JGR Biogeosciences

RESEARCH ARTICLE
10.1029/2019JG005107

On the Origin of Aged Sedimentary Organic Matter Along a River-Shelf-Deep Ocean Transect

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Key Points:
- Extensive degradation of pre-old OC occurs in the Yangtze River estuary
- Large OC ^{14}C age heterogeneity is found in northern Okinawa Trough sediment
- Hydrodynamic processes exert a strong influence on ^{14}C age distribution of OM in continental margin sediments

Supporting Information:
- Supporting Information S1

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Citation:
Bao, R., Zhao, M., McNichol, A., Wu, Y., Guo, X., Haghipour, N., & Eglinton, T. I. (2019). On the origin of aged sedimentary organic matter along a river-shelf-deep ocean transect. *Journal of Geophysical Research: Biogeosciences*, *124*, 2582–2594. https://doi.org/10.1029/2019JG005107

Received 25 FEB 2019
Accepted 17 JUL 2019
Accepted article online 12 AUG 2019
Published online 29 AUG 2019

Abstract To assess the influences of carbon sources and transport processes on the ^{14}C age of organic matter (OM) in continental margin sediments, we examined a suite of samples collected along a river-shelf-deep ocean transect in the East China Sea (ECS). Ramped pyrolysis-oxidiation was conducted on suspended particulate matter in the Yangtze River and on surface sediments from the ECS shelf and northern Okinawa Trough. ^{14}C ages were determined on OM decomposition products within different temperature windows. These measurements suggest that extensive amounts of pre-old (i.e., millennial age) organic carbon (OC) are subject to degradation within and beyond the Yangtze River Delta, and this process is accompanied by an exchange of terrestrial and marine OM. These results, combined with fatty acid concentration data, suggest that both the nature and extent of OM preservation/degradation as well as the modes of transport influence the ^{14}C ages of sedimentary OM. Additionally, we find that the age of (thermally) refractory OC increases during across-shelf transport and that the age offset between the lowest and highest temperature OC decomposition fractions also increases along the shelf-to-trough transect. Amplified interfraction spread or ^{14}C heterogeneity is the greatest in the Okinawa Trough. Aged sedimentary OM across the transect may be a consequence of several reasons including fossil OC input, selective degradation of younger OC, hydrodynamic sorting processes, and aging during lateral transport. Consequently, each of them should be considered in assessing the ^{14}C results of sedimentary OM and its implications for the carbon cycle and interpretation of sedimentary records.

1. Introduction

Continental margins comprise the interface between land and deep ocean systems. Examination of changes in content and composition of organic carbon (OC) along across-margin transects on the interface represents a critical approach for understanding its role in the carbon cycle. Radiocarbon (^{14}C) serves as a key property for investigating OC cycle in this complex and dynamic regime. For river-dominated margins, substantial amounts of terrestrial organic matter (OM) are discharged to accumulate in coastal and shelf sediments (Bauer et al., 2013; Bianchi et al., 2017; Keil et al., 1997). This exported OM may derive from different terrestrial OC pools (e.g., fresh vascular plant debris, soil, and fossil OC eroded from sedimentary rocks) that vastly differ in ^{14}C ages and residence times (Bao, Uchida, Zhao, et al., 2018; French et al., 2018; Galy et al., 2015; Wu et al., 2018). In addition, continental shelf sediments also receive amounts of marine OC derived from primary productivity that is generally younger in ^{14}C age by virtue of its recent synthesis and proximal source. The OM accumulating in continental shelf surface sediments can exhibit spatially diverse ^{14}C ages (Bao et al., 2016; Griffith et al., 2010; Tesi et al., 2014; Wu et al., 2013), partly reflecting codeposition of OM from the terrestrial and marine biosphere.

Lateral dispersal of sedimentary OM and associated hydrodynamic processes on continental shelves add another temporal dimension (Bian et al., 2013; Li et al., 2016), further influencing ^{14}C ages of sedimentary OC (Bao, van der Voort, Zhao, et al., 2018; Bao, Uchida, Zhao, et al., 2018; Bröder et al., 2018; Keil

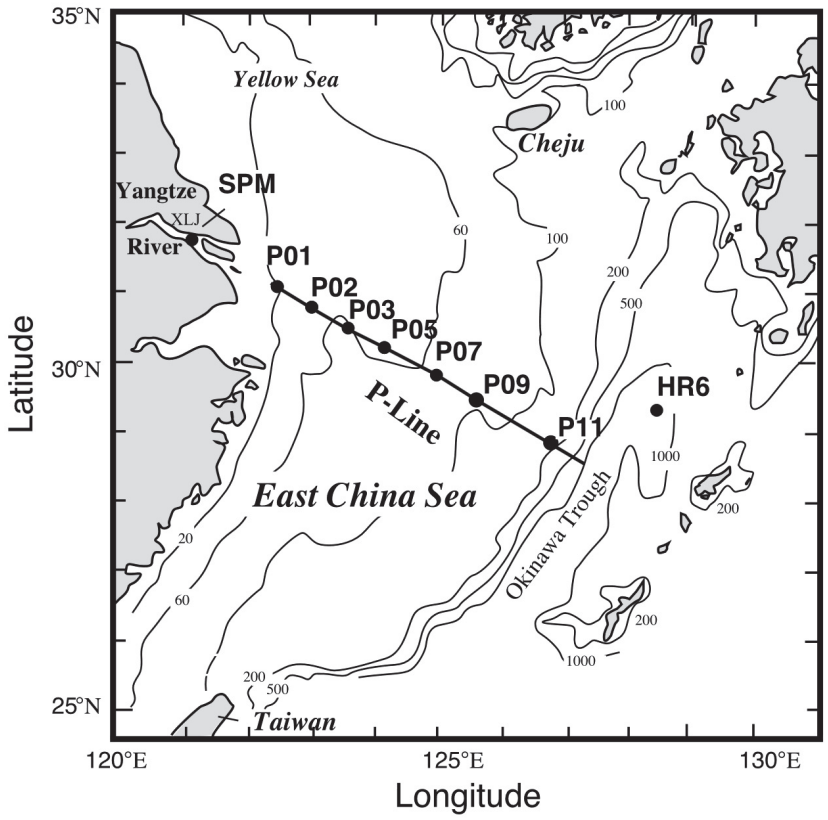

Figure 1. Sample locations in the Yangtze River, East China Sea, and Okinawa Trough (map modified from Oguri et al., 2003). The solid black line shows the P-Line.

et al., 2004; Mollenhauer et al., 2007; Tesi et al., 2016). These processes in marginal seas can increase ^{14}C ages of OM by several millennia; the magnitude of aging depends on many factors such as dispersal distance, current velocity, and sediment grain size (Bao, Zhao, et al., 2019; Bao, Uchida, Zhao, et al., 2018; Bröder et al., 2018). Due to complex and often temporally varying hydrodynamic conditions, these processes can result in markedly spatial variability in sedimentary OC ^{14}C in shallow marginal seas (Bao, Blattmann, et al., 2019). Translocation of sedimentary OM may occur from the shelf to the deep ocean, where it may encounter differing hydrodynamic and depositional regimes (Bao, Strasser, McNichol, et al., 2018; Blattmann et al., 2018; Hung et al., 2003; Hwang et al., 2010; Oguri et al., 2003; Zhu et al., 2006). This complex interplay between source inputs and transport processes renders it challenging to deconvolve underlying carbon cycling, solely based on bulk-level information (incl., ^{14}C) on sedimentary OM.

The East China Sea (ECS) forms an interface between the Yangtze River and Okinawa Trough, receiving substantial inputs of both terrestrial and marine OM (Deng et al., 2006; Li et al., 2012; Wu et al., 2013; Zhu et al., 2013). This broad and shallow marginal sea is characterized by redistribution and remobilization of sedimentary OM (Bao et al., 2016; Deng et al., 2006; Li et al., 2016; Wu et al., 2013; Zhu et al., 2006), and numerous studies have investigated regional changes in carbon biogeochemistry and dynamics along shelf-perpendicular transects (Figure 1, P-Line; Iseki et al., 2003; Oguri et al., 2003; Hung et al., 2003; Zhu et al., 2006). Prior studies have shown that OM in surface sediments along the P-Line primarily derives from marine sources as well as terrestrial materials discharged from the Yangtze River (Deng et al., 2006; Zhu et al., 2006) and that OM can be ultimately exported to the adjacent Okinawa Trough (Iseki et al., 2003; Kao et al., 2003; Oguri et al., 2003). We have conducted an extensive ^{14}C survey of bulk surface sediments at the ECS (Bao et al., 2016); however, ^{14}C-centric investigations of organic components within bulk sediments along such a typical river-shelf-deep

ocean transect have not yet been reported. In particular, in-depth examination of ^{14}C age characteristics of OM from "upstream" (Yangtze River) and "downstream" (Okinawa Trough) locations has not been attempted so far.

Such a systematically detailed ^{14}C investigation along the Yangtze River-ECS-Okinawa Trough transect can disentangle the influences of source- and transport-related factors on sedimentary OC content and composition. In this study, we applied ramped pyrolysis-oxidation (RPO; Rosenheim et al., 2008; Bianchi et al., 2015; Zigah et al., 2017; Bao, Strasser, McNichol, et al., 2018, Bao, McNichol, Hemingway, et al., 2018, Bao, McNichol, McIntyre, et al., 2018) coupled with ^{14}C analysis to suspended particulate matter (SPM) of the Yangtze River, four surface sediment samples along a shelf-perpendicular transect at the ECS (P-Line), and one surface sediment from the northern Okinawa Trough to assess and decipher the cause(s) of compositional and age variability within the source-to-sink system on a regional scale (Figure 1). In addition, we examined source-diagnostic biomarker (n-alkanoic acid) contents of ECS surface sediments along the transect to further constrain the fate of terrestrial OM. Findings from this in-depth assessment of underlying causes of OC ^{14}C age variability along a river-shelf-deep ocean transect carries important implications for our understanding of carbon dynamics on and beyond continental margins.

2. Sample Locations and Methods

2.1. Across-Shelf Transect and Sample Locations

Approximately 47% of the sediments exported by the Yangtze River are deposited in the deltaic area, with the remainder transported to the ECS shelf area (Du et al., 2016; Liu et al., 2006, 2007; Milliman et al., 1985). Although the majority of the material escaping the delta (~32%) is transported southward along the Min-Zhe coast (Du et al., 2016; Liu et al., 2006), a substantial amount (~20%) of terrestrial materials is dispersed offshore and across-shelf (Oguri et al., 2003). Fine sediments and associated OM accumulating in the prodeltaic area are subject to intensive resuspension by hyrodynamically driven processes, and parts of these resuspended materials can be transported to the deep ocean along across-shelf in winter (Oguri et al., 2003; Peng & Hu, 1999; Zhu et al., 2006). Substantial amount of coarse sediments deposits on the middle shelf under the control of tidal currents (Gao & Collins, 2014; Zhu & Chang, 2000). Additionally, sediments derived from the ECS shelf are accumulating in the Okinawa Trough, transported in the benthic layer during winter (Hoshika et al., 2003; Iseki et al., 2003; Oguri et al., 2003). Across-ECS shelf transect connecting to the deep ocean provides a region-scale OC source-to-sink system with a sharp contrast in hydrographic and depositional characteristics, compared to depositing environments of coastal mud belt along the ECS inner shelf (Bao, Zhao, et al., 2019; Li et al., 2012). The former exerts more complex dynamics of sedimentation and ambiguous sources of materials.

SPM from the Yangtze River was collected in July 2012 at Xuliujing (XLJ; 120.93°E, 31.78°N), ~100 km upstream from the Yangtze River mounth (Figure 1). Detailed information about sample site and collection methods was described in Wu et al. (2015). The seven ECS surface sediment (0–2 cm) samples were collected using a box corer along the P-Line from the prodelta to the upper slope on the R/V *Dongfanghong II* in July–August 2013 (Figure 1). The samples are divided into three representative groups by water depth: (i) prodeltaic sediment (P01), with water depths below 30 m and surface sediments mainly consisting of silt and clay. In this regime, sedimentation rate is ~4 cm/year and mixing layer depth is ~5–10 cm (Jia et al., 2018; Liu et al., 2006; Oguri et al., 2003; Su & Huh, 2002); (ii) middle-shelf sediments (P02, P03, P05, and P07), with water depths from 30 to ~100 m, mainly consisting of sands with low sedimentation rate (0.2–0.5 cm/year) and approximately 5-cm mixing layer depth (Huh & Su, 1999; Oguri et al., 2003; Qiao et al., 2017); (iii) outer-shelf and slope area sediments (P09 and P11), with water depths from ~100 to 200 m, consisting of silt and finer sand with lower sedimentation rate (~0.1 cm/year; Huh & Su, 1999). The water depth of the Okinawa Trough is generally greater than 800 m and underlying surface sediments consist of silt to clay, with the extremely low sedimentation and less bioturbation based on excess ^{210}Pb profile in the Okinawa Trough sediments (~0.1 cm/year; Li et al., 1996; Oguri et al., 2003). The surface sediment (0–1 cm,

HR6) sample was collected using a multicorer in the northern Okinawa Trough area (water depth, 1,065 m) on the R/V *Hakuhu Maru* in July 2013. All samples were then stored in the freezer on board until analysis.

2.2. Ramped Pyrolysis and Oxidation

RPO coupled with ^{14}C analysis was developed at the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) facility, Woods Hole Oceanographic Institution (WHOI; Rosenheim et al., 2008; Zigah et al., 2017; Hemingway et al., 2017; Bao, Strasser, McNichol, et al., 2018; Bao, McNichol, Hemingway, et al., 2018; Bao, McNichol, McIntyre, et al., 2018). The RPO technique is able to examine ^{14}C age distribution of different organic constituents within sedimentary OM as a function of their thermochemical stability. During RPO, samples are exposed to a linear temperature gradient; simultaneous oxidation of thermal decomposition products (as discrete temperature windows or fractions) yields CO_2 that can be trapped and subsequently measured for ^{14}C age. The thermogram fingerprint of evolved CO_2 and coupled ^{14}C ages of thermally resolved OC components enables assessment of age variability within sedimentary OM (Bao, McNichol, Hemingway, et al., 2018).

In this case, RPO of the six samples, comprising Yangtze River SPM as well as P-Line and northern Okinawa Trough (SPM, P01, P02, P05, P07, and HR6) surface sediments, was performed at NOSAMS (Figures 2 and 3). Freeze-dried samples were weighed into combusted petri dishes. A beaker filled with 37% HCl was placed at the bottom of a glass desiccator, and samples were placed on a ceramic tray above the acid. The desiccator was evacuated, and the samples were fumigated at 60 °C for 72 hr to remove carbonate. The HCl was subsequently replaced with a beaker containing NaOH pellets, and the dessicator was again evacuated and placed at 60°C, for 72 hr in order to neutralize any excess acid (Bao, McNichol, Hemingway, et al., 2018). The fumigated samples were weighed (~200 mg/sample, $n = 6$) and loaded into a quartz reactor, and heated using a linear temperature program (5°C/min) from 170°C until a maximum of 915°C. Evolved volatile products (thermal decomposition fractions [T_n]) were simultaneously oxidized and removed from the reactor using a carrier gas mixture of O_2 and He (~8% O_2, 35 ml/min total flow rate). A continuous record of evolved CO_2 was obtained via a flow-through infrared CO_2 analyzer (Sable Systems International Inc., CA-10a), before CO_2 derived from thermally volatilized components was sequentially collected in five temperature windows (intervals; 170–320°C [T_1], 320–391°C [T_2], 391–486°C [T_3]; 486–570°C [T_4], and 570–915°C [T_5]). Exceptions include the T_5 windows of P02 and P05, which were collected between 570–650°C and 570–720°C, respectively, due to blocked gas flow during the experiment, and the CO_2 evolved from the 570 to 915°C interval in the P07 was sequentially collected into two parts: 570–829°C and 829–915°C. Resulting CO_2 samples were trapped into precombusted glass tubes with ~50 mg CuO and ~10 mg Ag granules and combusted (525°C, 1 hr) as a final gas purification step at NOSAMS (Bao, McNichol, Hemingway, et al., 2018). The purified CO_2 samples ($n = 31$) were measured for ^{14}C ages using a mini radiocarbon dating System (MICADAS) at ETH Zurich (Ruff et al., 2007). All ^{14}C data are reported as ^{14}C age (year before present [BP]), as defined by Stuiver and Polach (1977).

2.3. Fatty Acid (*n*-Alkanoic acid) Analysis

Freeze-dried P-Line sediments (P01-P11, $n = 7$) were microwave-extracted for 20 min at 100°C (MARS, CEM Corporation) in 9:1 dichloromethane/methanol to yield corresponding total lipid extracts. Each total lipid extract was then saponified with 0.5 g KOH in methanol plus 0.1-ml Milli Q water for 2 hr at 70°C. NaCl (10 ml, 100g/L) was added to the saponified extracts, and then they were back-extracted using hexane (10 ml × 3) to yield "neutral" fractions. Following addition of concentrated HCl (until pH 2), the remaining liquids were then back-extracted using 4:1 hexane/dichloromethane to yield "acid fractions". The acid fractions were methylated to yield the corresponding fatty acid methyl esters (FAMEs) using a mixture of methanol/concentrated HCl (95:5, 70°C for 12 hr). The methylated products were back-extracted using hexane (10 ml × 3), and following blow-down under N_2 gas, the FAMEs were purified by silica gel column chromatography (fully activated) overlain by Na_2SO_4 and sequential elution with 4-ml hexane, 4-ml dichloromethane/hexane (2:1; containing FAMEs), and 40ml dichloromethane. The concentrations of FAMEs in the samples with extra standard (FAMEs) were determined by gas chromatography with flame ionization detector.

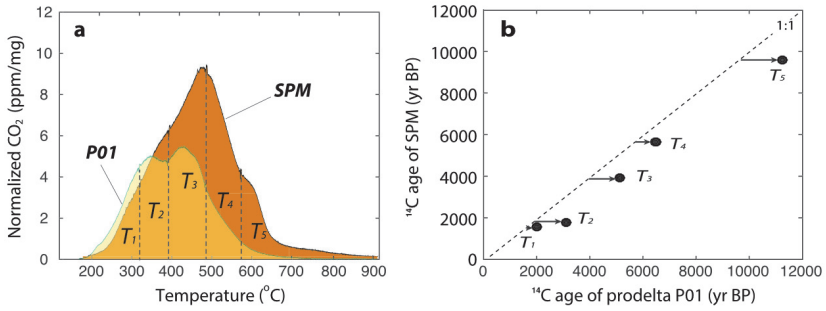

Figure 2. (a) The ramped pyrolysis-oxidation thermogram comparison between suspended particulate matter (SPM; brown) and P01 (yellow) samples. The CO_2 concentrations are normalized by the mass of sediments loaded into the ramped pyrolysis-oxidation instruments. (b) The ^{14}C age comparison of thermal decomposition fractions (T_1, T_2, T_3, T_4, and T_5) between SPM and P01 samples. The arrows show the ^{14}C age changes of corresponding thermal fractions from SPM to P01.

3. Results and Discussion

3.1. Aged OC in the Yangtze River Prodelta

The Yangtze River, the largest river draining into the ECS, discharges ~110 Mt/year sediment (Deng et al., 2006; Qiao et al., 2017; Yang et al., 2015), including ~2.1 Mt/year petrogenic OC (fossil OC, ^{14}C dead; Galy et al., 2015) and ~2.3 Mt/year terrestrial biogenic OC (Galy et al., 2015; Li et al., 2015). Although a substantial fraction of terrestrial OC is remineralized within the delta (Deng et al., 2006; Zhao et al., 2018; Zhu et al., 2013), substantial amounts of OC are delivered into the ECS. The OC loading on Yangtze River SPM (OC:SA, approximately 0.5 ±0.1 C mg/m^2; Wu et al., 2013) is higher than that of the inner shelf of the ECS (0.33 ± 0.11 C mg/m^2, n = 12; Wu et al., 2013) and reported $\delta^{13}C$ values for the former (e.g., < −24‰; Wang et al., 2012) are lower than that of the latter (−22.1‰, n = 16; Wu et al., 2013). This increase in $\delta^{13}C$ value and the decrease in OC:SA from SPM to prodeltaic sediments may be interpreted as both remineralization of terrestrial OM and replacement by marine OM (Keil et al., 1997). In addition, such compositional shifts between SPM and prodeltaic sediments have been used to estimate terrestrial OC burial efficiencies (Blair & Aller, 2012; Goñi et al., 2005; Keil et al., 1997; Wu et al., 2013).

However, we find a decrease in $\Delta^{14}C$ values (~ −110‰, Wang et al., 2012; ~ −200‰, Wu et al., 2018) from Yangtze River SPM to prodeltaic sediment at station P01 (−305‰; Bao et al., 2016), which is not consistent with the estimation of (aged) terrigenous OC loss and replacement with younger marine OC. Sedimentation rate and mixing layer depth in this prodeltaic area are high (~4 cm/year) and ~5–10 cm, respectively (Jia et al., 2018; Liu et al., 2006; Oguri et al., 2003; Su & Huh, 2002). Bioturbation should thus not be major cause for the decreasing in $\Delta^{14}C$ values. We propose that three processes may be potentially responsible for this carbon isotopic discord: (i) preferential remineralization of relatively young terrestrial OC, (ii) replacement with relatively old marine OM, and (iii) OC aging during the lateral transport. In order to examine these possibilities, we compare the thermograms of corresponding thermal decomposition fractions between SPM and P01 samples (Figure 2a). We find a decrease in entire integrated thermogram area (i.e., overall CO_2 yield), especially for higher temperature fractions (e.g., T_3, T_4, and T_5; brown area, Figure 2a) between the river SPM and deltaic sediment, reflecting the overall decrease in OC loadings presumably as a consequence of significant remineralization of OC in this dynamic regime. The OC contributing to the higher temperature fraction is considered to be predominantly refractory OC and fossil OC (Bao, McNichol, Hemingway, et al., 2018; Rosenheim & Galy, 2012), suggesting that a significant proportion of pre-old OC (e.g., ≳4,000 years, based on ages of the $T_{3–5}$, fractions of SPM; Figure 2b) is lost during translocation of sedimentary OM from river to delta. These results are consistent with the observations of previous studies (Bao et al., 2016; Wu et al., 2013). Our results support the possibility of remineralization of both young and old terrestrial OC. Overall, most of the OC loss and bulk-level ^{14}C depletion from SPM to prodeltaic area are accounted for by degradation of relatively younger (compared with remaining old OC) but pre-old (>4,000 years) terrestrial OC.

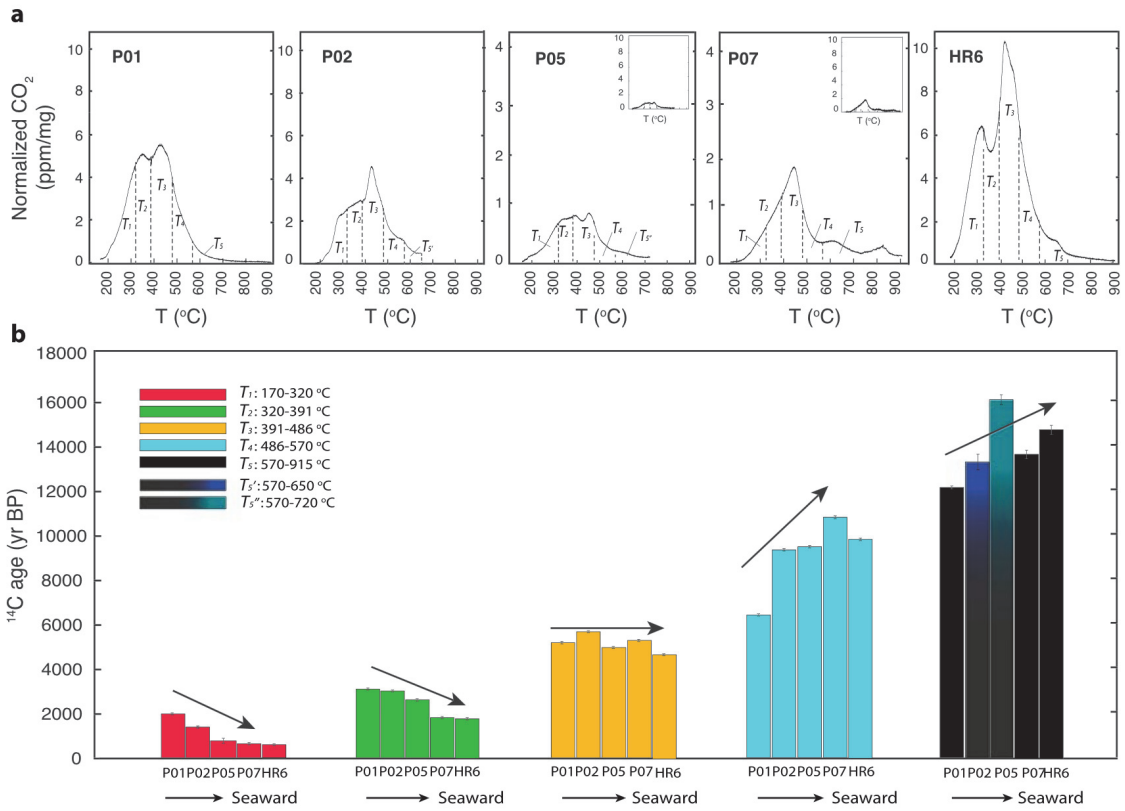

Figure 3. (a) Thermograms of OM from P01, P02, P05, P07, and HR6; inserts in P02 and P07 show the same scale of the *y* axis (mass-normalized CO_2 [ppm/mg]) as other samples. (b) The ^{14}C age variability among thermal windows along the across-shelf transects; the arrows show the general trends. Note that the highest temperature intervals of P02 and P05 were 570–650°C and 570–720°C, respectively. There was incomplete collection of CO_2 due to blocked gas flow during the experiments. The ^{14}C age of T_5 fraction in P07 sample is calculated based on the ^{14}C ages and amounts of two sub-intervals CO_2 for 570–829°C and 829–915°C.

Compared with refractory OC, labile OC tends to decompose at lower temperatures (Bao, McNichol, Hemingway, et al., 2018; Bao, McNichol, McIntyre, et al., 2018; Capel et al., 2005, 2006; Hemingway et al., 2017). In the low-temperature window (e.g., T_1), the CO_2 yield in the thermogram of the prodelta (P01) sample is slightly larger than for the corresponding fraction of river SPM (Figure 2a). This may suggest addition (loading) of labile OC, which decomposes at lower temperatures. Nevertheless, the corresponding ^{14}C age of the T_1 fraction in sample P01 (2005 ±84 year BP) is slightly older than that of SPM (1,560 ± 74 year B; Figure 2b). The radiocarbon age of dissolved inorganic carbon in the Yangtze River estuary is relatively young, ranging from modern to 1,380 ±25 year BP (Wang et al., 2016). These various lines of evidence suggest that the older OC age (lower ^{14}C value) of prodeltaic sediment relative to SPM is not mainly due to addition of pre-old marine OM.

Recent studies have revealed significant aging of sedimentary OC due to its prolonged lateral transport (~4–6 year per kilometer for finer [< 63 μm] sediments; Bröder et al., 2018; Bao, Uchida, Zhao, et al., 2018; Bao, Zhao, et al., 2019). Hydrodynamic processes have been attributed for widespread OC aging observed in marginal sea systems globally, regardless of carbon source (Bao, van der Voort, Zhao, et al., 2018, reference therein). The XLJ SPM sampling location is ~150 km away from P01 location. Station XLJ in the river is strongly tidally influenced; the sedimentary OM transport across the XLJ to P01 locations may be subject bi-directional flows depending on in-situ hydrology and tidal forcing (Zhang et al., 2007, 2017). This could contribute to aging during lateral transport as a consequence of sedimentary OM sloshing transport across the typical tide-dominated delta. Given sediment dynamics and associated hydrodynamic processes across the river-delta interface, it seems that aging of some organic components may occur due to the lateral transport. Overall, we infer that a combination of preferential removal of relatively younger terrestrial OM and hydrodynamically driven aging associated with lateral transport may be responsible for the increased age of sedimentary OM depositing in the prodeltaic area.

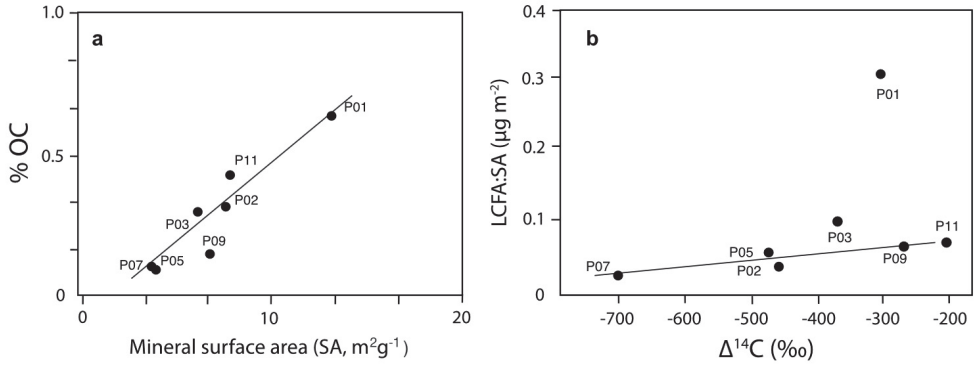

Figure 4. Relationships (a) between OC% and mineral surface area (SA) and (b) between long-chain $C_{24+26+28}$ FAs (LCFA) loadings (normalized to SA) and bulk OC ^{14}C values for P-Line sediment samples.

3.2. Aged OC Depositing Along the P-Line in the ECS

In general, the OC content (OC %) of ECS shelf sediments is related to mineral surface area (SA; Wu et al., 2013; Bao, van der Voort, Zhao, et al., 2018; Figure 4a). Thermograms of the four representative P-Line samples (P01, P02, P05, and P07; Figure 3a) resulted from RPO analyses are distinct, implying that organic constituents comprising each sample differ in both relative proportions and absolute abundances. The differences in OM content and composition influence their corresponding bulk ^{14}C ages. The relatively old ^{14}C ages of OC in ECS shelf surface sediments mainly attribute to inputs of pre-old terrestrial OM (Wu et al., 2013).

In order to assess the preservation of terrestrial OC and its impact on ^{14}C age, we examine the abundances of straight-chain (*n*-) fatty acids (FAs) in the P-Line sediments. Concentrations and distributions of n-FA homologues systematically vary along across the P-Line samples (Figure 5). The C_{16} FA is the most abundant compound, with maximum and minimum concentrations of 8.4 and 0.4 μg/gdw from prodeltaic (P01) and middle shelf (P07) samples, respectively. The concentrations of long-chain $C_{24+26+28}$ FAs (LCFAs), which are considered to mainly derive from terrestrial higher plants (Eglinton & Hamilton, 1967), are the highest in prodeltaic P01 sample, whereas the concentrations of LCFAs in middle-shelf samples (P05 and P07) are the lowest. The former are finer-grained sediments with higher mineral SA compared with the latter, likely suggesting that association with mineral surfaces may be an important factor in controlling the terrestrial OC preservation in sediments along the P-Line. With the exception of the river-proximal P01 sample, SA-normalized LCFA concentrations (i.e., loadings) exhibit a positive relationship with bulk OM ^{14}C values (Figure 4b). This suggests that proportion of (pre-old) terrestrial OM is one of the main reasons influencing on OC ^{14}C ages in ECS shelf sediments, and these bulk ^{14}C ages are thus constrained by similar mechanisms.

The abundance and composition of terrestrial OC in surface sediments are related not only to mineral SA but also to specific hydrodynamic processes associated with its across-shelf transport. The abundance of terrestrial OC has been linked to lateral transport time during pre-depositional processes of sediment remobilization and redistribution (Bao et al., 2016; Bao, Blattmann, et al., 2019; Bao, Uchida, Zhao, et al., 2018; Bao, Zhao, et al., 2019; Bröder et al., 2018; Keil et al., 2004). Since sedimentary OM is heterogeneous, OC constituents may experience different predepositional histories, thus influencing overall (bulk) OC ^{14}C ages in sediment. We find that the ^{14}C ages of the different thermal windows generally increase with increasing temperature (from T_1 to T_5), with a large range in ^{14}C ages between the lowest (T_1, 692 year BP) and highest (T_5, 16149 year BP) temperature intervals (Figure 3b and Table S2). ^{14}C ages of T_1 fractions decrease (i.e., increase in ^{14}C contents) along the seaward transect (red bars, Figure 3b). This likely reflects a combination of less resuspension influence (Bao, Blattmann, et al., 2019), and aged OC removal from labile fractions during lateral transport coupled with addition of young OM derived from vertical export of surface ocean productivity. The latter is consistent with the higher concentrations of short-chain (C_{14}, C_{16}, and C_{18}) FA in the outer shelf and slope settings (P09 and P11; Figure 5) that receive hemipelagic sedimentation (Bi et al., 2018; Li et al., 2016; Oguri et al., 2003; Qiao et al., 2017; Yang et al., 2007).

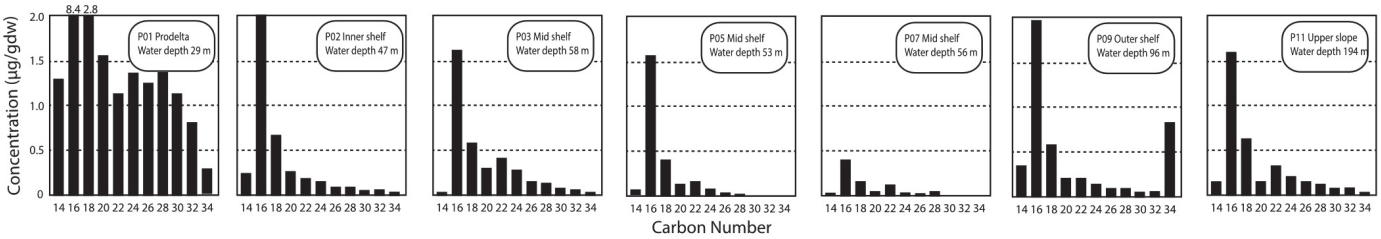

Figure 5. Concentrations of solvent-extractable C_{14}-C_{34} fatty acids (μg/g dry weight) in surface sediments (P01, P02, P03 P05, P07, P09, and P11) from the P-line transect in the East China Sea.

In contrast to the low-temperature windows, ^{14}C ages of higher temperature windows (e.g., T_4 and T_5) generally increase along this transect (Figure 3b). Middle-shelf sediments (P02, P03, P05, and P07) are coarser, and corresponding LCFA concentrations are very low, implying intensive degradation before and following deposition. The remaining OC within these coarser sediments is therefore likely to be relatively refractory compared with that associated with finer particles (Bao, McNichol, Hemingway, et al., 2018; Bao, McNichol, McIntyre, et al., 2018). Nevertheless, the P-Line sediments do not exhibit systematically increasing grain size with increasing across-margin distance. The mode of benthic transport may influence ages of refractory OM (e.g., near-bottom nepheloid layer transport; Bao, Blattmann, et al., 2019). The sediment transport seaward in the bottom layer is observed seasonally (Hoshika et al., 2003; Oguri et al., 2003; Zhu et al., 2006). The overall increasing age of higher temperature fractions (refractory OC) associated with lateral transport along the P-Line may reflect the hydrodynamic regime, although we cannot completely exclude the possibility of aged OC from different sources (e.g., old Yellow River (Saito & Yang, 1995) and relict sediments on the shelf (Deng et al., 2006)) influencing the ages of refractory fractions. To summarize, the contrasting ^{14}C age trends between the low- and high-temperature windows along the P-Line thus suggest that different transport modes influence preservation, distribution, and age of sedimentary OM, highlighting the important control of hydrodynamic processes on the fate of OM in the marginal seas.

3.3. Aged OC in the Okinawa Trough

OM produced and travelling on the ECS shelf can be exported down-slope to the Okinawa Trough (Iseki et al., 2003; Oguri et al., 2003). It has been estimated that at least 0.17 mg C cm^{-2} $year^{-1}$ of sedimentary OM is delivered by lateral transport and accumulates in the trough, compared with 0.25 mg C cm^{-2} $year^{-1}$ supplied via vertical export (Oguri et al., 2003). While the former may derive from several potential sources, including the Yellow River (Katayama & Watanabe, 2003), the old Yellow River (Yuan et al., 2008; Zhao et al., 2019), and Taiwan island (Kao et al., 2003, 2008; Li et al., 2018), some of OC deposited in the Okinawa Trough must originate from the adjacent ECS shelf area (Dou et al., 2010a, 2010b; Honda et al., 2000; Iseki et al., 2003; Oguri et al., 2003).

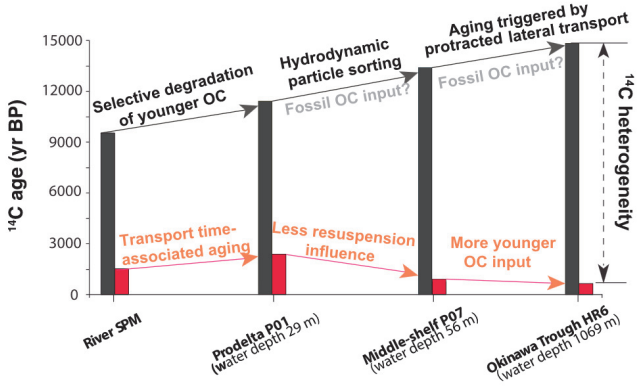

Figure 6. Comparison between T_1 (red bar) and T_5 (black bar) thermal fractions along the land-ocean transect. Due to the incomplete collection of evolved CO_2 for T_5 fractions from P02 and P05, the two samples are not presented.

The ^{14}C ages of sedimentary OM in the surface sediment of Okinawa Trough have not, to our knowledge, been investigated in detail (Honda et al., 2000). Our RPO investigation reveals a large range of ^{14}C ages among thermal fractions of the surface sediment investigated (HR6; Figure 3b). We find that the highest temperature window exhibits a very old age (T_5 fraction: 14,979 ±170 year BP), while its bulk OC ^{14}C is relatively young (2,264 ±82 year BP; Bao et al., 2016). We also find that the corresponding ^{14}C age of T_1 fraction in HR6 sediment is the youngest among those investigated in this study (594 ±75 year BP; Figure 3b). As a result, a large ^{14}C age heterogeneity (corresponding to a 14,385 ±245 years, ^{14}C age offset between minimum and maximum temperature windows) is found in the Okinawa Trough. Notably, the extent of this heterogeneity is larger than that observed from RPO analysis of hadal zone, Japan Trench (>7,500 m) sediments (~4,000 years; Bao, Strasser, McNichol, et al., 2018), suggesting complex OM compositions in the Okinawa Trough. Figure 6 reveals that the spread in ^{14}C ages among

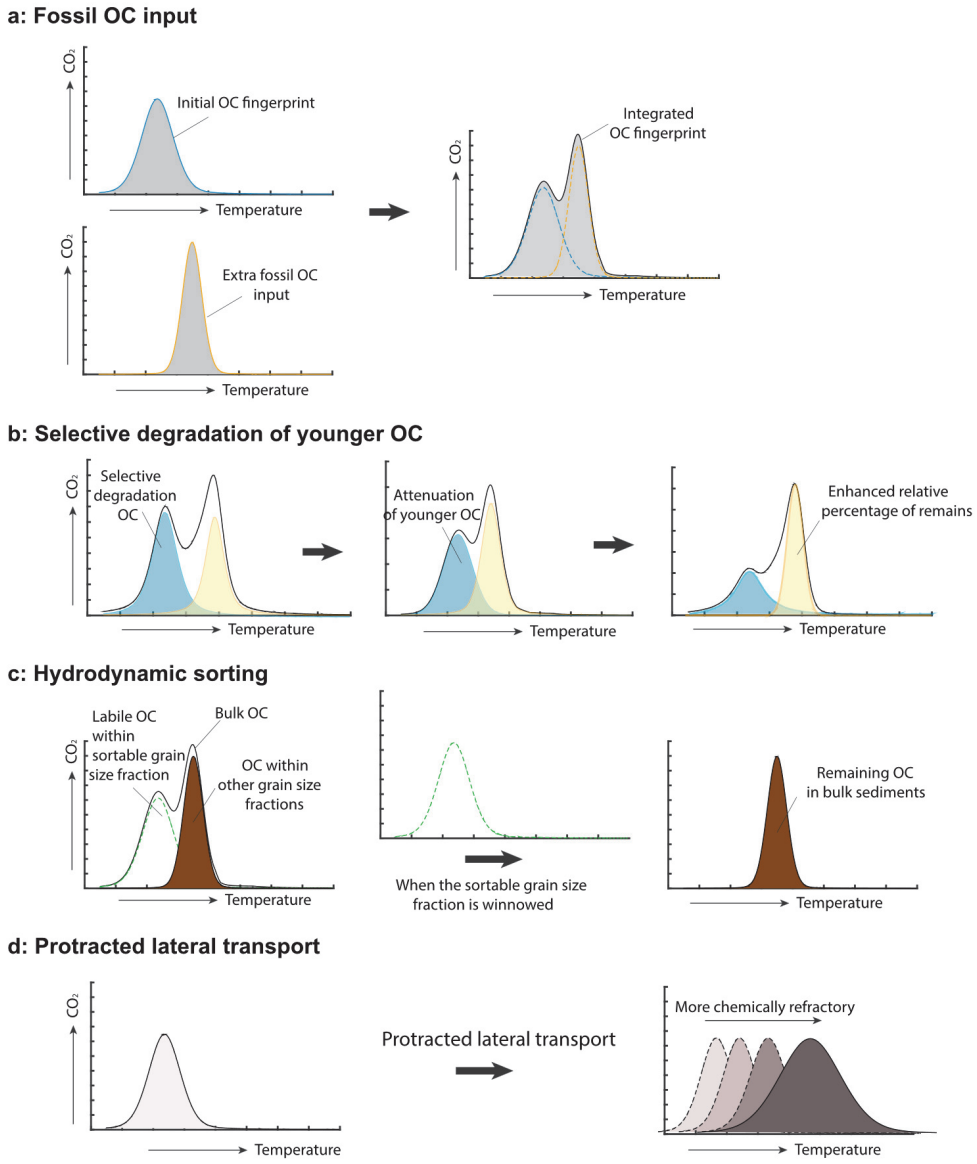

Figure 7. Conceptual schematics showing four scenarios to explain potential origin of increases aged sedimentary organic matter, and their potential manifestation in ramped pyrolysis-oxidation thermograms. (a) fossil organic carbon (OC) input, (b) selective degradation of younger OC, (c) hydrodynamic sorting, and (d) protracted lateral transport.

RPO fractions increases along the river-shelf-deep ocean transect. We thus argue that this amplified ^{14}C heterogeneity in the Okinawa Trough is due to a combination of varying hydrodynamic forcing and different OM inputs (e.g., pre-old OC derived from the ECS shelf/slope; marine OM produced in overlying water column; Bi et al., 2018; and potential aerosols input, Yu et al., 2018; Bao, Niggemann, Luo, et al., 2018).

During prolonged lateral transport, OM may experience ^{14}C aging as a consequence of both selective degradation of relatively labile and young OM, and lateral transport time. Together, these processes manifest themselves as an increase in the proportion and ^{14}C age of high temperature (T_5) fractions corresponding to refractory OM (top black arrows, Figure 6). In contrast, the lowest temperature fraction (T_1) fraction decreases in age, due primarily to addition of recently synthesized and vertically supplied marine OM as well as less-resuspension. Based on the comparison of thermogram of HR6 with those from the P-Line (Figure 3a), we suggest that pre-old and refractory sedimentary OM accumulates in the Okinawa Trough.

Consequently, the increase of aged OM in marginal sea and trough sediments largely depends on its relative abundance and the ^{14}C ages of refractory fractions. Based on our coupled RPO-^{14}C investigations, we propose four potential scenarios that may result in the observed aged sedimentary OM along the transect from the Yangtze River mouth to the Okinawa Trough, and over continental margins, in general. The manner in which these different scenarios may manifest themselves in the RPO thermograms is depicted in Figure 7:

1. Fossil OM: Fossil OC would manifest itself in the higher temperature thermal window from RPO of sedimentary OM (Bao, van der Voort, Zhao, et al., 2018; Kao et al., 2008; Rosenheim & Galy, 2012; Wu et al., 2013). Because of its highly refractory and ^{14}C-dead characteristics, the magnitude and age of this component corresponding to this largely unreactive material would be anticipated to increase, given fossil OM input (Figure 7a).
2. Selective degradation of younger OM: Selective degradation of OM in marine sediments may be influenced by several factors including lateral oxygen exposure time, degree, and mode of association with mineral surfaces, and hydrodynamic regime (Arnarson & Keil, 2007; Bao, Zhao, et al., 2019; Bröder et al., 2018; Keil et al., 2004; Zonneveld et al., 2010). In general, labile OC is preferentially removed and this OM tends to decompose and manifest itself in CO_2 evolved at lower RPO temperatures (Bao, McNichol, Hemingway, et al., 2018; Capel et al., 2005, 2006). This removal of labile OC during lateral transport would result in a relative increase in the proportion of refractory OC contributing to the thermogram (Figure 7b).
3. Hydrodynamic sorting: Specific grain size has a greater propensity for resuspension and redistribution than others during lateral transport (McCave & Hall, 2006), leading to sediment sorting and attendant changes in associated OM. For example, finer-grained sediments hosting younger OM may be preferentially winnowed with older OC retained on the outer shelf of the ECS (Bao, van der Voort, Zhao, et al., 2018), leading to apparent aging of remaining OC (Figure 7c).
4. Protracted lateral transport: During lateral transport, OM that is vulnerable for degradation is preferentially removed, while residual OM persists, the latter is presumably closely associated with mineral surfaces (or insides) (Arnarson & Keil, 2007). These OM-mineral associations may further strengthen as a consequence of protracted engagement (Bao, McNichol, McIntyre, et al., 2018), thereby enhancing OM thermochemical-stability and resulting in a shift of the corresponding thermogram peak to higher temperatures (Figure 7d; Bao, Strasser, McNichol, et al., 2018). More importantly, across- and along-shelf lateral transport times may range from hundreds to thousands of years, depending on transport distance and other factors (Bao, Uchida, Zhao, et al., 2018; Bao, Zhao, et al., 2019). The time component of protracted lateral transport would cause an increase in OC ^{14}C age as a consequence of radioactive decay. The transport-related aging (or decrease in ^{14}C content) of OC requires careful consideration in assessment of source apportionment of sedimentary OC calculated using ^{14}C mass balance approaches (Bao, Uchida, Zhao, et al., 2018).

4. Conclusions

Although the present analyses are mainly based on RPO and ^{14}C measurements of a limited number of sediments recovered along the typical river-shelf-deep ocean transect, they serve to assess the origin of aged sedimentary OM and constraint potentially which scenario is the most important for the fate of OM depositing in different marginal sea environments. We think that this selective degradation of younger OC processes may be main reason for aged OM observed in this interface between the Yangtze River and ECS. Contrastingly, coarser sediments on the middle shelf of ECS (mean grain size: $\gtrsim$200 μm) compared with finer deltaic sediments that are subject to hydrodynamic sorting, indirectly causing different carbon source proportions in sediments. The hydrodynamic sorting is considered as the most direct and important cause for the contrasting change of ^{14}C ages on the shelf compared with that in deltaic area. Fossil OC and OC derived from the ECS shelf both could be carbon source for contemporary sediments in the Okinawa Trough. Wherever the sources of aged OM depositing in the trough are from, nevertheless, they must be subject to long-distance transport. The hydrodynamically driven processes influencing in age should be seriously considered, in particular, for interpretation of millennial-scale paleoceanographic and paleoclimate signals by virtue of organic geochemical proxies in the Okinawa Trough.

Overall, this investigation of ^{14}C age distribution and heterogeneity in sedimentary OM extending from the Yangtze River mouth to the northern Okinawa Trough provides new insights into the cause(s) of compositional and age variability within the region-scale source-to-sink system. We find that apparent aging OM along the across-shelf transect are a consequence of a combination of source variations, selective degradation, and hydrodynamically driven processes (sorting and transport related aging). These causes, which can be closely interlinked, lead to substantial OC ^{14}C age heterogeneity within sediments, in particular, in the Okinawa Trough. The conceptual modes we propose serve as a framework for interpretation of potential causes of aged OC based coupled RPO-^{14}C data.

Acknowledgments
This study was supported by Doc. Mobility Fellowship (P1EZP2_159064; R. B.) from the Swiss National Science Foundation (SNSF). This study was also supported by SNF "CAPS-LOCK" project 200021_140850 (T. I. E.), by the National Natural Science Foundation of China (NSFC; grants 41520104009 and 41630966, M. Z.), and by the "111" project (B13030). We are grateful for support of the NOSAMS staff in the execution of this project. We also appreciate the assistance from Yushuang Zhang (Ocean University of China) at NOSAMS and members of the Laboratory for Ion Beam Physics at ETH Zurich for AMS measurements. We acknowledge Lei Xing, Haidong Zhang, Guodong Song, Meng Yu, Yonghao Jia, and Shanshan Duan (Ocean University of China) for sampling assistance on the cruises. Assistance at sea by the crews of R/V *Dongfanghong II* and R/V *Hakuhu Maru* is also acknowledged. Readers can access or find the data from figures and tables in the supporting information.

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