Please see below for our responses (in italics) to the detailed comments by the reviewers (normal font), outlining the changes we will make in our revisions. We appreciate their feedback which helped us to significantly improve our manuscript.

Reviewer1:

General comments

Bice et al have used a 1D reactive transport model to assess the impact of carbonate addition to benthic acidification and alkalinity release in a system broadly comparable to Yaquina Bay, OR (USA). To better constrain their mineral dissolution data, they used laboratory measurements of bio-calcite and bio-aragonite dissolution. The work in itself is interesting and deserves in my opinion to be published. As the authors also state, relatively little work has been done on mineral addition to sediments as a way of OAE, so that a study focusing on this topic would surely benefit the community. However, the manuscript itself needs a major revision, especially in the introduction and in the motivation for this study.

My biggest issue with the title, abstract and introduction is that they focus a lot on ocean acidification (OA), while the manuscript itself mostly focuses on sediment biogeochemistry. As the authors acknowledge and also show themselves in Fig 5, OM mineralization is by far the biggest source of acidity in sediments. And what they achieve by adding carbonate minerals is to counteract the acidity produced by OM mineralization, not by OA. If the authors really wanted to investigate how the effect of OA in coastal sediments is countered by carbonate mineral addition, as the title states, their baseline simulation before mineral addition should be compared to a) a simulation with only OA (i.e. lower overlying water pH due to higher DIC) and no mineral addition and b) a simulation with OA and mineral addition (shown in Figure 5, but not compared to both of the above scenarios). And then perhaps vary with the extent of OA. But this is not done In my opinion, the manuscript is much better framed solely in the context of OAE alone, and the links with OA only make it very confusing.

Besides this, there are some places in the introduction where the literature isn't properly cited, specifically on OAE; sediment buffering is determined without quantifying buffering factors / sensitivities, which are easy to calculate from the model results; and the budget construction is very difficult to understand without the supplementary information, so I suggest to move this part of the supplementary information to the main manuscript. I will elaborate on all of the above points in the detailed comments.

We thank the reviewer for the valuable feedback which helped us to improve the manuscript. As outlined in our responses to the detailed comments below, we have made numerous revisions aimed at clarifying our work and framing it in the context of OAE rather than ocean acidification.

Detailed comments

L. 13: "which can further lower both pH and the carbonate saturation state" → only in oxic conditions, else pH is increased due to TA generation

We changed this sentence to '...especially at the surface sediment where oxic conditions lower both pH and the carbonate saturation state'.

L. 13-14: This sentence, equal to the title, is one of the major issues I have with this work. Sediments are naturally lower in pH than the overlying water, due to the natural process of OM mineralization. Why is this framed as if this natural process is a bad thing? OA only leads to a slightly lower pH due to higher DIC in the bottom water, the majority still comes from OM mineralization and the impact of eutrophication (i.e. enhanced OM input) would be much higher. I feel the motivation that is presented here is not correct, and that it should be the potential of sediments to increase TA efflux to the overlying water and thus CO2 uptake (as the authors themselves also write in L. 24-25, but which should come out much more strongly in the abstract).

We generally agree with the comment (as reflected in our statement in the lower part of the abstract as pointed out by the reviewer) and address it by recasting the motivation toward sediment oxidation and moving away from ocean acidification. We also changed our title to:

'The effect of carbonate mineral additions on biogeochemical conditions in surface sediments and benthic-pelagic exchange fluxes'

L. 22: which "buffering effect"? The authors do not present any buffer factors / sensitivities

Addressed with rewording 'buffering effect' with 'the effect of mineral addition' (in addition, as outlined further below, in our revised manuscript we now also discuss buffer factors).

L. 30–31: "The effect of CO2 additions depends on the buffering capacity of the water. This is quantified by its alkalinity" → this is incorrect and not stated by these authors. Alkalinity plays a major role in buffering, but the buffering capacity is quantified by buffer factors (or rather their inverses, sensitivities). Please rephrase.

Rephrased by stating 'alkalinity plays a major role in quantifying buffering capacity'.

L. 49-61: I have many issues with this paragraph. First, it feels very disjointed from the previous and following paragraph, as if it's put in at a later stage. But mostly, even after reading multiple times, I really miss the purpose of it in the motivation for this study. Yes, benthos impact the overlying water (L.60-61), but how does the overlying water impact benthos? The line of reasoning in L.54-61 states that benthos live under lower pH conditions than in the overlying water, and this lower pH is not due to OA. My suggestion would be to strongly shorten this paragraph and only keep the essential information in, without trying to find a motivation for this study.

We follow this suggestion and addressed this comment by removing sentences in 49-54 and some rewording of this paragraph to focus more on porewater conditions.

L. 65: "OAE is mainly studied for its effect on ocean - atmosphere CO2 exchange" Renforth and Henderson (2017) actually looked at the biological side too, and provided a motivation to study the biological response to OAE.

Addressed by adding that Renforth & Henderson (2017) looked at the impact on organisms, too.

L. 65-68: This statement does not properly cite the literature. Taylor et al (2016) didn't investigate OAE, but enhanced weathering on land. While this ultimately also affects oceans through enhanced TA input, they did not investigate OAE.

Addressed by changing 'OAE' to 'increasing ocean alkalinity through enhanced weathering' when referring to Taylor et al. (2016).

L. 68-70: The work by Montserrat et al (2017) was also done in the context of coastal applications, so this citation does not match the statement.

Changed open ocean OAE statement reference to Fennel et al. (2023).

L. 71-72: Also Ferderer et al (BG, 2022) had a coastal focus, and recently Fuhr et al (Front Clim, 2024) was published.

Added suggested articles.

L. 77-78: Rather than assessing the extent and duration of buffering indirectly, why not simply calculate buffer factors or rather sensitivities as per Middelburg et al (2020)? With the data generated by the model, it should be easily possible to compute one or more of these terms, which actually shows how a system change its sensitivity to acidification following mineral addition

We follow this suggestion and now also compute dpH/dTA (see below).

L. 79: "Building on the previous work by Krumins et al. (2013) and Rassman et al. (2016)" → Rassman et al (2016) did not use an RTM, I would suggest to remove this citation here.

Removed Rassman et al. (2016) from this sentence.

L. 97: Figure S2 does not show a correlation coefficient (r) but a coefficient of determination (R2).

We are now referring to the coefficient of determination (R^2) in the text.

Table 1: on which scale is pH presented here? Which equilibrium constants used for this calculation (and the RTM in general)? The same as in L. 132-133?

We do not list the pH in Table 1, but we updated the pH in Table 2 to reflect the source of those values and that pH is in free proton scale as used in AquaEnv R package. Constants used in calculating DIC of overlying water were based on Lueker et al. (2000). The saturation states for calcite in Table S1 were also calculated using the equilibrium constants of Lueker et al. (2000).

Table 2: Are all these parameters based on Yaquina Bay, like Rc0? If yes, would be good to explicitly add to the start of section 2.3 that the RTM is meant to describe this site.

Our parameters are mostly taken from the literature as referenced in Table 2 to represent a near-coast environment. However, Rc0 was dependent on measured alkalinity fluxes in core incubations from Yaquina Bay. Thus, the modeled setting is more general, but loosely based on that location.

L. 188-190: This statement initially confused me, because alkalinity does not only include carbonate alkalinity, but also sulfide and ammonia alkalinity. Perhaps, for clarity, the equation of

L.180 should include only the acid-base species that were considered in the modelling, or L.188-190 should explicitly state which acid-base systems were included.

We appreciate this suggestion and removed this equation since the chemical species that impact TA in our work are evident from Table 3 where we explicitly list the impact of each reaction..

Table 3: I suggest to not only show dTA/dR but also dDIC/dR in this table. That will make part of the results and discussion easier to write because you can simply refer to this table when discussing the DIC/TA ratio (e.g. in L. 265 where you now refer to Thamdrup and Canfield (2000)).

Added dDIC/dR to the Table 3 as suggested.

L. 200-201: I miss a discussion in the manuscript (not per se here) on how this mineral addition would or could be done in practice. It now remains ambiguous how hypothetical this modelling exercise is.

We appreciate the suggestion and agree that the practical challenges of mineral addition are important. However, this is a complex topic that depends on the specific conditions at a site and beyond the scope of this paper. We now add that the model approach of mixing minerals into the upper 2 cm follows field experiments that our group is carrying out. ('Our model assumes that the minerals are mixed into the upper 2 cm of sediment, matching recent field manipulation experiments in Yaquina Bay.'). We also add a brief discussion mentioning the importance of how the minerals are applied. (' An important consideration is how the minerals are applied to the surface sediment. For example, a simple 'top-dressing' approach would involve less disturbance to the sediment and less labor than surface raking, but may be washed away more easily, and react less readily with the metabolic acids produced in the underlying sediment. Optimal mineral application and subsequent effects clearly requires field trials across a range of hydrodynamic settings')'

L. 201-203: This is not clear without presenting the impact of environmental factors first. I suggest to move it to after discussing the sensitivity to environmental parameters.

We moved this sentence to the end of the Application section.

L. 205-208: Why would minerals only dilute the organic matter and not the other solid phases in the sediment?

They would not. However, the goal of this assessment was to identify the likely impact on organic matter mineralization -since it drives mineral dissolution-, and its impact on saturation state, and we preferred to alter only this single factor in the model. Because this is a somewhat simplistic approximation to the full effect of adding minerals, we decided to clarify the statement and also move this entirely to the supplement.

L. 209-217: I think this information is better presented in a table. The information that also the minerals themselves were changes to aragonite, and that mineral dose is varied and also bottom-water pH is varied, is missing here. Finally, it should be made clear that the low OW pH scenario (term in Figure 5) is an OA scenario because TA is kept constant in the calculation, so DIC is increased.

We added a table summarizing parameters for different scenarios.

L. 219-230: I could not properly understand the method without the supplementary information, so I suggest to move here in a slightly shortened way, and at least include Figure S4.

We moved the mass balance equations – but not figure S4 - from the supplement to the main text, to make it easier to understand while keeping the focus on sediment biogeochemistry rather than giving too much weight to the rough estimates of CO_2 drawdown.

L. 266-267: this pH effect is also shown by the work of Soetaert et al (2007) and Hoffman et al. (2010) which are logical to cite.

Added citations.

L. 270-274: I'm not sure how to interpret this statement. So if you had used another k-value for sulfide oxidation, you would have obtained very different results? How (un)certain and sensitive was this parameter value in your model? Also, Brenner et al (2016) did not use an RTM or a kinetic approach, so that may also explain the difference.

We expanded the discussion of sulfide oxidation and its impact on alkalinity generation which we believe is a significant difference between our setting and that studied by Brenner et al. (2016). In our simulations, we used literature values for our reaction parameters, which leads to the outcome documented here. However, in settings where sulfide oxidation is more dominant, changing the interplay between e.g. Fe and S cycles, the alkalinity response can shift. We now address this explicitly by stating:

'This discrepancy may be due to the potentially substantial impact of sulfide oxidation on alkalinity dynamics in organically rich near shore sediments (Krumins et al. 2013). The aerobic oxidation of reduced products of organic matter mineralization such as ammonium, hydrogen sulfide and dissolved Fe^{2+} with O_2 all lower alkalinity. Conversely, anaerobic oxidation of hydrogen sulfide with iron oxide as electron donor produces a large amount of alkalinity, that either accumulates if the reduced iron produced is captured in the sediment (i.e., iron sulfur precipitates) or is removed if the Fe^{2+} subsequently reacts with O_2 in the oxic zone (see Table 3).'

L. 282-284: I think this is a nice result because if this is sustained and the FeS is long-term removed, it is a net TA source on the system scale, as shown by e.g. Hu and Cai (2011) and Brenner et al (2016).

We made the following changes to reflect this suggestion: '...which eventually can lead to long term removal of reduced sulfur through pyrite burial (Hu & Cai, 2011a).'

L. 292-294: In my opinion, this should come out much more clearly in the abstract, instead of the link with OA

We added the following sentence to the abstract to address this suggestion: '...demonstrating the potential of alkalinity enhancement in mitigating surface sediment acidification.'

L. 305: "lower dTA/DIC ratios" lower than what?

Clarified as 'lower dTA/dDIC ratios than mineral dissolution'.

L. 311-313: I'm not sure if I entirely follow this statement, because also in this system the majority of NH4 is produced by sulfate reduction. Maybe paraphrasing would help.

We removed that sentence as it was not adding a lot to the discussion.

L. 315: Figure 3 doesn't show the depth of the oxic layer, as it shows depth-integrated fluxes

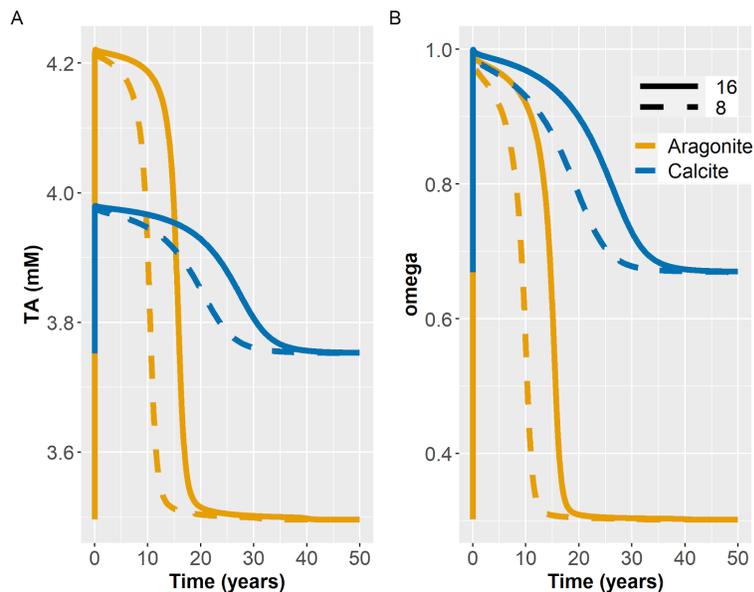
Figure 3 shows the contributions of reactions of reduced chemicals with O_2. We didn't explicitly refer to the depth of the oxic layer and hence to leave the manuscript as is.

L. 317-318: "added mineral dissolved at depth" → how deep? Or is it simply meant as opposed to at the top of the sediment?

Clarified by changing to '…added mineral dissolved in deeper layers…'.

L. 325-326: Why depth-averaged over 10 cm? The mixing depth is 4 cm (only 10 cm in Figure 5), and Figure 2 doesn't give a clear incentive for this choice since the conditions are still very different at the 0-10 cm depth range. I can imagine that choosing a different depth interval for the averaging would have yielded quite different results, have the authors checked this? And why not just make a 2D plot of depth versus time?

The choice of the depth interval is somewhat arbitrary. We chose 10 cm because this depth represents a meaningful layer relevant for many benthic infauna. But we also considered 4cm instead of 10cm and added the following sentence to the paragraph: 'We also tested averaging over 4cm which yielded the same patterns (with slightly lower TA concentrations) as shown in Figure 4 (not shown).'

(figure with averaging over 4cm for reference).

We also considered 2D plots. However, we found that the simpler 1D plots allow us to better convey the main message, and we opted to stick with this choice.

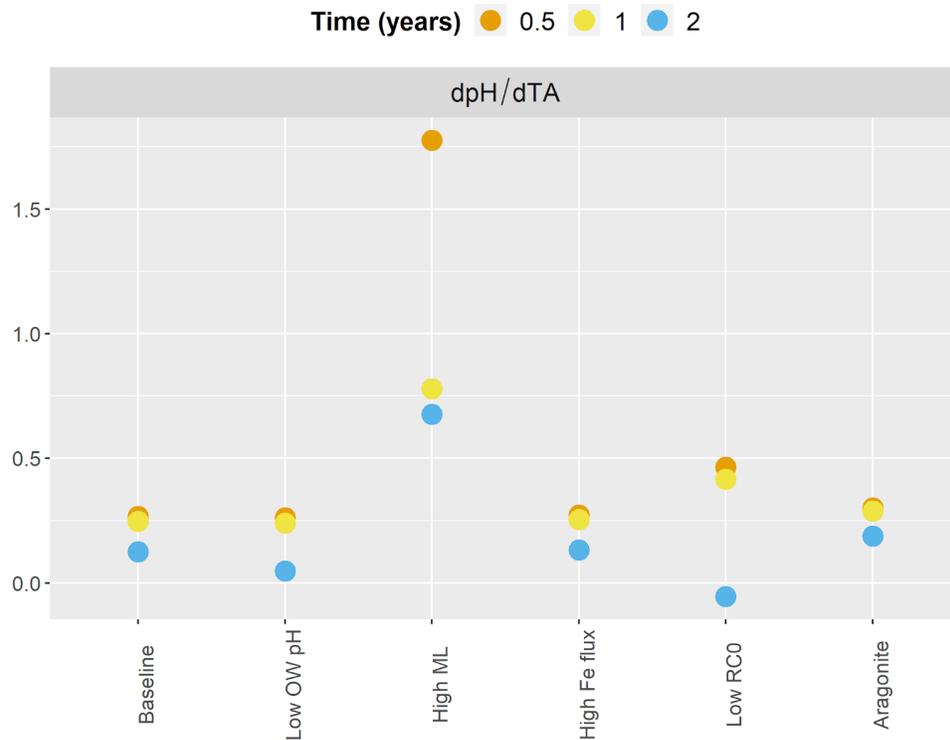
L. 326: "because the surficial sediment is most relevant for juvenile bivalves impacted by ocean acidification". I still don't follow, because the pH and saturation state are much lower in the top part of the sediment anyway, and not due to OA.

Changed 'ocean acidification' to 'acidification' to focus on lower pH in the upper parts of the sediment.

L. 346-347: "Burial of minerals" do you mean below 10 cm here?

Clarified by adding '...burial of carbonate minerals below 10 cm...'

L. 353: I am a bit critical when manuscripts talk about a "buffering response" without quantifying any buffer factors or better sensitivities. The authors have all parameters to compute many of the sensitivities in Table 1 of Middelburg et al. (2020). Perhaps in this case the Egleston et al. (2010) factor that looks at the response of CO_3^{2-} (and thus saturation state) to TA change ($\omega TA-1$) would make the most sense, or a general dpH/dTA. If you don't plan to include it, then maybe just better call this section a sensitivity analyses.

We changed the title of this section to sensitivity analyses as suggested. However, we also computed the change of pH with total alkalinity, as shown below for the top 10cm as in other figures. All scenarios show decreasing values of dpH/dTA with time illustrating that as buffering develops, increasing TA has a diminishing impact on changes in pH. High ML (i.e. deeper mixing) has larger values which are due to more efficient mixing of minerals. It is also interesting to see that after 2 years, low Rc0 scenario produces slightly negative dpH/dTA which might point to the suppression of mineral dissolution due to less acidification (i.e., although smaller, TA production is done by OM mineralization which also decreases pH). We plan on adding this figure to the supplement to support discussion around Figure 5.

L. 362-365: These results (and those of Figure 5d) so nicely show that OM mineralization is by far the dominating factor governing sedimentary carbonate system dynamics, and not OA. It boils down to my major comment.

Made changes accordingly throughout the paper reflecting the focus on sediment acidification rather than OA.

L. 365: "pH was buffered the most and increased the fastest in the aragonite implementation" that is a contradiction: stronger buffering means a smaller pH increase. Or am I misunderstanding this sentence?

Reworded by removing '… was buffered the most…'.

L. 367: "as buffering developed" so this is typically something you would like to see with sensitivity factors.

Changed 'as buffering developed' to 'as the impact of mineral addition developed'.

L. 380-382: perhaps nice to show some of the key process results related to Figure 5 (like this once) in the supplementary information.

We appreciate the suggestion. However, we don't think this is needed for all the scenarios since changing Fe input had more complex feedback therefore it was worth showing some numbers.

L. 394-396: I would still argue that the absolute changes are more important than the relative changes in pathways, but where can we see this? Perhaps also add to the supplementary information.

We appreciate the suggestion. However, we focus on partitioning between pathways and their relative impact on alkalinity therefore we considered relative changes.

L. 399-400: It's written as if this is a problem. But is it?

Reworded to clarify.

L. 403-404: I would like to see these results as well, because Figure 3 seems to suggest that the impact of mineral addition is relatively minor compared to e.g. sulfate reduction.

We appreciate this suggestion, and we did include changes in some important biogeochemical processes in the text as following: 'In our simulations, the lower OM mineralization rate led to depth-integrated reduction by 29% for aerobic mineralization, 19% for denitrification, 55% for sulfate reduction and 380% for mineral dissolution.' This supports the idea of lower OM mineralization rate not only changes the TA produced by OM mineralization, but also impacts dissolution/precipitation.

L. 407-408: The work by Cyronak et al. and Yamamato et al. is on coral reefs. How relevant is that in the context of your system?

We updated the references with a more relevant study that focuses on benthic fluxes in coastal shallow water environments. Change made is the following: 'These changes include the impact of short-term diel patterns and small scale spatial heterogeneity on benthic fluxes (Gadeken et al., 2023)...'

Gadeken, K. J., Lockridge, G., & Dorgan, K. M. (2023). An in situ benthic chamber system for improved temporal and spatial resolution measurement of sediment oxygen demand. Limnology and Oceanography: Methods, 21(11), 645-655.

L 407-409: I miss eutrophication / low-oxygen conditions in this list. See work by Cai et al (2011) or Hagens et al (2015). Perhaps it falls under anthropogenic impacts, but that is very broad.

We added eutrophication with a reference to Cai et al. (2011) as suggested.

L. 414-415: Rather than being lost, this movement of minerals may actually also stimulate dissolution and thus shorten the time of reach steady state again. See e.g. Meysman and Montserrat (2017).

Added '...washing away or enhancing dissolution (Meysman & Montserrat, 2017) ...'

L. 426-431: I suggest to put all numbers in a figure (similar to Figure S4) or table for clarity.

As we state in the supplement, estimates of mCDR are difficult to constrain and highly sensitive to the overlying water conditions making it difficult to report exact numbers. Therefore, we would like to keep numbers reported in supplementary text.

L. 431-434: Does this upscaling from an annual flux consider full mineral dissolution?

This upscaling is based on the TA and DIC fluxes during the peak impact of the dissolution (2 years) after which minerals/dissolution products start being buried. This has been clarified in the revised text.

L. 441-442: Also driven by primary production and aerobic respiration?

To address this, we added biogeochemical component to the end of the following sentence: '…total air-sea exchange flux, which is driven by tidal exchange of DIC and alkalinity between the bay and the coastal ocean and biogeochemical processes in water column.'

L. 447-448: Maybe you can speculate what this would mean for multiple applications. What would be the ideal timeframe for a second application?

We appreciate this comment which adds another practical component to our work. We added few words pointing to reapplication of this method to keep sediments saturated to the following sentence: 'We demonstrated that addition of minerals increased pH and saturation state of the sediment over the timespan of a few months and had the potential to persist over 30-40 years after which subsequent implementations might help keep the impact of mineral addition sustained. However, this would depend on the physics impacting the dissolution of minerals such as storms impacting the fate of added minerals and the impact of porewater advection that can enchance dissolution which were not considered in our model.'

L. 451-253: Is this simply because supersaturation would lead to mineral precipitation and these minerals would then dissolve again at a later stage? I'm not sure that I've read that somewhere explicitly.

Minerals dissolve fast and saturate the porewater. Therefore, we don't expect that adding more minerals results in more dissolution. However, when more minerals are added, it takes longer for minerals to get buried beneath the top 10 cm therefore the impact of dissolution dissipates slower as reflected in Figure 4.

Supplement L.77-78: And what about calcification? Is that assumed to be net zero in the water column?

We appreciate this comment pointing out an important aspect. In our approach, we assumed that calcification is trivial compared to other processes impacting TA/DIC and can be ignored especially considering the limited presence of calcifiers in our region (e.g., coccolithophores, pteropods).

Supplement L.83-85: This sentence does not read well, but do I understand correctly that RDIC, RTA and pH are all kept constant? Can you support that assumption?

We assumed that benthic fluxes would not impact biogeochemical processes in the water column because ocean sources have a larger effect. This is due to the large tidal exchange and decreased river flow in summer months (Hickey & Banas, 2003) and our study site is close to the estuary mouth with shorter residence times. Seasonal changes impact water column biogeochemical processes and pH which are not accounted for in our simple estimate that represent average surface water in the bay. To clarify we added the following sentence to the respective section in SI: 'This assumption was in line with Hickey & Banas (2003) which states that the estuarine water

column biogeochemical processes are driven by oceanic source water and process, given the large tidal prism and relatively small volume in this and other Pacific Northwest estuaries..'

Hickey, B. M., & Banas, N. S. (2003). Oceanography of the US Pacific Northwest coastal ocean and estuaries with application to coastal ecology. Estuaries, 26, 1010-1031.

Supplement L.92-94: This doesn't make much sense to present this way, because, pH is a logarithmic unit and so this value will change depending on the pH range you use. Better express in terms of [H+] change or add the pH range that is used.

Ranges added for clarification.

Minor and technical comments

General comment: "Alk" and "TA" are both used throughout. Please choose one of both for consistency.

Corrected with the use of TA.

L. 20: "total alkalinity"

Corrected as suggested.

L. 21: change "significant" to "substantial", no statistics done

Corrected as suggested.

L. 32: "aerobic respiration"

Corrected as suggested.

L. 32-34: this is a complex sentence, better split it in the impacts of primary production & aerobic respiration on the one hand, and calcium carbonate dissolution / precipitation on the other hand

Corrected as suggested. See following for revised sentence:

'Among these processes, both respiration and $CaCO_3$ dissolution produce total alkalinity (TA; Dickson 1981). However, pH is increased by $CaCO_3$ dissolution and decreased by respiration (Su et al., 2020; Green et al., 2009; Andersson et al., 2005).'

L. 36: "the decrease in pH" → change to "the OA-driven decrease in pH" for clarity

Corrected as suggested.

L. 38-39: remove "against ocean acidification", because the buffering capacity against any acidifying process is affected

Corrected as suggested.

L. 45: I'd remove "in particular" because TA generation is very minor under oxic conditions, and TA is even consumed if nitrification follows OM mineralization

Corrected as suggested.

L. 46: there are better references for this statement than Gimenez et al (2018). E.g. Chen & Wang (1999) for a first suggestion or Hu and Cai (2011) for a global estimate

Corrected as suggested with Hu and Cai (2011) reference added.

L. 65: in other parts of the manuscript you use mCDR, instead of CDR. I would define therefore mCDR here and stick to that abbreviation.

Corrected as suggested.

L. 97-99: Add a reference to Figure S2 at the end of the sentence.

Corrected as suggested.

Table 2: typos in Van Cappellen.

Corrected as suggested.

L. 183: "The OM mineralization rate" add that this is Rc0 for clarity

Corrected as suggested.

L. 184: "the resulting Rc0 value"

Corrected as suggested.

Table 3: the term γOM is nowhere in the manuscript defined (or I missed it)

Defined in the Table 3 caption as suggested.

Table 4: I would say that Rc0 is calibrated to match experimental TA data, but the parameter itself is not experimentally derived.

Corrected as suggested.

L. 235-236: Rephrase to "The results are in line with other results used to setup the RTM" or equivalent. The relationship is not an outcome of the cited RTMs.

Corrected as suggested.

Figure 1: please show the equations, correlation coefficient and p-values of the fits. Use different symbols for both colours for clarity.

Figure 1 was updated with the suggested changes.

L. 281: Rassman et al (2018) is not a modelling study, so I would remove the citation here

Removed as suggested.

L. 286-288: This sentence is not very clearly written, please rephrase.

Rephrased as: 'After 2 years, CO_2 production led to lower pH in the oxic layer and prevented full saturation.'

Figure 2: please put the depth in positive values, instead of negative values. And add in the caption how much calcite was added

Updated figure with positive depth.

L. 303: "Our results are in line"

Corrected as suggested.

L. 303: "calcite mineral dissolution"

Corrected as suggested.

L. 309: "in the marine environment"

Corrected as suggested.

Fig 3: The orange colour represents either dissolution or precipitation, depending on the sign of the DIC/TA response. So the legend needs to be altered here.

Updated figure with 'CaCO$_3$ - Diss/Prec'

L. 333-334: This cannot be clearly seen in the figure. Maybe add the baseline values as horizontal lines to the plots.

Added a sentence clarifying this as the following:

'Temporal impact of mineral addition was shown as TA and saturation state evolve from steady state conditions (t=0) then return back to steady state levels. (Figure 4).'

L. 343-344: also this is difficult to see in the figure. Adding vertical lines might help here, and/or a different figure design (2D plots as suggested earlier). Or the important times can be put in a table.

Added a sentence clarifying this as: '(i.e., orange curve reaches maximum slightly after the blue one)'.

We appreciate the suggestion however, as addressed in the previous comment above, we believe 1D plot delivers our message in a more concise way than a 2D one.

L. 346-347: "Burial of carbonate minerals"

Corrected as suggested.

L. 357: "the conditions" → which conditions?

Corrected as suggested.

L. 358: "with a low Rc0 scenario"

Corrected as suggested.

L. 360: "the deep bioturbation scenario" → i.e. with a deep mixed layer? Try to be consistent with terminology

Corrected as suggested.

L. 361-370: References to subfigures are missing here.

Corrected as suggested.

L. 366: "than the baseline and high FeOx flux scenarios"

Corrected as suggested.

L. 368-369: This is very difficult to see in the figure. I suggest a second y-axis for the low RC0 scenario.

We appreciate the suggestion. However, because we wanted to show the scale of change compared to other scenarios we decided to keep it this way.

L. 374: Add that lower overlying water is the OA scenario as TA is kept constant

Corrected as suggested.

L. 382-384: This sentence does not read very well.

Rephrased as: 'Overall, increased iron oxide supply led to slight decreases in TA concentrations and fluxes, and small increases in pH showing an effect similar to decrease in OM mineralization rate in a smaller magnitude (Figure 5).'

L. 390-392: Add that this is the OA scenario.

Corrected as suggested.

L. 396: "the low rate of aerobic mineralization" (because in the context of H+ production)

Corrected as suggested.

L. 439: "the Alk:DIC ratio"

Corrected as suggested.

L. 441: "which is drive by tidal exchange"

Corrected as suggested.

L. 450-451: True, but sulfate reduction is way more important then CaCO3 dissolution. Maybe paraphrase to show that.

Added: '...with sulfate reduction having a larger impact.'

L. 459-460: "higher dissolution rates"

Corrected as suggested.

Supplement L. 68: add that FaDIC is atmospheric CO2 uptake and that negative values indicate outgassing.

Corrected as suggested.

Reviewer 2:

Bice et al present a 1D reactive transport sediment diagenesis model, in which they test the effect of adding CaCO3 to sediments on a) porewater alkalinity and other relevant species, and b) the resulting net production (i.e., sediment flux) of alkalinity and DIC. They also describe lab-based CaCO3 dissolution experiments from which they derive the dissolution rate parameters employed in the model. While the porewater chemistry and related flux results are the primary focus of the study, they also use a box model to extend their modeled flux data to calculate the potential "marine CO2 uptake". At the porewater scale of analysis, they find that CaCO3 addition to sediment results in little change in species affected by redox reactions, but pH and omega are increased significantly 2 years after mineral addition. They also conclude (via a CDR = 1⁄2 Alk flux relationship) that the CaCO3 application can result in significant atmospheric CO2 uptake.

This topic is timely and interesting, and I think the model study contributes meaningfully to the conversation. However, I find three major issues with the paper that need to be addressed before consideration for publication: 1) Unclear justification and aim (amelioration of benthic acidification seems to be the main point, but it's not justified well; and mCDR via OAE is also discussed significantly); 2) the mCDR projections are coarse, misleading, and don't fit well with the rest of the data; and 3) the kinetic parameterization used for CaCO3 dissolution needs to be improved or better justified (modeling CaCO3 dissolution is the meat of the paper, but the parameterization is quite simple, and doesn't consider more thorough published data). Furthermore, the overall readability of the paper needs to be improved. A major issue on this point is that the lab methods are incompletely described, but reference an unpublished thesis. Despite these concerns, I think the topic is interesting enough that the study is valuable if these issues are addressed in a revised manuscript.

We thank the reviewer for the valuable feedback which helped us to improve the manuscript. As outlined in our responses below, we have made numerous revisions aimed at clarifying our work. In line with our response to feedback from reviewer 1, we more clearly delineate the effect of sediment mineralization from that of OA and improve the framing of our work, we better justify the use of our dissolution rate constants and clarify the value and limitations of the rough mCDR estimates.

Major issues:

Unclear aim: I found the aim of the paper unclear. Specifically, the focus on both mitigating the effects of OA on sediments, and mCDR (i.e., atmospheric CO2 uptake via OAE) is confusing. These are different enough topics to merit dedicated conversations and scales of analyses, yet neither are sufficiently explored. The jump between a porewater model and atmospheric CO2 uptake requires many intermediate assumptions, yet the assumptions made to calculate atmospheric CO2 uptake are coarse and inaccurate (described in subsequent point). I think this paper could be more valuable if it focused more on the sediment model, and limited discussion of atmospheric CO2 uptake. Second, the justification that sediment acidification (due to OA) is a problem, is not well established in the introduction. References to benthic organisms are scattered throughout the paper, and the Introduction does not convincingly establish that buffering porewater pH is indeed

an important objective for improving the health of benthic organisms. (Indeed, I imagine many benthic organisms are evolved to tolerate low pH conditions.) Related to this, I found the paper poorly grounded in OA, OAE (specifically accelerated weathering of limestone and coastal enhanced weathering), and CaCO3 dissolution research, with foundational citations missing, or others mis-referenced (e.g., L 70: Montserrat et al, 2017, is not about open ocean CDR). Reframing the discussion, with better references, could help here.

We appreciate the reviewer's suggestion. As detailed below, and in part in our response to reviewer 1, we addressed the above points by focusing our work on sediment biogeochemistry and removing most of the discussion of OA. We present CO_2 uptake as a simple extension to our sediment model, so we tried to make it clear that it is not the main focus of this study, but only a rough estimate of the impact of altered benthic DIC and TA exchange fluxes following the addition of carbonate minerals. As suggested, we shortened details about calcifying organisms but focused on sediment biogeochemistry. We also reference some additional recent publications.

CaCO3 dissolution kinetics: The rate parameters used for CaCO3 dissolution, based on four experimental data points, need to be better justified. First, they use a linear dependency with omega, and assume a rate order of 1. Much existing research exists on this topic, and some of the most comprehensive studies (using both biotic and abiotic CaCO3) have shown non-linear trends (due to dissolution mechanism transitioning from step-edge retreat to etch pit formation), and reaction rate orders not equal to 1. See, for example, Adkins et al, 2021 (Annual Reviews in Marine Science); Subhas et al, 2018 (Marine Chemistry)). While the simplified dissolution rates used in this paper may be adequate as rough approximations for the RTM, I find it strange that these other papers aren't considered, either using their published rate equations, or at least citing them. Second, aragonite dissolution has been shown to be slower than calcite dissolution (see same sources), but is comparable in the rate equations presented here. Furthermore, what are the dissolution constants used? They are not in Table 4, and only alluded to in Section 3.1.

We appreciate relevant articles suggested and by no means intended to minimize the vast existing literature on carbonate dissolution kinetics. However, this modeling study was carried out in part to inform a field study carried out in Yaquina Bay. Hence, we wanted to use dissolution kinetics directly measured on material that is being applied. The rate laws we used are commonly used approximation in early diagenetic models, and both rate constant and exponent derived from our data are in line with other reports. Thus, while we do not argue with the fundamental argument made by the reviewer, well documented by the literature they cite, we feel that our modeling approach is substantiated by both our data and has support in the existing literature.

However, we now discuss this in more detail. Specifically we now cite the suggested papers, and we state that the rate order is subject to debate and may change over time. We now also mention that this has implications for the long-term pattern for the buffering effect, and hence is identified as an important factor to consider with making predictions over the course of several years. Added the following after the Sensitivity analyses part where we talk about the limitations of our approach:

'We also acknowledge that changing the dissolution rate order would potentially lead to different outcomes which need to be explored in further studies. '

And as suggested, we added dissolution constants from 'Results' to Table 4.

CDR estimates: The authors calculate atmospheric CO_2 uptake as equivalent to ½ the sediment alkalinity flux resulting from $CaCO_3$ dissolution. This is not accurate for several reasons, first, the overlying water will be different from porewater (e.g., temperature, pH, etc.), requiring further thermodynamic considerations to assess CO_2 concentration in the water column. Second, a decrease in water column CO_2 does not necessarily translate to a 1:1 decrease in atmospheric CO_2, given air-sea flux time and potential subduction of water masses prior to equilibration. This is difficult to model. Third, the dependence on "unpublished data" to calculate CO_2 uptake (L 222) is problematic. If these data are available from the author team, why not include them in the paper or supplement? Furthermore, there are no uncertainties or temporal variability given, making any extrapolation dubious. While the presented estimate is a nice back of the envelope calculation, it is insufficient to present as a significant part of the paper's discussion, and should include many more caveats. Because of the public and commercial interest in CDR, studies like these should be particularly thoughtful and careful about making claims of CO_2 capture rates.

We accept that the model provides coarse estimates and demonstrate the potential link between sediment alkalinization and CO_2 uptake. The simplicity of our approach was based on previous work (Brenner et al., 2016). With regard to the specific issues raised: 1) We consider differences in thermodynamics (different sediment vs overlying water temperatures) however, and we also stress that the tidal exchange would dominate over these small changes. 2) We agree that CO_2 exchange is complex and not instantaneous. However, we also note that this is a shallow well-mixed shallow water environment. 3) We only use the unpublished data to estimate a reasonable system state for our rough flux estimates. We agree that more accurate estimates need to be supported by more extensive data.

In our revisions, we are clarifying these points, but we feel that establishing the link between benthic exchange and the fate of carbon fluxes is still a valuable point to make.

Other model concerns: the initial sediment conditions are not fully presented. Is there any initial $CaCO_3$ in the sediment? Or $CaCO_3$ deposition? Even a small amount of $CaCO_3$ can have a significant buffering effect, and adding $CaCO_3$ may not necessarily change alkalinity generation linearly. If Yaquina Bay sediments are 0% carbonate, that should be explained. Furthermore, seasonal variability is not considered. I imagine that changes in temperature / OM dissolution rates are important if considering year+ trends, but these are not considered or discussed.

Upper boundary conditions are presented in Table 1. In our model, we consider zero deposition flux of carbonate minerals to the sediment. This potentially does not reflect Yaquina conditions directly. However, considering average annual conditions we think the heterogenous presence of $CaCO_3$ would still be small compared to the relatively large mineral addition we are implementing. Seasonal dependencies and other factors that might impact our model results were included as limitations of the model in lines 405-425.

Readability: I found it difficult to follow both the study's motivation and methods, given some writing issues. The paper needs some significant editing, in overall structure, sentence structure, and word choice. I recommend having the revised manuscript reviewed by someone with scientific editing skills.

Clarified the manuscript in the light of comments.

Significant issues / clarifications needed:

The combination of existing data from an unpublished thesis (Myers, 2022) and the new sediment model is confusing as presented. The conclusion starts with "Our study successfully combines lab experiments and modeling". However, the lab data consist of only 4 data points (used to parameterize CaCO3 dissolution rates), which were not particularly critical to the model, given the availability of published equations for dissolution kinetics (e.g., Adkins et al, 2021). As written, the paper takes a middle ground where much of the methods from Myers, 2022, are presented as new, but important details are also skipped over with a reference (which made it seem as if Myers, 2022, was a published study). Specific examples:

Description of the mineral dissolution kinetic experiments (L 85 to 106).

We appreciate this valid point that the paper is model dominated and there is not much experimental focus. Therefore, our approach uses minimal field/experimental data (such as dissolution and TA flux data that are consistent with previous work) to anchor a computational model where computational experiments were used to answer research questions.

L 183: boundary condition for TA flux. This is a very important value, and is mentioned casually without much explanation.

the alkalinity flux value mentioned here is the measured alkalinity flux from lab experiments. (see lines ~195)

L 184: Why is the remineralization rate altered to match the TA flux? What about mineral precipitation/dissolution, and oxidation of reduced species? Also, need to explain what the rates / rate ranges are, instead of just saying they they're comparable to other studies. (E.g., Krumins 2013 provides values for both shallow banks/bays, and also deeper coastal sediments; why are you only comparing to the deeper sediments here?)

We use literature values to parameterize our model. However, to anchor our model to the site, we used measured alkalinity fluxes to parameterize OM rates which were comparable to values presented in a study in the same system (D'Andrea & Dewitt, 2009). The reviewer is of course correct that alkalinity fluxes not only depend on the mineralization rates, but also the partitioning of the pathways, and the extent of reoxidation reactions. Because these are part of the model, we feel that alkalinity fluxes can be used to tune the organic matter mineralization rate driving early diagenesis. We also added the range of OM mineralization from D'Andrea & Dewitt (2009) and clarified the sentence so that the other studies we compare our values to (Wang & Van Capellen, 1996; Krumins et al., 2013) are all from deeper environments.

L 235: "This [dissolution rate] is in line with previous reactive transport models focused on mineral dissolution." I find it strange that you're justifying your choice of dissolution rate equations by comparing to other RTM models (which surely cite other papers for their rate equations), instead of studies focused on CaCO3 dissolution. Furthermore, these references are all quite old, and progress has been made on constraining CaCO3 dissolution rates in the meantime.

We appreciate the concern and to address that, we reworded 'previous RTM models' to highlight that we are actually focusing on the data synthesis carried out by those studies.

The different model runs are not clearly explained or outlined anywhere. Perhaps there should be a table of the initial parameters, as well as subsequent tests. Examples:

L 202: I had no idea what the parameters used in the spin-up simulations were, and they're only mentioned as an "(e.g., ...)". They should be explicitly listed in a table somewhere.

Reaction parameters used in spin-up were presented in methods section. We also present baseline scenario parameters in Table 5.

L 217: surficial mixing was "varied" from 4 cm to 10 cm. Is this only two depths/tests? Or was a range of values tested?

We only considered 2 depths and we justify our selection of these values based on literature.

The different CaCO3 addition scenarios (8%, 16%) are not clearly discussed in the text. E.g., if 8% CaCO3 addition is the "baseline", it should be identified as such earlier (L 200). Which scenario is represented in the Results, Discussion, and Figs?

We appreciate the suggestion and added a table addressing the above points (see Table 5).

Presentation of both the experimental observations and model results is confusing. E.g., Section 3.1 presents measured data, but immediately follows the lengthy model methods description. You need to more clearly specify "experiment" and "model" results.

We made changes to the overall flow of the paper which we think might help with this issue.

Mixing discussion points about calcifying organisms into both the Methods and Results section is distracting. Example: L 288.

Removed sentence to make it clear.

Minor comments:

Throughout: abbreviations are not used consistently (e.g., "rate of OM mineralization" could be "R_C," or included as a reminder, e.g., "rate of OM mineralization (R_C)" (L 210)).

Not all variables are defined.

L 149 (Equation 4): What is s?

It is defined as the stoichiometric coefficient for each species

Subsequently R_C and R_C0 are used without definition.

Defined in Table 3 caption.

Table 3: What is RC? Is it supposed to be R_C?; What is R_M?

R_C and R_C^0 were defined in Table 3.

Table 4: kfm and kdm are not defined

Dissolution rates were presented in section 3.1. Both dissolution and formation rate constants are added to Table 4.

L 108: the 'volumetric dissolution rate' is confusing following directly after the Equation 1. It should be a new paragraph.

Done

L 110: The jump from equation 2 to equation 3 is not obvious and needs to be explained better.

We revised this part for clarification.

L 127: What do you mean by "validation samples"? How were these used, and what were they compared to?

We used the TCO2/PCO2 samples to ensure our seawater treatments were correct and to validate all the alkalinity titrations. The bottle samples were simply another layer of data checking. We removed 'validation samples' for clarification.

L 180: "other acids/bases" needs to be explained

We removed this equation since the information we want to present already exist in Table 3.

L 184: RC0 is not yet defined.

Defined in previous sentence.

L 201: "respectively" does not make sense here; not needed

Removed.

L 267: aerobic respiration consumes alkalinity, so saying it produces "significantly less TA" is confusing.

Removed.

L 359: "reached similar values" is unclear. Similar to what?

Clarified as: '...under all scenarios, mineral saturation states reached similar values...'

L 372: Specify that A is Omega_calcite. Also, caption should not say "2 years after" since data are for three timepoints

Removed '2 years'. We kept Omega as is since it represents saturation state for the mineral type added.

L 415: "Loosing"

Changed to 'washing away'

L 428: Reword "... in the bay most."

Removed 'in the bay'.

L 430: "Increase of CO2 uptake" is not accurate. The sediment is still a net source of CO2; "decrease in CO2 flux" would be closer to accurate

Changed as suggested.