



Radiocarbonscapes of Sedimentary Organic Carbon in the East Asian Seas

Rui Bao^{1,2,3*} and Thomas M. Blattmann^{3,4*}

¹ Frontiers Science Center for Deep Ocean Multispheres and Earth System, and Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education/Institute for Advanced Ocean Studies, Ocean University of China, Qingdao, China, ² Laboratory for Marine Ecology and Environmental Science, Qingdao National Laboratory for Marine Science and Technology, Qingdao, China, ³ Department of Earth Sciences, Geological Institute, ETH Zürich, Zurich, Switzerland, ⁴ Biogeochemistry Program, Research Institute for Marine Resources Utilization, Japan Agency for Marine-Earth Science and Technology, Yokosuka, Japan

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*Correspondence:

Rui Bao
baorui@ouc.edu.cn
Thomas M. Blattmann
blattmann@jamstec.go.jp

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Natural abundance radiocarbon (¹⁴C) is an increasingly widely used tool for investigating the organic carbon (OC) cycle in the contemporary ocean. Recent studies have provided extensive information on the ¹⁴C characteristics of organic matter (OM) in sinking particles and sediments in the East Asian Seas including studies from the Bohai Sea, Yellow Sea, East China Sea, South China Sea, Japan Sea, and Japan Trench. ¹⁴C investigations have provided insights into biogeochemical processes controlling the fate of sedimentary OM in these settings. Here, we highlight these insights from oceanic landscapes stretching across deltas, shelves, abyssal oceans, and the hadal zones of the East Asian Seas; share our perspectives on the source-to-sink dynamics of sedimentary OM in the ocean; and outline the challenges that need to be faced to make the most out of interpreting ¹⁴C signals in sedimentary OC.

Keywords: ¹⁴C, sediment, source-to-sink, shelf, margin, abyssal, hadal, turbidite

INTRODUCTION

The net amount of carbon that is reduced and buried in the form of sedimentary organic carbon (OC) is one key flux controlling global biogeochemical cycles over geologic timescales (Berner, 1990). The journey begins with fixation by autotrophs incorporating the radiocarbon (¹⁴C) signature of their sources. This carbon propagates through the food web until it is respired or enters non-living pools of marine OC, which are subject to ¹⁴C radioactive decay. Expressed as a large spread in ¹⁴C patterns in the ocean, sedimentary OC contains a spectrum of pools at varying states of (radioactive) decay (McNichol and Aluwihare, 2007). The cosmogenic origin and ¹⁴C decay provide insight into the timescales of source-to-sink carbon dynamics and the response of the carbon cycle to perturbations (Eglinton and Repeta, 2014). Natural abundance ¹⁴C has established itself as a cornerstone for assessing sources and processes governing the fate of sedimentary OC in the oceans (Williams et al., 1992; Wang et al., 1996; Eglinton et al., 1997; Masiello and Druffel, 1998; Drenzek et al., 2009; Griffith et al., 2010). Recent technical developments in accelerator mass spectrometry reducing sample size requirements (Synal et al., 2007; Xu et al., 2016; Yamane et al., 2019) and increasing throughput by interfacing with direct CO₂ intake systems by elemental analyzers (McIntyre et al., 2017) or gas bench-type systems (Wacker et al., 2013) are poised to continue this progress.

In marine environments, interpretations of ^{14}C in sedimentary OC often require going beyond a straightforward chronological approach and are convoluted owing to a series of factors. Across the vast expanses of oceanic landscapes stretching deltas, shelves, abyssal plains, and hadal trenches, “radiocarbonscapes” (here, defined as spatiotemporal variations, characteristics, and patterns of radiocarbon) of sedimentary OC are markedly different, providing insight into the stories that carbon has to tell as it traverses from sedimentary source to sink. The East Asian Seas host natural laboratories characterized by a range of depositional environments from vast shelves spanning hundreds of kilometers to the deepest trenches scarring the face of our planet. To date, extensive ^{14}C datasets exist for these natural laboratories with starkly contrasting underpinning controls, which we divide into four different themes shaping radiocarbonscapes including (A) provenance, (B) hydrodynamic processes, (C) event-driven sedimentation, and (D) organic matter (OM)–mineral interactions. In this contribution, we highlight several OC ^{14}C studies from the East Asian Seas and review these distinct radiocarbonscapes from the perspectives of these four themes.

DISCUSSION

Provenance

The East Asian marginal seas receive continentally derived sedimentary OM exported from some of the world’s largest rivers including the Yangtze, Yellow, and Pearl Rivers (Wang et al., 2012; Tao et al., 2015; Wu et al., 2018; Lin et al., 2019). After fixation from the atmosphere, terrigenous OC may undergo long storage times on land, reducing its ^{14}C content. For example, particulate OM from the Yellow River contains a higher percentage of aged OC (Tao et al., 2015, 2016; Yu et al., 2019); hence, the corresponding deltaic area shows lower ^{14}C content than elsewhere in the Bohai Sea (Bao et al., 2016; **Figure 1A**). Similarly, based on cluster analysis of combined sedimentary OC content and its ^{14}C concentration across the vast expanses of the East China Sea, the radiocarbonscape emanating from the Yangtze River reflects heavy terrestrial influence (Van der Voort et al., 2018). An extreme case of provenance-dominated oceanic radiocarbonscape is exemplified by the export of ^{14}C -free OC of petrogenic origin (i.e., kerogen) (Kao and Liu, 1996; Hilton et al., 2008; Lin et al., 2020). Surrounding Taiwan island, these contributions of petrogenic OC appear as anomalously low ^{14}C signatures in sedimentary OC (**Figure 1A**; Bao et al., 2016; Zheng et al., 2017). Therefore, low ^{14}C concentrations in sedimentary OC of deltaic and land-proximal settings may reflect a provenance-based effect with the addition of pre-aged sedimentary OM from terrestrial sources to recently synthesized marine OM (see also South China Sea investigation by Mollenhauer et al., 2005).

Hydrodynamic Processes

Often, the ^{14}C content of sedimentary OC is to a first-order determined by provenance; however, when viewing spatiotemporal patterns in radiocarbon contents of sedimentary

OC (**Figure 1A**), it is apparent that highly variable ^{14}C contents in distal shelf settings are prevalent. Sedimentary redistribution processes (e.g., resuspension and dispersal) affect particles with different hydrodynamic properties differently, resulting in these highly variable patterns. Thus, there is size-dependent redistribution, as well as lateral transport time, recorded by the ^{14}C “clock,” leading to a highly variable radiocarbonscape dependent on transport pathways (Bao et al., 2018c). Resuspension remobilizes aged sedimentary OM associated especially with intermediate grain size fractions in shallow inner-shelf settings, whereas in deeper regions and erosional areas, bedload transport exerts the strongest influence on redistribution of aged sedimentary OM, especially for the coarser fractions (Bao et al., 2016; **Figure 1B**). The aged sedimentary OM spreads into deep-sea settings that receive sedimentary input from these areas. In the East Asian Seas, based on ^{14}C analyses of specific compounds and OM thermal decomposition windows on specific grain size fractions, lateral transport of sedimentary OM over millennial year timescales is evident across hundreds of kilometers’ distance in the Bohai, Yellow Sea, and East China Sea (Bao et al., 2018c, 2019a,b). Such contributions of aged sedimentary OM are also apparent in deeper waters adjacent to and receiving detrital input from East Asian shelves, such as for aged suspended sedimentary OM arriving in the Okinawa trough (Honda et al., 2000). Laterally derived aged suspended sedimentary OM is also observed in the lower water column of the Japan Sea (Kim et al., 2017, 2020). Hydrodynamic processes redistribute sedimentary OM with aged ^{14}C signatures, thereby overprinting ^{14}C of OC in surface sediments into which redeposition takes place leading to considerable spatial variability in radiocarbonscapes and uncertainty in ^{14}C age-related information.

Event-Driven Sedimentation

Mass wasting events such as those triggered by earthquakes and typhoons mobilize enormous pulses of sediments into the deep sea (Carter et al., 2012; Kioka et al., 2019), carrying with it vast amounts of OM (Tsai et al., 2010; Liu et al., 2013). The heavily incised active margin shelves along the Japan Trench offer little accommodation space for sediment storage, facilitating landslides triggered by tectonic events. This old sedimentary OM, which is mobilized, a consequence of protracted storage in intermediate reservoirs on land (e.g., soils) and/or on the continental margin, strongly influences the ^{14}C contents of OC in the hadal zone such as the Japan Trench (~8,000-m water depth; Bao et al., 2018b). Such events blanket the seafloor with turbidites containing aged OM (**Figure 1C**; see also Nakamura et al., 1990). Similarly, within the Gaoping canyon, directly south of Taiwan, event-driven sedimentary inputs of terrestrial OC to the ocean are common. In the case of typhoons, hyperpycnal flow conditions directly export large contributions of land-derived biospheric and petrogenic OC into the South China Sea via submarine canyons (Hilton et al., 2008; Kao et al., 2014; Zheng et al., 2017; Lin et al., 2020). Over hundreds of kilometers’ distance, sedimentary OC is carried by turbidites (Zhang et al., 2018) or entrained in eddies (Zhang et al., 2014), reflecting an event-driven overlay observed

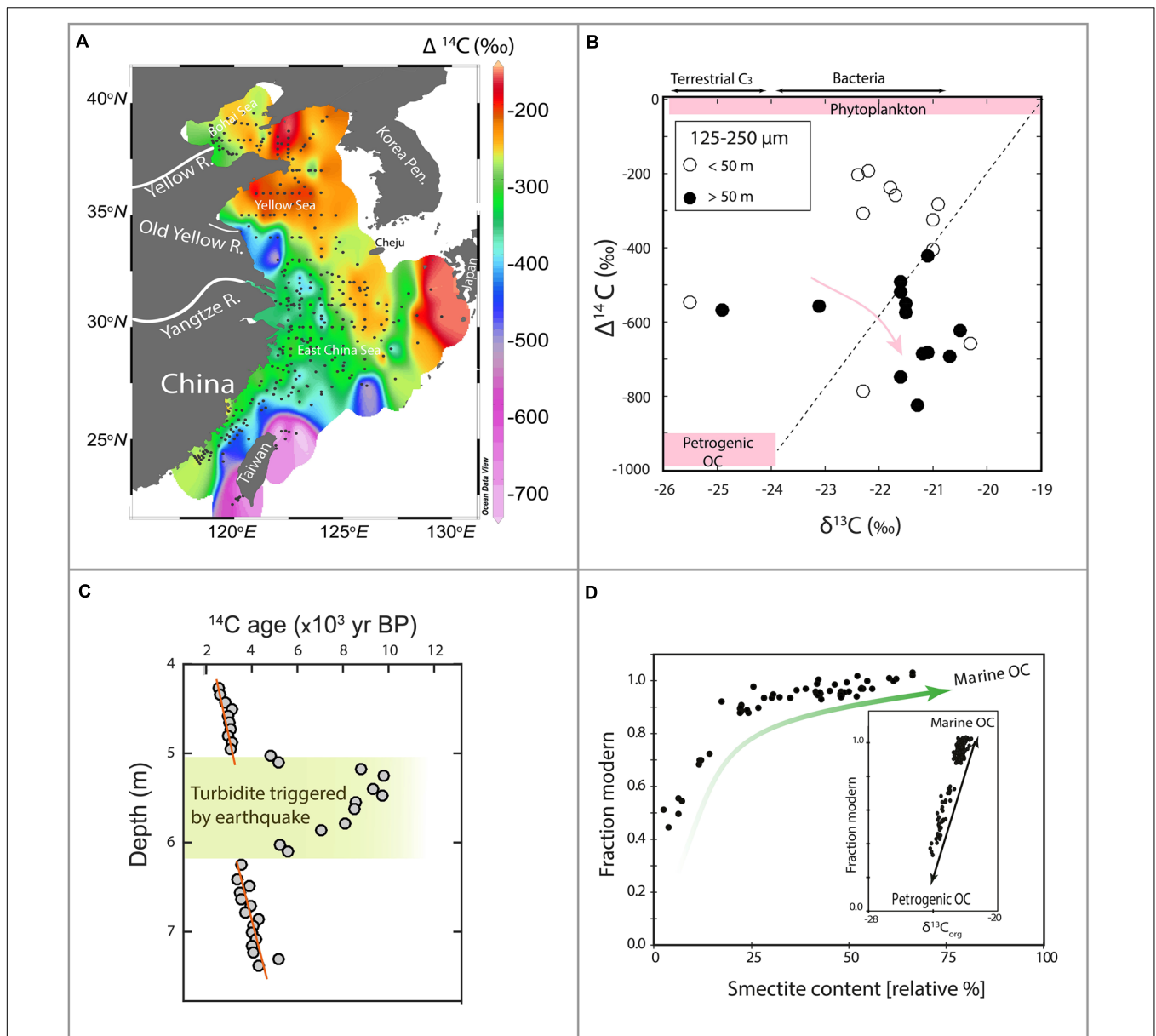


FIGURE 1 | ^{14}C study highlights from East Asian Seas with **(A)** illustrating the spatial distribution of ^{14}C concentration in sedimentary OC in the Bohai, Yellow, East China Seas (Bao et al., 2016). **(B)** Shows the ^{14}C concentrations of sedimentary OC in 125–250- μm grain size fractions of surface sediments in the East China Sea, with the dashed line separating the result of mixing of OC sources on the left from the effect of sedimentary reworking on the right (water depth >50 m; Bao et al., 2019a). **(C)** Illustrates a sedimentary profile in the Japan trench showing the ^{14}C imprint on sedimentary OC in a turbidite deposit triggered by one earthquake (after Bao et al., 2018b). **(D)** Shows the relationship between ^{14}C content of sedimentary OC and phyllosilicate composition in abyssal South China Sea sinking particles with modern marine OM associated with soil-derived smectite (after Blattmann et al., 2019).

over time series of sinking particles (Blattmann et al., 2018). Therefore, these episodic aged or even petrogenic OM exports, triggered by events such as earthquakes and typhoons, exert large-scale control on radiocarbonscapes in the deep ocean.

OM–Mineral Interactions

Mineral ballast exerts key control over the sedimentary transport and deposition of OM in aquatic environments (Ittekkot et al., 1990; Wakeham et al., 2009). Investigations have

revealed the association of marine OM with lithogenic minerals (Keil et al., 1997; Kennedy and Wagner, 2011), which is also observed in the sinking particles of the South China Sea, which impart a strong effect on ^{14}C contents of sedimentary OC (Blattmann et al., 2018). The systematic radiocarbonscapes with contrasting modern and ancient forms of sedimentary OM (see insert in **Figure 1D**), representing marine and petrogenic OM forms, respectively, display strong relationships with mineralogical composition, revealing mineral-specific behavior

on the retention and release of terrestrial OM in the marine environment by loss-and-replacement reactions (**Figure 1D**; Blattmann et al., 2019). Smectite, a pedogenic mineral, loses its association with pedogenic OM and associates with marine OM in distal marine settings. By way of this mechanism, sedimentary OM sourced from land is desorbed and/or degraded, leaving marine OM with a straightforward source-to-sink trajectory to repopulate the particulate phase (c.f., Zhang et al., 2019). Organic matter–mineral interactions impart a systematic effect on the fate of sedimentary OM in the ocean and govern the type of OM that is stabilized, thereby changing the overall radiocarbonscapes.

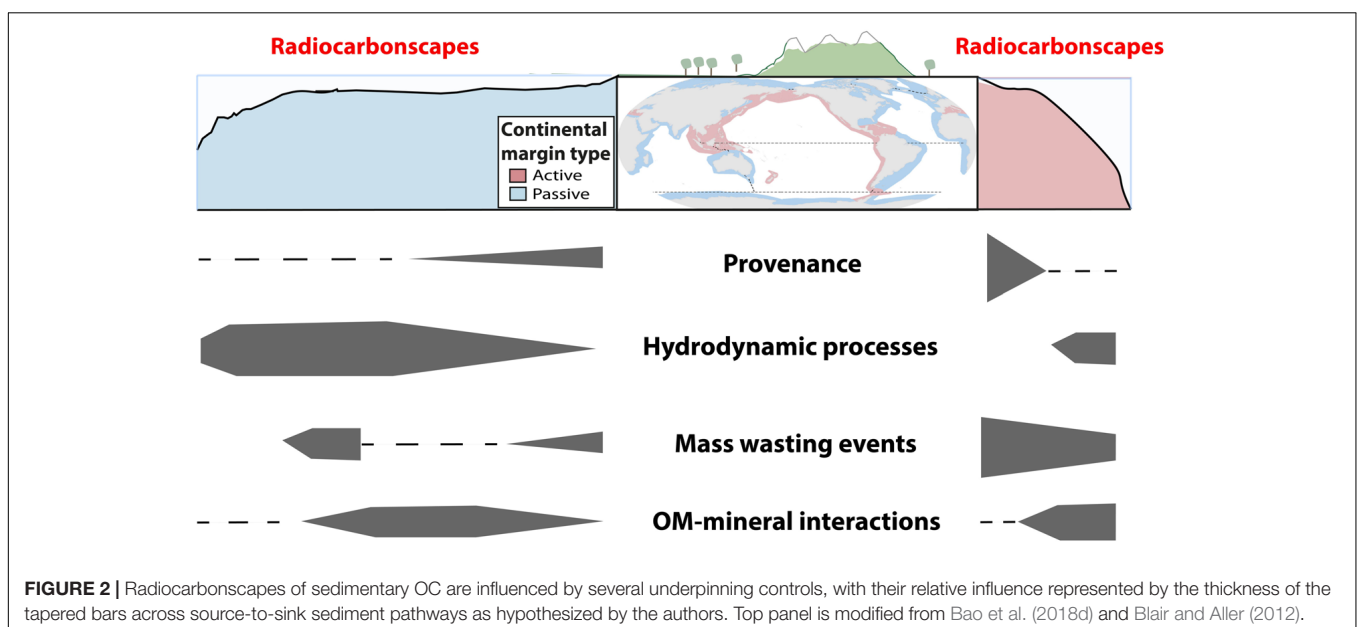
SYNTHESIS AND PERSPECTIVES

^{14}C is a one-dimensional value traditionally used to express age based on its decay constant (Libby et al., 1949). Rather than ^{14}C expressing a single property (i.e., age, which is commonly assumed), sedimentary OC in oceanic settings integrates an overlay of processes, which contribute to an overall radiocarbonscape. Delineating the manifold processes involved given the scalar nature of this measurement often arrives at a non-unique set of solutions. Toward resolving these ambiguities by extending ^{14}C into multidimensional space, sedimentary OM can be dissected into different physical fractions [e.g., density (Wakeham et al., 2009); grain size (Bao et al., 2019a), and hydrodynamic (Coppola et al., 2007)], reactivity pools [e.g., chemical oxidation (Ohkouchi and Eglinton, 2006), thermal treatment (Hemingway et al., 2019; Bao et al., 2018a)], and individual compounds that can be targeted (e.g., Eglinton et al., 1997; Tao et al., 2016). Based on the evidence accumulated thus far, the “radiocarbonscapes” of the East Asian Seas are controlled by a combination of (A) provenance, (B) hydrodynamics, (C) event-driven sedimentation, (D) OM–mineral interactions,

and/or other potentially important and less-well understood processes. ^{14}C is a tracer that intersects with all of these themes, and it is apparent that we need to continue our efforts to deconvolve the complex tapestry of radiocarbonscapes in the marine environment to resolve basic questions including the following:

1. How do the four themes provenance, hydrodynamics, event-driven sedimentation, and OM–mineral interactions weigh against each other in shaping radiocarbonscapes?
2. How can quantitative information on these four themes be extracted from radiocarbonscapes?

Based on the East Asian Sea natural laboratories, we have developed our own perspectives toward answering these questions and judged the relative influences of the four themes on seafloor radiocarbonscapes (**Figure 2**). It appears that the relative importance of these four themes varies greatly across delta to deep-sea transects for active and passive margin settings (c.f., Blair and Aller, 2012), with provenance-based influence strongest in deltas and more diffuse across passive margins with hydrodynamic processes, leading to extensive imprints on radiocarbonscapes on wide, high-energy shelf settings. In contrast, event-driven sedimentation markedly impacts radiocarbonscapes across active margin settings where episodic mobilization and redistribution of vast amounts of sediment occur along with a pronounced inheritance of a provenance signal. With their areas of greatest influence difficult to pinpoint, OM–mineral interactions control loss and replacement of terrestrial with marine OM, thereby rewriting the organic geochemical fingerprint of sedimentary OM. Based on individual case studies of the East Asian Seas natural laboratories, the operation of these four themes crystallizes out of radiocarbonscape patterns. However, quantitative approaches for disentangling



radiocarbonscapes are needed where these four themes are more intertwined, ultimately key for understanding marine biogeochemical cycles.

AUTHOR CONTRIBUTIONS

RB and TB contributed equally to discussion and writing. Both authors contributed to the article and approved the submitted version.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

The reviewer AM declared a past co-authorship with one of the authors RB to the handling editor.

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