Reviewer 1 (Charly Moras)

Overview:

The manuscript from Hashim et al. presents results from an OAE experiment conducted with liquid alkalinity (i.e., NaOH solution) during a research expedition from 2023. Using natural seawater, the alkalinity was increased using a 1M NaOH solution and the carbonate chemistry was measured throughout the experiment. Finally, the data explore the formation of CaCO3 and compare the results with available data from the literature.

Overall, the manuscript is very reader friendly. The setting, experimental design, results and discussion are easily understandable, and the data presented here match other data available from the literature. One very interesting aspect is that in the manuscript introduces for the first time (as far as the reviewer knows) the proposed carbonate chemistry sampling techniques from Schulz et al., 2023, available in the Guide to Best Practices in Ocean Alkalinity Enhancement Research. Furthermore, the authors focused on the mineralogy of precipitated material with continuous XRD analysis of the precipitated CaCO3. Finally, the data were fitted with other work available in the literature (Burton and Walter, 1987, Moras et al., 2022, and Mucci et al., 1989) which allow for an easy and effective comparison of the precipitation of CaCO3 under various seawater conditions. The reviewer is supportive of the publication of the research after discussing and/or addressing the various comments and questions below, and believes that after these minor revisions, the manuscript should be considered for publication.

> We thank the reviewer for his kind words, and we appreciate the useful and insightful feedback.

Comments:

Lines 37-38: I agree with the terminology "unseeded". However, I am questioning the term (pseudo)homogeneously. The use of unfiltered seawater (line 25) suggests that there may be some particles in suspension, which could have acted as seed. I think that the terminology should be slightly reviewed, emphasizing in the text that the term "unseeded" refers to the absence of CaCO3 seeds but that there might still be some resuspended particles that could have been used as precipitation nuclei for CaCO3.

> We generally agree with the reviewer and we will revise the sentence accordingly.

Line 38: I believe the right spelling would be "homogeneous" rather than "homogenous"

> Noted.

Line 40: I believe the word "correlated" was intended rather than "correlate"

Thank you. We agree and will make the change.

Line 85: for consistency, the TA unit should be reported as µmol kg-1 throughout the text

We will make sure the units are consistent throughout the manuscript.

Line 89: same as line 38

Noted.

Line 105: I rather use the term magnesium hydroxide here. While I agree that brucite is the mineral form of magnesium hydroxide, Mg(OH)2 can in some instances precipitate I an amorphous form which is not considered brucite. For ease, I would stick to magnesium hydroxide throughout the text

Noted. We will make sure to use the term "magnesium hydroxide" where appropriate. There are places in our manuscript where we specifically discuss minerals, their solubilities, saturation states, etc. In this case, we believe that the use of "brucite" is more appropriate but in other instances where we discuss precipitation in practical sense, we will make sure we use the term "magnesium hydroxide".

Line 115: I believe the sentence should read "… the ones that are more likely…"

We agree and we will change the sentence accordingly.

Line 132: were the incubated water in the bags exposed to any movement (floating around, boat rocking, etc.) or was it considered static? Such absence or presence of movement may have affected the CaCO3 precipitation kinetics and should be mentioned explicitly

Good point. We will add a sentence to explicitly state that the bags were floating and were allowed to move freely in the incubator.

Line 133: was the unfiltered seawater passed through a 1 or 2 mm mesh to get rid of bigger particles or was it fully unfiltered?

It was not filtered at all but given that we obtained the water from an open ocean setting, no sediment or large critters were present.

Line 147: I believe the standard notation for TA concentration is µmol kg-1 without the "." in between. May need to be edited throughout the text

Noted. We will make sure to update the units throughout.

Line 154: same as line 85; also, it would be beneficial to have a column with the measured ΔTA to show the maximum TA reached, as well as indicate whether there are some discrepancies (maybe from early CaCO3 precipitation after addition?)

We unfortunately didn’t measure the alkalinity immediately after addition. Accordingly, the first TA and DIC datapoints in each experiment are assumed rather than measured. We will explicitly explain this in the Method section.

Line 156: how was salinity measured? Because salinity does not have unit if measured on the practical salinity scale of 1978

The salinity was calculated from conductivity which was measured by the CTD. We will make sure to elaborate on how salinity was calculated.

Line 178-179: how exactly were the DIC samples taken? For stable DIC sampling, it is advised to sample the DIC in a borosilicate vial as described here using a peristaltic pump with the tubing placed at the bottom of the vial, and allowing at least half of the vial volume of overflow (Dickson et al., 2007). This section might need slightly more details.

The sampling for DIC was done using a plastic syringe and the sample was directly injected steadily into the borosilicate vial followed by poisoning with mercuric chloride. The whole procedure took less than 15 seconds and was done in a way that minimizes gas exchange. The use of peristaltic pump and overflowing were not possible as we wanted to avoid changing the volume of the experiments too much. We will make sure to add more details to this section.

Line 191: was the titrant ionic strength adjusted to match the samples’ ionic strength?

The titrant ionic strength was adjusted to match that of seawater but we did not consider the change in ionic strength as a result of alkalinity addition given that the change is small and variable due to precipitation.

Line 217: wouldn’t calculating the various Ω at 27 °C (line 132) instead of 25 °C more suitable considering the experiments were run at ~27 °C? Or are the differences negligible?

We believe that the difference is minor. Nonetheless, we will make sure to recalculate Ω using a temperature of 27 °C

Line 322: in caption, 3rd line, I believe there is a letter “r” missing, it should read “DIC decrease”

Thank you for pointing this out. We will correct this.

Line 336-337: the sentence reads that magnesite is both highly (Ω > 100) and moderately (Ω ~ 10) supersaturated. Please edit

Noted. We will edit this.

Line 354: here, the figure 3 is discussed. There is one pattern that I noticed and seems interesting to me. It appears that from figure 3, the aragonite A111 and A021 signals decrease at 8.8h and15.8h after TA addition. While I may not be the more familiar with XRD analyses, I would like to have some more details as to why there is such pattern? If all the XRD samples have been handled the same way, why is there a slight decrease at these points in time? Was CaCO3 precipitation halted during these times? Or is it only a sample artefact? I am not sure whether it is worth mentioning in the manuscript, but I would like to have the authors point of view on such pattern.

> Thank you for pointing this out and giving us the opportunity to elaborate on this. We believe that this is related to the higher amount of halite present in these two samples that makes the aragonite peaks smaller since the peak intensities reflect the relative percentage of minerals. Based on the Rietveld refinement results, amount of halite in the sample at 8.8h is 19% and the sample at 15.8h is 31% (see the associated Research data). These values are abnormally high and are likely related to residual of halite precipitated during filter drying. Rinsing the filters thoroughly with DI was challenging because the precipitate blocked the filters in most cases.

Line 372: see line 105 comments. It would be more justified to use the term magnesium hydroxide here as well

> Noted.

Line 388-389: if CaCO3 coated the Mg(OH)2 crystals, could this also explain why XRD did not reveal any? Even if the XRD covers the 2θ range of Mg(OH)2, if these are coated with CaCO3, the analysis result would only show CaCO3, right?

> We have thought about this possibility and made some calculations that consider the linear absorption coefficient, packing density, and sample crystal size, that revealed that the sample crystal sizes are too small and the XRD beam is capable of penetrating through them even if coating by other minerals was present. However, as you mentioned in other comments, the possibility that what precipitates is an amorphous $Mg(OH)_2$ and not brucite (mineral with a structure) could mean that the XRD would not be able to detect such as a phase. We will discuss this in our manuscript.

Line 402-410: this comment does not need to be addressed, but I wanted to highlight that I really appreciated the review of the methodology and the suggested work around this unexpected loss of DIC

> Thank you.

Line 426: same as line 38

> Noted.

Line 426-427: here, the work of Marion et al., 2009 could be used to determine a more accurate threshold for homogeneous precipitation given the experiment salinity and temperature

> Thank you for providing this reference. We will look into this and incorporate it into the manuscript as appropriate.

Line 433-434: same as line 38

> Noted.

Line 453: same as line 38

> Noted.

Line 463: same as line 38

> Noted.

Line 524-526: some works are available in the literature where they report on the inhibitory effect of various compounds under both natural and OAE setting, and could be considered as references in the manuscript (Chave and Suess, 1970, Moras et al., 2024, Pan et al., 2021, Pytkowicz, 1965)

> Thank you for sharing these references. We will incorporate these references into the manuscript.

Line 571: same as line 38.

> Noted.