

General comments

This study pairs two tracers (^{224}Ra and ^{228}Th disequilibrium for benthic exchange; $\delta^{13}\text{C}$ -DIC for the source of DIC) at three sites in Sansha Bay, and reports that CaCO_3 dissolution supplies a quarter to half of the benthic DIC efflux. They show that dissolution does not simply track the intensity of organic carbon degradation, and that the associated alkalinity buffers acidification caused by remineralisation. I think it is a well written and important paper. The tracer combination is a good idea and, as far as I know, has not been applied in this setting. The radium work is careful, the arithmetic is internally consistent, data and code are archived. My comments are about strengthening the paper rather than questioning the approach. The main gap is that the saturation state is never calculated, even though every ingredient is there. Beyond that, the flux magnitudes deserve a clearer benchmark, and the headline number needs an uncertainty.

We appreciate the suggestions. The proportion results were updated after the Monte Carlo simulations and the uncertainties were added when discussing the results. In this response, we reply the comments one by one. The text added to the revised MS are shown in italic.

The subject of the paper is CaCO_3 dissolution, but Ω_{calcite} and $\Omega_{\text{aragonite}}$ appear nowhere. You have porewater TA and DIC at 1 cm resolution, so this is a short calculation. It matters, because the reported porewater endpoints do not obviously permit the dissolution you infer. Running them through PyCO2SYS ($T = 18^\circ\text{C}$, $S = 30$), TA 2326 / DIC 2184 gives $\Omega_{\text{calcite}} = 2.9$ and TA 3081 / DIC 3007 gives 2.4. Both supersaturated. Only layers where DIC exceeds TA, as at SSW-B above 15 cm, fall below $\Omega_{\text{calcite}} = 1$. This should be discussed and interpreted.

We appreciate the comments. We added the CaCO_3 saturation in seawater in 4.3 to show the bottom conditions (lines 419-423). In 4.4, benthic influence on seawater Ω_{calcite} in different scenarios was discussed in the revised MS (lines 510-513). The value of Ω_{calcite} was shown in Table 4.

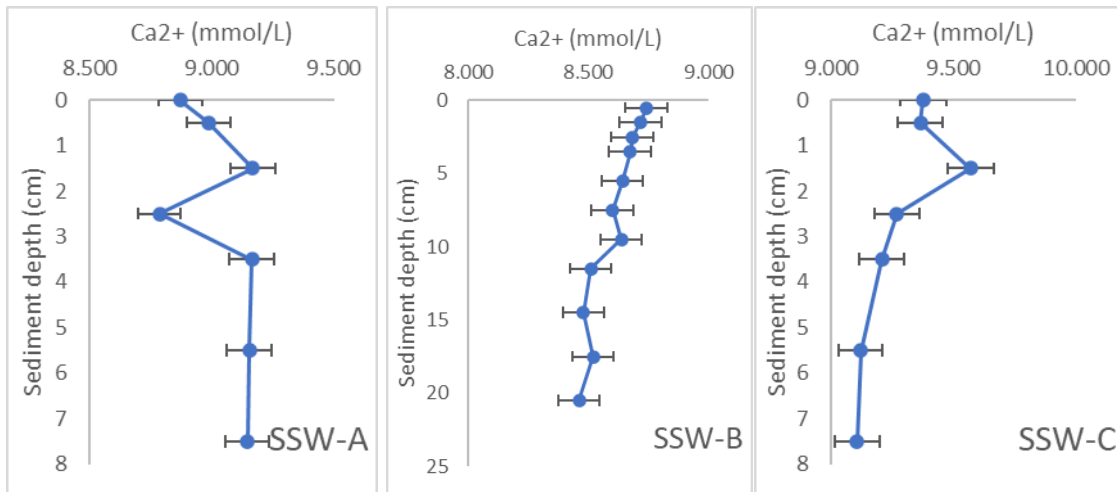
Although the concentrations of DIC and TA have indicated that the bottom water of Sansha Bay during sampling period presents calcite oversaturated ($\Omega_{\text{calcite}} = 1.15 \sim 3.50$ based on CO2SYS), CaCO_3 dissolution still likely occurs, as microbial remineralization and the oxidation of reduced sulfur species may modify porewater carbonate chemistry and generate localized acidic microenvironments (Ku et al., 1999; Hu and Burdige, 2007; Wang et al., 2025).

Your microenvironment argument may well resolve this (Wang et al., 2025, L&O Lett. do show shell dissolution at $\Omega > 1$). Another reading would be inspired by Hu and Burdige (2007), who proposed coupled dissolution and reprecipitation for exactly this

isotopic signature, which produces heavy porewater DIC with little net carbonate loss. A porewater Ca profile, if possible, would be enlightening and settle the question directly.

In previous studies, CO2SYS was generally used to calculate the carbonate parameters in seawater and hasn't been tried in porewater. The reactions between solid and liquid phases in sediments are complicated. We are not sure if the calculator for the CO₂ systems is available in porewater so we stick it to the carbonate system in seawater. Taking the station SSW-A for instance, Ω_{calcite} of 0-2 cm porewater was over 1.5 while the value of 2-6 cm porewater fell down to ~0.4; but at the bottom layer (6-8 cm), Ω_{calcite} was the highest, 4.5, even higher than the seawater (2.7). On one hand, it is hard to prove that the calculated Ω_{calcite} of porewater with CO2SYS are correct. On the other hand, Ω only presents the situation of seawater carbonate system, and $\Omega > 1$ doesn't mean that there is no CaCO₃ dissolution, and vice versa. Microbial remineralization and the oxidation of reduced sulfur species can modify porewater carbonate composition and generate localized acidic conditions. Such microenvironments can promote CaCO₃ dissolution at shell surfaces, even when $\Omega > 1$ (Ku et al., 1999; Wang et al., 2025).

We measured Ca²⁺ concentrations in porewater using ICP-OES. The variation in porewater Ca²⁺ concentrations at each site (8.5-9.0 mmol/kg) was below 5% and didn't show an increase with depth. In sediments, Ca²⁺ concentrations are affected by multiple processes other than CaCO₃ dissolution, including precipitation, authigenic mineral and even benthos growth. The profiles reflect all benthic processes acting on the porewater Ca²⁺ concentrations, not a specific one.



The Ca²⁺ concentrations in porewaters of SSW-A, SSW-B and SSW-C

You report $F_{\text{DIC}} = 624 \text{ mmol m}^{-2} \text{ d}^{-1}$ at SSW-A, that is $228 \text{ mol C m}^{-2} \text{ yr}^{-1}$. That is well above independent measurements: the global coastal benthic DIC efflux of Krumins et al. (2013), 126 Tmol yr^{-1} , corresponds to roughly $13 \text{ mmol m}^{-2} \text{ d}^{-1}$, and the highest in-situ chamber values I can find for organic-rich systems are 18 to $78 \text{ mmol m}^{-2} \text{ d}^{-1}$

(Rassmann et al., 2020) or around 100 to 120 (Fan et al., 2024). That said, the reported values are consistent with the ^{224}Ra literature, the method paper you follow, Cai et al. (2015), reports Pearl River Estuary DIC fluxes spanning two orders of magnitude, from -38 to 8500 (plus or minus 16000!) $\text{mmol m}^{-2} \text{d}^{-1}$, and Hong et al. (2017) give 26 to 1000 $\text{mmol m}^{-2} \text{d}^{-1}$ for the Jiulong River estuary. I would make this comparison explicitly and discuss it, including the fact that the largest ^{224}Ra -derived fluxes carry very large uncertainties.

In the revised MS, we clarified the approach when comparing the DIC fluxes and pointed out the uncertainty with ^{224}Ra - ^{228}Th disequilibrium (lines 353-354, 358-359). The comparison with benthic chamber method was also discussed (lines 365-368). According to Ortega et al. (2005), the DIC and TA fluxes were also in large ranges but mostly less than these from ^{224}Ra - ^{228}Th disequilibrium. Given the different conditions, we didn't discuss the flux values directly. In order to show the robustness of the estimate in our study, we calculated the DIC increment in water column and the results matched well with the measured DIC concentration. The limitation of ^{224}Ra - ^{228}Th disequilibrium was also mentioned (lines 379-395).

The DIC and TA fluxes estimated from the ^{224}Ra - ^{228}Th disequilibria are generally higher than those from in-situ chamber experiments. Ortega et al. (2005) compiled DIC and TA fluxes using benthic chambers in the ranges of 5–700 $\text{mmol m}^{-2} \text{d}^{-1}$ and 1–200 $\text{mmol m}^{-2} \text{d}^{-1}$, respectively, but most were less than 100 $\text{mmol m}^{-2} \text{d}^{-1}$

In addition, the spatial variations of DIC and TA concentrations in water column can also illustrate the robustness of the flux estimate. The increased DIC and TA derived from the benthic efflux ($\Delta\text{DIC}_{\text{sw}}$ and $\Delta\text{TA}_{\text{sw}}$) can be calculated with the modified Eq. (6). ...

No uncertainty is propagated onto f_{TOC} , f_{CaCO_3} or the headline range. A Monte Carlo over the endmember distributions (shells $n = 9$, TOC $n = 27$) is straightforward and would strengthen the paper. For instance, how sensitive are the results to the $\delta^{13}\text{C}$ signatures and their uncertainties? The measured TA flux gives an independent estimate. Inverting $F_{\text{TA}}/F_{\text{DIC}} = (2r + 0.14)/(r + 1)$ with your 0.95, 0.70, 0.73 gives $r = 0.77$, 0.43, 0.46, i.e. a CaCO_3 share of 44/30/32%. Presenting both sets of estimates would provide a better uncertainty envelope.

We appreciate the suggestion. We have used Monte Carlo simulations to estimate all proportions. The relevant results with uncertainties have been updated in the MS, including Fig. 7 and Appendix Table S4. The approach was shown in Sect. 2.4 and Sect. 4.3 (lines 236-243; lines 436-443). Uncertainties on proportions and fluxes has been added in abstract and tables (line 23; Table 3).

Considering the variabilities of $\delta^{13}\text{C}_{\text{CaCO}_3}$ and $\delta^{13}\text{C}_{\text{TOC}}$ in natural samples, a Monte Carlo simulation, performed with the software Oracle Crystal Ball (Release 11.1.2.4; Oracle Corporation), was used for the estimate of source proportions. In each

simulation, $\delta^{13}C_{CaCO_3}$ and $\delta^{13}C_{TOC}$ were independently sampled from their respective distributions, and then were substituted into the equations (4) and (5) to calculate f_{CaCO_3} and f_{TOC} . For each layer, this procedure was repeated 10000 times. The final results are reported with the average of the simulations, and the standard deviation (SD) represents the uncertainty.

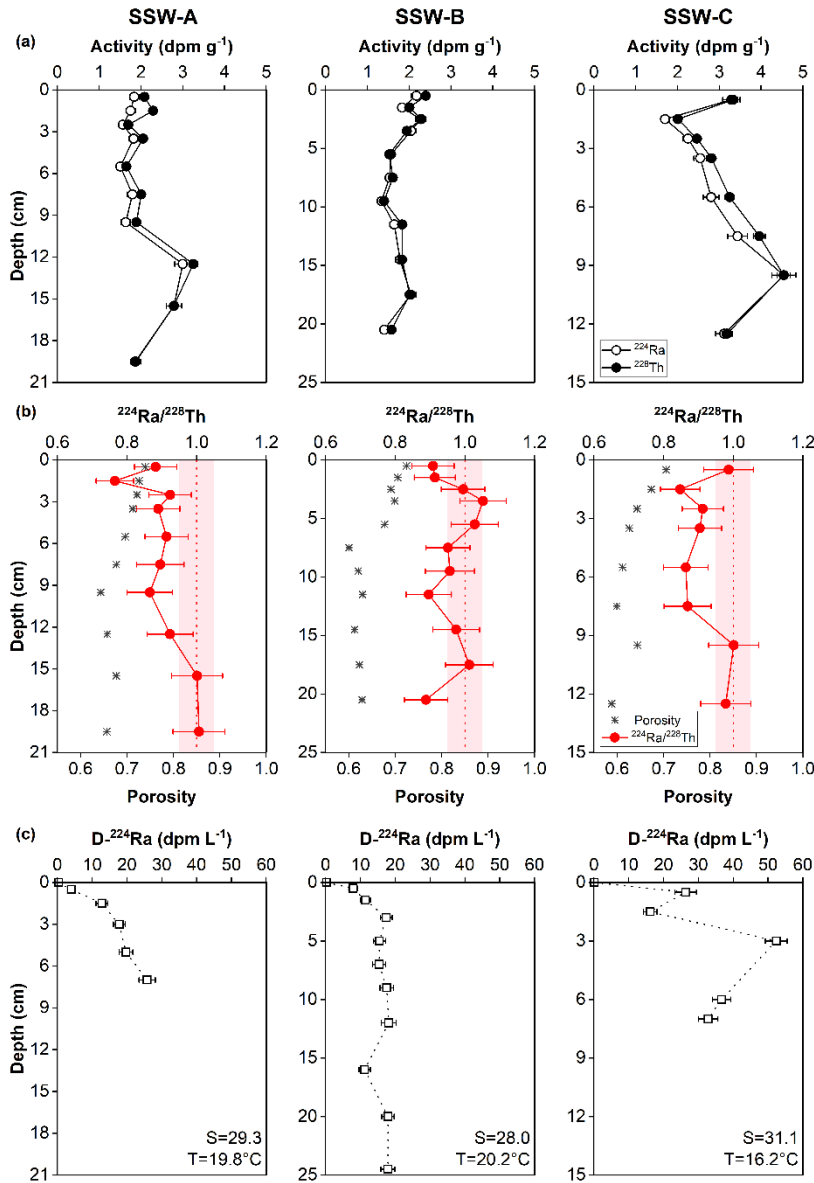
The contributions of TOC degradation (f_{TOC}) and $CaCO_3$ dissolution (f_{CaCO_3}) were quantified based on $\delta^{13}C_{CaCO_3}$ and $\delta^{13}C_{TOC}$ values derived from Monte Carlo simulations (see Sect. 2.4). The $\delta^{13}C_{CaCO_3}$ values were sampled in a range of -4.54 ~ -3.41‰, which was defined based on a uniform distribution of the shell-derived $CaCO_3$ ($n = 9$); and for $\delta^{13}C_{TOC}$, a triangular distribution was identified as the best-fitting model based on the measured values ($n = 27$) so the simulations were parameterized from -25.06‰ to -22.15‰ with the likeliest value of -22.86‰. With 10000-time repetition, the uncertainties of the proportions were under 0.7%. The f_{CaCO_3} and f_{TOC} in each sediment layer are listed in Table S4 (in Appendix) and are depicted in Fig. 7.

If the measured TA flux was used to estimate the proportions, i.e. $r = f_{CaCO_3} / f_{TOC}$, the assumption is that only aerobic mineralization and $CaCO_3$ dissolution produced TA. Although we used this assumption when quantifying the TA that sediment $CaCO_3$ dissolution could supply, the issues and the uncertainties still exist and have been addressed in the revised MS (lines 545-561) including lack of anaerobic pathways. Accordingly, the estimate based on $F_{TA}/F_{DIC} = (2r + 0.14)/(r + 1)$ will amplify the uncertainties. The Monte Carlo analysis can well indicate the uncertainty of estimate so we still keep the $\delta^{13}C$ signatures in the revised MS.

Minor comments

- Missing metadata: sampling month or season is never given, and temperature, salinity and porosity are never reported. All three are needed to reproduce the calculations.

We originally showed the metadata in Table S1 in Appendix, including salinity, temperature and porosities. In the revised MS, we added the information and porosity profiles in Fig. 3 but still kept the original data in Appendix.



- Anaerobic pathways are never mentioned. In organic-rich mariculture mud this needs a sentence, since sulfate reduction with sulfide burial gives $\Delta TA/\Delta DIC$ of about 1 rather than the 0.14 you assume.

Inspired by the comments, we reorganized the framework of Sect. 4.4. In previous section, the proportions were obtained from the $\delta^{13}C$ signatures. When we discussed the TA contributions from $CaCO_3$ dissolution, only f_{CaCO_3}/f_{TOC} ratios and the stoichiometry of R2 were used. One of the highlights of this study is to show the role of sediment $CaCO_3$ dissolution in regulating the carbonate system. Hence, we deleted the part discussing the TA contribution by TOC degradation in the revised MS, emphasizing the TA contributed by $CaCO_3$. The aerobic mineralization and its $\Delta TA/\Delta DIC$ ratio (0.14)

were used to show the assumption that TA from TOC degradation was negligible when setting the three scenarios (lines 500-508).

In the meantime, we addressed the limitation when quantifying the CaCO_3 -derived TA. The $\Delta\text{TA}/\Delta\text{DIC}$ for TOC degradation could be one of the reasons leading to the inconsistency of theoretical and practical $F_{\text{TA}}/F_{\text{DIC}}$. The issue was come up with in the revised MS (lines 557-561).

In addition, only aerobic mineralization (R1) was considered in organic carbon degradation, while anaerobic mineralization including denitrification or reduction of Fe and Mn oxides can also change the concentrations of TA and DIC in sediments, even providing a higher $\Delta\text{TA}/\Delta\text{DIC}$ than R1 (Middelburg et al., 2020). This may also lead to the inconsistency of theoretical and practical $F_{\text{TA}}/F_{\text{DIC}}$.

- CO2SYS constants: Lehrter et al. (2012) is a benthic flux study, not a source of K_1 and K_2 . Please name the parameterisation actually used. The canonical program citation is Lewis and Wallace (1998).

Corrected. We used the constants (K_1 and K_2) are from Millero et al. (2006).

Millero, F.J., T.B. Graham, F. Huang, H. Bustos-Serrano, and D. Pierrot: Dissociation constants of carbonic acid in seawater as a function of salinity and and temperature. Marine Chemistry 100 (2006) 80-94, <https://doi.org/10.1016/j.marchem.2005.12.001>, 2006.

- "Liner mixed-effects" (line 400): change to "linear".

Corrected.

- "rarely influenced by benthic inputs" (line 472): change to "little influenced".

Corrected.

- "sediments ... are in great heterogeneity" (line 96): change to "are highly heterogeneous".

Corrected.