

RESEARCH LETTER

10.1029/2018GL078904

Key Points:

- Significant OC ¹⁴C aging occurs due to lateral transport over continental shelves
- Different grain size fraction sediments exhibit variable degrees of OC ¹⁴C aging
- Hydrodynamic processes exert widespread influence on OC dynamics and burial on continental margins

Supporting Information:

- Supporting Information S1

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Citation:

Bao, R., Uchida, M., Zhao, M., Haghypour, N., Montlucon, D., McNichol, A., et al. (2018). Organic carbon aging during across-shelf transport. *Geophysical Research Letters*, 45, 8425–8434. <https://doi.org/10.1029/2018GL078904>

Received 26 MAY 2018

Accepted 31 JUL 2018

Accepted article online 7 AUG 2018

Published online 22 AUG 2018

Organic Carbon Aging During Across-Shelf Transport

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Abstract Compound-specific radiocarbon analysis was performed on different grain-size fractions of surficial sediments to examine and compare lateral transport times (LTTs) of organic carbon. ¹⁴C aging of long-chain leaf wax fatty acids along two dispersal pathways of fluvially derived material on adjacent continental margins implies LTTs over distances of ~30 to 500 km that range from hundreds to thousands of years. The magnitude of aging differs among grain size fractions. Our finding suggests that LTTs vary both temporally and spatially as a function of the specific properties of different continental shelf settings. Observations suggest that ¹⁴C aging is widespread during lateral transport over continental shelves, with hydrodynamic particle sorting inducing age variations among organic components residing in different grain sizes. Consideration of these phenomena is of importance for understanding carbon cycle processes and interpretation on sedimentary records on continental margins.

Plain Language Summary The radiocarbon age of organic matter accumulating in sediments on continental margins can shed light on processes that are involved in carbon cycling in these dynamic marine environments. However, it has proven challenging to constrain the role of hydrodynamic processes on observed radiocarbon ages of sedimentary organic matter. In this study, we examine the radiocarbon ages of source-specific biomarker compounds in different grain-size fractions of surface sediments from two shelf systems in the North Pacific Ocean. We find different patterns of aging among grain size fractions that are attributed to the contrasting hydrodynamic characteristics of the continental shelves. The aging of organic matter during lateral transport across continental shelves emerges as a widespread phenomenon, comprising an important facet of the marine carbon cycle.

1. Introduction

There is a growing body of evidence that organic matter (OM) accumulating in continental margin sediments is highly heterogeneous not only in terms of its origin and chemical composition but also with respect to radiocarbon (¹⁴C) contents (Bao et al., 2016; Blair & Aller, 2012; Eglinton et al., 1997; Griffith et al., 2010). While ¹⁴C contents in sedimentary organic carbon (OC) can shed light on OM ages and dynamics in the marine environment, numerous underlying factors may contribute to observed ¹⁴C signatures. For example, several studies have illustrated that different subfractions (e.g., grain-size fractions) of bulk sediment exhibit distinct OC ¹⁴C characteristics (Arnarson & Keil, 2007; Bao et al., 2016; Megens et al., 2002; Tesi et al., 2016; Wakeham & Canuel, 2016; Wakeham et al., 2009).

Although typically viewed in terms of vertical supply and accumulation of OM, there is increasing evidence that OM accumulating in continental shelf sediments experiences widespread lateral dispersal, resulting in translocation from source regions (Benthien & Müller, 2000; Inthorn et al., 2006; Mollenhauer et al., 2007; Ohkouchi et al., 2002; Uchida et al., 2001, 2005; Wagner et al., 2014). ¹⁴C contents of sedimentary OM may thus reflect not only sources with diverse ages (e.g., incorporation of ¹⁴C-rich marine OM and/or ¹⁴C-dead fossil OM; Hilton et al., 2008; McNichol & Aluwihare, 2007) but also redistribution processes that induce OC “aging” via radioactive decay prior to deposition. The magnitude of this latter, lateral transport-associated

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Table 1

Results of Compound-Specific Radiocarbon Analysis (CSRA) and Lateral Transport Times (LTTs) in This Study and Observed ^{14}C Aging of Alkenones in the Oceans Attributed to Lateral Transport

Locations	Transect distance (km)	Water depth (m)	Grain size fraction (μm)	Compound/group	^{14}C age (yr) in upstream (M1/WM3)	^{14}C age (yr) in downstream (H1/WM4)	LTT (yr)	Transport speed (km/yr)	Reference
(M1 and H1) BY transect	~500	~20	<20	$\text{C}_{26} + 28 + 30$	$3,392 \pm 61$	$3,624 \pm 56$	232 ± 117	2.16	This study
Bohai-Yellow Seas			20–32	$\text{C}_{26} + 28 + 30$	$2,165 \pm 90^{\text{b}}$	$3,467 \pm 130$	$1,302 \pm 220$	0.38	This study
			32–63	$\text{C}_{26} + 28 + 30$	$2,165 \pm 90^{\text{b}}$	$3,945 \pm 155$	$1,780 \pm 245$	0.28	This study
			>63	$\text{C}_{24} + 26 + 28 + 30$	763 ± 128	$1,495 \pm 202^{\text{c}}$	732 ± 328	0.68	This study
(WM3 and WM4) WM transect	~35	143–644	<38	C_{26}	$1,350 \pm 130$	$1,780 \pm 85$	430 ± 215	0.08	This study
Washington Margin			38–63	C_{26}	$1,460 \pm 110$	$2,030 \pm 100$	570 ± 210	0.06	This study
Laptev Sea Shelf	~600	4–92	63–250 < 63 μm	$\text{C}_{24} + 26 + 28$ $\text{C}_{24} + 26 + 28 + 30$	$2,230 \pm 110^{\text{a}}$	$3,070 \pm 230$	840 ± 340 $3,600 \pm 300$	0.04 0.17	This study Bröder et al., 2018
					Compound	Description of asynchronous ^{14}C aging of compound			
Bermuda Rise		4,517	Bulk	Alkenones	~7,000 years older than coexisting planktonic foraminifera				Ohkouchi et al., 2002
Namibian Margin		98–1,821	Bulk	Alkenones	1,000–4,500 years older than coexisting foraminifera				Mollenhauer et al., 2003
Chilean Margin		852	Bulk	Alkenones	~1,000 years older than coexisting foraminifera				Mollenhauer et al., 2005
California Borderlands		420–900	Bulk	Alkenones	~1,000 years older than coexisting foraminifera				Mollenhauer & Eglinton, 2007

^aSample fraction is 125–250 μm . Additionally, the ^{14}C ages of C_{26} in 63–150 μm fraction and bulk in WM3 sample are $1,750 \pm 150$ yr and $3,050 \pm 160$ yr, respectively. ^b ^{14}C age of $\text{C}_{24} + 26 + 28 + 30$ in combined 20–32 μm and 32–63 μm fractions. ^cIntegrated ^{14}C ages of C_{24} (876 ± 220 yr) and $\text{C}_{26} + 28 + 30$ ($2,732 \pm 165$ yr) in >63- μm fraction from H1 sample based on the similar mixing ratios (1:0.5) of combined compounds ($\text{C}_{24} + 26 + 28 + 30$) in >63- μm fraction from M1 sample.

aging, remains poorly constrained, despite its potential importance for carbon cycling and its ramifications for interpretation of sedimentary records. In particular, relationships between the extent of lateral transport-induced aging and continental margin setting remain unexplored. One key challenge in addressing this issue is the admixture of OM from marine and terrestrial productivity, as well as potential inputs from fossil (petrogenic or anthropogenic) sources to continental margin sediments that complicate the interpretation of bulk OC ^{14}C ages. Potentially large ^{14}C variations among these different components confound attempts to decipher lateral transport times (LTTs) of OC.

Furthermore, hydrodynamic particle sorting may induce OC ^{14}C variations among sediment grain-size fractions (Bao et al., 2016), implying that changes in grain-size distribution may also influence bulk OC ^{14}C ages. Consequently, while prior studies have established the occurrence of ^{14}C aging during redistribution of sedimentary OM on continental margins (Inthorn et al., 2006; Keil et al., 2004; Mollenhauer et al., 2007; Ohkouchi et al., 2002; Uchida et al., 2001, 2005; Table 1), direct assessment of LTTs has remained elusive (Keil et al., 2004). One promising approach is to determine changes in ^{14}C content of source-specific terrestrial biomarkers along a sediment dispersal pathway, such as emanating from a fluvial point source (Bröder et al., 2018; Keil et al., 2004). However, such molecular-level ^{14}C measurements should also be made on specific grain-size fractions in order to obviate hydrodynamic influences.

In an effort to constrain associated LTT of OM over continental margins, we examine ^{14}C age changes of terrestrial higher plant-derived long-chain leaf wax fatty acids (LCFAs) in specific grain-size fractions along two sediment dispersal pathways representative of passive and active margin settings, both bordering the north Pacific Ocean. One pathway follows Yellow River (Huanghe) sediments transported from the subaqueous pro-delta along the inner shelf of the Bohai and Yellow Seas (abbr.: BY transect, approx. longitudinal distance: ~500 km; Tao et al., 2016; Yang et al., 2007; Zeng et al., 2015); the second follows the

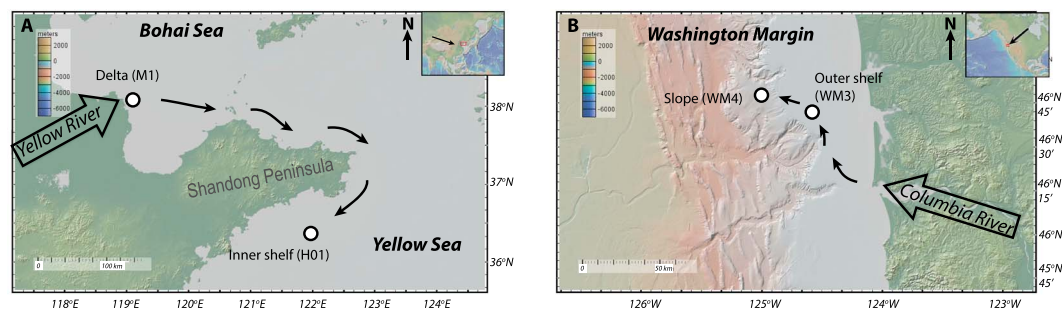


Figure 1. Sample locations in (a) the Bohai-Yellow Seas and (b) the Washington Margin. The arrows show general sediment transport directions.

dispersal pathway of the Columbia River sediment plume from the outer shelf (proximal to the river outflow) to the upper slope (abbr.: WM transect, shortest distance: ~35 km; Coppola et al., 2007; Keil et al., 2004; Nittrouer et al., 1979; Prahl et al., 1994) on the Washington margin (Figure 1). In both cases, the dominant fluvial point source of terrestrial sediment and associated OC implies that other inputs (e.g., direct runoff or eolian transport) are unlikely to influence observations. Compound-specific radiocarbon analysis on different grain-size fractions allows hydrodynamically driven particle sorting effects to be minimized and also limits potential effects of selective degradation of target biomarkers afforded by varying degrees of physical protection through association with mineral surfaces (e.g., Dickens et al., 2006; Keil et al., 1994).

2. Materials and Methods

Two surface sediment (0–2 cm) samples along the BY transect from the Yellow River pro-delta, Bohai Sea (M1: 119.04°E, 38.23°N, 20-m water depth) and the inner shelf along the coastal region of the Yellow Sea (H1: 122.12°E, 36.37°N, 21-m water depth) were collected using a box corer during cruises of R/V *Dongfanghong II* in June and August 2013. Surface sediment (0–2 cm) samples along the WM transect were collected from the outer shelf (WM3: 124.60°W, 46.70°N, 143-m water depth) and upper slope (WM4: 125.00°W, 46.80°N, 644-m water depth) of the Washington margin using a box corer during cruises of R/V *New Horizon* in May–June 2001.

Surface sediments (~2-kg wet weight) from BY transect were stored frozen (–20 °C) after collection. The wet sediment was passed through 20, 32, and 63- μ m sieves and resulting size fractions subsequently freeze-dried prior to analysis at ETH Zurich (Bao et al., 2016). For WM transect samples, sediment (>1-kg wet weight) was wet-sieved into different size fractions (<38, 38–63, 63–150, or 63–250 μ m) at Woods Hole Oceanographic Institution (WHOI) following protocol similar description of Coppola et al. (2007), and then freeze-dried prior to work-up and subsequent analysis at WHOI.

Sediment samples were solvent extracted, and the extract was subsequently purified to obtain a fraction containing straight-chain fatty acids (*n*-FAs). Following conversion to their corresponding fatty acid methyl esters, individual FAs or groups of FAs were then isolated by preparative capillary gas chromatography using established methods (cf., Dickens et al., 2006; Eglinton et al., 1996; Tao et al., 2016). Radiocarbon data of M1 and H1 samples were obtained using a MICADAS Accelerator Mass Spectrometry (AMS) system equipped with a gas ion source (Ruff et al., 2007) at the Laboratory for Ion Beam Physics, ETH Zurich (Synal et al., 2007; Wacker et al., 2010). Radiocarbon measurements of WM3 and WM4 samples were performed at the National Ocean Sciences AMS facility at WHOI. Details of experiment methods are provided in the Supplementary Materials (Drenzek et al., 2007; McNichol et al., 1994).

3. Results and Discussion

3.1. Aging of Sedimentary OM Along Dispersal Pathways

14 C ages of long-chain (> C_{24}) *n*-FAs isolated from different grain-size fractions of surface sediments increase from the subaqueous delta (M1) to inner shelf station H1 of the BY transect by an average of $1,011 \pm 673$ yr

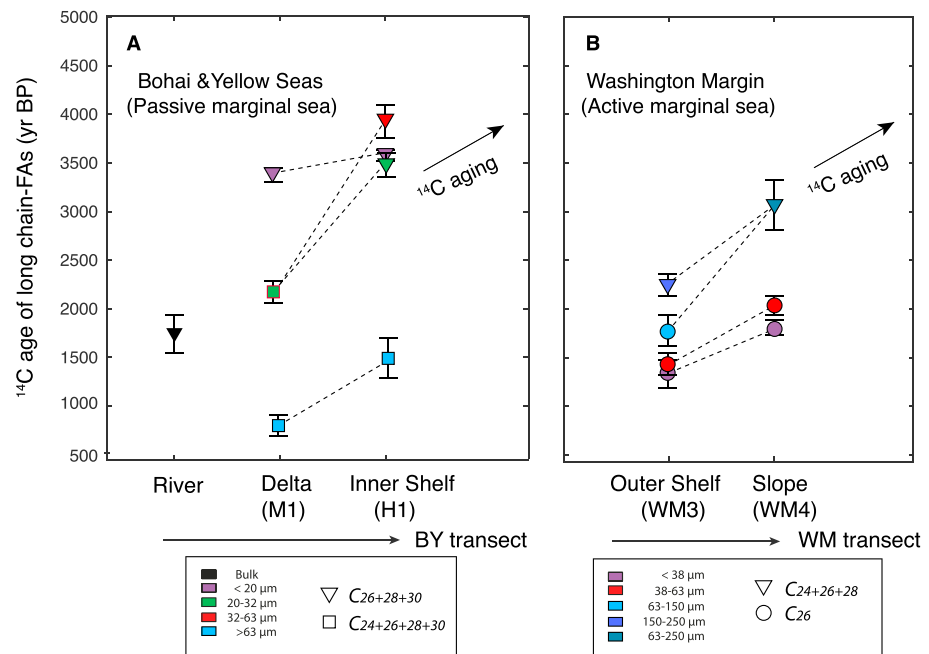


Figure 2. ^{14}C ages of long-chain (C_{26} , $C_{26} + 28 + 30$, $C_{24} + 26 + 28 + 30$, and $C_{24} + 26 + 28$) fatty acids measured in different grain-size fractions of surface sediments recovered along fluvial sediment dispersal pathways in (a) the Bohai-Yellow Seas (BY) and (b) the Washington margin (WM). The arrows depict ^{14}C “aging” of these terrestrially derived biomarkers in corresponding grain-size fractions along the BY transect from delta (M1) to inner shelf (H1), and the WM transect from outer shelf (WM3) to upper slope (WM4). Due to limited sample size, long-chain FAs from 63 to 150 μm and 150 to 250 μm fractions in WM4 were combined for FA extraction and ^{14}C analysis. Corresponding FA ^{14}C data for the Yellow River bulk suspended sediment (~ 50 km upstream of the Yellow River delta) are from Tao et al. (2015). 2σ errors are shown for each data point after calibration.

($n = 4$), and from the outer shelf (WM3) to the upper slope (WM4) along the WM transect by 613 ± 208 yr ($n = 3$; Figure 2). In fluvially dominated sediments, this group of compounds is generally considered to derive almost exclusively from terrestrial higher plant leaf waxes (Tao et al., 2015, 2016, references therein). Thus, by measuring ^{14}C ages of these biomarkers, interferences from fossil (petrogenic OC) and marine OC inputs along the transport pathway can be excluded. The dispersal pathways are located in two river-dominated continental shelf settings where coastal currents dominate circulation and advective processes (Coppola et al., 2007; Dickens et al., 2004; Li et al., 2016). Consequently, the observed offset of LCFA ^{14}C ages at different locations along the dispersal pathway is considered to directly reflect OC aging by radioactive decay during lateral transport.

The observed increase in OC ages accompanying cross-shelf transport is consistent with estimates of transport-related oxygen exposure times for sedimentary OM of up to 1,800 years associated with ~ 150 -km transit from the shelf (water depth: ~ 100 m) to the continental rise ($> 2,740$ m) on the Washington Margin (Keil et al., 2004). Recently, Bröder et al. (2018) measured plant wax ^{14}C ages in surface sediments spanning the 600-km-wide Laptev Sea shelf and calculated a LTT for terrestrially derived OC of $\sim 3,600$ years. Both of these prior studies, suggesting that LTTs over continental margins may be millennial in scale, were carried several assumptions. In particular, hydrodynamic effects that may induce differential particle transport (particle sorting) and selective degradation/preservation of OM associated with grain-size fractions were not considered. Given (i) the changes in hydrodynamic regime between shallower (coastal) and deeper (more distal) depositional settings (Nittrouer & Wright, 1994), (ii) the established relationships between current velocity (shear stress) and particle resuspension as a function of grain size (Thomsen & Gust, 2000), and (iii) the strong links between OM reactivity and mineral surface area and grain size (Mayer, 1994), such effects may exert significant influence on ^{14}C changes observed in OM in bulk sediment phases.

In the shallow Chinese marginal seas, transport-related aging of associated OM as a consequence of cyclic deposition-resuspension processes is likely induced by seasonally oscillating southward and northward currents (Chen, 2009), with this bidirectional flow prolonging transport times of suspended sediments and associated OM. Similar hydrodynamic processes are likely operative on the Washington margin (Coppola et al., 2007) where sediments emanating from the Columbia River are hydrodynamically sorted during dispersal and deposited as texturally distinct bands orientated parallel to the coastline and finally transported to the deep ocean, driven by prevailing bottom currents (Keil et al., 1994; Prahl et al., 1994; Sternberg, 1986). Taken together, these observations suggest that hydrodynamic processes could trigger marked ^{14}C aging of sedimentary OM on continental margins.

The magnitude of OC aging induced by lateral transport determined from ^{14}C ages of OM differs sharply from the estimates of material transport determined from other approaches. For example, by tracing fluvial (Columbia River) dispersal of ash derived from the 1980 Mount St. Helens eruption, Ridge and Carson (1987) found evidence for very rapid transport of fine-grained ($<22\ \mu\text{m}$) sediment (i.e., $>70\ \text{km/yr}$) across the Washington margin. These contrasting findings suggest either that OM transport is at least partially decoupled from that of inorganic matter or that there are different populations of components that exhibit a broad spectrum of transit velocities. We consider each of these in turn below.

With respect to the first explanation, aggregation/disaggregation and association/dissociation of OM with inorganic materials during sediment deposition and resuspension (Thomsen & Van Weering, 1998) may retard OM transport. Enhanced aging may occur due to ^{14}C radioactive decay of sedimentary OM entrained in repeated deposition-resuspension loops in the benthic boundary layer induced by local hydrodynamic processes. A single deposition-resuspension loop may be up to 1–2 months in duration (van der Loeff et al., 2002), and mean residence times of freshly deposited OM in surface seabed sediments prior to resuspension may be on the order of years (McCave, 2009; McKee et al., 1983). Resuspended particles can experience numerous such episodes of emplacement/displacement between OM and inorganic particles due to sediment winnowing and reworking following short-term deposition (Hedges et al., 1999; Tao et al., 2016) and associated benthic nepheloid layer transport prior to permanent sedimentation (McKee et al., 1983).

Regarding the second explanation, terrestrial storage times and mobilization pathways may vary for different components of higher plant-derived OM, resulting in a spectrum of ages for different organic components (e.g., Feng et al., 2015). Riverine export of bomb ^{14}C incorporated into vascular plant biomass, including leaf wax FAs, can be rapidly mobilized (years) or retained within terrestrial watersheds for millennia prior to export to the ocean. The former stems from direct, surface runoff and transfer to fluvial networks, whereas the latter reflects protracted storage in mineral soils prior to mobilization and discharge (e.g., French et al., 2018; Tao et al., 2015). This mineral-associated component of terrestrial OM is considered to predominate in BY (Tao et al., 2016) as well as other river-influenced continental margin sediments (e.g., French et al., 2018; Holtvoeth et al., 2005). It is likely that ashfall from a volcanic eruption may be subject to a similar fate, with some material rapidly mobilized and exported offshore, giving rise to the observation of rapid transit times, while other material retained on the landscape. There may be a similar pool of vascular plant derived OM; however, the ^{14}C signature for this pool is likely overwhelmed by the much larger, more refractory pool of mineral-associated plant waxes (French et al., 2018) that becomes entrained in benthic nepheloid layer transport. Consequently, the LTT estimated from the offset of ^{14}C ages of LCFAs should be considered to reflect an integrated signature of more rapidly and slowly traveling components, highlighting the complexity and multiple pathways of lateral transport of OM.

Sediment mixing due to bioturbation could also serve as an explanation for the observed offsets in plant wax ^{14}C ages between stations (Griffith et al., 2010). For the YS transect, the inner shelf sample (H1) exhibits lower sedimentation rates ($\sim 50\ \text{mm/yr}$) than that of delta sample ($>200\ \text{mm/yr}$; Qiao et al., 2017). However, these high sedimentation rates and relatively shallow sediment mixed layer depths ($\sim 5\ \text{cm}$; Su & Huh, 2002) suggest that bioturbational mixing is unlikely to result in any significant time offset (Alexander et al., 1991; Li et al., 2006; Qiao et al., 2017). Furthermore, OM (incl. biomarker) concentrations typically decrease exponentially over the upper few centimeters of marine sediments due to degradation accompanying early diagenesis (Guo et al., 2015), suggesting that OM mixed upward from deeper sediment layers would be overwhelmed by that of freshly deposited material. For the WM transect, relatively low bottom water oxygen concentrations are observed at stations WM3 and WM4 (2.3 mg/L, 1.1 mg/L, respectively), limiting selective

OC degradation during benthic transport of sedimentary OM (McKee et al., 2004; Zonneveld et al., 2010), and underlying sediments are characterized by shallow oxygen penetration depths (4 and 6 mm, respectively, Coppola et al., 2007). Given sediment accumulation rates of 2.9 ± 0.3 and 1.1 ± 0.1 mm/yr at the outer shelf and upper slope sites, respectively (Coppola et al., 2007), this corresponds to relatively short oxygen exposure times (~ 1.5 and 5.5 years, respectively). We therefore conclude that potential time offsets resulting from bioturbation at both locations should be much shorter (< 10 years) than expected LTT and ^{14}C age measurement error. Moreover, given that mineral surface area-normalized concentrations or loadings of high-molecular weight (FAs: $\text{C}_{20}\text{--}\text{C}_{31}$) among the grain-size fractions do not exhibit decreasing along two BY and WM transects (Table S1 in the supporting information), selective degradation of plant wax FAs during bioturbation is considered unlikely to contribute significantly to the observed ^{14}C aging. We therefore conclude that lateral transport as the most plausible explanation for the observed age offsets.

3.2. Variable OC Aging Among Grain-Size Fractions

Given that settling velocity and propensity for resuspension varies with grain size, OM associated with different sedimentary components may be subject to differential transport under the same hydrodynamic conditions (McCave, 1988). For the BY transect, LCFA ^{14}C ages ($\text{C}_{26+28+30}$, $\text{C}_{24+26+28+30}$) systematically increase from delta to inner shelf; however, those associated with the sortable silt (e.g., $20\text{--}32\ \mu\text{m}$, $32\text{--}63\ \mu\text{m}$ fraction) exhibit the largest age increase (1,300 years, equivalent to an average transport speed of $0.38\ \text{km/yr}$ and $\sim 1,800$ years $0.28\ \text{km/yr}$, respectively), while those from the $< 20\text{-}$ and $> 63\text{-}\mu\text{m}$ fraction exhibit the smallest age increase (~ 200 years, $2.16\ \text{km/yr}$; ~ 700 years; $0.68\ \text{km/yr}$, respectively; Table 1). We attribute this differential aging of LCFAs to the dynamic properties of their host grain-size fractions, with enhanced aging of OM associated with the “sortable silt” ($20\text{--}63\ \mu\text{m}$) fraction reflecting its greater propensity to undergo resuspension and mobilization relative to both smaller ($< 20\ \mu\text{m}$) and larger ($> 63\ \mu\text{m}$) grain-size fractions under the hydrodynamic regime of this shallow and energetic marginal sea setting (Bao et al., 2016). Sediment sorting by seasonally oscillating currents may enhance this preaged signal for OM associated with the sortable silt fractions.

The observations from the BY transect contrast with those from the WM transect, where coarser fractions ($63\text{--}250\ \mu\text{m}$) of the latter exhibit a greater age increase (~ 800 years; $0.04\ \text{km/yr}$) from outer shelf to slope than finer ($< 38\ \mu\text{m}$) fractions (~ 400 years; $0.08\ \text{km/yr}$; Table 1). These deeper, lower energy and less oscillatory hydrodynamic regimes are less conducive to rapid seaward export of coarser-grained sediments, and sluggish bed load movement prevails as the dominant transport mechanism (Hickey & Banas, 2003). The results in a greater ^{14}C age increase in OM associated with coarser compared to finer-grained sediments in distal settings (Bao et al., 2018). Indeed, the relatively young C_{26} FA ^{14}C age of the $< 150\text{-}\mu\text{m}$ fractions compared to that of bulk sediment ($3,050 \pm 160\ ^{14}\text{C yr}$) suggests that the ^{14}C age of C_{26} FA in coarser ($> 150\text{-}\mu\text{m}$) fractions—while too low in concentration to measure—should be substantially older. We conclude, therefore, that the local hydrodynamic regime exerts an important influence on the extent, nature, and variation of ^{14}C aging among grain-size fractions of continental margin sediments.

Seasonal variations in the ^{14}C contents of OC in suspended particle matter exported from the Yellow River to the Bohai Sea are relatively invariant ($\Delta^{14}\text{C}$: -417 ± 17 [SD]‰, Tao et al., 2015), despite significant variability in flux (the latter is highest in summer). The median grain size of suspended particle matter collected over 1 year is $\sim 8\text{--}17\ \mu\text{m}$ and found to be predominantly composed of clay-rich soil particles (Tao et al., 2015). Yellow River sediments accumulate in the deltaic area in summer where sediment reworking and mixing processes dampen any seasonal variations in sediment supply. In winter, sediments are resuspended and mobilized under the influence of strong hydrodynamic forcing driven by winter storms and tides and transported along the Shandong Peninsula by currents driven by regional influences of the East Asian monsoon (Li et al., 2016). Together these processes serve to dampen the amplitude of any seasonal variability in OM exported from the Yellow River. Given that the YS transect is overwhelmingly dominated by and serves as the major transport conduit for Yellow River sediments (Li et al., 2016), seasonal variations in the magnitude and location of sediment supply are not considered to influence apparent LTTs along the BY transect.

Sediments exported from the Columbia River experience intensive reworking following initial deposition on the adjacent shelf (Hedges et al., 1999). Here also, physical mixing processes are likely to homogenize and mask seasonal variations in the flux and composition of fluvially derived sediments (Coppola et al., 2007;

Hedges et al., 1999; Keil et al., 2004). Furthermore, since the WM samples lie at the more distal the end of the WM transect, seasonal variations in sediment supply from the Columbia River are unlikely to exert strong influence. While the influence of spatial variations cannot be discounted, the general movement of sediment from outer shelf to slope (Coppola et al., 2007; Hickey & Banas, 2003) implies that the offset of ages (LTTs) between WM3 and WM4 reflects aging due to basinward transport and that variability in LTTs reflects hydrodynamic behavior of different grain sizes. Further sampling and measurements would be needed to further constrain spatial variability in LTTs, along the WM transect and over other continental margins.

Given frequent observations of aged OC in surface sediments from continental margin settings (Griffith et al., 2010), our findings suggest that lateral transport-associated ^{14}C aging of OM may be a widely occurring phenomenon. Energetic conditions that drive lateral transport are prevalent on continental margins, and local hydrodynamic conditions dictate transport velocities of different grain-size sediments, the result being dispersal of variably aged OC on, and beyond, the continental shelves. While the magnitude and nature of OM aging likely depend on specific characteristics (e.g., transport distance and shear stress) that induce sediment mobilization and sorting (Nittrouer & Wright, 1994), grain size appears to be a key modulator of lateral transport-associated aging of OM. Thus, assessment of LTTs of OC residing in different grain-size fractions provides key constraints on aging of OM during across shelf-transport that complement those derived from ^{14}C measurements on bulk sediments.

4. Implications for OC Cycling and Burial in Ocean Sediments

While our compound-specific radiocarbon data imply millennial-scale aging of higher plant wax biomarkers during along/across shelf sediment transport, this process is not restricted to terrestrial OC. Other organic components associated with the sediment derived from different sources (i.e., marine biogenic OC) should experience similar ^{14}C aging during lateral transport, with the magnitude of aging depending on the timing and site of production, the physical disposition (e.g., grain-size association) within the sediment, and on regional hydrographic and geomorphic characteristics. For example, Mollenhauer et al. (2007) observed sharply contrasting ^{14}C ages of marine algal biomarker lipids (alkenones) from inner shelf (modern) and slope surficial sediments on the Namibian margin and attributed this age contrast to result from across-shelf (nepheloid layer) transport (Table 1). Moreover, such aging processes likely extend beyond continental shelves and may be prevalent in any oceanic settings where hydrodynamic conditions promote sediment dispersal. Ohkouchi et al. (2002) found, for example, that alkenones in surficial sediments from a abyssal contourite deposit (Bermuda Rise, NW Atlantic Ocean, water depth: 4,517 m) were preaged by up to ~7,000 years, indicating extensive reworking and redistribution. Therefore, lateral sediment redistribution processes may exert widespread influence on the ^{14}C age OM in ocean sediments.

The aging (or decrease in ^{14}C content) of OM requires consideration in calculations of terrestrial OC burial on continental margins utilizing isotope mass balance approaches. Changes in ^{14}C content resulting from transport are typically not considered in assignments of isotopic end-member values (e.g., Tao et al., 2016). A decrease in ^{14}C contents of OC due to lateral transport may lead to overestimates of terrestrial OC burial and corresponding underestimates of the marine autotrophic OC burial when a traditional ^{14}C mass balance approach such as the following is applied: $\Delta^{14}\text{C}_s = f_t * \Delta^{14}\text{C}_t + f_m * \Delta^{14}\text{C}_m$; $1 = f_t + f_m$ (where $\Delta^{14}\text{C}_s$ is ^{14}C content of sample, $\Delta^{14}\text{C}_t$ and $\Delta^{14}\text{C}_m$ are ^{14}C contents values from terrestrial and marine OC end-member, and f_t and f_m are corresponding fractions in the sample). Since $\Delta^{14}\text{C}_t$ is typically assumed to be lower than $\Delta^{14}\text{C}_m$, a decrease in $\Delta^{14}\text{C}_s$ values as a result of lateral transport would result in an artificial increase in calculated f_t (decrease in f_m).

In addition, lateral transport, as well as hydrodynamically driven variations in ^{14}C contents of organic components among different grain-size fractions, influence temporal phasing of biomarker signals in sediments. Consequently, the size dependence of sediment transport processes should be carefully considered in interpretations of molecular proxy records (McCave, 2002).

The eastern Chinese marginal seas (including the BY) and the Washington margin can be considered broadly representative of continental margins globally (Harris et al., 2014) due to their sharp contrast in spatial extent and environments for OC lateral transport. There is growing evidence for pervasive OC aging during lateral transport (Table 1); however, it is likely that LTTs vary between different margin systems as a function of

geomorphologic, physical oceanographic (e.g., tidal forcing and storm activity) characteristics. With respect to the former, for example, margins with very narrow shelves or those incised by submarine canyons (e.g., Harris & Whiteway, 2011) may not provide enough accommodation space for significant hydrodynamically driven OM aging to occur. Regarding the latter, extreme climate events (e.g., typhoons) may result in rapid export of OC to deeper ocean settings (e.g., Zheng et al., 2017).

Extensive remineralization of OC to dissolved inorganic carbon occurs during lateral transport (Bröder et al., 2018; Keil et al., 2004; Tesi et al., 2016), resulting in release CO₂ into the atmosphere and thereby influencing climate on longer time scales (Leithold et al., 2016). In the context of ongoing global warming, changes in the occurrence and intensity of extreme climate events and/or shifts in regional climate may influence ocean current velocities (Toggweiler & Russell, 2008), altering the dynamics of across-shelf OC transport. Interrelationships between climate variability and lateral transport processes on continental shelves require further consideration in the context of both carbon burial and interpretation of marine sedimentary records. Overall, aging of OM during hydrodynamically driven lateral transport forms an integral and dynamic component of carbon cycling on continental shelves.

Acknowledgments

We thank the captain, crew, and scientific parties aboard the R/V *Dongfanghong II* and R/V *New Horizon* for onboard assistance. This study is supported by the SNSF“CAPS-LOCK” project 200021_140850 (T.I.E.), the National Natural Science Foundation of China (grants 41520104009 and 41521064, M.Z.), and the “111” project (B13030). This study is part of scholarship to M.U. by the JAMSTEC and was also supported partly from NIES grant (Kibanseibihi, Rijityotouseihi, Syoureikenkyu, and Tokubetukenkyu, M.U.), JSPS (KAKENHI 25550020, 23651021, and 22310014, M.U.), and from MOE (Suishinhi B-0903, B-0904, 2-1304, and A-1003, M.U.). We thank the staff and members of the Laboratory for Ion Beam Physics (ETH) for the support from National Ocean Sciences AMS (WHOI) in all aspects of the AMS measurements. Data used to generate in figure and table are reported in the paper.

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