

AC1

General Comments:

This paper is both well-written and timely and warrants publication with some minor revisions. I think the paper could benefit from further exploration of the rate of exchange between the OLW and pore water in terms of driving dissolution and the resultant enhanced alkalinity fluxes in their incubations. The authors address the importance of exchange rate and pore water residence time, so it would be interesting to see an estimate of their exchange rate, how it relates to measured exchange rates along the continental shelf, and how pore water chemistry evolves with depth in the sediment according to pore water residence time.

Additionally, when discussing optimal locations for shell amendment, some discussion of natural sediment deposition and burial rate could be useful. When these shells are buried below the oxygenated zone, they could result in enhanced CaCO_3 precipitation rather than dissolution. However, overall I think this is a very good and interesting paper. See some specific comments below.

We thank the reviewer for their positive feedback and appreciate the valuable suggestions.

Unfortunately, we were unable to determine robust exchange rates in our experiment because the available uranine tracer measurements were insufficient for reliable quantification (see also below). Consequently, we are unable to discuss the contribution of pore-water exchange to dissolution and alkalinity fluxes in our incubations (or compare these rates with those reported for continental shelf sediments).

We agree that the residence time of amended shell material within the reactive surface sediment layer is an important consideration for determining the effectiveness of shell amendment. Section 4.3 has now been expanded to provide a more quantitative assessment of the timescales over which amended shell material may remain exposed to conditions favorable for dissolution.

Specific Comments:

L24: Looking at Figure 5b, is it correct to say there is a new steady-state dissolution rate in the treatments? Seems like it's still decreasing.

We thank the reviewer for this observation. Although the mean dissolution rates in week 24 ($2.2 \pm 1.1 \text{ mmol m}^{-2} \text{ d}^{-1}$) and week 21 ($7.0 \pm 3.3 \text{ mmol m}^{-2} \text{ d}^{-1}$) were comparable within their associated uncertainties, we agree that the data do not support describing the dissolution rate at the end of the experiment as a new quasi-steady state, as it was still decreasing at the end of the experiment. We have therefore revised the text to remove the reference to a quasi-steady state. Instead, we now describe the dissolution rates in the treatment chambers as gradually approaching those observed in the control chambers, while remaining slightly elevated at the end of the experiment.

L78- The placement of “(i.e. the reverse of Eq. 1)” is a little confusing here. Should be moved to the end of the sentence, otherwise it sounds like calcification is the reverse of Eq. 1.

We thank the reviewer for pointing this out. We agree that the original phrasing was ambiguous. We have revised the text by replacing “(i.e. the reverse of Eq. 1)” with “(Eq. 1)”, which directly refers to the calcification reaction and avoids confusion.

L108-109: Was the sediment sieved to remove macrofauna?

The sediment was not sieved to remove macrofauna due to the large volume of sediment required for the experiment. Instead, the sediment was carefully inspected during homogenization for the presence of macrofauna. No visible macrofauna were observed during homogenization, throughout the incubation, or during sediment slicing for porewater sampling at the end of the experiment. We added a line in the manuscript to clarify this.

L145-146: Were you able to determine an OLW-pore water exchange rate?

Uranine tracer was included in the experimental design to quantify pore-water exchange; however, the design of the incubation chamber lid did not allow for continuous monitoring of tracer concentrations. Instead, only discrete measurements could be obtained by removing the lid during incubation, which proved insufficient to derive reliable exchange rates.

L165: How long were AT samples stored unpoisoned before analysis?

Both A_T and DIC samples were analyzed within the same week or, at most, the following week after collection (we now mention this in the Methods text). Samples were intentionally analyzed within this timeframe to ensure accurate monitoring of fluxes and to minimize potential changes in sample composition during storage.

L248: Should the MCDE be $100 \times 2 \times f_{carb}$? Looks like it is reported as % in results rather than a fraction.

We have corrected the equation by including the factor of 100.

Table 1- Why don't you report isotopic values for the organic and inorganic carbon from the mussel shells? If not measured, can you calculate values based on the 8% amendment and how much the isotopic composition of the top 2cm changed following amendment? Also, why is there no error on the mussel shell solid analysis results?

We thank the reviewer for pointing this out. We have added the isotopic values of POC and PIC from the mussel shells to Table 1 and have included uncertainties for all mussel shell solid-phase analyses.

We would like to clarify that, for the Keeling plot analysis, the POC and PIC endmembers were derived from the bulk sediment layer following shell amendment and therefore represent a mixture of natural sediment and added shell material. Because shell-derived and sediment-derived carbonate have similar $\delta^{13}C$ values, $\delta^{13}C$ alone cannot be used to separate their individual contributions to DIC. Therefore, we used the bulk sediment values as the most appropriate endmembers for the Keeling analysis. For carbonate dissolution, we consider this approach appropriate because both natural and shell-derived $CaCO_3$ dissolve in response to the same surrounding porewater saturation state. Although differences in mineral surface properties and mineralogies may influence dissolution kinetics, these cannot be separated using $\delta^{13}C$ measurements. Furthermore, the added shell material was crushed to a particle size comparable to the surrounding sediment, minimizing particle-size-related differences in surface area.

Figure 3: Why are you only showing results from one set of control and treatment incubations rather than all three of each? Can you show a plot of how OLW Ω changed in the control and treatment incubations? Also, the starting $\delta^{13}C$ of the OLW seems pretty light. Do you know why that is the case?

We thank the reviewer for this suggestion. In light of comments from all three reviewers, we have decided to remove Figure 3 from the revised manuscript. Although the original intention was to provide an example of the individual measurements during a single incubation session, we agree that the figure generated more confusion than additional insight. Instead, we have chosen to focus on the calculated benthic fluxes throughout the manuscript, as these represent the primary results of the study and provide a more comprehensive overview of the experimental findings.

The relatively low initial $\delta^{13}C$ -DIC value reflects the coastal seawater used for the incubations, which was collected from the Eastern Scheldt. Compared to open-ocean waters, coastal systems can exhibit lower $\delta^{13}C$ -DIC values due to enhanced organic matter remineralization and the accumulation of ^{13}C -depleted DIC. Since Figure 3 has been removed, these initial $\delta^{13}C$ -DIC values are no longer presented in the revised manuscript.

L325: Is there any correlation between the depth of the grey/black transition depth in the sediment and the $CaCO_3$ dissolution rate/alkalinity flux?

The development of the grey/black transition was tracked qualitatively through photographs, but we did not quantify the depth of this transition and therefore we cannot quantitatively assess its correlation with dissolution rates or alkalinity fluxes.

Nevertheless, we agree that the depth of the black layer provides useful indications on the redox conditions within the sediment. A shallower black layer indicates stronger reducing conditions and likely reflects enhanced iron sulfide formation. Under such conditions, aerobic organic matter mineralization and the associated carbonate dissolution are expected to decrease. However, anaerobic processes such as sulfate reduction and iron sulfide formation can contribute to alkalinity production independently of carbonate dissolution. This is consistent with the observed offset between total alkalinity fluxes and dissolution-derived alkalinity fluxes (Fig. 7b), suggesting that additional alkalinity-generating processes contributed to the measured alkalinity fluxes.

L401-405: Was there any attempt to determine pore water residence time with depth in the sediment column? Especially over the oxygenated zone?

As described in our response to the general comments, we were unable to determine a robust overlying water–porewater exchange rate because the available uranine tracer measurements were insufficient for reliable quantification.

L410-411: Could be helpful to readers if you show the reaction stoichiometry.

We thank the reviewer for this suggestion. We have included the relevant reactions to the discussion.

L492-495: The deposition of fresh organic matter is clearly important, however some note about burial would be useful as natural sediment deposition will eventually bury the added shells below the oxygenated zone and where precipitation of CaCO_3 would be more likely.

We thank the reviewer for this suggestion. As described in our response to the general comments, we have added a discussion of this aspect in the revised manuscript.

We now consider sediment accumulation and bioturbation as important controls on shell residence time within reactive surface sediments. Sediment accumulation rates on continental shelves commonly range from approximately 0.1 to 1 cm yr^{-1} (Restrepo et al., 2020). Assuming an oxygen penetration depth of 2 cm in permeable sediments, corresponding to the thickness of the amended layer in our experiment, this corresponds to estimated residence times of approximately 2–20 years before burial below the oxygenated zone. However, bioturbation can substantially increase the depth of oxygen penetration through bio-irrigation, and extend the residence time of particulate material within the oxygenated zone. Assuming an average bioturbated layer depth of 10 cm (Boudreau, 1998), residence times may increase to approximately 10–100 years (based on Kuderer & Middelburg, 2024). Given that the estimated time required for complete shell dissolution in our experiment was approximately 38 years, these estimates suggest that a substantial fraction of amended shell material may dissolve before permanent burial, particularly in bioturbated sediments. Furthermore, continuous supply of organic matter from the water column in natural scenarios would likely decrease the dissolution timescale by sustaining metabolic dissolution. This further suggests that most shell material may dissolve before burial, even in sediments with limited bioturbation.

Technical:

L23- Try to consistently use weeks or months rather than switch between the two.

We have revised the manuscript to ensure consistent use of the term “weeks” throughout the text.

L71- Consistently use CaCO_3 or calcium carbonate.

We have revised the manuscript to ensure consistent use of the term “ CaCO_3 ” throughout the text.

L204- Redundant to say flux calculations were performed using the software that allows for the calculation of sediment-water fluxes

We have revised the text to remove this redundant statement.

L393- “as such” seems out of place

We have removed these words from the revised manuscript.

Figure 1- somewhat difficult to see the green arrows on the blue background.

We have changed the color of the green arrows to black and modified them to a dashed line style in the revised manuscript to improve visibility.