



Unveiling carbonate dissolution in coastal sediments and its influence on seawater buffering capacity with $\delta^{13}\text{C}_{\text{DIC}}$ and ^{224}Ra – ^{228}Th disequilibria

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Abstract

Organic carbon mineralization is generally recognized as the primary source of dissolved inorganic carbon (DIC) released from sediments in coastal seas. The CO_2 accumulation or the formation of corrosive microenvironment induced by organic carbon degradation can promote the dissolution of calcium carbonate (CaCO_3) in sediments, complicating the efficiency of carbon burial and total alkalinity (TA) inputs to aquatic environments. However, quantitative assessments of sediment CaCO_3 dissolution and its impacts on the seawater carbonate remain poorly constrained. In this study, we selected typical high-productivity regions, mariculture farms, and applied the ^{224}Ra – ^{228}Th disequilibrium approach to quantify the effluxes of DIC and TA across the sediment-water interface. Stable carbon isotopes of DIC ($\delta^{13}\text{C}_{\text{DIC}}$) were employed to trace DIC sources in porewater. The results showed that CaCO_3 dissolution in sediments accounted for 27–56% of the benthic DIC efflux. Notably, a high contribution of CaCO_3 dissolution did not coincide with strong organic carbon degradation across sites, suggesting that dynamic disturbance on sediments, which weakened the metabolic CO_2 accumulation in porewater, was also a crucial factor affecting carbonate dissolution. According to the evaluation of the influence that benthic DIC and TA efflux exerted on the seawater CO_2 content, the TA supplied by the CaCO_3 dissolution was identified to enhance the carbonate buffering capacity of seawater and counteracted the acidification driven by organic matter remineralization. This indicates that CaCO_3 dissolution in sediments should be involved in coastal carbon cycling and assessments on coastal ecosystem resilience under the risk of CO_2 elevation.

Keywords: Sediment CaCO_3 dissolution; DIC- $\delta^{13}\text{C}$; Benthic efflux; ^{224}Ra – ^{228}Th disequilibria



1 Introduction

Over half of atmospheric carbon dioxide (CO₂) is estimated to be fixed by marine primary production and the coastal ocean contributes over 1.0 Pg C y⁻¹ (Bauer et al., 2013; Mathis et al., 2024). Consequently, the coastal sediment becomes a crucial carbon reservoir as a result of particulate carbon deposition, particularly in high production regions such as estuaries and seaweed farms (Friedlingstein et al., 2025; Duarte et al., 2025). Some of the buried carbon undergoes early diagenetic processes, e.g., aerobic mineralization, and is transformed into dissolved inorganic carbon (DIC), reducing the long-term carbon burial (Burdige et al., 2016; Silverberg et al., 2000). For instance, approximately 35% of the deposited carbon in the surface sediment was returned to the water column in the Baltic Sea (Wakeham and Canuel, 2006); and studies in China estuaries demonstrated that DIC effluxes from sediments were in the same order of magnitude with those from the riverine or submarine groundwater discharges (Cai et al., 2015; Hong et al., 2017). Meanwhile, the accumulation of DIC may decrease porewater pH, potentially degrading the ecological environment.

In general, the degradation of organic carbon is considered the dominant process to add DIC to sediment porewater in coastal seas (Berner, 1980; Middelburg, 2018). Some studies have demonstrated that carbonate dissolution can be triggered by microbial respiration even in the calcite-oversaturated waters (Cai et al., 2006; Wang et al., 2025). The role of calcium carbonate (CaCO₃) dissolution in absorbing CO₂ and increasing total alkalinity (TA) in marine systems is recognized (Jahnke et al., 1997; Morse et al., 2006). Marine calcifying organisms are widely distributed throughout the ocean so surface sediments above the saturation horizon generally contain abundant CaCO₃ debris (Harikrishnan and Nathan, 2023; Ying et al., 2024), implying that both CaCO₃ dissolution and organic matter degradation likely occur ubiquitously within sediments. Some chamber experiments have demonstrated that the CO₂ produced from organic matter degradation drove CaCO₃ dissolution in marginal sea sediments. However, the extent of such CO₂-induced dissolution varied widely in different regions. As the chamber experiments mostly focus on biochemical influences on CaCO₃ dissolution, the constraints of dynamic processes are still unclear (Jahnke et al., 1994; 1997; Ortega et al., 2005). Moreover, the TA increased by CaCO₃ dissolution is demonstrated to mitigate the risks of water acidification induced by organic matter remineralization (Millero, 2007; Boudreau et al., 2018; LaRowe et al., 2020), but quantitative influence on the carbonate buffering capacity of seawater is also lack of constraint. Therefore, it is crucial to clarify the mechanism on biogeochemical cycling of inorganic carbon at the sediment-water interface, in particular when CaCO₃ dissolution may act as an important regulator maintaining the long-term stability of marine carbonate systems.

Sansha Bay in Fujian Province (China) represents one of major mariculture regions in the world and has undergone rapid expansion of seaweed farming in recent decades (FAO, 2024). Previous studies have



shown that the sediment carbon inventories under the farming zones have been ten times higher than those in non-mariculture seas (Duarte et al., 2025; Zhao, 2023; Wang et al., 2023). Despite such high productivity, the seawater in the bay does not deteriorate into acidification. In addition to active tidal exchange with shelf waters, CaCO₃ dissolution is hypothesized to play a key role in counteracting acidification (Wang et al., 2025). However, the regulating mechanism and quantitative influence on seawater are still unclear. In this study, a natural radionuclide approach (²²⁴Ra–²²⁸Th disequilibrium, Cai et al., 2015) was applied to quantify benthic fluxes of DIC and TA in Sansha Bay. By integrating stable carbon isotopic signatures (δ¹³C) of sedimentary carbon (Alling et al., 2012), we further identify the relative contributions of organic carbon degradation and CaCO₃ dissolution to benthic DIC release under different benthic environments, delineating the mechanism controlling sediment CaCO₃ dissolution. At last, the processes improving the seawater buffering capacity were recognized by comparing the changes of CO₂ and pH induced by various processes, elucidating the benthic influence on the carbonate system of seawater.

80 **2 Materials and methods**

2.1 Study sites and sample collection

Sansha Bay (26.5–26.8°N, 119.5–120.1°E) is a semi-enclosed bay located in southeastern China with water depths ranging from several meters to ~50 m. The local currents are dominated by tides and the rates of tidal currents are over 1 m s⁻¹ near channel zones (Lin et al., 2019). The seaweed farms are widely distributed throughout Dongwuyang and the tidal zones adjacent to the main channels, and are controlled by regular semi-diurnal tides. Macroalgae-shellfish co-culture is the main practice of farms in Dongwuyang. The dominant macroalgae species are *Saccharina japonica* and *Gracilariopsis lemaneiformis*, and shellfish species are *Haliotis discus hannai* and *Crassostrea* (Wang et al., 2019).

Field work was conducted in 2024 and three stations were selected to represent contrasting mariculture settings (Fig. 1). Station SSW-A is located within the Dongwuyang seaweed farms, where the sediment contains abundant shell debris and polychaetes fragments (see Fig. S1 in Appendix). SSW-B is located in a sea cucumber culture zone adjacent to the main channel, with comparatively low farming intensity. SSW-C, near the bay mouth and outside the seaweed-farming zone, serves as a reference site for a non-mariculture condition. Water depth varied with tides and the specific depths during core collection ranged from 4–9 m. Detailed sampling information was listed in Appendix Table S1. In particular, sediments in the embayment are in great heterogeneity due to the mariculture behaviors. Human activities such as channel dredging and seasonal feeding can also change the sediment characters. The sampling sites in this study were mainly considered for the different hydrodynamic environments (inner and outer bay) and anthropogenic influences (with and without mariculture), not for the



100 representative of the whole embayment.

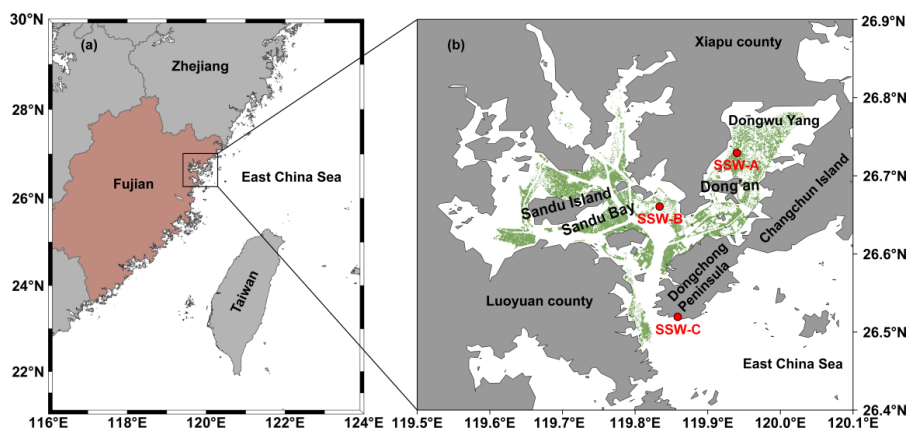


Figure 1: Sampling stations in Sansha Bay (red circles). The green-shaded area within the bay indicates the distribution of mariculture.

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Bottom water was collected with a 5-liter Niskin bottle. Samples for carbonate parameters (DIC, TA and $\delta^{13}\text{C}_{\text{DIC}}$) were stored in 250 mL borosilicate glass bottles and ~5 L of water was transferred into a clean cubitainer for ^{224}Ra enrichment. Sediment cores were collected by a single-tube sampler, using polycarbonate tubes (i.d. 65 mm) with pre-drilled holes. After siphoning off the overlying water, sediment porewater was extracted using Rhizon samplers via the holes at 1–2 cm intervals (Seeberg-Elverfeldt et al., 2005). Porewater from two cores was stored for carbonate analysis, ensuring the total volume >10 mL; the same depth porewater from different cores was combined so >30 mL porewater was used for a ^{224}Ra measurement. An intact core longer than 15 cm was sliced at a 1-cm interval for ^{224}Ra – ^{228}Th disequilibria and particle analysis (Cai et al., 2015). The sediment was characterized by silty clay and clayey silt. Seawater and porewater samples for DIC and TA were killed with saturated HgCl_2 solution by 0.5‰ (vol./vol.). Water samples for dissolved ^{224}Ra ($\text{D-}^{224}\text{Ra}$) were filtered through 142-mm glass fiber filters (GF/F) and stored at room temperature for radium enrichment.

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2.2 Sample analysis

2.2.1 ^{224}Ra and ^{228}Th

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Each of the sediment slices was transferred into a 250 mL Teflon beaker and mixed with approximately 200 mL of Milli-Q water. The slurry was thoroughly stirred and subjected to ultrasonication for 5–10 minutes to ensure complete dispersion of sediment particles. Subsequently, concentrated ammonia solution was added to the slurry to adjust the pH to 8–9. Thereafter 1.00 mL of



125 KMnO₄ solution (3.0 g L⁻¹) and 1.00 mL of MnCl₂ solution (8.0 g L⁻¹ MnCl₂·4H₂O) were sequentially
added to form a MnO₂ suspension that coprecipitated Ra from the slurry. The particles of the slurry were
filtered through a 142-mm GF/F filter (0.7-μm nominal pore size), and the bulk ²²⁴Ra activity of the
sediment was measured using a radium delayed coincidence counting system (RaDeCC, Scientific
Instrument Inc., USA). After ~10 days and ~25 days, when ²²⁴Ra had ingrown to secular equilibrium with
its parent ²²⁸Th, the second and third measurements were conducted in the same system. The final
130 activities of both ²²⁴Ra and ²²⁸Th in the sediment were calculated from the first and the average of the last
two measurements (Cai et al., 2012). A comparison between the last two runs was performed to check the
reproducibility, yielding a mean ratio of 1.018 ± 0.026 ($R^2 = 0.98$, $n = 29$), suggesting that the RaDeCCs'
efficiency kept stable during the analysis.

135 In the enrichment of D-²²⁴Ra, 30–40 mL of porewater was transferred into a 100 mL Teflon beaker
and ²²⁴Ra was coprecipitated with MnO₂ following the similar procedure to the bulk ²²⁴Ra. The
precipitates were collected on a 47-mm GF/F filter. For the bottom water (~5 L), 5 mL of KMnO₄ (3.0 g
L⁻¹) and MnCl₂ (8.0 g L⁻¹ MnCl₂·4H₂O) solutions were sequentially added into the sample and mixed for
full reaction. The formed MnO₂ coprecipitates were then filtered onto a 142-mm GF/F filter. D-²²⁴Ra
activities were determined by counting the filters in RaDeCCs for more than 8 hours (Cai et al., 2015).

140 The final ²²⁴Ra and ²²⁸Th activities were corrected with chance coincidences, detector efficiencies
and self-decay (Cai et al., 2012), and the uncertainties, propagated from counting statistics, were
approximately 5% for total ²²⁴Ra-²²⁸Th and 8–10% for D-²²⁴Ra. According to the standard deviation of
the activity ratios of ²²⁴Ra to ²²⁸Th (²²⁴Ra/²²⁸Th) in aged sediment samples ($n = 29$), sediment layers with
²²⁴Ra/²²⁸Th of 1.00 ± 0.05 were considered to be in secular equilibrium between ²²⁴Ra and ²²⁸Th.

145 2.2.2 DIC, δ¹³C_{DIC} and TA

The DIC concentrations in seawater and porewater were determined with a non-dispersive infrared
analyzer (Li-Cor 7000, USA) after acidification with a mixture of 3% H₃PO₄ and 7% NaCl (Sun et al.,
2024). The relative standard deviation (RSD) of replicates was well controlled less than 0.2%. The δ¹³C_{DIC}
(‰) was analyzed with an automatic DIC-δ¹³C analyzer with an accuracy (1σ) of ±0.1‰ (Picarro G2131-
150 i, USA). The TA was measured following the Gran titration (Gran, 1952). Porewater samples were first
diluted with TA-known aged seawater. The titrant was 0.1 mol L⁻¹ HCl and the titration was automatically
terminated using a titrator (Apollo AS-ALK3, USA) with an Orion AR1-17304 pH electrode. Each TA
sample was measured 2–3 times, yielding the RSD within ±0.5% after volume correction. Both DIC and
TA concentrations were calibrated with Scripps Certified Reference Material (CRM, Batch #198)
155 (Dickson et al., 2003).



2.2.3 The $\delta^{13}\text{C}$ isotope of sediment TOC ($\delta^{13}\text{C}_{\text{TOC}}$)

Dried sediment samples were ground and homogenized with a mortar and pestle. Approximate 1 g of each sample was weighed and acidified with 1.2 N HCl (guaranteed reagent, GR). The mixture was subjected to ultrasonication for 1 h, followed by a 5-h water bath at 60°C to completely remove the inorganic carbon. Subsequently, the samples were repeatedly rinsed with Milli-Q water until the pH of the supernatant was neutral. The remaining particles were then dried at 60°C and accurately weighed. The mass loss relative to the dry sediment was assumed as total CaCO_3 in this study. Total inorganic carbon (TIC) was calculated from CaCO_3 and the precision (0.1 mg) was based on that of the weight scale. The TOC content was determined using an elemental analyzer (vario ISOTOPE, Elementar., Germany) and calibrated with an acetanilide standard (Sigma-Aldrich). The $\delta^{13}\text{C}_{\text{TOC}}$ was simultaneously measured with a continuous-flow isotope ratio mass spectrometer and the accuracy (1σ) was $\pm 0.1\%$.

2.3 Benthic effluxes estimated from ^{224}Ra – ^{228}Th disequilibria

In Sansha Bay, intensive aquaculture activities introduce shell fragments and coarse particles into the sediments (see Appendix Fig. S1). Influenced by tidal currents and biological activities, the sediment in the bay is generally permeable. The exchangeable interface between sediment and seawater is further extended by active benthic faunal irrigation. In such a dynamic system, traditional methods such as diffusion or incubation experiments may fail to accurately estimate benthic exchange fluxes (Shi et al., 2018). In contrast, the ^{224}Ra – ^{228}Th disequilibrium approach has been confirmed as a robust method for capturing complex interfacial processes in most China coastal seas (Shi et al., 2019).

In the ^{232}Th decay series, ^{224}Ra is the daughter of ^{228}Th . Owing to the distinct particle reactivities, ^{224}Ra can be desorbed from particles in saline waters, whereas ^{228}Th remains strongly bound on sediment particles. With exchanges between bottom waters and porewaters, total ^{224}Ra generally presents deficits relative to its parent ^{228}Th in surface sediments (Cai et al., 2015). Therefore, the ^{224}Ra efflux (F_{Ra}) across the sediment-water interface can be quantified from the disequilibria between ^{224}Ra and ^{228}Th (Eq. 1, Cai et al., 2015):

$$F_{\text{Ra}} = \lambda_{\text{Ra}} \int_0^z (A_{\text{Th}} - A_{\text{Ra}}) dz \quad (1)$$

where λ_{Ra} represents the decay constant of ^{224}Ra (0.189 d^{-1}), A_{Ra} and A_{Th} represent the ^{224}Ra and ^{228}Th activities in bulk sediments (dpm cm^{-3}) corrected with porosity (Φ), respectively, and z denotes the depth at which ^{224}Ra and ^{228}Th approach secular equilibrium. This efflux reflects the synergistic effect with molecular diffusion, bioturbation and irrigation at the interface so a diffusion model embedding increased surface area can be applied to stand for the exchange:

$$F_{\text{Ra}} = \xi \Phi D_s^{\text{Ra}} \frac{\partial A_{\text{DRa}}}{\partial z} \quad (2)$$

where ξ denotes the factor indicating the enhancement of exchangeable area, D_s^{Ra} represents the



190 molecular diffusion coefficient of ^{224}Ra that has been corrected with in-situ temperature and sediment porosity, and $\frac{\partial A_{\text{DRa}}}{\partial z}$ is the ^{224}Ra activity gradient at the sediment-water interface.

Under the same circumstance, the area-enhancement model should be appropriate to other solutes so the benthic fluxes of carbonate parameters (DIC and TA) can be estimated as

$$F_m = \xi \Phi D_s^m \frac{\partial C^m}{\partial z} \quad (3)$$

195 where F_m refers to the benthic flux of DIC or TA across the sediment-water interface, D_s^m represents the diffusion coefficient of bicarbonate (HCO_3^-), and $\frac{\partial C^m}{\partial z}$ denotes the concentration gradient of DIC or TA at the interface.

2.4 The model for tracing sources of porewater DIC

In marine environments, $\delta^{13}\text{C}_{\text{TOC}}$ is generally around -20‰ (Jahnke et al., 1997; Hu and Burdige, 2007), whereas CaCO_3 in shells exhibits $\delta^{13}\text{C}$ values ($\delta^{13}\text{C}_{\text{CaCO}_3}$) ranging from -7.7‰ to -2.6‰ (Surge et al., 2001; Gillikin et al., 2006; Hadden et al., 2018). Although sediment porewater is derived from bottom seawater, the degradation of organic matter and the dissolution of CaCO_3 in sediments can add lighter carbon to porewater DIC (DIC_{PW}), resulting in the $\delta^{13}\text{C}$ of DIC_{PW} more negative than those of bottom water (DIC_{BW}). Assuming that the differences in DIC concentrations between bottom water and porewater are solely attributed to the two processes (organic carbon degradation and CaCO_3 dissolution), their relative contributions to the porewater DIC can be defined as f_{TOC} and f_{CaCO_3} , namely

$$f_{\text{CaCO}_3} + f_{\text{TOC}} = \frac{\text{DIC}_{\text{PW}} - \text{DIC}_{\text{BW}}}{\text{DIC}_{\text{PW}}} \quad (4)$$

The isotopic composition of DIC in porewater ($\delta^{13}\text{C}_{\text{PW}}$) can be expressed as (Alling et al., 2012)

$$\delta^{13}\text{C}_{\text{PW}} \cdot \text{DIC}_{\text{PW}} = \delta^{13}\text{C}_{\text{BW}} \cdot \text{DIC}_{\text{BW}} + \delta^{13}\text{C}_{\text{CaCO}_3} \cdot f_{\text{CaCO}_3} \cdot \text{DIC}_{\text{PW}} + \delta^{13}\text{C}_{\text{TOC}} \cdot f_{\text{TOC}} \cdot \text{DIC}_{\text{PW}} \quad (5)$$

210 where $\delta^{13}\text{C}_{\text{BW}}$ represents the $\delta^{13}\text{C}$ -DIC in bottom water; and $\delta^{13}\text{C}_{\text{CaCO}_3}$ and $\delta^{13}\text{C}_{\text{TOC}}$ stand for the $\delta^{13}\text{C}$ of shell debris and organic carbon (TOC) in the sediment, respectively. By combining Eq. (4) and Eq. (5), f_{TOC} and f_{CaCO_3} can be calculated.

2.5 Significance Tests

Significance tests were performed using R (R Core Team, 2023). The Shapiro–Wilk test (shapiro.test) was used to assess the normality of $\delta^{13}\text{C}_{\text{DIC}}$ in porewaters and bottom waters, and a t -test was applied for the value comparison. In Sect. 4.3, a linear mixed-effects model (from the package 'lme4') was employed to test whether the theoretical $\delta^{13}\text{C}_{\text{DIC}}$ derived solely from TOC degradation matched the measured porewater $\delta^{13}\text{C}_{\text{DIC}}$.



3 Results

3.1 The distributions of ^{224}Ra and ^{228}Th in the sediment

220 The total ^{224}Ra and ^{228}Th present similar vertical distributions in sediments as ^{224}Ra is derived from
the radioactive decay of ^{228}Th (Fig. 2a). At SSW-A and SSW-C, deficits of ^{224}Ra , with the average
 $^{224}\text{Ra}/^{228}\text{Th}$ of 0.89 ± 0.05 ($n = 14$, 1SD), were observed in the upper 10 cm, whereas equilibrium between
 ^{224}Ra and ^{228}Th was characterized below 12 cm ($^{224}\text{Ra}/^{228}\text{Th} = 0.98 \pm 0.03$, $n = 4$). For SSW-B, the total
225 ^{224}Ra was nearly in equilibrium with ^{228}Th (Fig. 2b). In the outer-bay sediments (SSW-C), the ^{224}Ra and
 ^{228}Th activities were in the range of 1.70 ± 0.10 – 4.55 ± 0.29 dpm g^{-1} and 2.01 ± 0.06 – 4.54 ± 0.16 dpm g^{-1} ,
respectively. In comparison, the sediment in the inner bay (SSW-A and SSW-B) had lower radionuclide
activities (1.34 ± 0.09 – 3.00 ± 0.20 dpm g^{-1} for ^{224}Ra and 1.40 ± 0.05 – 3.25 ± 0.10 dpm g^{-1} for ^{228}Th , Table
S1 in Appendix).

Vertical profiles of D- ^{224}Ra activities in porewater and bottom water (depth = 0) are presented in Fig.
230 2c. The activities of D- ^{224}Ra in bottom water, ranging from 0.11 ± 0.01 to 0.39 ± 0.02 dpm L^{-1} , were 1–2
orders of magnitude lower than those in porewaters (Table S2 in Appendix). The differences in porewater
D- ^{224}Ra between stations were consistent with the total ^{224}Ra in sediments. Namely, D- ^{224}Ra activities at
SSW-C (16.2 ± 1.9 – 52.4 ± 3.1 dpm L^{-1}) were higher than those at inner-bay stations (4.0 ± 0.8 – 25.8 ± 2.4
dpm L^{-1}). At depths where total ^{224}Ra was deficient relative to ^{228}Th , D- ^{224}Ra activities exhibited obvious
235 variations. Correspondingly, layers where ^{224}Ra – ^{228}Th activities approached equilibria, such as 4–10 cm
at SSW-B, coincided with stable distributions of D- ^{224}Ra (RSD < 7%).

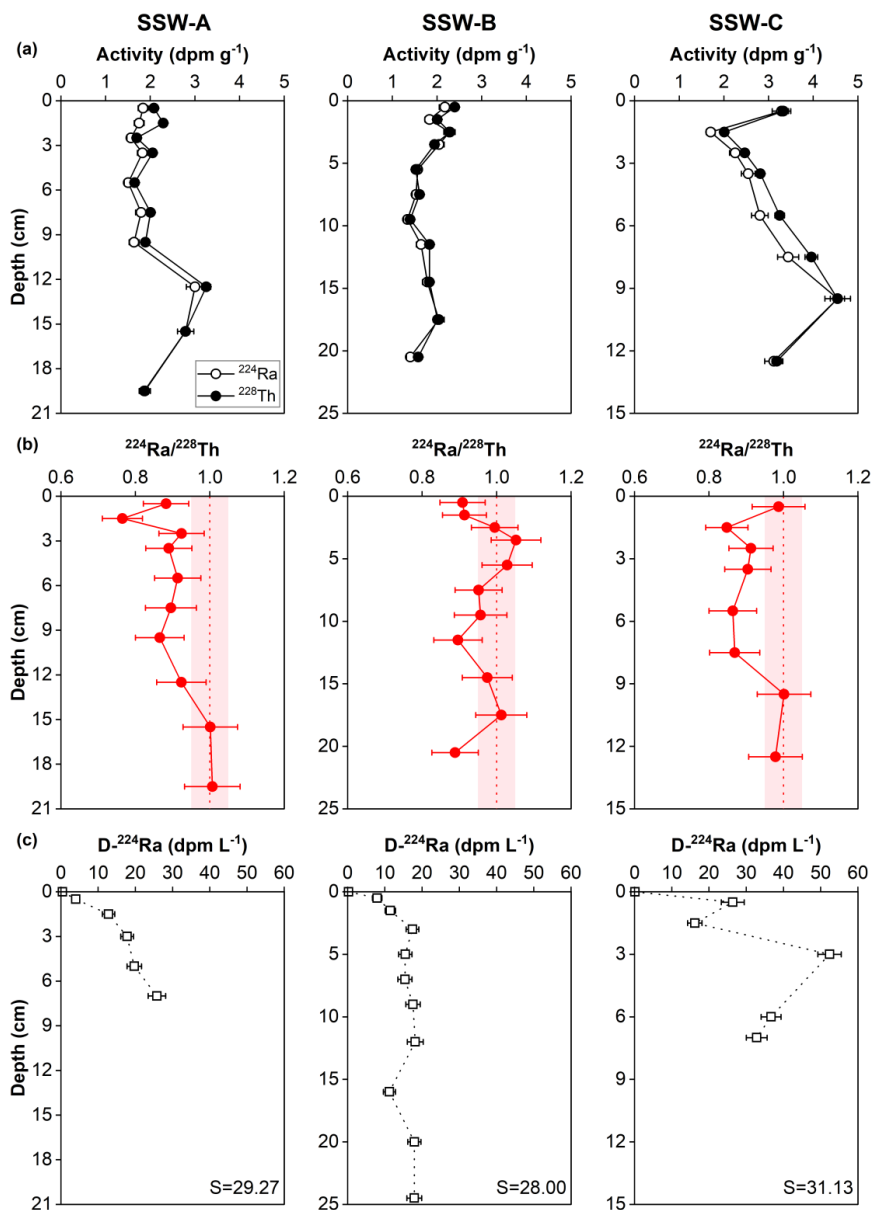


Figure 2: (a) Vertical profiles of bulk ²²⁴Ra and ²²⁸Th (error = ± 1σ) activities in the sediment. (b) Activity ratios of ²²⁴Ra to ²²⁸Th, with the shaded indicating secular equilibrium between ²²⁴Ra and ²²⁸Th (²²⁴Ra/²²⁸Th = 1.0 ± 0.05); (c) Vertical profiles of D-²²⁴Ra (error = ± 1σ) in porewater (the symbol at depth = 0 represents the bottom water). Note that the activity ranges and depth ranges differ slightly among stations.



3.2 The distributions of DIC, TA, and $\delta^{13}\text{C}_{\text{DIC}}$ in the porewater

245 In the bottom water, the DIC and TA concentrations were in the range of 1964–2229 $\mu\text{mol kg}^{-1}$ and
 1982–2357 $\mu\text{mol kg}^{-1}$, respectively. The porewater exhibited particularly higher DIC and TA (ranging
 from 2184 to 3007 $\mu\text{mol kg}^{-1}$, from 2326 to 3081 $\mu\text{mol kg}^{-1}$, respectively) than the bottom water (Table
 S3 in Appendix). At SSW-A and SSW-C, the DIC and TA concentrations generally increased with depth
 (Fig. 3). At SSW-B, unexpectedly, both DIC and TA concentrations decreased with depth below 3 cm,
 250 and the concentrations of DIC above 15 cm were higher than TA.

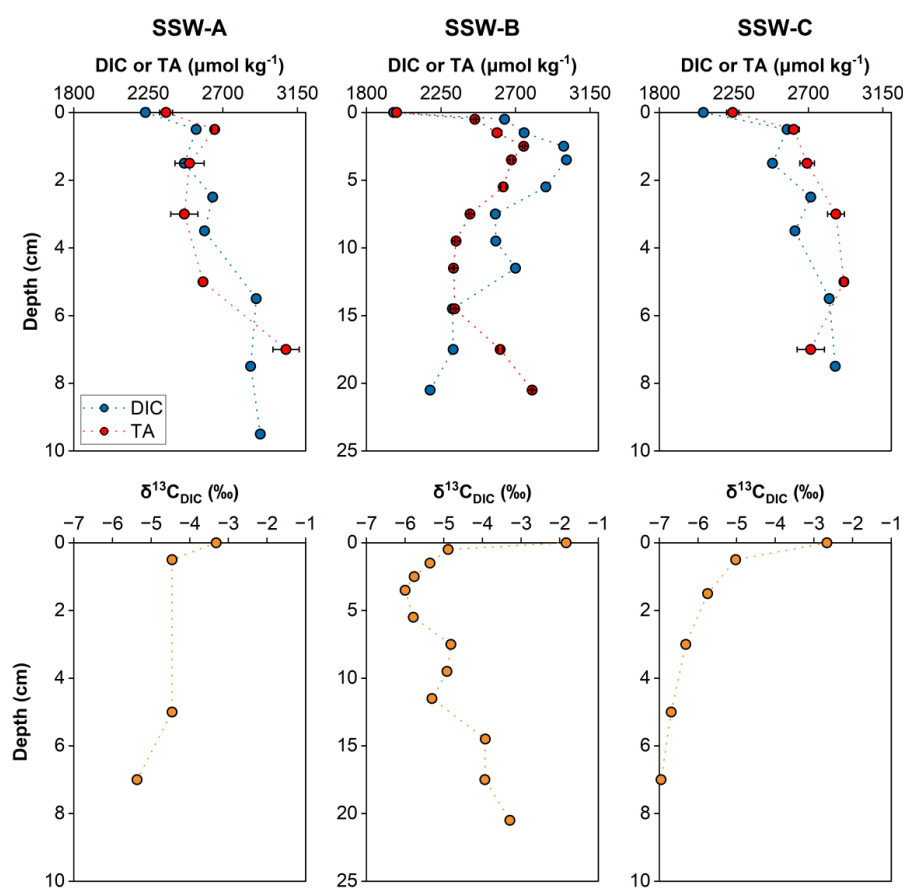


Figure 3: Vertical profiles of DIC, TA (error = $\pm 1\text{SD}$), and $\delta^{13}\text{C}_{\text{DIC}}$ in porewater, where depth = 0 represents the bottom water layer. Note that the depth ranges at Station SSW-B are different from the other stations.

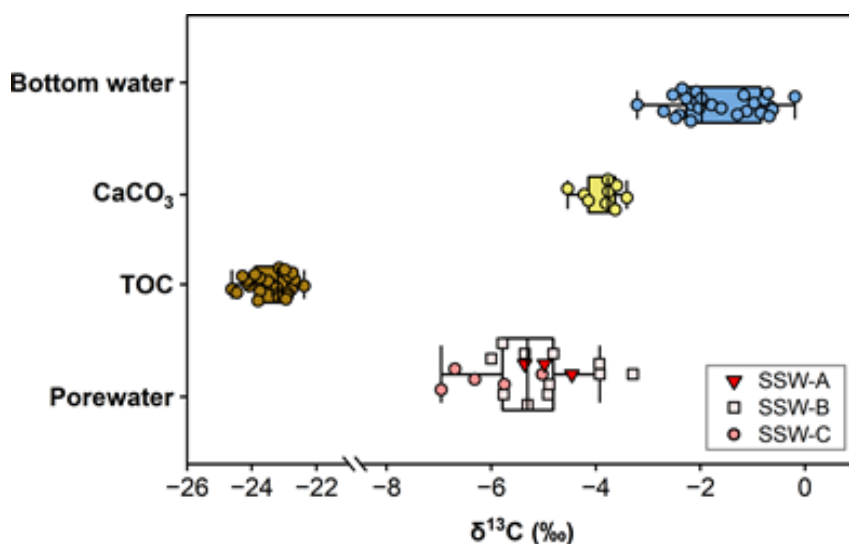
In comparison, the profiles of porewater $\delta^{13}\text{C}_{\text{DIC}}$ and DIC overall exhibited opposite trends although



the $\delta^{13}\text{C}_{\text{DIC}}$ values at station SSW-A were discrete due to the restricted number of samples (Fig. 3). The $\delta^{13}\text{C}_{\text{DIC}}$ of the bottom water in our study area ranged from -3.3‰ to -1.8‰, more negative than the values of offshore waters in the East China Sea (-2.0–0.5‰) (Wang et al., 2016; Duan et al., 2022). The $\delta^{13}\text{C}_{\text{DIC}}$ values of the porewater varied between -7.0‰ and -3.3‰, significantly lower than those of the bottom water ($p < 0.001$, t -test, Fig. 4). Especially, the porewater $\delta^{13}\text{C}_{\text{DIC}}$ at SSW-A was the heaviest while that at SSW-C contained the lightest $\delta^{13}\text{C}_{\text{DIC}}$ although the DIC concentrations of the two stations were comparable.

265 3.3 The $\delta^{13}\text{C}$ in shells and sediment TOC

Shells for $\delta^{13}\text{C}_{\text{CaCO}_3}$ were collected from oysters and abalones and the values ranged from -4.5‰ to -3.4‰ ($n = 9$) with a median of -3.8‰ (Fig. 4). The sediment $\delta^{13}\text{C}_{\text{TOC}}$ of all subsamples was in the range of -24.6–-22.4‰ ($n = 27$), the median of which was -23.3‰. In particular, the TIC contents were highest in the sediment of SSW-A (15.4 to 35.4 mg g^{-1} , Table S1 in Appendix), coincident with the abundant shell debris in the sediment and the relatively positive porewater $\delta^{13}\text{C}_{\text{DIC}}$ at this station (Fig. S1 in Appendix).



275 **Figure 4: The $\delta^{13}\text{C}$ values in different matrix. CaCO_3 represents shells collected from oysters and abalones. TOC refers to the sediment TOC at our sampling sites.**



4 Discussion

4.1 The mechanism of interfacial exchange revealed by the ^{224}Ra – ^{228}Th disequilibria

In comparison of ^{224}Ra profiles, the high D - ^{224}Ra in porewater corresponds to the high bulk ^{224}Ra in
280 sediment (SSW-C). The layers where ^{224}Ra – ^{228}Th activities presented disequilibria coincided with great
variations of D - ^{224}Ra (RSD of three adjacent D - ^{224}Ra larger than 10%), indicating that the sediment served
as the major source of D - ^{224}Ra . As the ^{224}Ra – ^{228}Th disequilibria provided a reliable indicator of D - ^{224}Ra
transfer, the ^{224}Ra effluxes (F_{Ra}), as well as the interface area factor (ξ), were calculated with the method
in Sect. 2.3 (Eq. 1 and Eq. 2) based on the activity gradients of D - ^{224}Ra at the sediment-water interface
285 (Table 1). The integration depth of SSW-A and SSW-C was set at the first layer where the ^{224}Ra and ^{228}Th
reached equilibrium. For SSW-B, the full core was integrated as the radionuclides were basically in
equilibrium. The gradients were calculated from the D - ^{224}Ra activity differences between 0-1 cm
porewater and bottom water.

Although the ^{224}Ra fluxes at SSW-A and SSW-C were similar, the ξ at SSW-A was 6–7 times higher.
290 Given that SSW-A was located in seaweed farms where dense macrobenthic communities existed (Wang
et al., 2019), the sediment contained abundant burrows and tunnels generated by benthic fauna activities,
effectively increasing the area of the sediment-water interface and thereby enhancing the ^{224}Ra flux. In
contrast, the ξ values at SSW-B and SSW-C were comparable but the ^{224}Ra flux at SSW-C was
approximately three times higher, implying that processes other than bioturbation stimulated the benthic
exchange at SSW-C. As the Dongchong Channel is dominantly affected by semi-diurnal tides, the
295 sediment-water interface at SSW-C nearby is subject to stronger hydrodynamics than SSW-B (Lin et al.,
2017). Fine particles were entrained to open waters by tidal currents and the coarser ones were retained
so the sediment at SSW-C was of permeability. Therefore, the benthic effluxes were dominated by
bioactivities in inner-bay mariculture zones while tidal currents and flow turbulence accounted for the
300 active interfacial exchange in outer-bay zones.



310 **Table 1: The molecular diffusion coefficient of ^{224}Ra (D_s^{Ra}), the activity gradient of D- ^{224}Ra ($\frac{\partial A^{\text{Ra}}}{\partial z}$), the integration depth (z_{int}) used to calculate the benthic flux of ^{224}Ra , the ^{224}Ra flux (F_{Ra}) and the interface area enhancement factor (ξ) at each sampling station. Uncertainties for all parameters are reported as $\pm 1\sigma$.**

Station	D_s^{Ra} ($10^{-6} \text{ cm}^2 \text{ s}^{-1}$)	$\frac{\partial A^{\text{Ra}}}{\partial z}$ ($\text{dpm L}^{-1} \text{ cm}^{-1}$)	z_{int} (cm)	F_{Ra} ($\text{dpm cm}^{-2} \text{ d}^{-1}$)	ξ
SSWA	4.5	7.16 ± 1.54	16	0.53 ± 0.11	260.1 ± 78.44
SSWB	4.4	15.4 ± 2.50	21	0.17 ± 0.08	41.22 ± 19.49
SSWC	3.8	52.5 ± 6.15	10	0.54 ± 0.13	44.75 ± 11.64

315 Moreover, the ^{224}Ra effluxes were utilized to quantify the residence time (τ) of the bay water if we assumed that the addition of ^{224}Ra in the inner bay were all derived from the sediment. According to the ^{224}Ra activities of water column in Sansha Bay (Wang et al., 2018), we constrained the endmembers in Sandu Bay and selected the ^{224}Ra activities of mid-salinity tidal water and bay-mouth seawaters to plot the conservative mixing line (Fig. S2a in Appendix). Consequently, low-salinity inputs from rivers or submarine groundwater discharge were not included in this model. Excess ^{224}Ra above the conservative line (ΔRa) was assumed to be derived from the sediment so τ can be estimated as

320
$$\tau = \frac{\Delta\text{Ra}}{F_{\text{Ra}}} h \quad (6)$$

where h denotes the average water depth of the bay, and a value of 14 ± 3 m was assigned in consideration of the tidal ranges and the water depths measured during the field work. Since all of seawater ^{224}Ra referred to were collected within Sandu Bay (Wang et al., 2018; Fig. S2b in Appendix), we adopted the ^{224}Ra flux of SSW-B, which is located in the middle of the bay, to represent the average benthic ^{224}Ra input to the bay (F_{Ra}). It should be noted that the ^{224}Ra released from the sediment was assumed to be rapidly mixed into seawater due to the influence of semi-diurnal tides, and that the radioactive decay of ^{224}Ra was not considered due to the frequent water exchange in this model. Therefore, the τ estimated here represents the low limit of the water residence time. By substituting these values into Eq. (6), the seawater residence time based on ΔRa of Sandu Bay ranged from 0.11 to 3.7 days (Fig. S2b), comparable to those of the wet season (0.2–3.0 days) in Sandu Bay estimated by the seawater $^{224}\text{Ra}/^{228}\text{Ra}$ (Wang et al., 2018).

4.2 Fluxes of DIC and TA across the sediment-water interface

By applying the area-enhancement model to other solutes (Eq. 3 in Sect. 2.3), the benthic fluxes of



DIC and TA at each station were estimated. The gradients of DIC and TA were calculated from the
335 concentration differences between 0-1 cm porewater and bottom water (Table 2). The largest DIC and
TA fluxes, approximately $600 \text{ mmol m}^{-2} \text{ d}^{-1}$, were exhibited in the farming zone (SSW-A) although the
porewater DIC and TA concentrations were not the highest. The fluxes at the other two stations ranged
from 105 to $207 \text{ mmol m}^{-2} \text{ d}^{-1}$. The magnitude of these DIC fluxes is comparable to the estimates in the
intermediate-salinity zones of the nearby Jiulong River estuary ($26\text{--}1000 \text{ mmol m}^{-2} \text{ d}^{-1}$, Hong et al., 2017).
340 In particular, the high benthic DIC and TA fluxes were dominated by the enlarged interface areas (ξ). If
only molecular diffusion was considered, benthic DIC flux at SSW-A ($\sim 2.4 \text{ mmol m}^{-2} \text{ d}^{-1}$) fell within the
reported range derived from the porewater gradient model ($0.54\text{--}2.65 \text{ mmol m}^{-2} \text{ d}^{-1}$, Jahnke et al., 1997),
a method that has been confirmed to underestimate benthic exchanges (Glud et al., 2016). The decoupling
between fluxes and concentration gradients (Table 2) indicates that dynamic processes are important in
345 interfacial exchanges and must therefore be involved in the quantification of benthic effluxes.

Ortega et al. (2005) compiled DIC and TA fluxes measured using benthic chambers, reporting ranges
of $5\text{--}700 \text{ mmol m}^{-2} \text{ d}^{-1}$ and $1\text{--}200 \text{ mmol m}^{-2} \text{ d}^{-1}$, respectively. The relatively low fluxes were attributed to
the exclusion of advective transport by chambers. In their studies of three Cantabrian estuaries, despite
in-situ flow rates differing by an order of magnitude, the average of benthic DIC effluxes, 342, 252 and
350 $217 \text{ mmol m}^{-2} \text{ d}^{-1}$, were similar. Although the porewater DIC and TA in Sansha Bay were not particularly
high, the fast currents in the bay ($\sim 1 \text{ m s}^{-1}$; Lin et al., 2017) strengthened the benthic exchange by irrigating
the permeable sediment. Consequently, the DIC effluxes at stations SSW-B and -C were comparable to
the chamber results in Cantabrian estuaries, where organic carbon oxidation was considerable.
Furthermore, the current influence was amplified when the sediment was also subject to intensive benthos
355 disturbance. As a result, the DIC and TA fluxes in the farming zone (SSW-A) were higher by a factor of
3–5 than others.

To illustrate the benthic influence on water columns, we used modified Eq. (6) to estimate the
increased DIC and TA derived from the sediment ($\Delta\text{DIC}_{\text{sw}}$ and $\Delta\text{TA}_{\text{sw}}$). Here the residence time (τ) from
Sandu Bay were adopted owing to the similar hydrodynamic conditions (Table 2). As a result, the benthic
360 release at SSW-A contributed the largest amounts of DIC and TA to the water column ($>150 \mu\text{mol kg}^{-1}$,
Table 2). In the meantime, elevated DIC and TA concentrations were observed in Dongwuyang, where
the average concentrations of DIC and TA were higher by $122 \mu\text{mol kg}^{-1}$ and $64 \mu\text{mol kg}^{-1}$ than those in
the main channel, respectively (Fig. S3 in Appendix). The elevations were consistent with the magnitudes
of $\Delta\text{DIC}_{\text{sw}}$ and $\Delta\text{TA}_{\text{sw}}$, confirming that the sediment served as the dominant source of DIC and TA in the
365 inner bay. In contrast, stations close to channels showed minimal sediment influences on seawater
concentrations ($< 15 \mu\text{mol kg}^{-1}$), indicating that tidal exchange was the dominant factor for the variations
of DIC and TA in the outer bay. This corresponds to the dynamic mechanism regulating the interfacial
exchange at SSW-C illustrated by the $^{224}\text{Ra}\text{--}^{228}\text{Th}$ disequilibria. However, we need to point out that the



estimates here only reflect an average efflux over two weeks around sampling time as the time scale of
 370 the ^{224}Ra - ^{228}Th disequilibrium approach is constrained by the half-life of ^{224}Ra (3.66 days). Given that
 there are great spatial and temporal heterogeneity on sediments in Sansha Bay, these effluxes are not
 suitable for assessments of annual exchange or carbon budget of the whole bay.

**Table 2: The summary of parameters for flux calculation including the molecular diffusion
 375 coefficients of DIC (D_s^{DIC}) and the concentration gradients of DIC and TA at the interface ($\frac{\partial C^m}{\partial z}$), and
 benthic flux of DIC and TA (F_m) at each sampling station. The last three rows represent the
 residence time (τ) and increased concentrations of DIC and TA to the water column ($\Delta\text{DIC}_{\text{sw}}$ and
 $\Delta\text{TA}_{\text{sw}}$) estimated from F_m and τ . Uncertainties for all parameters are reported as $\pm 1\sigma$.**

Parameters	Stations		
	SSW-A	SSW-B	SSW-C
$*D_s^{\text{DIC}}$ ($10^{-6} \text{ cm}^2 \text{ s}^{-1}$)	6.0	5.9	5.1
$\frac{\partial C^{\text{DIC}}}{\partial z}$ ($\mu\text{mol L}^{-1} \text{ cm}^{-1}$)	626.5	1367	1033
$\frac{\partial C^{\text{TA}}}{\partial z}$ ($\mu\text{mol L}^{-1} \text{ cm}^{-1}$)	598.2	963.7	754.8
F_{DIC} ($\text{mmol m}^{-2} \text{ d}^{-1}$)	624 ± 188	207 ± 97.4	143 ± 37.3
F_{TA} ($\text{mmol m}^{-2} \text{ d}^{-1}$)	595 ± 180	146 ± 68.7	105 ± 27.3
$F_{\text{TA}}/F_{\text{DIC}}$	0.95 ± 0.40	0.70 ± 0.47	0.73 ± 0.27
τ (d)	3.73 ± 0.38	0.86 ± 0.09	0.24 ± 0.03
$\Delta\text{DIC}_{\text{sw}}$ ($\mu\text{mol kg}^{-1}$)	163 ± 62.5	12.6 ± 6.7	2.4 ± 0.9
$\Delta\text{TA}_{\text{sw}}$ ($\mu\text{mol kg}^{-1}$)	156 ± 59.6	8.9 ± 4.7	1.8 ± 0.6

* The dominant component of DIC and TA is HCO_3^- so the same diffusion coefficient is used.

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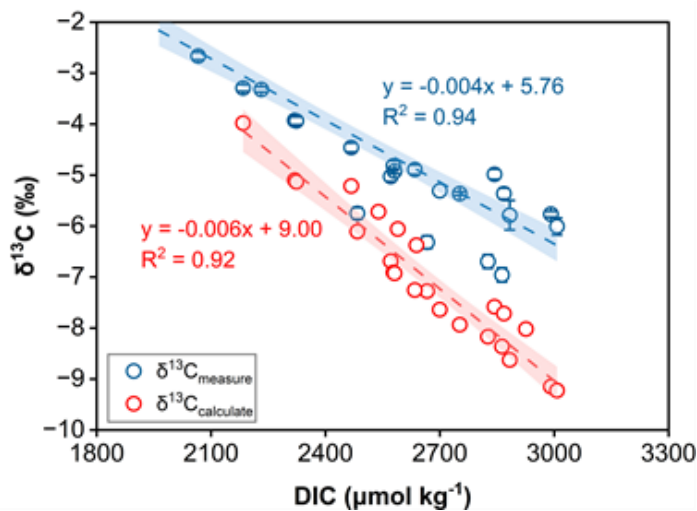
In a semi-enclosed embayment, Luoyuan Bay, located close to our study region, the settling fluxes
 of particulate organic carbon (POC) estimated using sediment traps were reported to range from 92 to 375
 $\text{mmol C m}^{-2} \text{ d}^{-1}$, comparable with those on the inner shelf of the East China Sea (60–608 $\text{mmol C m}^{-2} \text{ d}^{-1}$)
 (Hung et al., 2013; Wang et al., 2018). As the sediment trap was deployed in a shallow aquaculture
 385 zone during a macroalgae blooming, we supposed that Sansha Bay exhibited a similar range of POC flux.
 Correspondingly, the DIC released from sediment were equal or even higher than the deposited POC.
 This indicates, on one hand, that the sediment was characterized with intense microbial activities,
 allowing rapid remineralization of the deposited POC. Furthermore, the higher DIC efflux relative to POC



flux suggests that organic carbon degradation was not the sole process responsible for DIC increment in
 390 the sediment, particularly in the inner bay (SSW-A).

4.3 The contributions of organic carbon degradation and CaCO₃ dissolution to porewater DIC

With the method in Sect. 2.4, conservative DIC addition to porewater relative to the bottom water and porewater $\delta^{13}\text{C}_{\text{DIC}}$ variations were used to quantify the contributions of organic carbon degradation
 395 and CaCO₃ dissolution. The medians of $\delta^{13}\text{C}$ in shells and sediment TOC were selected to represent the characteristics of the processes that added DIC to porewaters, namely $\delta^{13}\text{C}_{\text{CaCO}_3} = -3.77\text{‰}$ and $\delta^{13}\text{C}_{\text{TOC}} = -23.19\text{‰}$ (Fig. 4). In order to prove the necessity of including CaCO₃ dissolution, TOC degradation as a sole DIC addition was modelled with the modified Eq. (5), $\delta^{13}\text{C}_{\text{PW}} = \delta^{13}\text{C}_{\text{BW}} \frac{\text{DIC}_{\text{BW}}}{\text{DIC}_{\text{PW}}} + \delta^{13}\text{C}_{\text{TOC}} \frac{\text{DIC}_{\text{PW}} - \text{DIC}_{\text{BW}}}{\text{DIC}_{\text{PW}}}$. The theoretical porewater $\delta^{13}\text{C}_{\text{DIC}}$ as a result of TOC degradation only would range
 400 from -9.2‰ to -4.0‰ (red plots in Fig. 5, data in Table S3), significantly different from the measured $\delta^{13}\text{C}_{\text{DIC}}$ in the porewater ($p < 0.001$, Linear mixed-effects model; blue plots in Fig. 5). The negative deviation indicates that other source with heavier carbon than TOC, i.e., CaCO₃, collectively influenced the porewater DIC concentrations.



405 **Figure 5: The correlations between $\delta^{13}\text{C}$ and DIC concentrations, where $\delta^{13}\text{C}_{\text{measure}}$ (error = $\pm 1\sigma$) represents the laboratory-measured porewater $\delta^{13}\text{C}_{\text{DIC}}$, and $\delta^{13}\text{C}_{\text{calculate}}$ represents the theoretical $\delta^{13}\text{C}_{\text{DIC}}$ values influenced solely by organic carbon degradation. The shaded areas represent the 95% confidence intervals.**



410 According to the original Eq. (5), the contributions of TOC degradation (f_{TOC}) and CaCO_3 dissolution
(f_{CaCO_3}) were quantified (Table S4 in Appendix). Overall, bottom water dominated the composition of
porewater. The TOC degradation contributed 5.6–20.5% of the porewater DIC and f_{CaCO_3} were in the
range of 1.8–17.5% while the relative contributions between f_{TOC} and f_{CaCO_3} varied between stations
(Fig. 6). It is evident that CaCO_3 dissolution played a greater role than TOC degradation at SSW-A,
415 consistent with the phenomenon of abundant shell debris and the highest TIC content in the sediment
(Table S1 in Appendix). At SSW-B where sea cucumber mariculture existed, f_{TOC} was elevated and the
DIC concentrations of the porewater were also higher than others. This enhancement is likely associated
with the decomposition of organic matters accumulated from local feeding. In contrast, f_{TOC} was
significantly higher than f_{CaCO_3} at SSW-C ($p < 0.005$, t -test) where TIC content was relatively low. It
420 seems that high degradation of organic carbon cannot always lead to a high CaCO_3 dissolution, suggesting
that except for the sediment matrix, the bottom hydrodynamics may influence the relative proportions.
As SSW-C was located near the bay mouth and the water exchange was active, the oxic environment
promoted the organic matter degradation in the surface sediment. But the strong interfacial exchange
limited the CO_2 accumulation, consequently weakening the CaCO_3 dissolution in the sediment. By
425 comparison, the DIC was enriched in the sediment porewater at the inner-bay stations so more CaCO_3
dissolution was triggered. The inference is also supported by the vertical variation of SSW-B: the highest
 f_{CaCO_3} and f_{TOC} were presented in the subsurface sediment where DIC was highest; and meanwhile, the
lowering f_{CaCO_3} with depth showed the opposite effect that CaCO_3 dissolution was weakened when the
porewater DIC was diluted as a result of seawater irrigation.

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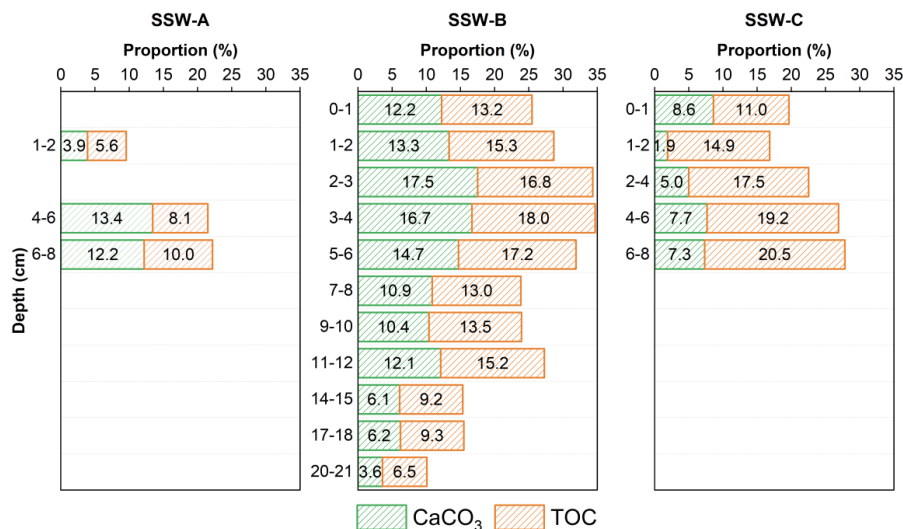


Figure 6: The contribution proportions of organic carbon degradation (f_{TOC} , orange bars) and CaCO_3 dissolution (f_{CaCO_3} , green bars) to porewater DIC in different sediment layers. Values within bars indicate the respective proportion.

The average ratios between f_{CaCO_3} and f_{TOC} ($f_{\text{CaCO}_3}/f_{\text{TOC}}$) of SSW-A, SSW-B and SSW-C were 1.2, 0.8 and 0.4, respectively (Table S4). Hence, the DIC efflux contributed by CaCO_3 dissolution at each station was estimated to be 339 ± 102 , 92.7 ± 43.6 and $40.2 \pm 10.4 \text{ mmol m}^{-2} \text{ d}^{-1}$, respectively ($F_{\text{CaCO}_3} = F_{\text{DIC}} \frac{1}{1 + \frac{f_{\text{TOC}}}{f_{\text{CaCO}_3}}}$), contributing 27–56% of the total DIC efflux. These ratios are in agreement with the results of previous studies that about one third to half of DIC fluxes were derived from CaCO_3 dissolution in benthic chambers (Ortega et al., 2005; Jahnke et al., 1997). The chamber studies well demonstrated that the CO_2 produced by aerobic and anaerobic degradation was vital for CaCO_3 dissolution in sediments and supported the trigger mechanism in this study. Furthermore, we introduced the hydrodynamic conditions in explaining the different extents of CaCO_3 dissolution that were hardly simulated in past research: except for the metabolic CO_2 , the intensity of interfacial exchange determined whether the CO_2 could be accumulated to produce a microenvironment for continuous dissolution. This study provides a new perspective on the reason why the role of metabolic CO_2 in CaCO_3 dissolution varied regionally in previous studies.

4.4 Sediment influence on the carbonate buffering capacity of seawater

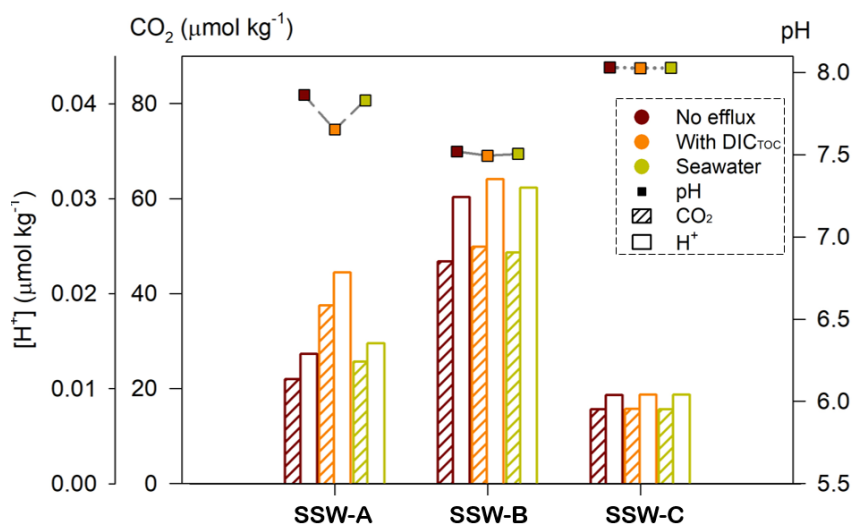
As the sources of porewater DIC have been illustrated in Sect. 4.3, the same ratios $f_{\text{CaCO}_3}/f_{\text{TOC}}$ (1.2, 0.8 and 0.4 for SSW-A/B/C) were applied to reflect the benthic influence on the seawater. Based on the



benthic addition to the water column estimated in Sect. 4.2, the seawater DIC and TA without sediment
455 influence (DIC_0 and TA_0) can be calculated by deducting ΔDIC_{sw} and ΔTA_{sw} from the measured DIC and
TA of seawater (Table 2). Similar with the efflux contributions, the changes of DIC concentrations in
seawater that was attributed to organic matter degradation (ΔDIC_{TOC}) was estimated as 71.5, 7.9 and 1.8
 $\mu\text{mol kg}^{-1}$ for SSW-A, SSW-B and SSW-C, respectively ($\Delta DIC_{TOC} = \Delta DIC_{sw} \frac{1}{1 + \frac{f_{CaCO_3}}{f_{TOC}}}$). If there was only

aerobic mineralization occurring in the sediment, the DIC in seawater would be $DIC_0 + \Delta DIC_{TOC}$ while
460 the change of TA was assumed to be negligible. The DIC and TA concentrations we measured in seawater
are reckoned as the results influenced by both aerobic mineralization and $CaCO_3$ dissolution. Therefore,
the influence of the two processes can be distinguished by pairwise comparison (Table 3).

In order to directly exhibit the change of the buffering capacity in different scenarios, we applied
CO2SYS program (Lewis et al., 1998) to calculate H^+ and CO_2 concentrations from assumed seawater
465 DIC and TA (Table 3) and depicted them in Figure 7. Notably, the absolute concentrations of H^+ and CO_2
are mainly determined by the initial concentrations of DIC and TA, not available as buffering capacity
indicators. Hence, we focused on the concentration changes between these scenarios. At SSW-A, the TOC
degradation increased CO_2 and H^+ , leading to pH being lowered by 0.2; while when additional TA was
involved, CO_2 and H^+ decreased by approximate 30%. Although the variation in different scenarios was
470 not obvious at SSW-B as CO_2 and H^+ concentrations were high, the H^+ induced by TOC degradation was
still slightly lowered by ~2% via adding extra TA. In contrast, the parameters at SSW-C kept consistent
in the scenarios, indicating that the water was rarely influenced by benthic inputs. However, the absolute
amount of TA added is not the primary factor in elevating pH. Although the TA level in seawater groups
is elevated to a similar extent relative to the other two groups, the pH shows opposite response: the pH of
475 seawater group is higher than that in the DIC-TOC-only group but lower than that in the no-efflux group.



480 **Figure 7: The variations of pH (plots), CO₂ (shadowed bars) and H⁺ (filled bars) concentrations at each station in different scenarios which are shown in colors (red: without benthic input; orange: only with DIC derived from organic degradation; yellow: with all of benthic inputs). The axes on the left are for the bars and the right axis is for the plots.**

485 Instead, the ratio of added TA and DIC (i.e., $\Delta\text{TA}/\Delta\text{DIC}$) is likely the determining factor for whether the acidification is mitigated. Thus, $\Delta\text{TA}/\Delta\text{DIC}$ between seawater and other two groups (no-efflux and DIC-TOC-only) are calculated respectively and the results are shown in Table 3. When $\frac{\Delta\text{TA}}{\Delta\text{DIC}} > 1$, pH increases and the extent of pH rise becomes larger with the increasing ratio. Conversely, when $\frac{\Delta\text{TA}}{\Delta\text{DIC}} < 1$, pH exhibits a decreasing trend, and the variation in pH is primarily governed by the amount of DIC added to the system. Given that DIC release from organic carbon degradation is often inevitable in coastal seas, processes leading to $\frac{\Delta\text{TA}}{\Delta\text{DIC}} > 1$ are critical to counteract the negative effects induced by excess DIC.

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Table 3: The concentrations of TA and DIC in seawater influenced by different processes. The pH and CO₂ were calculated from the DIC and TA ahead using the CO2SYS program[#].

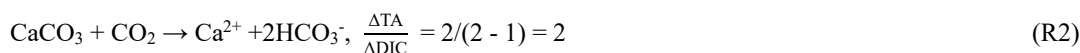
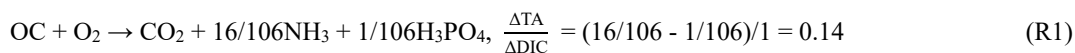
Station	Scenarios	TA ($\mu\text{mol kg}^{-1}$)	DIC ($\mu\text{mol kg}^{-1}$)	pH	CO ₂ ($\mu\text{mol kg}^{-1}$)	$\frac{\Delta\text{TA}}{\Delta\text{DIC}}$
SSW-A	No benthic input	2201	2066	7.86	21.9	0.96 [§]
	DIC-TOC only	2201	2137	7.65	37.5	2.18 [*]
	Seawater	2357	2229	7.83	25.7	—
SSW-B	No benthic input	1973	1951	7.52	46.8	0.70 [§]
	DIC-TOC only	1973	1958	7.49	49.9	1.27 [*]
	Seawater	1982	1964	7.51	48.6	—
SSW-C	No benthic input	2241	2048	8.03	15.6	0.73 [§]
	DIC-TOC only	2241	2050	8.03	15.8	0.99 [*]
	Seawater	2243	2051	8.03	15.7	—

[#] The carbonic acid dissociation constants (K_1 and K_2) are from Lehrter et al. (2012), K_{HSO_4} dissociation constants are from Dickson (1990), and total borate-salinity relationship is from Lee et al. (2010). All pH values are reported on the total hydrogen scale (pH_T).

[§] The ratio is calculated by comparing "No benthic input" and "Seawater" scenarios.

^{*} The ratio is calculated by comparing "DIC-TOC only" and "Seawater" scenarios.

Early diagenetic processes in sediment generally exhibit specific $\frac{\Delta\text{TA}}{\Delta\text{DIC}}$ ratios like R1 and R2 if we assume that the composition of labile organic matter in the sediment follows the Redfield ratio (C:N:P = 106:16:1) (Middelburg et al., 2020). The $\frac{\Delta\text{TA}}{\Delta\text{DIC}}$ derived from aerobic mineralization and CaCO₃ dissolution, according to the stoichiometric amount of the reactions, are 0.14 and 2, respectively.



Based on the contributions to DIC effluxes in Sect. 4.3, the TA release via sediment CaCO₃ dissolution (R2) was consequently approximately 678 ± 205 , 185 ± 87.1 and 80.4 ± 20.9 mmol m⁻² d⁻¹ for SSW-A, SSW-B and SSW-C, respectively. These results are comparable with the benthic TA fluxes estimated from ²²⁴Ra-²²⁸Th disequilibria (see Table 2). Following the $f_{\text{CaCO}_3}/f_{\text{TOC}}$ ratios (1.2, 0.8 and 0.4 for SSW-A/B/C) and the stoichiometry of R1 and R2, the theoretical $F_{\text{TA}}/F_{\text{DIC}}$ ratios can be calculated as 1.16, 0.97 and



515 0.68 for SSW-A/B/C. In fact, the practical ratios for these sites were 0.95, 0.70 and 0.73, respectively (see
Table 2). The lower practical F_{TA}/F_{DIC} ratios indicate that employing the average f_{CaCO_3}/f_{TOC} of the entire
core introduced bias when quantifying process influences on benthic effluxes. Therefore, high-resolution
 f_{CaCO_3}/f_{TOC} ratios of surface sediment are essential in future studies to ensure accurate assessments.

Overall, $CaCO_3$ dissolution is the key process that enhanced TA and promoted the benthic F_{TA}/F_{DIC}
520 above 1. As demonstrated by the scenario comparison, an addition with a high $\frac{\Delta TA}{\Delta DIC}$ ratio can effectively
elevate the pH of water column, indicating that $CaCO_3$ dissolution is a viable approach to enhance the
buffering capacity. In coastal seas where organic matter accumulation in sediments is high, this process
serves as a vital regulator of the carbonate system in bottom water, mitigating the risk of acidification
induced by organic matter degradation. Accordingly, the implementation of a co-culture model with
525 shellfish is necessary in maricultural farms for additional $CaCO_3$ supply.

5 Conclusions

This study chose a coastal bay as a high-productivity representative and quantified the benthic
processes affecting the carbonate system. Using the ^{224}Ra – ^{228}Th disequilibrium approach, the highest
fluxes of DIC and TA were found in the regions of seaweed farms, demonstrating that the benthic input
530 accounted for the DIC and TA hotspots observed in water columns. Intensive bioturbation in the sediment
and the hydrodynamics were confirmed to regulate the exchange across the sediment-water interface in
the bay. Based on the $\delta^{13}C_{DIC}$ signatures of different sources, the roles of organic matter degradation and
 $CaCO_3$ dissolution in elevating porewater DIC concentrations were quantified. The results illustrated that
 $CaCO_3$ dissolution contributed 27–56% of the increased DIC in porewater and dominated the TA release
535 from the sediment, balancing the H^+ increment induced by aerobic mineralization.

This research sheds light on the self-regulating mechanism of the carbonate system in a high-
productivity coastal zone, and for the first time, illustrates the dynamic impact on sediment $CaCO_3$
dissolution: intensive disturbance affects the accumulation of metabolic CO_2 from organic carbon
degradation, thereby influencing the extent of $CaCO_3$ dissolution. The benthic TA input from the sediment
540 $CaCO_3$ dissolution is validated as a key role in maintaining the buffering capacity of seawater, lowering
the potential acidification associated with the organic carbon remineralization. As biogenic $CaCO_3$
commonly exist in sediments, the sediment $CaCO_3$ dissolution was likely a ubiquitous process in the
ocean, even in $CaCO_3$ supersaturated surroundings. This provides an insight into the carbon cycling of
coastal and marginal seas that sediment $CaCO_3$ dissolution needs to be included in assessing the budget
545 of inorganic carbon.



Appendix

Supporting information in appendix includes Tables S1 to S4 and Figures S1 to S3.

550 **Code and data Availability**

The sediment and porewater data, the R codes for statistical testing and analysis results are available through Mendeley Data, V2, doi: 10.17632/wd5xyszzjv.2.

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565 Writing - Reviewing and Editing, Funding acquisition. Yingxu Wu: Writing - Reviewing and Editing. Xiangqi Yi: Statistical Analysis, Writing - Reviewing and Editing. Biqi Zheng: Writing - Reviewing and Editing. Di Qi: Project administration, Writing - Reviewing and Editing, Supervision.

Conflict of Interest

- 570 The authors declare there are no conflicts of interest for this manuscript.

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