendant degradation processes promoting attenuation, aging, and ^{13}C depletion of associated OM. In offshore and deeper water settings, relatively high $\Delta^{14}C_{org}$ values suggest translocation of sediments to distal regions of the CMS by other processes (e.g., near-surface transport; Milliman et al., 1985; Chen, 2009) or direct supply from overlying waters. With respect to the latter, the muddy area southwest of Cheju Island may reflect vertical settling and accumulation of marine OM, as supported by observations of higher chlorophyll-*a* concentrations in surface waters (Fu et al., 2015) and higher $\delta^{13}C$ values of underlying sediments (data not shown).

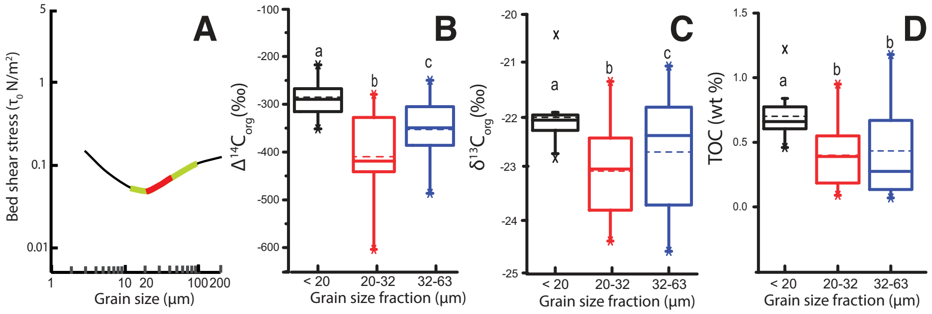

Figure 3. A: Black line shows the idealized behavior of bed shear stress (*y* axis) as a function of grain size (*x* axis) (modified from McCave and Hall, 2006), and green and red curves highlight 10–63 and 20–32 μm grain-size ranges, respectively. B–D: Box and whisker plots of <20, 20–32, and 32–63 μm grain-size fractions in surface sediments from shallow, nearshore regions of the Chinese marginal seas (see text) (Fig. 2B). Dashed and solid horizontal lines in the boxes indicate mean and median value, respectively; upper and lower x symbols and box represent 1% and 99%, 25%, and 75% statistics. Letters (a, b, c) above the boxes indicate a significant statistical difference at the level of $p < 0.05$. B: $\Delta^{14}C_{org}$ (n = 12; Fig. 2B, samples denoted with black circles). C: $\delta^{13}C_{org}$ (n = 16). D: Total organic carbon (TOC, %) values (n = 16).

Overall, our investigation reveals that marked spatial heterogeneity exists in ^{14}C ages of bulk OC and in grain-size fractions from surface sediments of the CMS. This heterogeneity reflects both modern and relict material, and sedimentological influences on OC content and composition. Enrichment of aged OC in the sortable silt fraction of inner shelf sediments is attributed to cyclic resuspension-deposition processes occurring within the BBL. These results shed new light on processes that control the fate and composition of OM delivered to and produced in continental shelf seas, and have implications for carbon cycling and burial in other shallow marginal sea systems (e.g., Dauwe and Middelburg, 1998). With respect to the latter, aging and chemical transformations of OC on shallow and wide continental shelves will confound assessments of OC burial based on simple isotopic mixing models. Assignment of end members based on $\Delta^{14}C$ or $\delta^{13}C$ values of bulk OC and/or specific molecular tracers of source carbon pools may fail to account for these processes during transport, leading to potential errors in source apportionment and in corresponding budgets for OM burial. These processes may also lead to aliasing in organic geochemical proxies in sedimentary records. Moreover, because the degree of transport-associated aging likely varies with sea-level stand due to changing time and length scales of sediment resuspension and redistribution, the magnitude of temporal and spatial offsets between proxy signals associated with different sedimentary phases may also vary. Such processes occurring in continental shelf seas also likely influence the nature of sedimentary OM that is ultimately exported to and buried in sediments accumulating in adjacent ocean basins. Sediment and OC redistribution is by no means restricted to shallow marginal seas (McCave and Hall, 2006; Inthorn et al., 2006).

OM-mineral interactions play a key role on continental shelves, influencing OM reactivity and hydrodynamic properties. Protracted sediment entrainment in cyclic resuspension-deposition loops enhances remineralization of OC, prompting these systems to serve as sources of carbon to the atmosphere, while the refractory OC that does not undergo remineralization is likely to serve as a long-term carbon sink. Overall, the net influence of sediment redistribution processes over continental margins on the carbon cycle, and on continental margin and deep ocean sedimentary archives, remains poorly understood, as does the manner in which it may vary under changing ocean and climate conditions.

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