AC2

General comments:

Overall, I want to express my respect for the huge amount of work that went into this study. Such experiments are very work-intense and the authors backed this up with thorough analytical efforts. Hence, I am convinced that the study and related data definitely should be published.

The result and methods section are in overall good shape and only need minor editing (see line-by-line comments).

The discussion is in principle ok, but needs some rephrasing in section 4.2

Section 4.3 is also ok, but lacks rigor. The authors start nicely in laying out the rates they calculated, how the fraction of compensated emissions can be estimated and where an application is possible. But then they don't present an actual number. A quick statement about 6.1 % in 24 weeks, but no explanation as to what that implies. It would be very interesting to get a better idea over what time which amount of mussel shell could dissolve given typical residence times in the bioturbated mixed layer of sediments.

Likewise, I miss the comparison with other studies that investigated CaCO3 dissolution in sediments. Are the rates per surface comparable? Is the efficiency higher or lower? Also, a quick calculation of the emissions created by grinding the mussels would be helpful.

The speculations about possible application areas is comparably long and somewhat detached from the rest of the study as the implications for an application and the global potential are not really clarified.

Last, I miss a better estimate of the limitations of this study. What effect would resuspension have? How often would a sediment of this grainsize be resuspended in a natural environment? Were there any bioturbators in the incubated sediment?

Maybe this could be covered in a section together with the side-effects that are discussed in section 4.3. This would lead to a more comprehensive structure like: What have we found (also in comparison to previous findings), how do we have to interpret these new findings and what are the implications.

Again, I want to highlight the scientific quality of this study and encourage the authors to elevate the (already not bad at all) discussion to a level that this study deserves.

We thank the reviewer for their positive feedback and appreciate the valuable suggestions. We have revised the manuscript following the line-by-line comments and have substantially revised section 4.2 to better distinguish between observations and interpretations.

Section 4.3 has been expanded to provide a more quantitative assessment of the timescales over which amended shell material may remain exposed to conditions favorable for dissolution. We refer the reviewer to our response to the comment regarding lines 492-495 from the first reviewer. We have also added a discussion on sediment resuspension as an important limitation when extrapolating laboratory results to natural environments. The frequency and intensity of resuspension events depend strongly on local hydrodynamic conditions. Resuspension may have contrasting effects on shell dissolution by enhancing sediment-water exchange and reoxygenation, while potentially also transporting shell fragments away from the application site or exposing them to oversaturated water.

We considered including a quantitative comparison with previously reported $CaCO_3$ dissolution rates. However, direct comparison is challenging due to substantial differences in carbonate material (e.g., biogenic shells versus sedimentary carbonate minerals or pure calcite/aragonite phases), as well as environmental conditions such as sediment type, carbonate saturation state, and organic matter availability. To our knowledge, no previous studies have reported dissolution timescales for biogenic shell material under comparable conditions, precluding a direct comparison with our estimated timescale for complete dissolution. The most relevant comparison would be with studies investigating biogenic $CaCO_3$ amendments in natural permeable sediments, but such studies are currently unavailable.

We agree with the reviewer that an assessment of the emissions associated with shell processing would be valuable. However, we consider this to be beyond the scope of the current study, which focuses on quantifying the dissolution potential and controlling processes of reintroduced mussel shells in marine sediments. A comprehensive life-cycle

assessment would need to consider multiple additional components, including shell collection, cleaning, drying, grinding, transportation, and potential changes in waste management pathways. Since these factors are highly system- and location-dependent, a simplified estimate based only on grinding emissions could provide a misleading estimate of the overall environmental impact. We have therefore clarified in the revised Summary and Outlook section that our CO_2 compensation estimate represents a first-order assessment of the circularity potential of shell reuse, while a complete life-cycle assessment is required in future work.

Finally, while we did not add a separate subsection dedicated to study limitations, we have incorporated a more comprehensive discussion of potential limitations and environmental controls throughout the revised Section 4.3. Specifically, we now discuss sediment burial, bioturbation, resuspension, and hydrodynamic conditions, and their potential influence on the persistence and effectiveness of enhanced mussel shell dissolution under natural conditions.

Specific Comments:

Line 42: Add Fuhr et al. 2025.

We thank the reviewer for the suggestion. We have added the suggested reference to the revised manuscript.

Line 44: Here You should include all studies that have looked into olivine as a potential feed stock for OAE.

We thank the reviewer for the suggestion. We have added additional references that investigated olivine addition to sediments as a potential feedstock for OAE. As this section specifically discusses mineral addition to sediments, we focused on studies addressing sediment-based olivine applications rather than studies investigating olivine dissolution and weathering in the water column.

Line 55: Not sure, figure 1a is really needed. This correlation has been shown in many publications. I would rather try and improve Figure 1b in a way that it clearly encompasses the information contained in Fig. 1a.

We thank the reviewer for this suggestion. While we agree that the relationship illustrated in Figure 1a has been presented in previous studies, we have chosen to retain the figure because it provides important background for a broad readership. In particular, there is still considerable discussion regarding whether bivalve aquaculture acts as a net CO_2 sink or source (Pernet et al., 2025), and Figure 1a provides the context necessary to introduce this topic before presenting the waste stream proposed in Figure 1b.

Line 74: The formula appears a bit lost here.

We thank the reviewer for this comment. We agree that the motivation for including the incineration pathway was not sufficiently clear. We have revised this section to clarify that the purpose is to highlight existing shell waste management pathways and their potential implications for CO_2 emissions. We have also improved the transition between incineration, ash disposal, and potential subsequent carbonation, and have reformatted the reaction equation to improve readability.

Line 75: The entire explanations around incineration are a bit difficult to follow. Why is this done anyways, and why is the resulting product buried?

As described in our response to the previous comment, we have revised this section to clarify the relevant processes.

Line 86: Here you should cite work from Aller et al. 1980 and 1981 where they investigated these processes at FOAM.

We thank the reviewer for this suggestion. We have added a citation to Aller (1982), which provides a comprehensive description of this process.

Line 94/95: This sentence is repeating what you stated in line 78.

We thank the reviewer for pointing this out. We agree that the final sentence repeated part of the motivation already introduced earlier in the introduction. We have therefore revised the final paragraph to focus more specifically on the objective of the present study and the selected target species, while retaining the initial explanation of the overall concept of shell reintroduction.

Line 149/150: I like the detail with which the authors describe their experimental procedure. At the same time, I believe the SOP for calibrating an oxygen sensor should be clear.

We thank the reviewer for this comment. We agree that the two-point calibration procedure is a standard procedure for oxygen optodes and that readers familiar with oxygen measurements will be aware of this approach. However, we have retained this information in the revised manuscript because we consider these details useful for ensuring transparency and reproducibility of the oxygen measurements.

Line 163: Was the removed volume replaced?

We thank the reviewer for pointing this out. We have clarified in section 2.5 that solute concentrations were corrected for dilution resulting from sample volume replacement in both closed and open incubations.

Lines 225 ff.: Please provide units for all parameters that are used in these equations. Is the final RDiss reported per m2 of surface area of added material or per m2 of sediment? If the latter please report the values also relative to the reactive surface of the added mineral for comparability.

We thank the reviewer for this valuable suggestion. We have now included the units for all parameters when they are first introduced in the equations and have clarified in the Methods section that all dissolution rates (R_{diss}) are normalized per sediment surface area.

We also considered normalizing R_{diss} to the BET surface area of the added shell material. However, we ultimately decided not to include these values because the calculated dissolution rates represent the total $CaCO_3$ dissolution within the sediment, including both the added shell material and the naturally occurring sedimentary carbonate. While the BET surface area of the added mussel shells is available, the reactive surface area of the natural sedimentary carbonate is unknown. Consequently, normalizing the total R_{diss} to the BET surface area of the added shells would be incorrect.

We could alternatively estimate shell-specific dissolution by subtracting the control R_{diss} from those of the shell-amended treatments. However, this approach is also not appropriate because the added shell-associated organic matter stimulated mineralization and thereby likely enhanced the dissolution of both the added shell material and the natural sedimentary $CaCO_3$. As a result, the difference between treatments and controls cannot be attributed solely to dissolution of the added shells. We therefore believe that expressing R_{diss} per unit sediment surface area is the most appropriate representation.

Line 273: Figure 3e: Why this range on the y-axis? Please add information on which incubation session this is.

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 collected 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.

Lines 265 – 270: Here you only describe data that is all in the supplement. While I can understand that the amount of data is very large, it is not very convenient to switch to the supplementary information every other sentence. Is this amount of detail needed? If it is really necessary, find a way to present all relevant data in the main script, if it is not, concentrate on the relevant data in the main script.

We thank the reviewer for this suggestion. We agree that the original text contained excessive detail on data presented in the Supplementary Information. We have therefore shortened this section to focus on the main finding that most incubations exhibited linear changes in solute concentrations over time, while briefly highlighting the nonlinear behaviour observed during week 1, which is relevant for the interpretation of the calculated fluxes in the following sections.

Line 288: I would prefer to see the NH4+ fluxes in the main script as part of Fig. 3. And having seen Fig. 4 I would transfer Fig 3 and the section that describes it (lines 262 – 273) entirely to the supplement section. Eventually, the fluxes are the important bit. The concentrations in the single sessions are not necessary to understand the discussion.

We thank the reviewer for this suggestion. As explained in our response to the comment on line 273, we have decided to remove Figure 3 from the revised manuscript. We have also considered including nutrient fluxes in the

main figure (Figure 4). However, we decided to retain these results in the Supplementary Information because clear nutrient flux signals were only observed during the first week of incubation, whereas fluxes during the remainder of the experiment were generally close to zero. We therefore believe that including nutrient fluxes in the main figure would add limited additional information compared with the primary focus of the figure on oxygen, DIC, and alkalinity fluxes.

Line 326: How exactly can pore-water analysis at the end confirm a development over time? Furthermore, this sentence and partly those before, have a quite discussing character. I suggest focusing on results here, and leaving discussions, explanations and interpretations for the discussion section.

We thank the reviewer for this comment. We agree that the pore-water analysis performed at the end of the experiment should be described more cautiously, as these measurements provide an end-point snapshot rather than direct evidence of temporal development. We have therefore revised the text to clarify this distinction. In addition, we have moved the discussion and interpretation of these pore-water results to Section 4.2 and have restricted the corresponding Results section to the description of the observations.

Lines 337 ff. This is discussion of the data. Please move these ideas to a suitable section.

We thank the reviewer for pointing this out. We agree that this discussion represents interpretation of the results. We have therefore moved these to the discussion section (Section 4.1) in the revised manuscript.

Line 399: This opening statement is not correct as it is: There are several studies that have shown undersaturation of pore waters in anoxic pore-waters with both either oxygenated or anoxic overlaying bottom waters. Please clarify under which conditions you believe this statement holds true and provide a suitable reference.

We thank the reviewer for this comment. We agree that our original wording was too general. Our intention was to reintroduce the mechanism of metabolic carbonate dissolution in permeable sediments, where aerobic mineralization in the oxic sediment layer is the primary driver of pore-water undersaturation. We have therefore revised the opening sentence to: "Undersaturation of pore waters with respect to $CaCO_3$ is promoted by oxic conditions, which drive CO_2 production through aerobic mineralization." This wording better reflects the scope of our discussion.

Lines 406 ff: This paragraph nicely describes findings from Dale et al. 2024.

We thank the reviewer for pointing this out. We have added the reference to the revised manuscript.

Line 417: Why do you believe it was oxygen limitation in the first weeks? Is there any data supporting this (except from black colour and apparently lower dissolution rates)?

We thank the reviewer for raising this point. Our interpretation of oxygen limitation during the first weeks was based on nitrogen cycling signals. During the first part of week 1, NH_4^+ accumulated in the overlying water, with measured NH_4^+ fluxes of 5.1 ± 1.5 mmol m^{-2} d^{-1}, while NO_3^- fluxes were approximately zero. This indicates that nitrification was strongly inhibited, consistent with oxygen depletion in the overlying water and sediment. During the second part of week 1, NH_4^+ fluxes declined to approximately zero, while NO_3^- fluxes increased to 5.8 ± 1.0 mmol m^{-2} d^{-1}, indicating the re-establishment of oxygen availability and the onset of nitrification. This reoxygenation allowed metabolic carbonate dissolution to occur. The temporal evolution of and NO_3^- and NH_4^+ concentrations is shown in Supplementary Figures S4 and S5, while the corresponding fluxes are provided in Supplementary Table S1. This interpretation is provided in lines 424–426, but we agree that line 417 introduced a mechanistic interpretation before the underlying evidence was presented. We have therefore revised this section to first describe the three distinct phases based on the observed MCDE patterns, while the potential mechanisms controlling each phase are discussed in the following paragraphs.

Lines 421-431: Nice explanation, but try to state things more carefully. As long as there is no measurable evidence of FeS formation, it is just a possible explanation. Likewise, it is possible, that the saturation state in pore waters has changed, but there is no evidence for this. Micro-profiling and subsequent calculation of integrated ΩCal values would have resolved this. The authors should consider this for their next experiments.

We thank the reviewer for this valuable comment. We agree that some of the mechanistic interpretations in this section should be stated more cautiously, as we did not directly measure iron sulfide formation or pore-water

saturation states throughout the experiment. We have therefore revised the text to clarify that iron sulfide formation and increased pore-water $CaCO_3$ saturation are possible explanations rather than direct observations.

We appreciate the suggestion regarding microsensor profiling and agree that calculation of pore-water saturation state would provide valuable additional information. However, performing microsensor profiling in our experimental system would be challenging, as the coarse sediment containing shell fragments poses a risk of damaging the sensors. An alternative approach for future experiments would be the installation of rhizon samplers to allow repeated pore-water sampling throughout the experiment. However, sufficient pore-water volumes would need to be collected to allow simultaneous analysis of DIC and A_T, which proved challenging during our post-experiment pore-water sampling due to the low sediment porosity.

Line 433: Again, is there any evidence for more oxygenated porewaters? Not really. It is a plausible and likely explanation for the observed fluxes and colour of the sediments.

We refer the reviewer to the response to the comment of line 417. During the first part of week 1, NH_4^+ accumulated in the overlying water, while NO_3^- fluxes were approximately zero. This indicates that nitrification was strongly inhibited, consistent with oxygen depletion in the overlying water and sediment. During the second part of week 1, NH_4^+ fluxes declined to approximately zero, while NO_3^- fluxes increased considerably, indicating the re-establishment of oxygen availability and the onset of nitrification. Together with the observed sediment color changes, these observations suggest that more oxygenated conditions likely developed within the sediment, thereby promoting metabolic carbonate dissolution.

Line 443: A data point obtained after 24 weeks does not inform at which point in time the process responsible for this data point has taken place. It informs what has happened, not when it has happened. Please rephrase this in a factually correct way.

We thank the reviewer for this important comment. We agree that the pore-water samples were collected only at the end of the experiment and therefore cannot provide information on the exact timing of the processes contributing to the observed δ^{13}C-DIC profiles. Rather, these measurements represent an integrated signal of DIC production throughout the experimental period. We have revised the text accordingly to clarify that the δ^{13}C-DIC profiles are interpreted as an end-point observation and not as direct evidence for when processes occurred.

The interpretation that metabolic carbonate dissolution during the second phase was an important contributor to the pore-water DIC pool is based on the intermediate δ^{13}C-DIC signature, which reflects approximately equal contributions from organic matter mineralization and $CaCO_3$ dissolution. Furthermore, because dissolution rates were highest during weeks 9-15, this phase likely contributed substantially to the integrated isotopic signal observed at the end of the experiment. We have revised the text to make this distinction clearer.

For entire section 4.2.2: I agree with the authors explanations as they appear most likely. Still, the way the authors state these explanations is too strong.

There is no evidence, of FeS formation, the abundance of labile or non-labile Corg. Please rephrase this section in a way that it becomes clear what is fact-based and what is the most likely explanation for the observations.

We thank the reviewer for this comment and appreciate the agreement with the proposed interpretations. We agree that some of the statements in section 4.2.2 were phrased too strongly given that several mechanisms are inferred from indirect evidence rather than directly measured. As described in our responses to the previous comments related to section 4.2.2, we have revised the text throughout this section to more clearly distinguish between observations, supporting evidence, and the proposed interpretations/hypotheses.

Reviewer 3

General comments:

Overall, the manuscript is well written and clearly structured. The methodology is straightforward, appropriately explained, and well suited to address the key questions. I only have minor comments, mainly regarding the structure and clarity of the results and discussion sections.

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

Specific Comments:

Line 149: Please add some details on the specific optodes used here.

We refer the reviewer to line 131, which provides details on the specific optodes used.

Lines 152-154: Can you give a range here for the duration of the 'closed' part (e.g., 1-3 days)?

We thank the reviewer for this suggestion. We have added the duration of both incubation periods to the revised manuscript (specifically to line 145), specifying that the closed part lasted around 3 days, and the open part around 7 days.

Line 155: Please state somewhere the volume of seawater enclosed in the chambers and whether any correction for seawater dilution was done for the closed incubations.

We thank the reviewer for pointing this out. We have stated the volume of enclosed seawater in the chambers in section 2.1, and we have clarified in section 2.5 that solute concentrations were corrected for dilution resulting from sample volume replacement in both closed and open incubations.

Line 156: Can you be more specific here; e.g., sampled every time O2 dropped by 10 %?

We have clarified the sampling strategy in the revised manuscript by specifying that sampling occurred after approximately every 8% decrease in O_2 saturation, as determined from the continuous optode measurements.

Line 210: Move definition of ICP-OES to line 171 (first occurrence).

We thank the reviewer for pointing this out. We have moved the definition of ICP-OES to its first occurrence in line 171.

Line 263: I recommend changing to 'in the closed (a, b) and open (c-e) incubations'…

We thank the reviewer for this suggestion. In light of comments from all three reviewers (see responses to similar comments above), we have decided to remove Figure 3 from the revised manuscript.

Figure 3: I find the setup of Figure 3 slightly confusing (e.g., mix between open and closed incubations). Maybe move c) to middle panel and clearly state that upper panel shows closed incubations and middle panel open incubations. I would also recommend reducing the range for ammonium concentrations (y axis in e). Keeping it at the same level as nitrate is somewhat meaningless here. Also, why did you choose to only depict a representative incubation rather than showing the variability as well? Wouldn't mean plus standard deviation be more meaningful here?

We thank the reviewer for the suggestions. As explained in our response to the comment on line 263, we have decided to remove Figure 3 from the revised manuscript.

Figure 4: I find using positive fluxes to indicate both a flux into the overlying water (DIC, alkalinity) as well as a flux into the sediment (oxygen) confusing. Consider changing oxygen fluxes to negative values. Also, consider adding nutrient fluxes to the main figure here rather than the supplements.

We thank the reviewer for this suggestion. We have revised the oxygen flux figure to report oxygen uptake as negative fluxes. We have also considered including nutrient fluxes in the main figure. However, we decided to retain these results in the Supplementary Information because clear nutrient flux signals were only observed during the first week of incubation, whereas fluxes during the remainder of the experiment were generally close to zero. We therefore believe that including nutrient fluxes in the main figure would add limited additional information compared with the primary focus of the figure on oxygen, DIC, and alkalinity fluxes.

Lines 372-373: This sentence feels out of place and raises more questions than it answers. Please provide a better transition to sections 4.2 and elaborate on these additional alkalinity sources and sinks here or elsewhere in the discussion.

We thank the reviewer for this helpful suggestion. We agree that the original transition was abrupt. We have revised the text to explicitly state that the deviations observed in weeks 3, 11, and 13 indicate the presence of additional alkalinity-generating or alkalinity-consuming processes, and we now refer the reader to the following section where these processes are further discussed.

Lines 406-407: Can you be more specific here, i.e., state specific reactions and/or metabolites.

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

Lines 422-424: Very fine shell material was likely also added to the sediment given that all ground shell material smaller than 0.25mm was retained for the experiment. Can you speculate on potential impacts of clogging of pore space on oxygen penetration and observed solute fluxes? Was the grain size distribution of the ground shell material measured?

We thank the reviewer for raising this important point. The grain size distribution of the crushed shell material was unfortunately not measured separately. We acknowledge that the addition of fine shell particles could potentially influence sediment permeability and pore-water exchange by partially filling pore spaces. However, we expect this effect to have been limited because shell material represented only approximately 8 wt% of the amended sediment layer and was homogenized with the surrounding sediment. Furthermore, the rapid oxygen consumption observed following shell addition, together with the accumulation of ammonium in the overlying water, indicates that enhanced oxygen demand from mineralization of the organic matter associated with the shells was likely the dominant driver of oxygen depletion. This interpretation is further supported by the occurrence of oxygen depletion in the overlying water in the first week, which would not be expected from reduced pore-water exchange alone. Nevertheless, we acknowledge that changes in sediment permeability caused by fine shell particles cannot be completely excluded.

Line 460: Is this a new steady-state? It seems that additional timepoints would be required to show whether this is a new steady-state or if it decreases further to meet control values.

We thank the reviewer for this observation. Although the mean dissolution rates in week 24 (2.2 ± 1.1 mmol m^{-2} d^{-1}) and week 21 (7.0 ± 3.3 mmol m^{-2} 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 gradually 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.