

Point by point edits - Referee #1: Gunter Flipkens, gunter.flipkens@uantwerpen.be

The authors have put substantial effort into rewriting the manuscript and addressing the reviewers' comments. Therefore, I only have a limited number of minor comments on the corrections, which are listed below.

Line 21: Add “these” after “but” for grammatical improvement.

We edited accordingly and it now reads: “Mg(OH)₂ flakes were observed but **these** eventually redissolved in the bottles exposed to advection” (line 21)

Line 30: The conversion “1 Gt CO₂ = 10¹⁵ g CO₂” can be omitted. Readers can look up unit conversions themselves. You would also not state that 2350 μmol kg⁻¹ corresponds to 2.35 × 10⁻³ mol kg⁻¹, for example.

We understand and removed it as suggested.

Line 32: Add “the question” before “whether”.

We edited accordingly and it now reads: “However, **the question** whether added alkalinity remains in the surface ocean...” (line 32)

Line 39: Is the 500 μmol kg⁻¹ threshold only applicable under average ocean conditions, or is it also valid for regions with elevated temperature and/or salinity? Please briefly clarify this in the sentence.

Available research reporting on the 500 μmol kg⁻¹ value result from laboratory studies at room temperature (20-25 °C). They mostly used natural seawater (salinity ~35), though some also used lower salinities. We therefore cannot safely say whether it is applicable under other conditions than average open ocean conditions and refrained from saying so. Furthermore, we cannot safely state that this is a threshold which is why we avoided using this word, but this value has been repeatedly used as ballpark number where unwanted secondary formation occurs. We edited the text as suggested and it now reads: “**Laboratory** studies using more reactive solid feedstocks such as calcium hydroxide (Ca(OH)₂) and magnesium hydroxide (Mg(OH)₂), **mostly conducted in natural seawater at room temperature**, have revealed CaCO₃ precipitation once alkalinity was increased by ~500 μmol kg⁻¹” (lines 37-39)

Line 48: Add “formation” after “inhibit CaCO₃”.

We edited accordingly (line 48)

Line 53: Since this is a newly introduced term in the OAE field, I think the definition of the CAP could be formulated somewhat more sharply. I would suggest something along the following lines:

“This transient period following alkalinity addition, during which carbonate chemistry remains substantially perturbed relative to ambient conditions, potentially promoting secondary mineral

precipitation and affecting biological processes, is hereafter referred to as the critical alkalinity period (CAP).”

I think explicitly stating that the perturbations are relative to the background conditions, as well as mentioning the potential implications of the CAP (i.e., secondary precipitation and adverse effects on biological processes), would make the definition clearer.

We agree and have incorporated the suggestions. It now reads: “This transient period following alkalinity addition, during which carbonate chemistry **remains** substantially perturbed **relative to ambient conditions promotes secondary mineral formation, may affect biological processes**, and is hereafter referred to as the critical alkalinity period (CAP)” (lines 53-55)

Line 51: I feel that the comment from Reviewer 3 regarding biological risk has not yet been fully addressed. As far as I understand, Bach et al. (2019) discusses changes in the overall water-column carbonate chemistry rather than the diffusive boundary layer (DBL) surrounding feedstock particles. These represent different exposure scenarios and may therefore entail different ecological risks. A transient 500 $\mu\text{mol kg}^{-1}$ increase in TA within the microenvironment surrounding a dissolving particle is a very different exposure scenario from a 500 $\mu\text{mol kg}^{-1}$ increase throughout a large water parcel.

I therefore reiterate Reviewer 3’s question: What is the biological risk associated with perturbed microenvironments around feedstock particles? My understanding is that the risk is likely to be relatively low, except perhaps for particle-associated microorganisms that may experience these conditions directly. Please rephrase this sentence to clearly indicate whether you refer to biological effects resulting from changes in bulk water-column carbonate chemistry or those associated with changes within the DBL surrounding feedstock particles.

We understand the comment and acknowledge that our reformulation did not efficiently explain our train of thought. Microenvironments around particles are not primarily affecting biology, bulk seawater conditions are. We have reformulated to clearly differentiate the risk for CaCO_3 formation created by micro-environments around particles from the environmental pressure on biology created from changes in bulk seawater. It now reads: “Overall, the persistence of high alkalinity and ΩA **microenvironments around dissolving particles** is expected to enhance the likelihood of CaCO_3 formation, particularly through secondary precipitation in the diffusive boundary layer of dissolving particles or on suspended solids (Moras et al., 2022). Also, the concomitant high pH and low CO_2 **bulk seawater conditions resulting from alkalinity addition** may impact biological processes because CO_2 is essential for photosynthesis (Bach et al., 2019).” (lines 49-53)

Section “TA manipulations and sampling procedure”: I would briefly reiterate here that two sets of experimental bottles were used for static and advected conditions, respectively. I would also mention this in the header of Table 1 to make the experimental setup clearer.

We agree and edited as: “Each **of the two** 42-bottle experimental sets” (line 125) and “Experimental design for one **of two sets** of 42 bottles” in Table 1 header (line 141)

Line 227: Remove “after six months” and add “Furthermore,”. Additionally, add “at the end of the experiment” after “and mineral analyses” in line 228. This would improve clarity. Otherwise, the current wording could suggest that the TA budget was derived only after six months, whereas I understand that it was derived from the data obtained at the final sampling point.

We edited as suggested (lines 230-231)

Lines 239–241: The sentence states twice that TA generation was higher in the advected bottles than in the static bottles. Please make this more concise.

We understand and removed the first sentence. It now reads: “Stark differences emerged in realised TA based on the material used and the advection regime within the first 24 hours. In the advected bottles, TA generation was higher, with %TA_{realised} values that were 2.5 ± 1.0 and 4.3 ± 2.1 times those in the static treatments for $\text{Ca}(\text{OH})_2$ and $\text{Mg}(\text{OH})_2$, respectively.” (lines 241-243)

Line 262: I would move the reference to Figure 3 to the first sentence of this paragraph, since this sentence already presents the main results shown in the figure. The same applies to the first reference to Figure 2 in the first Results section, “Differences between expected and measured alkalinity using solid feedstocks.”

We have moved the reference to Figure 3 as suggested (line 265) and moved the reference to Figure 2 to the second sentence as the first does not refer to the figure yet (line 241).

Line 296: “Respectively” suggests that two different ranges or values are being given for the two feedstocks ($\text{Ca}(\text{OH})_2$ and $\text{Mg}(\text{OH})_2$), which does not appear to be the case. I would therefore remove “respectively” and instead explicitly state when the lowest pCO₂ was observed (i.e. after 1 day).

We agree and removed “respectively” (line 298)

Line 298: Please add a reference to Figure 7 here, since the NaOH results are discussed in the text but are not shown in Figure 4, which is the figure currently discussed in this paragraph.

We have added it at line 301 as suggested.

Line 341: The k values have relatively large uncertainties due to variability in the experimental data. This uncertainty does not appear to be propagated into the calculated wind speeds (U), although I think it should be, or at least the variability in k should be acknowledged when discussing the calculated wind speeds. If you prefer to present only average values, I suggest mentioning this variability explicitly in the Discussion.

We understand and have propagated the uncertainty. It now reads: “yielded corresponding U wind speeds of 1.2 ± 1.4 and 3.0 ± 2.5 m s⁻¹.” (lines 343-344). This was also done later (line 589-590) and now reads: “estimated to be 1.7 ± 1.0 m s⁻¹ and 3.1 ± 0.7 m s⁻¹, respectively.”

Line 580: The calculated wind speeds here do not appear to match those reported in line 341. Please verify these values.

We understand the question, but this is intentional. The start of the section “Implications of the CAP duration for CO₂ uptake and environmental impacts” discusses the role of CaCO₃ precipitation on the estimates of CO₂ fluxes. This part of the discussion reviews the calculation performed earlier and now introduces a correction accounting for CaCO₃ precipitation (as suggested by Reviewer 3). The new values are now referred to as “corrected fluxes” (line 583) and “corrected equivalent wind speeds” (line 589). Accounting for CaCO₃ precipitation reveals higher values and lower uncertainties.

Lines 407–409: These sentences could be formulated more clearly. In the current version, the second sentence seems to imply that a thicker DBL reduces the concentration gradient driving diffusion and lowers particle dissolution rates. I do not think this is correct. A thicker DBL would primarily reduce the diffusive flux of dissolution products away from the particle surface, potentially resulting in higher concentrations of dissolved species at the particle surface (i.e. not a lower concentration gradient as mentioned in the text). This could, in turn, increase the local saturation state and reduce the thermodynamic driving force for further particle dissolution as identified by the authors. Please revise these sentences to clarify this mechanism.

We agree with the comment, and this is what we wanted to express. It has now been edited and reads: “The reduced advection could have increased the thickness of the diffusive boundary layer surrounding particles, **reducing the diffusive flux of dissolved products away from the particles surface**. This in turn could have created micro-environments enriched in Ca^{2+} , Mg^{2+} and OH^- , increasing local saturation state and ultimately decreased dissolution rates.” (lines 413-416)

Line 411: “At 0 RPM, TA generation was up to 50% lower” — lower compared with what? Please specify the reference condition.

We understand and reformulated as follows: “At 0 RPM, TA generation was **consistently lower in the static conditions compared to the advected conditions**” (line 418)

Line 434: Add “There,” before “the maximum” to establish a clearer link with the preceding sentence.

We have edited accordingly (line 441)

Lines 456–460: I am still missing a more quantitative discussion of the importance of CO_2 ingassing for secondary precipitation. I think your calculated Ω values for the bulk solution would be useful to refer to here. In the 125 RPM treatment, for example, Ω decreases from approximately 4–5 toward the control value of approximately 2.5 somewhat more rapidly than in the 0 RPM treatment, although the difference does not appear to be very large.

Given that the difference in saturation state is relatively modest, my interpretation is that the effect of advection on the DBL is likely (much) more important than the effect of CO_2 ingassing for the risk and extent of secondary precipitation. If you agree with this interpretation, I suggest making the sentences revolving around the general conclusion I seem to get from this paragraph that faster CO_2 ingassing leads to substantially less secondary precipitation more nuanced.

We agree and have emphasised in the comment below that CO_2 ingassing but most importantly the larger DBL from weak or absent advection enhanced CaCO_3 precipitation.

Line 478: How could particle settling have affected CO_2 invasion? Wouldn't advection be the more direct factor affecting CO_2 invasion? Please rephrase this sentence.

We understand and did not express our ideas convincingly. We reformulated this end of section to also incorporate the comment above. It now reads: “In the absence of advection, however, particle settling was likely more important, which may have reduced dissolution, **increased the diffusive boundary layer around the particles and increased high saturation micro-environment at the bottom of the**

bottles. The smaller gradient of $p\text{CO}_2$ between air and water, coupled to the decreased invasion rate also likely enhanced the persistence of the CAP. Therefore, the buffering by CO_2 ingassing but most importantly the larger diffusive boundary layer resulting from the absence of advection eventually led to further CaCO_3 precipitation and lower final Ω_A values, i.e., from 1.8 to 2.6.” (lines 484-489)

Line 539: “Addresses” should be “addressed”.

Thank you for noticing, we edited accordingly (line 548)