REVIEWER 2

General comments

The manuscript addresses an interesting topic; the potential of ocean alkalinity enhancement in removing CO_2 using laboratory experiments. However, in its current form, the manuscript requires substantial revision before it can be considered for publication.

The introduction would benefit from a more comprehensive and critical engagement with the existing literature. At present, it does not sufficiently synthesize prior work or clearly articulate how this study advances the field. Expanding the literature review, deepening the discussion of competing models, and broadening the citation base beyond a limited subset of recurring studies would significantly strengthen the manuscript. Currently the manuscript relies on a relatively small subset of studies, including multiple self-citations.

Thank you for the comments. We understand that the introduction as of now is not sufficient to articulate the relevance of the study. We suggest adding more information in the paragraph to explain the relevance of such study, such as the concept of CAP and the idea behind the use of the circular advection: "Overall, the persistence of high alkalinity and Ω_A (micro-)environments will dictate the likeliness of alkalinity loss to $CaCO_3$ and potential impacts on biology. This period of time is therefore critical to the overall safe and effective implementation of OAE to limit negative impacts of OAE and maintain high CDR potential, which is later referred to as the critical alkalinity period (CAP). […] In a new approach, the dissolution of $Ca(OH)_2$ and $Mg(OH)_2$ in natural seawater were addressed both in static and rotating reaction vessels for 6 months. Circular advection regimes were chosen to try and represent open ocean conditions, while gas exchanges were allowed to occur during the experiments. Carbonate chemistry was monitored throughout, particle analyses were conducted at the end of the experiments, and gas velocities for CO_2 and equivalent open ocean wind speeds were derived, allowing direct comparison between laboratory data and global modelling studies."

Regarding the multiple self-citation comment, we understand the concerns of the reviewer. However, the few studies mentioned have been the first to test the suitability of the materials of interest in this manuscript, i.e., $Ca(OH)_2$, $Mg(OH)_2$ and NaOH, and therefore, the more relevant ones. Nevertheless, in an effort to address the reviewer comments, the choice of citation will be reviewed to incorporate other works such as:

- Hashim, M. S., Marx, L., Klein, F., Dean, C. L., Burdige, E., Hayden, M., McCorkle, D. C., and Subhas, A. V.: Mineral formation during shipboard ocean alkalinity enhancement experiments in the North Atlantic, Biogeosciences, 22, 7149-7165, 10.5194/bg-22-7149-2025, 2025.
- Yang, B., Leonard, J., and Langdon, C.: Seawater alkalinity enhancement with magnesium hydroxide and its implication for carbon dioxide removal, Marine Chemistry, 253, 104251, 10.1016/j.marchem.2023.104251, 2023.

More broadly, while the authors introduce new experimental variables that move in a promising and appropriate direction, the methodological rigor with which these experiments were conducted and documented is currently insufficient to clearly demonstrate advancement beyond previous studies. As presented, it remains unclear whether the modifications—such as extended experimental duration and altered mixing conditions—translate into meaningful new insights or improved constraints on the processes under investigation. Strengthening the experimental design, documentation, and quantitative analysis would help clarify the extent to which this study advances our understanding of these issues.

Thank you for the comment. We understand that the novelty may not be sufficiently discussed, and we suggest addressing this issue by emphasising in the discussion for example that the results of our experiments clearly differ from those reported in the literature with the same alkaline feedstocks. We also consider incorporating a figure representing the experimental setup to further enhance the clarity of the material and methods section.

Several aspects of terminology and conceptual framing require refinement. In multiple instances, terms are used in ways that may not align with their standard definitions. Improving precision in terminology and clarifying key concepts would enhance the scientific rigor of the manuscript. For example, “advection” and “laboratory stirring” are not the same thing. Similarly, phrases such as “due to the non-dissolution of CaCO3 in seawater” is both unclear and strange.

We understand the comment and agree that the terms may have been misinterpreted in the text. During the revision, we will ensure consistency when discussing advection. Regarding the second part of the comment, we will also revise the sentences accordingly and suggest: “as $CaCO_3$ does not dissolve naturally in open ocean seawater.”

The experimental design and quality control procedures also require clearer documentation. Additional methodological detail and justification would strengthen confidence in the robustness and reproducibility of the results. There are certain aspects of the methodology that left me wondering if these experiments were done in the most rigorous way possible. For example, I was shocked to read that that MilliQ was added to compensate for evaporation. Why MilliQ and not seawater? How much was added? No documentation of this effect is presented. There are also no controls to compare to. These are all serious problems.

We understand the concerns of the reviewer. We can ensure that the experiments have been rigorously conducted and that all available precautions were taken when handling the bottles. All treatments have been conducted in triplicate and the consistent trends between triplicates suggest that the results can be reproducible. We also believe that adding the schematic of the setup below may allow the reader to better visualise it.

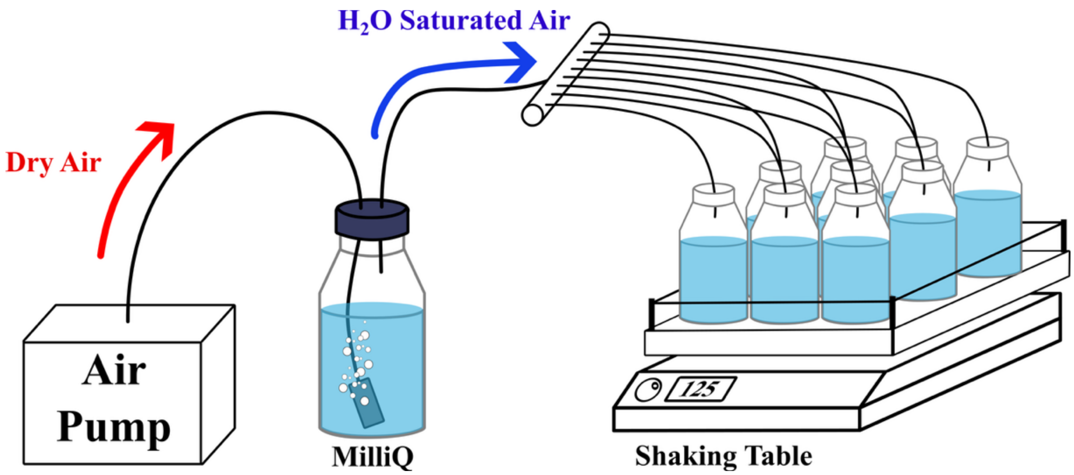

Figure A 1: Schematic of the experimental setup. An air pump is taking up dry air and pushes it into a closed Schott bottle where MilliQ is bubbled. The new H_2O saturated air is then carried to an air diffuser from which individual tubing are connected to each experimental bottle, constantly and equally supplying each open bottle with fresh atmospheric CO_2.

Regarding the MilliQ comment, we are not fully sure why the reviewer was shocked by such procedure and consider it a serious problem. Evaporation was unfortunately occurring as all bottles were kept open to the air. Preliminary tests revealed that the setup used for the experiments (with H_2O saturated air) was efficient at decreasing evaporation, which was estimated to be only about 1% of the weight per day. In the event of evaporation, which was controlled twice a week, spontaneous and precise addition of MilliQ was conducted. The weight of the bottle and water was recorded at the start of the experiment and after every sampling campaign. Hence, when a decrease in weight was observed due to evaporation, MilliQ addition was performed until the total weight was identical to the previously recorded one. Throughout the experiment, conductivity was also recorded to control for evaporation and MilliQ addition, and changes observed were negligible (~1%) and within the calibration standard certified precision of ± 0.5%.

During evaporation, the water "portion" of the sample is lost, i.e., H_2O, which concentrates the sample with respect to TA for example. The purest water available to us in our lab was MilliQ. We therefore proceeded with fresh MilliQ. If we were to have used seawater, the addition of alkalinity will ultimately lead to a more concentrated sample. Let's assume a sample with a TA of ~2480 µmol kg^{-1} (similar to day 1 of the $Mg(OH)_2$ experiment at 125 RPM and targeted ΔTA of 600 µmol kg^{-1}, figure 2). If this sample was to have lost 0.5% of its water content (deceasing from 1000g to 995g), the TA would now be 2480 x 1000 / 995 = 2492.5 µmol kg^{-1}. By adding seawater (assuming open ocean, 2300 µmol kg^{-1}), 5g of such TA will be added, yielding a new 1000g sample with a TA of: 2492.5 x 0.995 + 2300 x 0.005 = 2491.5 µmol kg^{-1}. The new sample TA is 11.5 µmol kg^{-1} higher than the starting value, which is therefore not the same as it was before evaporation. However, adding pure water without alkalinity would yield the original alkalinity before evaporation.

Another thing about the structure of the manuscript, I am unclear why the figures are placed in an Appendix rather than integrated into the main text. Presenting figures within the main manuscript would improve readability and allow readers to more easily follow the argument.

Thank you for the comment. However, we are puzzled by it as the submitted version already contains all relevant figures. There are 5 figures and 1 table in the main text, and only 3 figures in the appendix.

The section discussing wind speed and advection rate is a promising step toward connecting laboratory experiments with real-world conditions. However, this analysis currently feels incomplete. It would be valuable to fully develop this approach—potentially by solving the mentioned equations and presenting a figure that explicitly relates the experimental mixing conditions to realistic environmental scenarios. This would substantially strengthen the broader relevance of the study.

Thank you for pointing this out. The equations provided are those developed by Wanninkhof 2024 (based on decade-long and quasi-standard research based on the pioneering work in Wanninkhof 1992 with more than 5000 citations). The first equation for CO_2 flux is used in our case to derive the k value as the CO_2 flux F is calculated from our experiments (i.e., 21.3 ad 39.9 µmol kg^{-1} d^{-1}), K_0 is obtained from Weiss (1974) and the pCO_2 values are obtained from our experiments. Then using the calculated k values (1.4 and 2.7 cm h^{-1}, line 469), we can solve the second equation. In our case, we were after the U value, given that we have a Schmidt number at 24.6 ^{o}C as per Wanninkhof, R. (1992). Then, we end up with U values of 2.2 and 3.1 m s^{-1} (line 477).

Detailed comments

Line 15 The terms "needles" and "broccolis" are informal and not standard terminology for crystal morphology. Please revise using conventional mineralogical or crystallographic terms.

Thank you for pointing this out. However, the terminology "needle" and "broccoli" have been used several times in the literature, and especially by John W. Morse, who did extensive research on $CaCO_3$ nucleation in seawater. This is mentioned in his 2007 work "Calcium Carbonate Formation and Dissolution" (doi: https://doi.org/10.1021/cr050358j)

Line 37 The description of "runaway precipitation" needs clarification. This term typically refers to the extent of reaction and the resulting decrease in total alkalinity, rather than necessarily implying an increase in precipitation rate over time. Please clarify whether there is direct evidence for changing reaction rates.

Here we do not fully agree with the reviewer. The concept was named and described first in Moras et al., 2022. It is correct that runway precipitation refers to a decrease in total alkalinity over time, but it is also clearly explained that it refers to the acceleration of the decrease in TA resulting from $CaCO_3$ precipitation in a logarithmic-like fashion. We suggest adding a sentence in the manuscript advising the reader to refer to Moras et al., 2022 for further details.

How was it determined that two hours of UV exposure was sufficient to prevent microbial growth? Were control experiments performed? Providing more detail on these points would strengthen the methodological rigor of the study.

We agree with the comment. The use of UV exposure was only an extra step taken to prevent potential bacterial growth, though sterile filtration only would have been enough. The supplier suggests that 30min of exposure is sufficient for 99% killing efficiency. Furthermore, seawater disinfection with such UV-C lamp has been reported to be efficient (https://doi.org/10.1080/01919512.2012.722050).

Line 101 Please clarify the reference to "Carl Roth" (e.g., supplier name and location).

Thank you for pointing this out. Carl Roth is the supplier's name, we will edit as follow: "Carl Roth (Germany)"

Line 123 The values reported here appear to represent variability rather than analytical error. Were samples analyzed in replicate? Please report the standard deviation (or other relevant metric) of replicate analyses.

The values are calculated based on successive measurements of certified reference materials from Prof. Dickson. All these measurements conducted for this manuscript have been compiled and the error propagation calculated. The standard deviation of the repeated measurements yielded 2.4 µmol kg^{-1}, corresponding to an offset of ~0.1%.

Line 151 The discussion of quality control procedures is difficult to follow. Please clarify how analytical precision, accuracy, and reproducibility were assessed. I have no idea what the authors are talking about here.

We understand the confusion and thank you for pointing it out. We suggest reformulating the section to emphasise that three parameters of the carbonate system were measured throughout and compared with each other. This allowed us to prevent unexpected loss of data due to instrument issue or miscalibration. Alkalinity titration is a highly reliable method allowing for precise measurements of samples TA (0.1%, line 123) and accurate estimations once calibrated against CRMs. DIC measurement with a TOC-L/CPH analyser is also precise (3.5%, line 126) and accurate once calibrated with CRMs. pH measurement with a probe was used in parallel to track carbonate chemistry changes without having to samples water. While the pH data are not directly used, they allowed to reconstruct the whole carbonate system on top of TA and DIC measurements and were used for quality control purposes. For example, a stable TA and a decreasing pH should correlate with an increasing DIC. In the event where such pattern was under or overestimated, the cross-check with these three parameters allowed to identify potential issues. This was the case for DIC (lines 151-161). We will reformulate this section to incorporate such knowledge.

Line 290 The statement regarding previous studies and the use of stir bars may need revision. Not all prior studies relied on stir bars, and the use of a shaking table may not constitute a substantial methodological innovation.

We agree with the reviewer and will adjust the formulation to be less radical, such as: "Such an outcome was to be expected as the milder, yet likely more realistic approach of the shaking table, relying on water diffusion and mild vertical mixing compared to the vortex created by stir bars, decreases the dissolution effect and disturbance created with a magnetic stir bar."