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Differential Regulation of Hydrodynamics and Anthropogenic Mariculture Activities on the Fate of Sedimentary Organic Carbon

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Abstract The spatial evolution of sedimentary organic carbon (OC) along transport pathways reflects burial and remineralization processes, traditionally attributed to hydrodynamic transport. However, direct evidence verifying the mechanism or examining whether the evolution pattern can be disrupted particularly by the input of marine OC from anthropogenic sources remains limited. This study integrates sediment resuspension experiment with sediment and OC characteristics analyses to elucidate hydrodynamic regulation on OC and additional mariculture-induced variability. Hydrodynamic resuspension regulates OC content through grain-size and mineral sorting but does not directly alter its thermochemical properties, regardless of hydrodynamic intensity. In comparison, mariculture directly modulates OC content and properties by the input of labile OC. We address a previously overlooked aspect of the thermochemical properties of mariculture-derived OC and underscore the importance of comprehensively evaluating the impacts of mariculture activities on OC, particularly regarding their long-term biogeochemical consequences.

Plain Language Summary Decreasing the global carbon degradation to increase the sequestration to mitigate climate change is a significant area of research. To explore and compare the impact of hydrodynamics and human activities on organic carbon (OC) burial and degradation, we strategically positioned sampling stations based on the presence of mariculture areas along the transport pathway and employed advanced multidisciplinary methods. These methods yield detailed insights into the spatial variation of OC, the hydrodynamic resuspension processes, and the dynamics of sediments and OC. Erosion microcosm experiment proved the correlation between hydrodynamic intensity and OC release due to the hydrodynamic sorting of grain size and minerals. This study indicates that over long transport distances, the influence of hydrodynamics on OC properties is indirect and widespread, whereas the impact of mariculture is direct and profound, altering the spatial distribution of OC properties under natural conditions. We highlight that the effects of anthropogenic activities on OC have been underestimated.

Key Points:

- Hydrodynamic sediment sorting governs the redistribution of organic carbon (OC) content via its strong coupling with grain size and mineral associations
- Transient resuspension has limited impact on OC thermochemical properties, despite notable grain-size sorting
- Labile OC input from mariculture overrides preferential degradation, disrupting sedimentary OC stabilization along transport pathways

Supporting Information:

Supporting Information may be found in the online version of this article.

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1. Introduction

Hydrodynamic regulation of sedimentary organic carbon (OC) primarily operates through transport processes closely linked to OC burial and remineralization, which are critical components of global carbon cycling and atmospheric CO_2 balance (Z. Chen et al., 2022; M. Yu et al., 2024). This is especially evident in marginal seas with substantial terrestrial OC (OC_{terr}) input (Bianchi et al., 2016; Hedges & Keil, 1995; Walsh, 1991) and elevated remineralization rates driven by persistent sediment resuspension (Bianchi et al., 2018; Z. Chen et al., 2022; X. Liu & Bao, 2024), a process regarded as a prelude to sediment transport (Gargett et al., 2004).

OC exhibits a trend of spatial heterogeneity due to the transport process. Hydrodynamic sorting, caused by the differential response of sediment particles to resuspension, is regarded as one of the predominant processes and thus regulates the dynamics of sedimentary OC, which is mostly mineral-associated (Babakhani et al., 2025). Generally, finer grains, in correlation with low-density mineral components (Garzanti et al., 2008), are more susceptible to erosion and tend to travel longer distances due to the stronger flow resistance and lower settling velocity. In contrast, coarse-grained, high-density particles are typically transported nearer to the bottom layer in

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water flow (Li et al., 2020; van Rijn, 1993). Although factors like cohesion in clay fractions may influence erosion characteristics, leading to unique OC characteristics in certain particle size intervals (e.g., 20–32 μm) (Bao et al., 2016), the overall trend of grain-size fining along the transport transect remains observable. The process is followed by the selective dispersion and lateral transport of sedimentary OC. Specifically, larger surface area of minerals and higher clay mineral composition provide more reactive interfaces which also offer an advantageous microhabitat for microorganisms and thus make it more likely for fine-grained sediments to adsorb, aggregate, and stabilize OC (Blair & Aller, 2012; Vogel et al., 2014), leading to a relatively higher OC content (Bao, Voort, et al., 2018; Bianchi et al., 2018; Blanca et al., 2021; Hedges & Keil, 1995). Grain-size fining manipulates an increase in OC content and might also contribute to the variation of OC properties (Bao, McNichol, Mcintyre, et al., 2018; Bao, et al., 2019).

Moreover, the hydrodynamic force may contribute to the development of the OC spatial heterogeneity pattern regardless of grain size and mineral sorting. Sediment resuspension alters the biochemical environment of mineral-bound OC or physically breaks the mineral-OC bond that is essential for mineral protection. The process exposes OC to an environment rich in dissolved oxygen and microorganisms, increasing the oxygen exposure time (OET) of OC and promoting its decomposition (X. Liu & Bao, 2024). The process is also coupled with the mixing of different OC sources (Che et al., 2024; X. Liu et al., 2024) and desorption-reabsorption of OC, which leads to the mutual transfer between particulate OC (POC) and dissolved OC (DOC) (He et al., 2016; X. Liu et al., 2024). During transport, preferential degradation of labile OC components emerges as the principal driver of the spatial evolution pattern (Bao et al., 2019). Both mechanisms lead to a theoretical pattern of heterogeneity during prolonged along- and across-shelf transport characterized by grain-size fining, an increase in OC content, and thermochemical stabilization, as well as OC aging (Bao, Uchida, et al., 2018; Bao et al., 2019; Huang et al., 2023; M. Yu et al., 2024; Zhang et al., 2022).

Despite the well-documented spatial evolution of sedimentary OC driven by hydrodynamic processes, this pattern could be disturbed by external organic inputs from anthropogenic activities, notably in nearshore regions where mariculture is concentrated (C. Liu et al., 2024; J. Yu et al., 2025). In particular, in areas lacking dominant inputs from major river mouths, such local sources can become key regulators of spatial heterogeneity in OC properties (C. Liu et al., 2024; M. Yu et al., 2021). Primarily via biologically mediated processes such as photosynthesis and biodeposition, mariculture contributes to carbon sequestration (Forrest et al., 2009; Xu et al., 2024) and alters the biogeochemical reactivity of sedimentary OC due to the high lability of fresh marine OC (OC_{mar}; B. Yang et al., 2023). In addition, mariculture-induced alterations in hydrodynamic conditions, oxygen availability, and microbial activity potentially reshape environmental interactions and influence sedimentary OC cycling (C. Liu et al., 2024; B. Yang et al., 2023). Given the influence of mariculture on multiple aspects of sedimentary OC dynamics, particularly its effects on OC content and properties, characterizing these changes is crucial to understanding the mechanism and magnitude of the potential of mariculture activities to reshape OC spatial heterogeneity. Such characterization also informs assessments of its contribution to global carbon sequestration and the sustainability of mariculture-induced carbon sequestration.

In this study, sediment samples were systematically collected along transects aligned with the Yellow Sea Coastal Current (YSCC) direction within the mariculture zones offshore of the Shandong Peninsula (Figure 1a). We conducted an integrated analysis comprising three components: continuous erosion experiment using the UMCES–Gust Erosion Microcosm System, grain size characterization, and OC analysis through ramped pyrolysis/oxidation (RPO) combined with excitation-emission matrix spectra (EEMs). By analyzing OC variations under staged hydrodynamic forcing and comparing the spatial evolution pattern in OC content and composition between natural and mariculture-impacted sites, we aim to clarify how hydrodynamics regulate sedimentary OC and the potential of mariculture-induced disruptions.

2. Materials and Methods

2.1. Sampling

The Yellow River (YR) sediment is discharged at the Yellow River estuary (YRE) and transported along the YSCC, which contributes to the muddy depositional area of the Yellow Sea (YS) in the nearshore of the Shandong Peninsula (J. P. Liu et al., 2004; Qin et al., 2018; Qiu et al., 2014; Z. S. Yang & Liu, 2007; Zhu et al., 2022). Significant differences in sediment characteristics have been observed between sites near and distal to the YRE along the YSCC (Bao et al., 2019; M. Yu et al., 2024). The high mariculture production (C. Liu et al., 2024),

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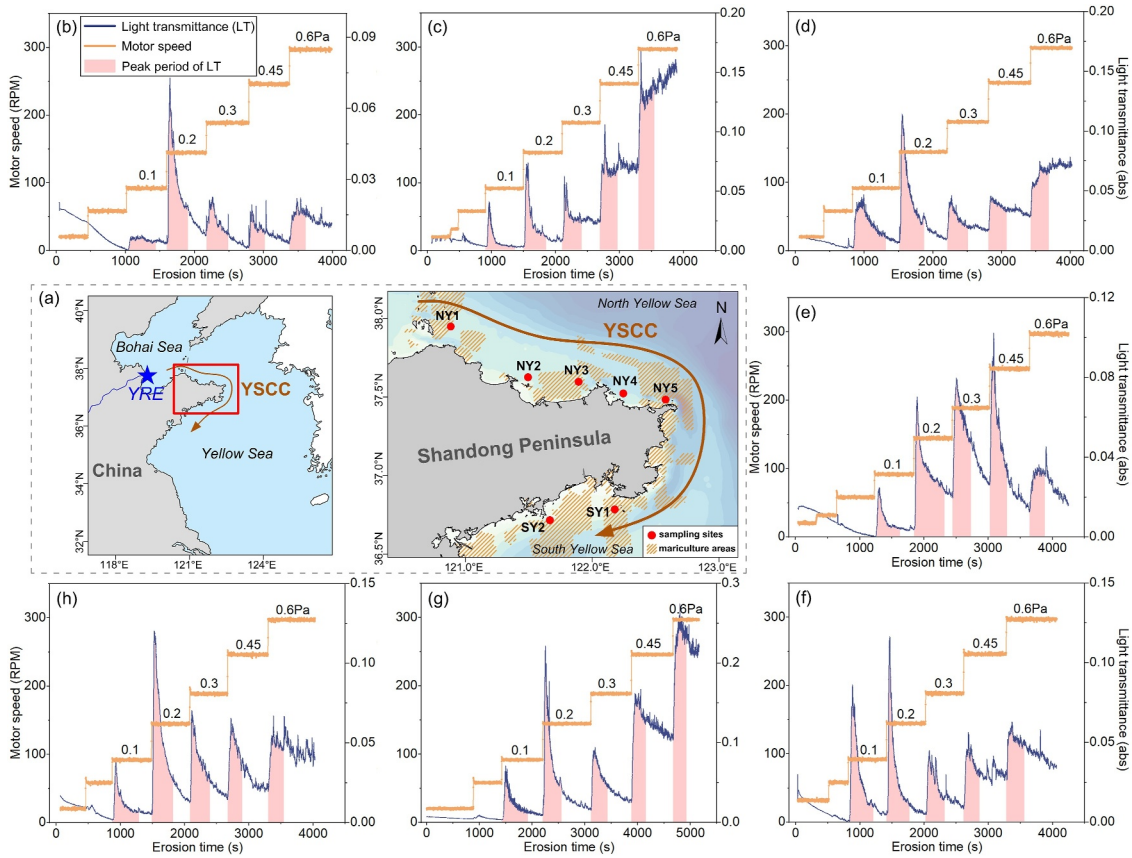


Figure 1. (a) Sampling stations and the distribution of mariculture areas along the Yellow Sea Coastal Current (indicated by the brown arrow). Light transmittance (LT) variation under varying bottom shear stress in micro-erosion experiments in (b) NY1, (c) NY2, (d) NY3, (e) NY4, (f) NY5, (g) SY1, (h) SY2. The red shadows represent each peak period of LT used for analysis of dissolved organic carbon characteristics.

comparable sedimentary environments, and relatively elevated OC content (Bao, Voort, et al., 2018; Shi et al., 2024) render the area well suited for investigating the impact of hydrodynamics and mariculture on OC cycling. Surface sediments (0–2 cm) sampling stations along the YSCC were strategically positioned primarily according to the distribution of the mariculture area (Figure 1a; see details in Text S1 and Table S1 in Supporting Information S1).

2.2. Micro-Erosion Experiment

U-GEMS (Green Eyes, USA) was employed to simulate sediment resuspension under progressively increasing shear stress. This system enables stepwise erosion of sediment under controlled hydrodynamic conditions, allowing real-time monitoring of light transmittance (LT, used as a proxy for suspended sediment concentration [SSC], reflecting sediment resuspension intensity) and sequential collection of sediment-laden water at each shear stress level (see system schematic in Che et al., 2024). Shear stress, representing the hydrodynamic forcing that mobilizes sediment and associated OC, was applied in incremental steps (0.1–0.6 Pa) to encompass shear stresses induced by typical wind-wave events and tidal currents in shallow YS environment(Che et al., 2024), with each stage lasting about 10 min until the erosion rate no longer decreased significantly, ensuring that the integrated sample collected constituted a definitive sample of the total sediment fraction erodible at that stress before progressing to the next higher level. At each stress level, sediment-laden water was collected and further filtered through 0.45 μm cellulose acetate membranes and 0.7 μm GF/F filters (pre-combusted at 450°C for 5 hr and pre-weighed). Bulk sediments, resuspended sediments (RS) and filtrate were used for the analysis of grain size distribution (GSD), POC, and DOC (see details in Text S2 and S3 in Supporting Information S1). The erosion experiments were conducted once per site.

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2.3. POC Content, Composition, and Thermochemical Reactivity Analyses

POC content and components were measured using a modified Organic and Elemental Carbon analyzer, adapted from the RPO method, with analytical precision, expressed as the relative standard deviation, better than 0.03% (Wu et al., 2023). RPO method provides the quantitative relationship between evolved carbon dioxide (CO_2) concentration and heating temperature, reflecting the thermochemical stability and components of POC (Hemingway et al., 2017). The sediments loaded on the membranes were freeze-dried, transferred, homogenized and inorganic carbon was removed using the $Rinse_{HCl}$ method (Bao, McNichol, Hemingway, et al., 2018) prior to instrumental analysis. Exposed to a linear temperature gradient, OC fractions with different thermal stabilities were sequentially evolved. Activation energy (E) was calculated using the OC E inversion method derived from the Arrhenius formula (Hemingway et al., 2017) and components of POC were classified according to commonly used E ranges (see details in Text S4 in Supporting Information S1; Huang et al., 2023).

2.4. DOC Content and Composition Analyses

The content and composition of DOC are analyzed using EEMs obtained from a fluorometer (Duetta, Horiba Scientific, Japan). The excitation and emission wavelengths were set in the 250–600 nm range, with scanning intervals of 5 and 2 nm, respectively. The fixed slit width was set at 5 nm. The data was processed using Horiba EzSpec software for scattering correction, followed by Raman normalization with Milli-Q water as the blank. The fluorescence regional integration method was used to calculate the total fluorescence volume (TFV) and the FV for each region. FV reflects the abundance of specific class of DOC, while TFV provides an overall measure of DOC fluorescence load (W. Chen et al., 2003; Niu et al., 2022; Song et al., 2019).

3. Results and Discussion

3.1. Dominant Control of Sedimentary OC Properties: Hydrodynamic Sorting

Although the erosion intensity varies among samples, the LT variation (representing SSC, Figure S1 in Supporting Information S1) exhibits a broadly consistent hydrodynamic response pattern (Figures 1b–1h). At each shear stress level, erodible sediments are resuspended, causing the SSC to escalate and reach a peak, followed by a gradual decline as suspended sediments is discharged and equilibrium is approached. This periodic pattern results from the prompt response of eroded sediments to the rapidly changing hydrodynamic conditions (Che et al., 2024).

The combined analysis of thermochemical and grain size characteristics reveals the hydrodynamic response and regulation mechanism of sedimentary POC content and components. At each station, the thermochemical profiles of RS closely resemble the bulk sediments, as evidenced by overlapping peak morphologies in both the CO_2 frequency distributions (pCO_2) and E spectra (NY1-SY2 and insets in Figure 2a). Despite limited variation in both low and high temperature peak (~390°C; ~540°C) under hydrodynamic forcing, the former exhibits a more recognizable variation. Specifically, either a shoulder near ~360°C (NY1, NY3–NY5, SY2) or a change in main peak height (NY2, SY1) is observed in RS samples, suggesting that hydrodynamic forcing differentially mobilizes and exposes labile OC fractions with distinct reactivities. The wide GSD (2φ to 11.4φ) allows examination of grain size fraction responses to hydrodynamic forcing (Figure 3a). Due to the limited upward transport of sand ($<4\varphi$) by water flow (van Rijn, 1993), sand is generally depleted in RS compared to bulk sediments. RS at the lowest shear stresses (0.1 and 0.2 Pa) are finest, with the highest proportion of clay fraction ($>8\varphi$). This selective resuspension of fine grains is particularly evident at stations NY3, NY4, and NY5, where sediments contain more sand, less clay, and have broader GSD (Figure S2a in Supporting Information S1). As shear stress increases beyond 0.3 Pa, sorting weakens and sediment fractions resuspend more uniformly. In contrast, selective resuspension is not observed in the fine-grained SY1 sample due to its high clay content (29.5%) and narrow GSD. Differential sorting effect implies that the clay and finer silt can be winnowed from the sand and coarser silt except in sediments with high clay content (Law et al., 2007; Lin et al., 2024). Despite the theoretical highest potential to resuspension of silt (McCave & Hall, 2006; Thomsen & Gust, 2000), lower density and finer grain size facilitate transportation (Li et al., 2020; Mclaren & Bowles, 1985), causing fine-grain particles to be preferentially transported under weak hydrodynamics (Li et al., 2020; Liu et al., 2015; Marisa et al., 2022).

A consistent trend between POC content and median grain size (φ_{50}) suggests that particles serve as a conduit for hydrodynamic regulation on POC content (Figure 3b). The gradually declined slope of the regression line of POC

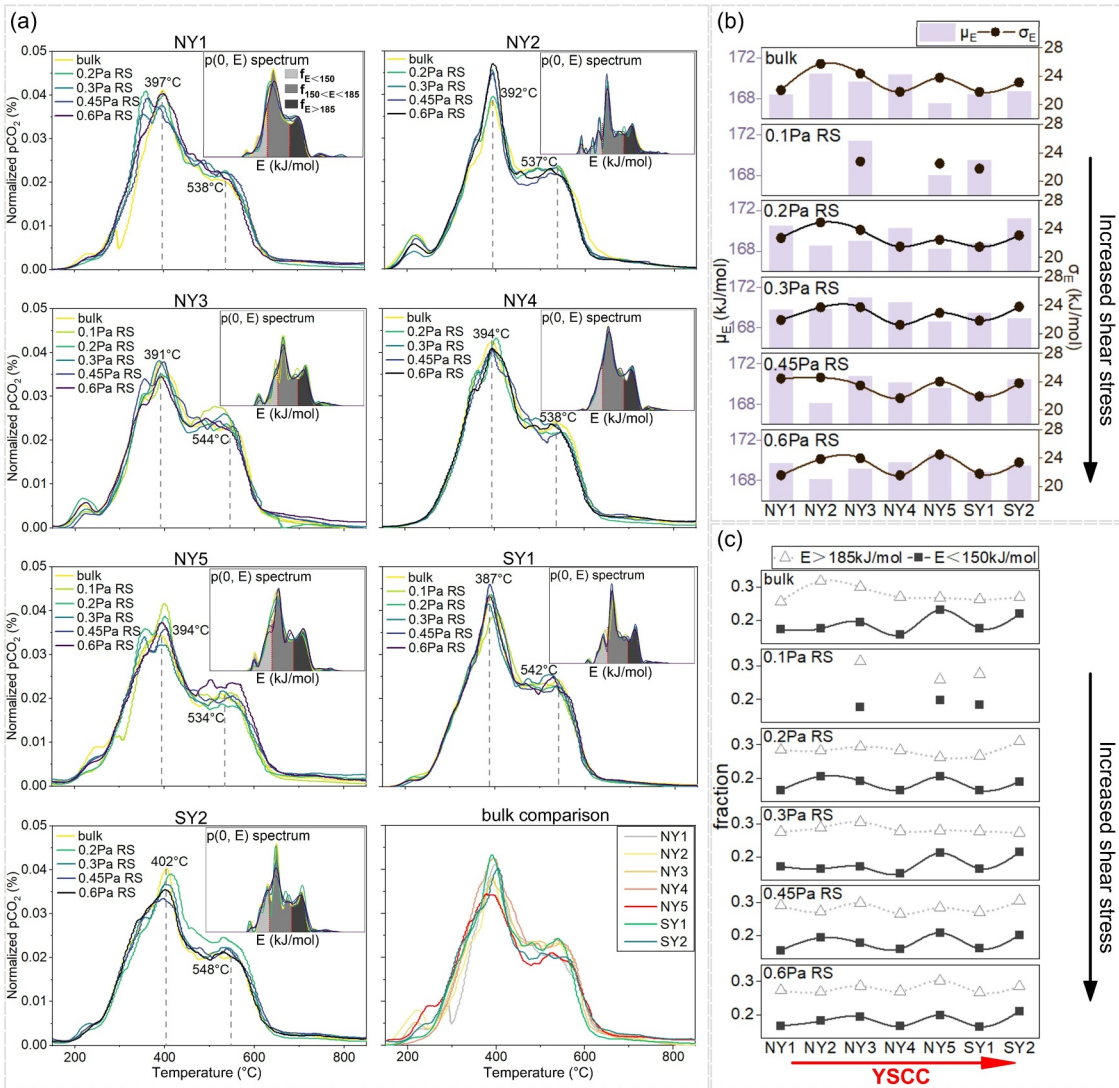


Figure 2. (a) Normalized pCO_2 spectra of bulk sediments and resuspended sediments, and the comparison of pCO_2 among bulk samples. Inset on the right of each figure shows corresponding activations energy spectrum. (b, c) Variations in mean activation energy (μ_E), standard deviation (σ_E), and the fractions of organic carbon with $E < 150$ kJ/mol ($f_{E<150}$) and $E > 185$ kJ/mol ($f_{E>185}$) under increasing shear stress and along the Yellow Sea Coastal Current.

content and φ_{50} of RS under increasing shear stress (Figure 3c) verifies the preferential resuspension of fine grain size-related (Garzanti et al., 2008; Laceby et al., 2017), low-density (Wakeham et al., 2009; Wang et al., 2015) and high-OC content (Wang et al., 2015) minerals. The non-monotonic spatial trend, characterized by an initial decrease and subsequent increase in both POC content and φ_{50} along the YSCC (Figure S2b in Supporting Information S1), is likely shaped by regional variations in hydrodynamic intensity and depositional settings (Bao, Uchida, et al., 2018). The persistence of this trend under uniform shear stress further highlights the dominance of background hydrodynamic conditions regimes over instantaneous forcing in controlling sediment grain size and OC content.

Despite the verified correlation between POC content and grain size, no significant change in thermochemical reactivity under hydrodynamic forcing occurs as previously mentioned (Figure 2a), likely due to the consistent absorption of different OC components. In the study area, clay and silt make up the majority of sediments (Figure S2a in Supporting Information S1). Previous studies have shown that OC within the <63 μm fraction generally displays minor variability in both ^{14}C age and thermochemical properties (e.g., Bao, McNichol, Mcintyre, et al., 2018; Ge et al., 2020; M. Yu et al., 2019). Given the established link between increasing OC activation energy and older ^{14}C ages (Zhang et al., 2022), we infer that fine grain-associated (<63 μm) OC in nearshore

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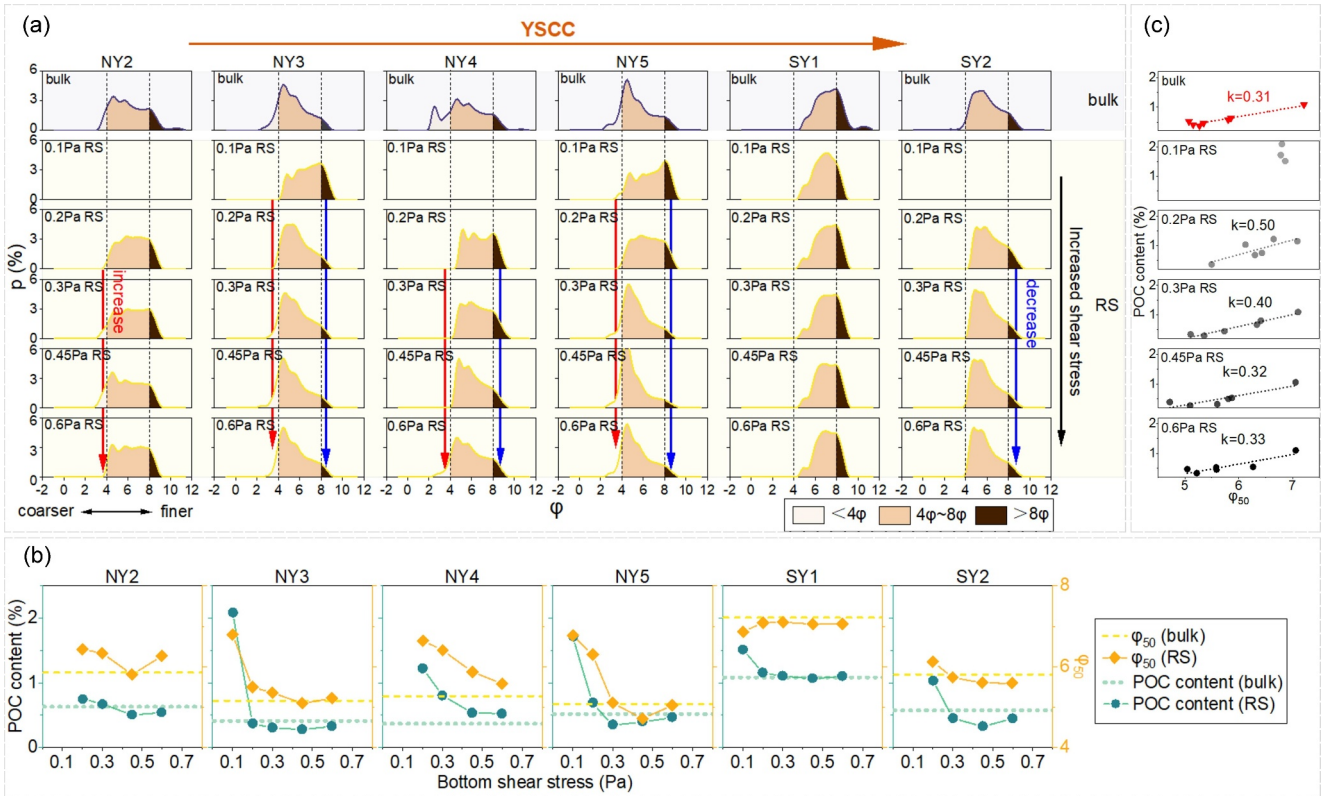


Figure 3. (a) The variation trend of grain size distribution under increasing shear stress and along the Yellow Sea Coastal Current. (b) Variation of particulate organic carbon (POC) content and medium grain size (φ_{50}) under varying bottom shear stress. (c) Linear correlation between POC content and φ_{50}. The slope is marked with the k value. Some samples lack data due to insufficient resuspension mass.

sediments tends to exhibit relatively uniform thermochemical stability. Distinguished from coarser fractions (sand) associated with discrete OC, clay and silt fractions are dominated by OC-mineral aggregation (Blanca et al., 2021) which helps preserve OC through close association and stabilization in aggregates (Babakhani et al., 2025; Bao, McNichol, Mcintyre, et al., 2018). We propose that short-term sediment resuspension does not prominently exhibit selective mobilization of mineral-bound OC fractions with distinct thermochemical stability, suggesting that in this case, hydrodynamic sorting does not directly alter the intrinsic properties of sedimentary OC. Based on the invariable OC thermochemical reactivity in sediment resuspension, the amount of OC released during resuspension can be reasonably estimated from bulk OC content, simplifying source-to-sink carbon budgets.

The characteristics of released DOC during resuspension reflects the release amount of porewater (Santos et al., 2012) and transfer dynamics between POC and DOC (X. Liu et al., 2024). Fluorescence intensity, positively correlated with fluorescence DOC concentration, can semi-quantitatively determine the content and composition of DOC (W. Chen et al., 2003; X. Liu et al., 2024; C. Zhao et al., 2023). The limited DOC release (all TFV <1,500 ru·nm^2, ru represents Raman units, Figure 4) indicates the desorption of weakly bound OC during sediment resuspension (He et al., 2016; X. Liu et al., 2024). The TFV, SSC and FV ratio of region V and region IV ($FV_{(V:IV)}$, commonly used as proxies for the relative proportion of humic-like OC and protein-like OC; W. Chen et al., 2003; Shutova et al., 2014) shows a generally consistent increasing trend under elevated shear stress (Figure 4), implying the enhanced release of DOC, especially humic-like OC due to intensified sediment erosion. Our data show that the concentration of DOC in the water column did not increase linearly with rising shear stress (Figure 4). This nonlinear release pattern suggests a rapid shift in the erosion process. We propose that under low shear stress, the preferential resuspension of fine, low-permeability particles (Henning et al., 2021; Santos et al., 2012) may have rapidly increased the overall permeability of the sediment bed (Huettel et al., 2014), thereby resulting in reduced subsequent porewater release. Concurrently, we observed a significant increase of the humic-like OC (Figure 4). Given that sediment minerals are known to preferentially sorb humic-like compounds (Hur &

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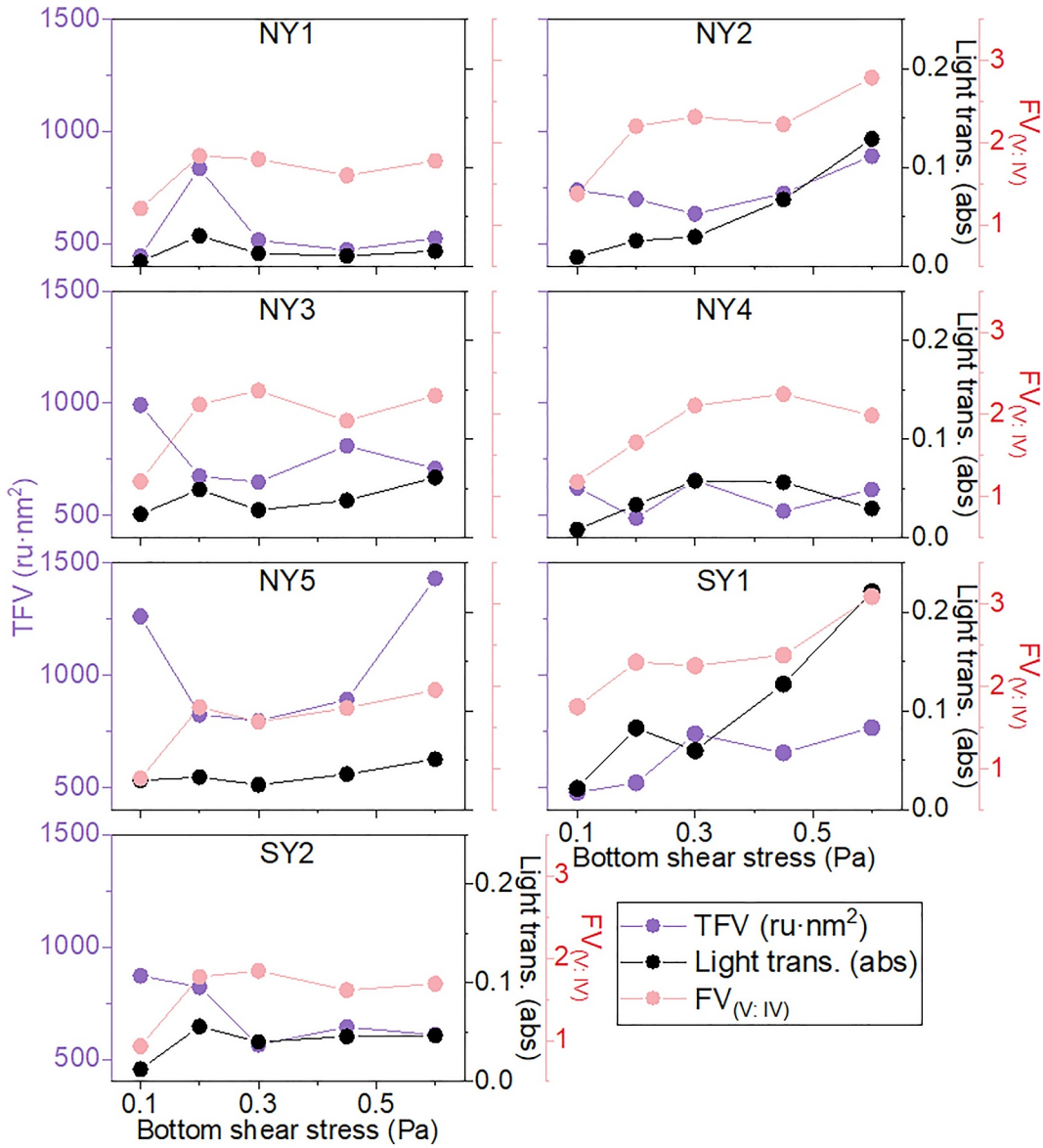


Figure 4. Variation of total fluorescence volume, the ratio of FV of humic-like organic carbon (OC) to protein-like OC ($FV_{(IV:V)}$), and light transmittance under varying bottom shear stress.

Schlautman, 2003; X. Liu et al., 2024), we interpret that the elevated proportion of humic-like OC resulted from the hydrodynamic disruption of organo-mineral bonds, which promoted OC desorption. This is also evidenced by the unusually high TFV and the relatively elevated proportion of protein-like OC observed under 0.1 Pa shear stress (Figure 4). Therefore, the enhanced transfer from POC to DOC with increased shear stress may stem from increased RS (Figure 1) providing a larger pool of convertible OC, intensified hydrodynamic disruption of organo-mineral bonds (X. Liu et al., 2024) promoting OC desorption, or uneven porewater release (Santos et al., 2012) affecting relative proportion of DOC components. Their relative contributions require further investigation.

Our coupled DOC and POC component investigations captures their differential short-term responses to hydrodynamic forcing. Considering the limited duration of hydrodynamic forcing and the short residence time of sediments in the resuspended state, despite the relatively minor variation of POC components (NY1-SY2 in Figure 2a), the generally monotonic trend of DOC components under increasing shear stress (Figure 4) may imply

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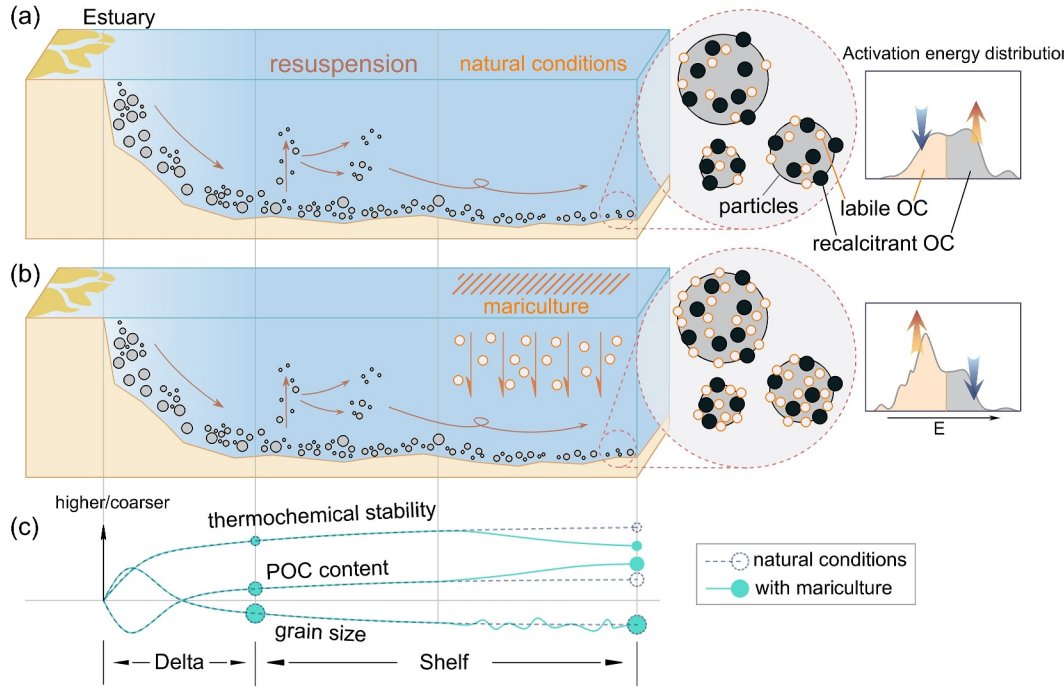


Figure 5. Conceptual diagram shows the divergent evolution pattern of sedimentary organic carbon characteristics (a) under natural conditions and (b) with mariculture. (c) The variation of thermochemical stability, particulate organic carbon content, and grain size along the transport pathway.

that the cumulative effect of repeated resuspension events may progressively alter OC composition and stability during sediment transport, particularly under sustained hydrodynamic regulation (Bao et al., 2019).

3.2. Divergent Controls on OC Spatial Heterogeneity: Mariculture Versus Hydrodynamics

The significant degradation of OC_{terr} typically occurs during the initial stages of transport in estuarine areas (M. Yu et al., 2024) and due to selective degradation of labile young OC, thermally refractory OC increases during protracted lateral transport (Figure 5a; Bao, Uchida, et al., 2018, 2019). However, no monotonic trend in OC thermalchemical stability is observed along the YSCC, and instead, spatial patterns exhibit moderate fluctuation among bulk OC (bulk comparison in Figure 2a). Specifically, the μ_E (mean activation energy, serves as a comprehensive indicator of overall thermochemical stability; Text S4 in Supporting Information S1) and fractions of recalcitrant OC_{terr} (OC with E values > 185 kJ/mol, $f_{E>185}$, Text S4 in Supporting Information S1) is relatively high in the former stations, while fractions of labile OC_{mar} ($f_{E<150}$) shows an opposite pattern (Figures 2b and 2c). No statistically significant impact on the thermochemical properties of POC due to resuspension was observed (NY1-SY2 in Figure 2a). Overall, despite varying stress levels, the trend of spatial variation in thermal stability parameters remains consistent with bulk sediment, especially the standard deviation of E (σ_E, Text S4 in Supporting Information S1), which exhibits relatively more significant spatial heterogeneity (21.3–25.7 kJ/mol; Figure 2b). The observed μ_E values are comparable to global averages (168.9 kJ/mol; Hemingway et al., 2019), yet lower than those reported for surface sediments in other deltaic systems (Huang et al., 2023), consistent with the phenomenon in earlier reports (Zhang et al., 2022), indicating a higher proportion of labile OC. The non-monotonic variation of thermalchemical properties along the transport pathway and overall elevated thermal lability may result from regional environment factors, especially the intensive mariculture actives (Zhou et al., 2024), particularly under the circumstance of the recent diminishment in sediment supply of YR, which might facilitate other OC sources to assume greater prominence (M. Yu et al., 2021). In the study area, mariculture is dominated by seaweed and shellfish cultivation, which are known for their high ecosystem multifunctionality, including efficient carbon sequestration (C. Liu et al., 2024). The prominence of these farming types may help explain the pronounced mariculture-driven signal in sedimentary OC characteristics. This is exemplified by observations from Sungo Bay, located off the Shandong Peninsula, which indicate a prominent export of POC to

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the open sea across the offshore mariculture boundary, estimated at approximately 12.07 Gg C per year (J. Yu et al., 2025). These findings provide initial evidence that mariculture activities may exert a considerable influence on the spatial heterogeneity of OC thermochemical stability.

The additional thermochemical indicators of OC further verifies the contribute of mariculture activities (Figures 2b and 2c; Huang et al., 2023). The bulk of NY4 contains the lowest $f_{E<150}$ and σ_E, which implies the least marine-derived, fresh, and recently fixed biogenic OC input (Sarwar et al., 2020) and simplified OC sources, corresponding to the circumstance of being distal to mariculture areas, in concordance with the relatively higher μ_E. NY2-NY3 and NY4-NY5 constitute two internally contrasted groups to represent being outside and within mariculture zones within a relatively homogeneous natural environment, which facilitates the discrimination of mariculture-derived OC loading. The mariculture sites (NY3, NY5) have a relatively reduced μ_E (by 0.48% and 1.70%) coupled with a relatively elevated labile OC (by 10.85% and 47.66%), relative to their natural controls (NY2 and NY4). Besides, the proportion of recalcitrant OC_{terr} is relatively reduced in NY3 and NY5, by 5.60% and 0.87% respectively. Moreover, the difference between NY4 and NY5 is more pronounced than that between the other pairs, which can be contributed to the port fairway zones near NY4, where mariculture activities are strictly restrained, with a distance of 12.2 km from NY4 to the nearest mariculture boundary compared to NY2 (5.8 km). Although E trends of OC can be influenced by the physical disposition of particles, such as grain-size association (Bao, McNichol, Mcintyre, et al., 2018), this factor does not sufficiently account for the observed thermochemical differences: coarser grain size fractions above 63 μm (sand) are known to affect OC thermal reactivity (Bao, McNichol, Mcintyre, et al., 2018; Blanca et al., 2021), however, the sand content within sample groups shows inconsistent comparison. For instance, NY3 contains more sand than NY2 (13.9% vs. 7.5%), while NY4 contains more sand than NY5 (23.5% vs. 13.8%) (Figure S2a in Supporting Information S1). Combined with the pronounced grain-size sorting and limited alteration of OC thermochemical properties in sediment resuspension results, these divergent trends suggest that mariculture, rather than hydrodynamic-induced conditions, serves as the primary control on the observed spatial variation in thermochemical stability of sedimentary OC.

Given that thermochemical properties exhibit a spatial association with mariculture distribution (Figure 1) rather than with transport distance and hydrodynamic forcing (Figures 2b and 2c), we propose that the magnitude of mariculture-induced effect surpasses that of hydrodynamics sorting and transport in the nearshore area, as evidenced by the diminished heterogeneity (Shi et al., 2024; M. Yu et al., 2024; S. Zhao et al., 2024). The impact of mariculture primarily occurs by direct input of external labile OC sources to lower seawater and sediments through the biological activities of aquaculture (J. Yu et al., 2025). It directly modulates OC content and properties, manifested as elevated marine-derived contributions, a younger ^{14}C age, and lower thermochemical stability. The scope of impact is restrained near the mariculture area and depends on the production and type of mariculture (Zhou et al., 2024).

In contrast, the hydrodynamic processes exert chronic and pervasive controls on OC, while its mechanism differs between the delta and shelf regions (M. Yu et al., 2024). Initially, the selective deposition of coarse particles and rapid remineralization of the labile composition of OC_{terr} lead to sediment fining, an elevated OC content, a reduced OC_{terr} contribution, and rapid aging (M. Yu et al., 2024; S. Zhao et al., 2024; Zhong et al., 2017). Along the shelf transect, the residual OC_{terr} is recalcitrant, and the labile young OC_{mar} is presumed to be selectively degraded (Bao et al., 2019). Though sorting of particles and minerals (Figure 3), as well as prolonging the OET (Blanca et al., 2021) and physically breaking the mineral-OC bond (X. Liu et al., 2024), hydrodynamics modulate the OC content, while the variation of thermochemical properties of OC requires a protracted transport process driven by hydrodynamics. Particularly in regions distal to major river mouths, such reworking-related characteristic enables local OC inputs to be key regulators of spatial heterogeneity in thermochemical stability, evidenced by the influence of coastal streams around the Shandong Peninsula. Stations NY2 and NY3, located near multiple stream estuaries, receive more OC_{terr} input (Figure S3 in Supporting Information S1; B. Yang & Gao, 2019), resulting in elevated μ_E and σ_E values, indicative of enhanced stability and more heterogeneous OC provenance. The differential observations in variation of thermalchemical properties along the transport pathway and under hydrodynamic forcing imply that the mechanism, the scope, and the magnitude of the influence differ from mariculture and hydrodynamics. As for the nearshore area along the transport pathway, the aforementioned analytical synthesis compellingly demonstrates that extensive mariculture activities can supersede the influence of hydrodynamics on OC heterogeneity by replenishing additional labile OC_{mar} (Figure 5b; C. Liu et al., 2024; Xu et al., 2024).

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3.3. Implications for Anthropogenic OC Modulation

In recent years, mariculture activities, proposed as a means to enhance carbon sequestration efficiency (C. Liu et al., 2024; Xu et al., 2024), have become increasingly controversial (Forrest et al., 2009; L. Yu et al., 2025). The biogeochemical effects associated with these activities are complex, profound, and remain uncertain in terms of their magnitude (Q. Chen et al., 2016; C. Liu et al., 2024). Notably, negative environmental impacts caused by mariculture such as water pollution and biodiversity reduction (C. Liu et al., 2024) may reduce the sustainability of the carbon sink efficacy. While rapid expansion of mariculture suggests a potentially enhanced carbon sequestration capacity, such large-scale OC enrichment may pose risks on ecosystem (L. Yu et al., 2025). Particularly, amid escalating global climate challenges, it is imperative to conduct more detailed investigations into the impacts of mariculture activities on sedimentary OC dynamics. This study provides a proper perspective to distinguish the effect of mariculture activities from hydrodynamics through a comparison between the spatial heterogeneity of bulk OC and the content and components of OC in RS subjected to hydrodynamic forcing. The combined approach offers a valuable framework for multidimensional assessments of mariculture-induced alterations in OC properties. Unlike previous studies that primarily focused on quantifying OC fluxes and source apportionment associated with mariculture (Xu et al., 2024), our results demonstrate that mariculture can exert a decisive influence on the thermochemical reactivity of sedimentary OC, particularly in areas distant from river mouths where lack major terrigenous input, thereby reshaping the spatial heterogeneity of OC content and thermochemical stability. Mariculture activities enhance the downward transport of OC to the underlying water column and sediments, in a manner functionally analogous to the biological carbon pump, but mariculture-derived OC tends to exhibit higher thermochemical reactivity, resulting in a overall elevated lability of sedimentary OC. Unless buried, mariculture-enhanced OC sink may not be stable as natural process of biological carbon pump.

Our findings highlight the strong contrast between natural and mariculture conditions in shaping the spatial heterogeneity of sedimentary OC. Hydrodynamic resuspension experiments indicate that labile OC, regardless of origin, is not readily mobilized (Figure 2; X. Liu et al., 2024). This suggests that fine-grained sediments may play a key role in retaining OC through mineral-organic associations, which can limit desorption and contribute to the observed uniform thermochemical properties of the <63 μm fraction (Bao, McNichol, Mcintyre, et al., 2018). While direct evidence of mineral-bound OC microstructure was not obtained in this study, our results are consistent with the interpretation that such associations contribute to the consistent resistance of different OC components during hydrodynamic resuspension under shear stresses up to 0.6 Pa. This demonstrates the substantial capacity of anthropogenic activities to reshape sedimentary OC, and indicates that human influence on OC dynamics is already widespread (Pan et al., 2021), which warrant particular attention in regions markedly influenced by local environmental factors. In addition, this study offer support for further exploration and distinction of mariculture-derived influence on OC dynamics. Results from thermochemical analysis highlight the sensitivity of various thermal indices in evaluating the impact of mariculture on OC stability. For instance, σ_E shows stronger spatial variability and a closer correlation with the distribution of mariculture than μ_E (Figure 2b), making it a more sensitive and effective indicator for regional environmental variation. Furthermore, given the globally observed correlation between OC age and thermal reactivity (Bao et al., 2019), mariculture activities may also modulate OC ^{14}C age, making OC ^{14}C a potential auxiliary indicator to identify the contribute of mariculture. To improved quantification of mariculture OC stability and enhance estimates of sediment carbon sources to sink, further investigation is required to elucidate the quantitative links between mariculture-derived OC and mariculture characteristics, including species cultivated and culture density (J. Yu et al., 2025). Additionally, although labile and recalcitrant OC exhibit only minor differences in resistance to short-term hydrodynamic forcing, the threshold conditions under which their behaviors diverge remain unclear. Identifying the threshold combinations of hydrodynamic intensity and duration is essential for understanding mariculture-derived OC preservation.

4. Conclusions

We combined erosion microcosm experiment and analysis of characteristics of sediments and OC to investigate the differential impact of mariculture and hydrodynamics on OC. We propose that hydrodynamic sorting of grain size and minerals enables weak hydrodynamic-induced sediment resuspension to cause a disproportionate release of OC relative to the resuspended mass. However, this sorting does not directly alter the thermochemical properties of OC, regardless of hydrodynamic intensity. The mechanism is jointly governed by the properties of particles and OC. The coarse-grained sand fraction, which exhibits OC thermochemical difference associated

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with discrete OC, revealed resistance to resuspension into the water column, and thus lead to minor variation of thermochemical properties of OC, while the consistent thermochemical properties of OC in finer fractions reflect the stability of mineral-OC aggregates under hydrodynamic forcing. Due to rapid sediment deposition and OC degradation in estuarine regions, the combined influence of grain size characteristics and their correlation with OC collectively governs the evolution pattern of OC properties along the transport pathway, characterized by the initial rapid degradation of labile, discrete OC followed by a trend toward stabilization. In comparison, mariculture directly modulates OC content and properties by the input of labile OC, manifested as elevated marine-derived contributions, a younger ^{14}C age, and lower thermochemical stability.

We suggest that mariculture can supersede the influence of hydrodynamics on OC heterogeneity, leading to the offset of preferential degradation of labile OC, diminished OC aging along transport pathways, and disruption of the progressive stabilization trend of sedimentary OC along transport pathways under natural conditions. The reshape potential of mariculture on OC heterogeneity is particularly prominent in areas lacking major terrigenous input, supported by evidence showing that variations in thermochemical properties are linked to the distribution of mariculture area which addresses a previously overlooked aspect of the thermochemical properties of mariculture-derived OC. The impact of mariculture on OC properties is profound since OC components with different thermochemical reactivity demonstrates consistent resistance during hydrodynamic resuspension. Further investigation into the carbon sink benefits of mariculture is necessary to comprehensively assess its ecological impact and needs to integrate multidimensional analysis to disaggregate and quantify mariculture impacts on OC-ecosystem feedback loops, providing critical insights for optimizing carbon sequestration strategies.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

Data reported in this study are available at https://doi.org/10.5281/zenodo.15773665.

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