

Dear Editor and Reviewers,

We would like to thank you for the time and work put into reviewing our manuscript. Following the open discussion, we have addressed your comments and edited the manuscript accordingly. As shown in the Track Changes version, we have put a strong effort into reviewing the procedures, rewriting hard-to-follow passages and improve the clarity throughout the text. We have also incorporated reviewer 2's comments and have moved all relevant figures from the supplementary materials to the main body. We also incorporated a new figure of the experimental setup and incorporated the grain size analysis suggested by reviewer 1.

We have provided all the reviewers' comments below from the open discussion and our specific answers and changes made to the manuscript. For each comment, we have added the corresponding lines in the Manuscript file.

We thank you for your consideration and if our changes are accepted, we are looking forward to the next round of comments and reviews from you.

With kind regards,

Charly A. Moras,

On behalf of all co-authors

REVIEWER 1

General comments:

This study assesses the long-term alkalinity generation and CO₂ ingassing following the addition of Ca(OH)₂, Mg(OH)₂, and liquid NaOH to seawater under slow advective and stagnant conditions. The experimental design and findings are novel and provide a valuable contribution to the growing body of literature on ocean alkalinity enhancement (OAE), particularly regarding the role of hydrodynamic conditions in determining alkalinity realization and CO₂ uptake. The results are relevant for informing implementation strategies and parameterization in larger-scale models. However, the manuscript contains numerous minor grammatical errors and some unclear sentence constructions, which make parts of the text, especially outside the Discussion, difficult to read fluently. In addition, the Materials and Methods section lacks a clear and logical structure, making it challenging to easily understand the experimental setup and workflow. I believe that moderate revisions are therefore required before the manuscript can be considered for publication in Biogeosciences.

Thank you for the time and efforts put into the review, as well as the good words. We will address the comments as suggested unless otherwise stated.

Specific Comments

Abstract & Introduction

Line 4: “Long term modelling studies have focused on this strategy, but most laboratory experiments focus on shorter term with strong advection...”. This sentence is strangely formulated. Shorter-term what? Experiments? Timescales? Please clarify and reformulate.

We understand the confusion. We reformulated as follows: “Long term implementation has been assessed in modelling studies, but most laboratory experiments focused on shorter timeframes with strong advection, which may not be representative of natural systems.” (lines 7-8).

Line 10: Replace “have been” with “were” to be grammatically correct, since the six-month experiment has presumably been terminated. The abbreviation TA is not introduced in the abstract. Please introduce total alkalinity before using TA.

Thank you, this will be done.

Line 14: What is the critical alkalinity period (CAP)? This is not a commonly used term in OAE research and should therefore be explained in more detail.

We understand. To address the comment, we rephrased as: “The concept of critical alkalinity period (CAP) is introduced here to represent such critical period of high alkalinity where unwanted secondary calcium carbonate (CaCO₃) formation may occur, significantly altering the CDR potential of the approach.” (lines 15-17).

Line 18: “Using NaOH revealed buffering by Mg(OH)₂ precipitation, which ultimately redissolved in the bottles exposed to advection.”. The meaning of this sentence is unclear. Please rephrase for clarity.

Thank you, we reformulated as: “In addition, when using NaOH as alkaline feedstock, $\text{Mg}(\text{OH})_2$ precipitation was observed but eventually redissolved in the bottles exposed to advection” (lines 20-21).

Line 19: “Finally, the bottles exposed to advection represented open ocean conditions with wind speeds of $\sim 3 \text{ m s}^{-1}$...”. This is a somewhat unusual concluding sentence. I suggest mentioning earlier in the abstract (around line 11) that slow advection represents open ocean conditions ($\sim 3 \text{ m s}^{-1}$ wind speeds). You can then conclude that advection is important to consider and that these data may serve as lower thresholds for inclusion in global models.

Thank you for pointing this out. We edited the abstract as requested by adding the following text earlier: “Bottles were either kept stagnant or exposed to slow circular advection, similar to open ocean conditions with wind speed of $\sim 3 \text{ m.s}^{-1}$ ” (lines 11-12). We also concluded the abstract as follows: “The study emphasises that water advection is important to consider and the data presented here may serve as a lower threshold for implementation in global models.” (lines 21-22).

Line 21: Either “carbon dioxide” is missing before (CO_2), or the brackets should be removed.

This is indeed a mistake from our side, we edited it as suggested.

Line 22: This sentence does not read fluently and is missing a verb before “the” and after “atmosphere.” I suggest: “While strong efforts to reduce CO_2 emissions are required, current projections report that these efforts are insufficient to slow global warming, with active CO_2 removal from the atmosphere being the only solution.”

Thank you pointing this out, we edited to: “While strong efforts to reduce CO_2 emissions are required, current projections report that these are insufficient to slow down global warming, with active CO_2 removal from the atmosphere being the only solution” (lines 25-27).

Line 26: Insert “seawater” before “total alkalinity” so it is clear to non-experts where alkalinity is added.

This is noted and was added.

Line 36: Does precipitation always lead to a runaway pattern? For example, if dilution occurs (leading to a decrease in Ω_a) after precipitation has started, would precipitation continue or stop? Please rephrase this statement more cautiously if necessary.

We agree with the suggestion, we therefore added a more cautious formulation such as: “Once precipitation starts, it may enter a runaway pattern where the precipitation speed increases, sometimes reaching TA values lower than the starting conditions” (lines 41-42).

Line 43: Since CAP is not a commonly used term in OAE research, I miss a clear definition of its meaning here.

We understand and reformulated as: “Overall, the persistence of high alkalinity and Ω_A (micro-)environments will dictate the likeliness of alkalinity loss to CaCO_3 , as well as high pH and low CO_2

conditions, potentially impacting biological processes. This period of high alkalinity 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).” (lines 46-50).

Line 48: Is the word “rates” missing after “dissolution”? The sentence reads awkwardly.

We noted the comment and edited to: “While these settings allow for easy and standardised comparison between the various dissolution rates of alkaline minerals, and possible precipitation of CaCO_3 , they usually omit the role of CO_2 ingassing on the stability of alkalinity.” (lines 55-57).

Line 49: “More important” is not the right word here. I believe “stronger” would be more appropriate.

Thank you, we edited accordingly.

Lines 50–54: The research aim is not explicitly mentioned. Please clearly state the objective of the study. The rationale for selecting a specific rotation speed would likely fit better in the Materials and Methods section.

We understand and reformulated as: “Here, in a new approach, the dissolution of Ca(OH)_2 and Mg(OH)_2 in natural seawater was addressed both in static and advected reaction vessels for 6 months. Circular advection regimes were chosen to try and represent open ocean conditions with vertical diffusion but limiting surface tension disturbance, 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.” (lines 59-64).

Materials and Methods

The Materials and Methods section starts in a very unconventional way, with the duration of the experiment mentioned in the first sentence and the actual setup described only later. Please rewrite this section in a more logical, chronological order reflecting how the experiments were conducted. This will make it easier for readers to understand the experimental setup, sampling, and analyses. Furthermore, adding a figure of the experimental setup would improve clarity.

We understand the reviewer comment. We edited accordingly by first introducing the setup from line 67 to 79 (type of bottles, shaker, temperature and RPM selection). We then proceed with the preparation of the experiments from line 80 to 88 (bottles cleaning, seawater preparation and equilibration). The section finishes by highlighting forecasted issues that may have arisen during the experiments, such as evaporation, and how to control it, from line 89 to 102. A figure was also added.

Line 58: Define “room temperature.” I see it is mentioned again in line 132. I suggest combining this information so that all relevant details are presented together.

Thank you for pointing this out, this moved it up and mentioned it in the first lines of the section (lines 66-67).

Line 59: Why was circular motion used? Does this represent seawater movement under natural hydrodynamic conditions? Please clarify.

Thank you for pointing this out. We have introduced the concept of Reynolds number which gives information on turbulence, and edited to: “Similarly, the tests revealed that a circular motion was more appropriate to resuspend the dye without breaking water tension. Using linear motion, the dye resuspension was insufficient, and the water tension was significantly disturbed compared to the static bottles. Chosen conditions yielded a Reynolds number $\sim 1.6 \cdot 10^4$, above the threshold considered for turbulent systems (Thorpe, 2007). Such setup was then used for a total of 6 months to assess the long-term response of alkalinity addition to seawater.” (lines 75-59)

Line 68: A small schematic of the experimental setup would be useful for clarity.

We understand. The schematic below was added in line 104.

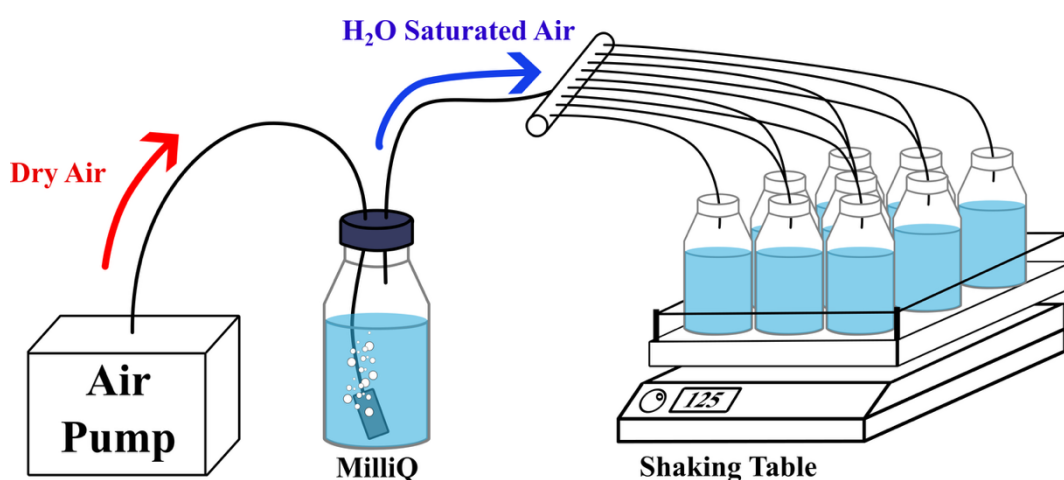


Figure 1: Schematic of the experimental setup. The air pump takes up dry air and pushes it into a closed Schott bottle where MilliQ is bubbled. The newly H₂O saturated air is then carried to an air diffuser from which individual tubing are connected to each experimental bottle, constantly and equally supplying each open bottle with fresh air equilibrated to atmospheric CO₂.

Line 72: There should be a space between “90” and “L.” Please ensure consistent spacing throughout the manuscript.

Thank you for pointing this out. This has been noted and edited throughout the text.

Lines 85–86: This sentence is unclear. Was MilliQ only added if the difference in weight was between 5 and 10% (and not if higher or lower)? Please clarify.

We understand the confusion, we reformulated as: “If a difference in weight was observed to be higher than 5% after these 2-3 days, a precise amount of fresh MilliQ was added by weighing to compensate for the slight evaporation.” (lines 96-98)

Line 88: What were the light conditions? Was the experiment conducted in the dark or under normal laboratory light? Please specify.

This is a good point, we added: “Hence, experiments were mostly conducted in the dark except during sampling where laboratory lights were tuned on.” (lines 99-100)

Line 90: Change “organisms” to “microorganism.”

This was edited in line 102.

Line 100: “Powders were sieved to allow for direct comparison.” Even after sieving, the powders can still differ significantly in grain size distribution and reactive surface area. Please elaborate on this limitation.

Thank you for the comment. To tackle this limitation, we have sent the samples for grain size distribution analysis which we will add to the manuscript as soon as the results are available and if those are of relevance to the interpretation. The powders revealed similar D50 (31.98 and 29.79 μm , respectively) and specific surface areas (59.90 and 62.82 $\text{m}^2 \text{kg}^{-1}$, respectively). The following was added: “The $\text{Ca}(\text{OH})_2$ and $\text{Mg}(\text{OH})_2$ powders were analysed for grain size distribution with a Betsizer S3 Plus, exhibiting similar D50 values, i.e., 31.98 and 29.79 μm , respectively, as well as comparable specific surface areas, i.e., 58.90 and 62.82 $\text{m}^2 \text{kg}^{-1}$.” (lines 120-122)

Line 108: Write dissolved inorganic carbon (DIC) in full before using the abbreviation.

This was edited as suggested (line 128).

Line 109: Does “3.3” refer to the type of TOC vials? The manufacturer name is typically mentioned in brackets. Please apply this consistently throughout the manuscript.

The 3.3 refers to the type of borosilicate, which is named “borosilicate 3.3”.

Line 138: This is already a result and should not be included in the Materials and Methods section.

We understand and reformulated as: “Secondary mineral formation was expected and suspected to occur on the surface of added material, i.e., $\text{Ca}(\text{OH})_2$ and $\text{Mg}(\text{OH})_2$.” (lines 159-160)

Line 144: How does Ca and Mg analysis of the water help determine whether undissolved $\text{Mg}(\text{OH})_2$ (solid phase) could have acted as a substrate for CaCO_3 precipitation? Please clarify.

We understand and added: “In the case where CaCO_3 would have precipitated on yet to be dissolved $\text{Mg}(\text{OH})_2$ particles, a clear signal of Mg^{2+} would be observed. Otherwise, if CaCO_3 precipitated from the water column only, the Mg^{2+} signal would be absent.” (lines 166-168)

Line 145: Why was the remaining water volume weighed and dried in a 50-mL Falcon tube? I assume this was to estimate the precipitated CaCO_3 amount, but please explicitly state this. Were filters rinsed with MilliQ before drying? Please elaborate.

This is a good point, we added more details: “The weighing of the remaining liquid allowed to determine the exact amount of seawater filtered for the analysis, which is required to report on the concentration after analysis.” (lines 169-171)

Line 151: Quality control for what? To compare measured and calculated carbonate system parameters? Please clarify.

We understand the question and added: “Throughout the experiments, quality control of the carbonate chemistry measurements was conducted using the TA, DIC and pH data from each set to prevent loss of data to unexpected instrument misbehaviour.” (lines 179-180)

Results

Figure 1: Instead of showing only average values, also display variation (standard deviations). Was one day expected to be sufficient to dissolve $\text{Ca}(\text{OH})_2$ and $\text{Mg}(\text{OH})_2$ under stagnant or rotation conditions? Perhaps the percentage is higher after several days?

The figure was adjusted accordingly in line 215:

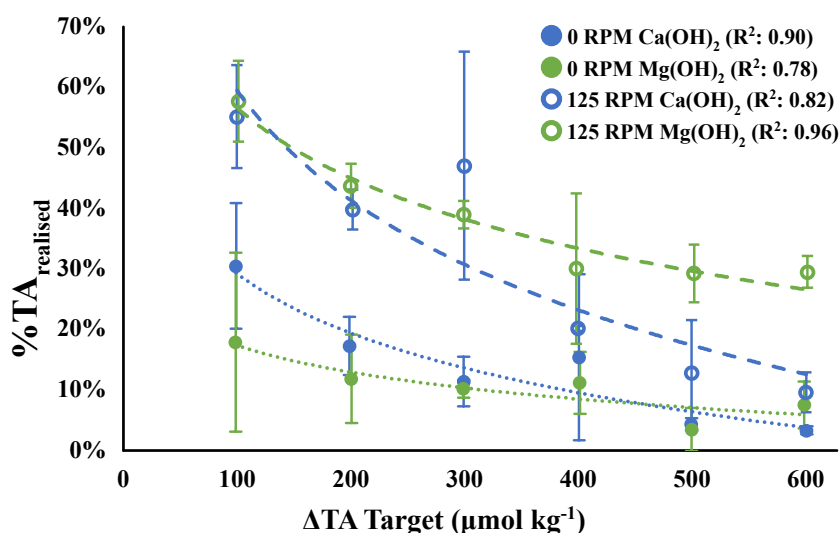


Figure 1: %TA realised after 1 day, i.e., maximum ΔTA measured, as a function of the ΔTA target (in $\mu\text{mol kg}^{-1}$). Each point represents the average of each triplicate, with blue markers representing the $\text{Ca}(\text{OH})_2$ treatments and the green markers representing the $\text{Mg}(\text{OH})_2$ treatments. Dotted lines represent logarithmic fits of the 0 RPM treatments while the dashed lines represent the logarithmic fits of the 125 RPM treatments. Corresponding R^2 are provided in the legend.

Line 160: Space missing between “125” and “RPM.” Please check spacing throughout.

This was noted and edited throughout the manuscript.

Line 168: After “%TA realised,” “which was” is missing.

This was added in line 202.

Line 173: The statement comparing $\text{Mg}(\text{OH})_2$ and $\text{Ca}(\text{OH})_2$ TA generation without advection needs reformulation. $\text{Mg}(\text{OH})_2$ values (3–18%) appear mostly lower than $\text{Ca}(\text{OH})_2$ (3–30%) relative to expected TA increase. $\text{Mg}(\text{OH})_2$ seems only significantly higher than CaCO_3 in at most half of the treatments (100, 200, 400 $\mu\text{mol kg}^{-1}$). Please revise accordingly.

We do not fully agree here. In fact, figure 1 clearly shows that the plain green data points ($\text{Mg}(\text{OH})_2$, no advection) are lower than the plain blue data points ($\text{Ca}(\text{OH})_2$, no advection) in nearly all experiments, i.e., ΔTA targets from +100 to +500. The only target with a higher %TAre realised with $\text{Mg}(\text{OH})_2$ and no advection occurred with a target of +600.

Line 187: “Overtime” should be “over time.”

This is noted and edited as suggested (line 221).

Line 188: Add standard deviations to the 100 and 180 $\mu\text{mol kg}^{-1}$ averages and to other averages throughout the text.

This was edited as suggested (lined 222-223).

Line 197: Provide actual numbers instead of “slightly negative ΔTA .”

We understand and edited accordingly (line 231).

Line 199: Add “values” after “positive.”

We understand and edited it accordingly (line 233).

Line 202: The “l” is missing in “slight.”

Thank you for pointing this out.

Line 203: Provide exact values instead of “about 100 $\mu\text{mol kg}^{-1}$.” Also, error bars are large after one month in the 600 $\mu\text{mol kg}^{-1}$ treatment. Is there actually a statistically significant decrease compared to day 6? Statistical analysis would strengthen this section.

This is noted and edited accordingly (line 237). Regarding the error bars after one months in the 600 $\mu\text{mol kg}^{-1}$ treatment, we are not sure what the reviewer means but assumed it was for the bottles exposed to no advection. There is indeed a decrease compared to day 6, both with $\text{Ca}(\text{OH})_2$ and $\text{Mg}(\text{OH})_2$. While we agree with the statistics comment below, we are not convinced a statistical analysis here will change the interpretation. In our case, we are mostly interested in the pattern and the clear difference following varying alkalinity addition rather than a statistically significant change. Nevertheless, we have conducted a statistical test and included it in the text (see below).

Line 215: “In most cases...” — this sentence needs improvement; it does not read fluently and contains errors.

We understand the point and edited to: “In most cases, the decrease in pCO_2 was correlated to the targeted ΔTA , i.e., higher targeted ΔTA led to lower pCO_2 . This was the case in most treatments except for the 0 RPM $\text{Ca}(\text{OH})_2$ experiment, where the lower pCO_2 was observed in the +400 $\mu\text{mol kg}^{-1}$ experiment.” (lines 249-252)

Line 220: The trend in pCO₂ values for 0 RPM Mg(OH)₂ treatments appears very similar to 0 RPM Ca(OH)₂ treatments. I do not agree with the current interpretation. Statistical analysis would help reduce subjectivity.

We agree with the comment and have run a simple t test, to identify from which time point the treatments pCO₂ were not statistically significantly lower than the control pCO₂. We created a p-value table (see below) that was added to the appendix of the manuscript.

Table 1: Compiled p-values from a two samples t-test to determine whether a given treatment pCO₂ is statistically significantly lower than the control pCO₂ for a specific day. The treatments statistically significantly lower with a confidence level of 95% (p-value < 0.05) are bolded, and those with a confidence level of 90% (p-value ≤ 0.1) are indicated by a *.

0 RPM	Ca(OH) ₂						Mg(OH) ₂						NaOH
ΔTA	100	200	300	400	500	600	100	200	300	400	500	600	600
Day 1	0.02	0.01	0.00	0.00	0.00	0.00	0.01	0.01	0.01	0.01	0.01	0.00	0.00
Day 3	0.02	0.01	0.01	0.00	0.00	0.00	0.07*	0.02	0.01	0.00	0.00	0.00	0.00
Day 6	0.27	0.06*	0.33	0.60	0.00	0.05	0.33	0.10	0.01	0.01	0.04	0.00	0.00
Day 35	0.07*	0.04	0.04	0.02	0.02	0.04	0.02	0.01	0.01	0.01	0.01	0.01	0.00
Day 69	0.06*	0.32	0.17	0.24	0.30	0.56	0.29	0.84	0.39	0.80	0.45	0.79	0.03
Day 108	0.57	0.52	0.88	1.00	1.00	0.83	0.53	0.78	0.71	0.88	0.87	0.17	0.52
Day 174	0.32	0.95	0.96	0.99	1.00	0.99	0.97	0.99	0.95	1.00	1.00	0.99	0.16

125 RPM	Ca(OH) ₂						Mg(OH) ₂						NaOH
ΔTA	100	200	300	400	500	600	100	200	300	400	500	600	600
Day 1	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00
Day 3	0.68	0.60	0.97	0.76	0.21	0.17	0.79	0.29	0.00	0.00	0.00	0.01	0.00
Day 6	0.75	0.73	0.75	0.55	0.64	0.81	0.75	0.78	0.74	0.66	0.77	0.68	0.60
Day 35	0.31	0.34	0.55	0.61	0.19	0.32	0.15	0.40	0.27	0.21	0.20	0.16	0.07*
Day 69	0.55	0.53	0.80	0.71	0.51	0.36	0.60	0.72	0.63	0.63	0.67	0.44	0.19
Day 108	0.13	0.06*	0.05*	0.04	0.13	0.26	0.16	0.16	0.01	0.03	0.34	0.10	0.87
Day 174	0.28	0.47	0.87	0.64	0.66	0.12	0.38	0.45	0.49	0.22	0.59	0.93	0.34

We therefore edited as follow: “With both Ca(OH)₂ and Mg(OH)₂, pCO₂ slowly increased over 1 month and remained statistically significantly lower than the control pCO₂ values. After 2 months, the difference was not statistically significant anymore (Table A1). Over the following 4 months, pCO₂ slightly varied, but followed the control batch fluctuations.” (lines 253-255)

Line 223: There is no day 2 data point; 24 h should likely be 48 h (day 3).

This is correct. We edited as: “In the case of Ca(OH)_2 , the pCO_2 values were not statistically significantly lower than those of the control incubations after only 72 h.” (lines 267-268)

Line 224: Please rephrase. For low alkalinity treatments, Mg(OH)_2 pCO_2 values were already similar to the control at day 3.

Thank you for pointing this out, we suggest being clearer as: “The Mg(OH)_2 incubations' pCO_2 also quickly increased, becoming non statistically significantly lower than the controls pCO_2 after 3 days for additions of 100 and 200 $\mu\text{mol kg}^{-1}$, and after 6 days for additions of 300-600 $\mu\text{mol kg}^{-1}$ ” (lines 258-260)

Lines 236 and 337: The first two sentences of this paragraph appear contradictory. Please rephrase.

We agree and edited to: “The NaOH experiments revealed differing patterns to those from the Ca(OH)_2 and Mg(OH)_2 incubations. While both setups are meant to be identical, a clear difference in ΔTA emerges” (lines 272-273)

Section: Effects of advection on pCO_2 changes: Statistical analysis would be useful to identify when pCO_2 values are not significantly different from the control.

We agree and have provided some statistical tests results above. We incorporatde them throughout the section as suggested. (lines 280-282)

Figure 2 caption: Replace “range” with “error bars.”

This was noted and changed to: “Each point represents the average of triplicates, with error bars representing the uncertainty based on the standard deviation of these three measurements.” (lines 245-246)

Lines 256–258: Move this calculation to Materials and Methods and explain how it was calculated.

We agree and moved it up in the text (lines 173-177).

Line 259: Add “more” after “about 194 $\mu\text{mol kg}^{-1}$.”

This is a good point and was edited accordingly (line 294).

Table 1: Consider whether this could be presented as a figure for improved clarity.

We have discussed further regarding such issue, and the table appears to be more suitable than a figure here (line 302)

Table 1 caption: Specify that it refers to the 600 $\mu\text{mol kg}^{-1}$ treatment.

This is a valuable point, we edited accordingly (line 304).

Line 261: Add the formula for estimated Ca^{2+} in Materials and Methods.

We agree and moved it up in the text (lines 173-177).

Lines 264–266: Improve fluency.

We agree and will considered keeping it much simpler, such as: “Finally, another major difference emerges from the measurements of Mg^{2+} , where the 0 RPM has nearly twice as much Mg^{2+} as the 125 RPM. Overall, the particles collected from the 0 RPM experiments were composed of $3.8\% \pm 0.4\%$ of magnesium, while the particles collected from the 125 RPM experiments, were about $3.0\% \pm 0.3\%$ magnesium.” (lines 297-300)

Figure 5 caption: Mention exact treatment (e.g., $600 \mu\text{mol kg}^{-1}$), not “higher addition.”

We agree and incorporated it (line 318).

Discussion

Line 293: Specify $\text{Ca}(\text{OH})_2$ and $\text{Mg}(\text{OH})_2$ instead of “such materials.”

We agree and edited to: “A very general observation regarding alkalinity generation is that the dissolution of $\text{Ca}(\text{OH})_2$ and $\text{Mg}(\text{OH})_2$ here was lower than reported in the literature” (lines 326-327)

Line 295: Explain why this incubation is more realistic and which coastal conditions are being mimicked. The impact of advection on CO_2 ingassing rates is not sufficiently discussed despite being in the section title.

This is a good point and we edited to: “Such an outcome was to be expected as the milder 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” (lines 328-330)

Line 298: Flipkens et al. (2023, GCA, 359, 84–99) reached similar conclusions in an olivine incubation under different advective regimes and could therefore be cited.

Thank you for providing the reference, this was incorporated (line 333).

Line 313: Provide percentage of total expected TA increase rather than absolute concentration.

We agree and edited accordingly e.g., lines 331, 335 and 336).

Line 338: Change “loss of alkalinity to CaCO_3 ” to “loss of alkalinity through CaCO_3 precipitation.”

This was slightly edited to “However, the loss of alkalinity to CaCO_3 precipitation in the 125 RPM differs” (lines 372-373) as we believe it to be more grammatically correct to use “loss to CaCO_3 ” rather than “loss from CaCO_3 ”.

Line 349: Improve sentence fluency.

We agree and edited to: “Hence, allowing for fast CO₂ ingassing limits and decreases the risk for runaway CaCO₃ precipitation by shortening the CAP” (lines 383-384)

Line 353: Change “figure 2, Figure3” to “Figure 2 and 3.”

This is a good point and it was edited to “Figs. 3 and 4” as per BG guidelines (line 388).

Line 358: Specify which figure is being referenced.

Thank you for pointing this out, we added references to Fig. 7 and 8 where appropriate (lines 390 and 393).

Line 405: Should this be “upper limit of effects” rather than “lower limit”?

It is indeed upper limit, thank you (line 447).

Line 406: The statement that real-world OAE applications may only have small effects, if any, on living organisms is too bold. Results from a 1-L bottle cannot necessarily be extrapolated to a 50-m water column. CAP duration and impacts could differ. Also specify which OAE applications are meant. This conclusion cannot yet be drawn for all OAE approaches, especially those involving trace-metal-releasing materials such as olivine or steel slags. Please nuance this statement.

We understand and edited as: “In open ocean application, the quick dilution and subsequent CO₂ ingassing will most likely significantly decrease the CAP duration, decreasing OAE effects. However, such hypothesis is yet to be tested as open ocean OAE trials are very scarce.” (lines 447-449)

Line 413: Add “drastically” after “decreased.” CCS/CCU of calcination associated CO₂ emissions will be essential for ocean liming (see Foteinis et al., 2022).

This is noted and was edited (line 457).

Line 429: Export to deeper ocean is not the only possibility; settling onto coastal sediments is also possible.

We agree and edited to: “Hence, the crystals will ultimately sink and be transported onto coastal sediments or into the deeper ocean” (lines 473-474)

Line 431: “Buried in sediments without further dissolution” is incorrect. The cited studies show CaCO₃ dissolution in surface sediments under undersaturated conditions. Dissolution would only stop if buried below the reactive layer. Please correct.

Thank you for the details, we edited to: “Once transported in the deeper waters, CaCO_3 particles may redissolve in areas of undersaturated seawater (like below the carbonate compensation depth) or be buried in sediments where further dissolution may occur until transported below the reactive layer” (lines 475-477)

Line 443: It is unclear why slower $\text{Mg}(\text{OH})_2$ dissolution would allow a higher CAP in the diffusive boundary layer. Wouldn't slower dissolution instead prolong but reduce oversaturation intensity?

This is a fair point. We agree that with the faster dissolving $\text{Ca}(\text{OH})_2$ particles, the oversaturation intensity is likely to be higher. However, it is most likely “washed away” faster when diffusion and mixing occurs. In the case of $\text{Mg}(\text{OH})_2$, the slower dissolution would likely prolong the CAP as the reviewer points out. The level of oversaturation as well as the time at which such level is maintained are both key factors to trigger CaCO_3 precipitation.

It is safe to assume that in the diffusive boundary layer, the Ω_A is reaching very high levels, much higher than levels required to trigger homogeneous precipitation. We can therefore assume that the oversaturation intensity is, in both cases, critically high, leading to instant nucleation. As discussed above, the second important factor for CaCO_3 precipitation kinetics is the length of time during which such levels are maintained. Therefore, when $\text{Mg}(\text{OH})_2$ dissolves, instant CaCO_3 formation is triggered but also critical conditions maintained for longer than with $\text{Ca}(\text{OH})_2$, which ultimately results in further CaCO_3 precipitation.

Line 461: Add “seawater” before “the bottle.”

This is a good point and was edited (line 509).

Line 470: Add typical open-ocean gas transfer velocities in brackets for comparison.

We agree and added: “These gas velocities are significantly lower than those observed naturally in the open ocean (averaging between 14 and 20 cm h^{-1} , though ranging from nearly 0 to roughly 50 cm h^{-1}) but may be correlated to wind speed (Frew et al., 2007; Yu et al., 2013; Zhou et al., 2023).” (lines 517-519)

Line 483: This is not purely hypothetical. Your data and other studies (England & Bach, 2025; Flipkens et al., 2023) support this claim. Please cite accordingly.

We understand and agree, we edited to: “While the effect of wave height on the dissolution of alkaline materials is not fully constrained, results presented here and published research (England & Bach, 2025; Flipkens et al., 2023) support that the stronger the water movement and disturbance, the quicker the mineral dissolution” (lines 531-533)

Line 492: Other long-term OAE experiments exist (e.g., Fuhr et al., 2025), but not with fast-dissolving minerals like $\text{Ca}(\text{OH})_2$ or $\text{Mg}(\text{OH})_2$. Please rephrase.

We agree and edited to: “The experiments presented here report on the first long-term (6 months) laboratory incubations for OAE with varying alkalinity increases using $\text{Ca}(\text{OH})_2$, $\text{Mg}(\text{OH})_2$ and NaOH, in stagnant and advected seawater.” (lines 541-542)

Lines 497–499: Rephrase this sentence; it is currently too complex.

We agree and edited as: “CaCO₃ formation was directly correlated to the amount of Mg(OH)₂ added at 125 RPM. However, above targets of 300 μmol kg⁻¹ with Ca(OH)₂, the TA generation decreased while CaCO₃ precipitation rate increased at 125 RPM.” (lines 546-547)

REVIEWER 2

General comments

The manuscript addresses an interesting topic; the potential of ocean alkalinity enhancement in removing CO₂ 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 added 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 CaCO₃, as well as high pH and low CO₂ conditions, potentially impacting biological processes. This period of high alkalinity 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). [...] Here, in a new approach, the dissolution of Ca(OH)₂ and Mg(OH)₂ in natural seawater was addressed both in static and advected reaction vessels for 6 months. Circular advection regimes were chosen to try and represent open ocean conditions with vertical diffusion but limiting surface tension disturbance, 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 CO₂ and equivalent open ocean wind speeds were derived, allowing direct comparison between laboratory data and global modelling studies.” (lines 46-50 and 59-64)

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, new relevant citations have been added 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 addressed 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 (lines 327-328). We also incorporated a figure representing the experimental setup to further enhance the clarity of the material and methods section (line 105).

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 CaCO_3 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 ensured consistency when discussing advection. Regarding the second part of the comment, we also revised the sentences accordingly and suggest: “as CaCO_3 does not dissolve naturally in open ocean seawater.” (lines 338-339)

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.

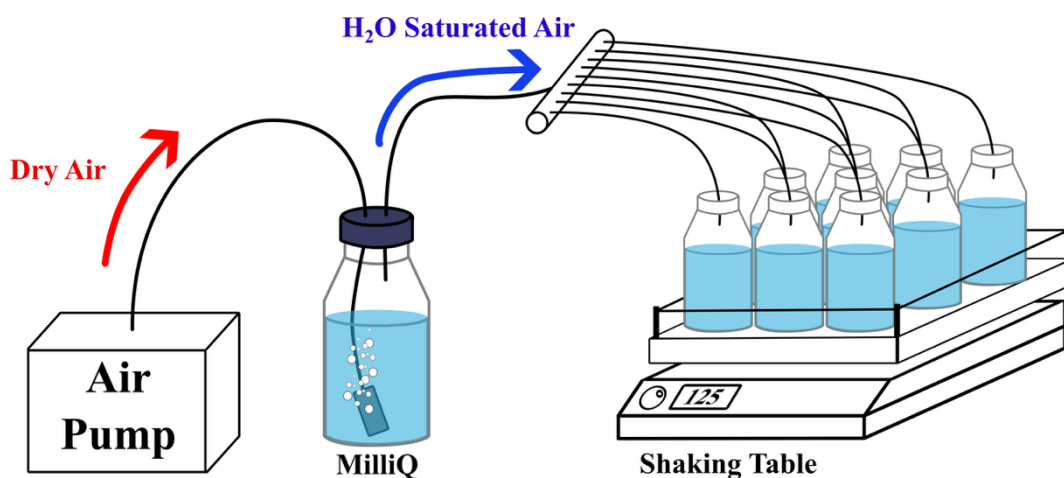


Figure 1: Schematic of the experimental setup. The air pump takes up dry air and pushes it into a closed Schott bottle where MilliQ is bubbled. The newly H₂O saturated air is then carried to an air diffuser from which individual tubing are connected to each experimental bottle, constantly and equally supplying each open bottle with fresh air equilibrated to atmospheric CO₂.

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 H₂O 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 $\pm 0.5\%$.

During evaporation, the water “portion” of the sample is lost, i.e., H₂O, 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 $\sim 2480 \mu\text{mol kg}^{-1}$ (similar to day 1 of the Mg(OH)₂ experiment at 125 RPM and targeted ΔTA of $600 \mu\text{mol kg}^{-1}$, figure 2). If this sample was to have lost 0.5% of its water content (decreasing from 1000g to 995g), the TA would now be $2480 \times 1000 / 995 = 2492.5 \mu\text{mol kg}^{-1}$. By adding seawater (assuming open ocean, $2300 \mu\text{mol kg}^{-1}$), 5g of such TA will be added, yielding a new 1000g sample with a TA of: $2492.5 \times 0.995 + 2300 \times 0.005 = 2491.5 \mu\text{mol kg}^{-1}$. The new sample TA is $11.5 \mu\text{mol kg}^{-1}$ 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. Nevertheless, we have reviewed all figures and added all relevant ones inside the text. Only a figure with calculated parameters and a longer table which are both not needed for the understanding of the manuscript have been kept in supplementary materials.

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 CO₂ flux is used in our case to derive the k value as the CO₂ flux F is calculated from our experiments (i.e., 21.3 ad $39.9 \mu\text{mol kg}^{-1} \text{ d}^{-1}$), K_0 is obtained from Weiss (1974) and the pCO₂ values are obtained from our experiments. Then using the calculated k values (1.4 and 2.7 cm h^{-1} , line 516), 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°C as per Wanninkhof, R. (1992). Then, we end up with U values of 2.2 and 3.1 m s^{-1} (line 525)

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 runaway 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 edited the following sentence in the manuscript: “Once precipitation starts, it may enter a runaway pattern where the precipitation speed increases, sometimes reaching TA values lower than the starting conditions (Moras et al., 2022)” (lines 41-42)

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 edited as follow: “Carl Roth (Germany)” (line 120)

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 $2.4 \mu\text{mol kg}^{-1}$, corresponding to an offset of $\sim 0.1\%$.

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 reformulated 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 144) and accurate estimations once calibrated against CRMs. DIC measurement with a TOC-L/CPH analyser is also precise (3.5%, line 147) and accurate once calibrated with CRMs. pH measurement with a probe was used in parallel to track carbonate chemistry changes without having to sample 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. We reformulated this section (lines 179-190).

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 adjusted the formulation to be less radical, such as: “Such an outcome was to be expected as the milder 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. The low (125 RPM, Reynolds number of $\sim 1.6 \cdot 10^4$) or absent (0 RPM) advection decreased the speed of diffusion around the particles.” (lines 328-331).