

Biogeosciences Discuss., referee comment RC2
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Comment on bg-2021-330

Anonymous Referee #2

Referee comment on "Ocean alkalinity enhancement – avoiding runaway CaCO₃ precipitation during quick and hydrated lime dissolution" by Charly A. Moras et al., Biogeosciences Discuss., <https://doi.org/10.5194/bg-2021-330-RC2>, 2021

General Comments:

Overall, this Reviewer finds this study to be an important contribution to the field and the manuscript is well-structured. However, the Reviewer has some major concerns about the study and minor concerns regarding the manuscript.

The primary concern this Reviewer has is the lack of replication of experiments and the absence of justification for this. This concern is compounded by the differences between the 500 $\mu\text{mol kg}^{-1}$ hydrated lime incubations presented in Figures 2 and 4; furthermore, the end-points of these incubations cannot be compared because the second experiment was not continued for the same length of time. The inconsistent length of experiments and frequencies of sampling/ measurements is noted across the reported experiments. Additionally, the range of TA additions tested is limited for the broad conclusions given. The result of these concerns leaves the Reviewer with a strong feeling that there is insufficient evidence for the conclusions presented.

Regarding the manuscript itself, the author has a tendency to use vague and non-specific language at points, contrasting with the majority of the manuscript which is meticulously and clearly written. Additionally, the in-text citations are not systematically organised, it would be useful if these could be organised in a consistent manner (e.g. youngest to oldest).

In section 1, a lot of background information is given before the author indicates the significance and focus of the current study; it would be advantageous to the reader if this was indicated earlier.

The methods presented in section 2 vary between thorough and detailed descriptions and

vague indications, the Reviewer would like this to be more consistent throughout.

The Reviewer found some inconsistency between the motivation given in Section 1 and the results presented in Section 3, particularly that there are no quantifications of the influence of CaCO_3 on pH. The author also presents results as approximations (i.e. using tilde) where no replicates were performed and some values were not consistently used throughout the manuscript. In Section 3.5, the author refers to pairs of subfigures from Fig. 4 which does not always appear relevant when the context suggests one subfigure is being discussed. It could also be more clear to the reader, if the changes discussed in Section 3.5 were relative to the initial state before dilution. Section 3.6 also feels abrupt and it is not clear how much the particulate organic carbon investigation contributes to the manuscript.

The first paragraph of Section 4.1 is a good paragraph which clearly explains the precipitation pathways and therefore what the author was investigating as sources of precipitation. However, this information may have benefited the reader earlier in the manuscript to justify the methods used.

In Section 5, the author gives both the expanded and abbreviated form of terms that were defined through the paper and it is not clear why this is necessary.

Finally, regarding figures and tables, it would be useful to the reader if the measured/ actual TA addition was also included in Table 1. The author also divides the temporal resolution of Figures 1-3 and Figure 4 differently, which is not immediately clear to the reader. This difference should be clarified in the Figure 4 caption to prevent confusion.

Specific Comments:

■

Line 13: Does "their" refer to alkaline minerals in general or specifically to the investigated minerals, quick and hydrated lime?

■

Line 14: "...how much..." is again rather vague, is the author referring to mass of alkaline mineral or only the investigated minerals?

■

Line 19: The author uses “so-called” here but later in the discussion at line 292 indicates that this is an established form of CaCO_3 precipitation described in multiple literature sources; unclear why it is referred to in this manner in the abstract.

■

Line 21: “...until Ω_{ar} reached levels below 2.”; Does this mean that the runaway precipitation spontaneously stops at this threshold but does not start again until the threshold of 5?

■

Line 23: “...ideally within hours of the addition...” is vague and the significance of the statement to the work is not made clear in the abstract (i.e. that the authors investigated the significance of dilution at different timescales.)

■

Line 24: The term “...model considerations...” is unclear and no description of any modelling is given in Section 2 and the model results presented in Table 3 are only discussed in Section 4. More detail should be included about what was modelled and how, if the author wants to include these results. Furthermore, “considerations” does not indicate if the authors considered using models or whether simulations were performed.

■

Line 26: The authors indicate that equilibration of seawater to atmospheric CO_2 mixing ratios would enhance mineral dissolution but do not indicate what CO_2 partial pressures are being referenced or what the factors given are in reference to.

■

Line 36: “...ideally around $+1.5^\circ\text{C}$...” makes it sound as though, even if less were possible, that Governments should still aim for $+1.5^\circ\text{C}$ because of the use of “ideally”

■

Line 39: “...one...” is vague, can the author be more specific as to what OAE is one of the highest of...? e.g. “...additional mitigation strategies...”

■

Line 40: The authors give an extreme example of OAE (Feng et al., 2017) and do not address other examples of OAE discussed in literature with arguably more realistic application strategies i.e. Köhler et al. (2013), Keller et al. (2014), Lenton et al. (2018) or Burt et al. (2021)

■

Line 42: Is the use of "...typically..." appropriate? Are there OAE methods which do not use alkaline minerals?

■

Line 43: "In this regard..." is vague and not clear that the author is introducing alkaline minerals which are considered for AOE.

■

Line 44: "The last two minerals..." is vague and could be replaced with "Quick and hydrated lime..." for greater clarity.

■

Line 47: Is the additional release of CO₂ considered with respect to the final CO₂ drawdown?

■

Line 51: Is it appropriate to state that the non-linearities of the seawater carbonate system are ignored as Equation 1 gives the bicarbonate ion which is only the major dissociation product at typical seawater conditions due to the non-linearities?

■

Line 56-58: Is the repetition of the equation explanation required? Furthermore, the DIC concentration only remains unchanged if there is no uptake of atmospheric CO₂ while the statement at line 51 "Including the subsequent oceanic uptake of atmospheric CO₂..." indicates that Equation 1 considers atmospheric CO₂ rather than aqueous CO₂. How did the author intend the understanding of these statements be reconciled?

■

Line 58: "This increases the pH...", the author does not explain why this is the case and neither does Equation 1 explicitly indicate the change which produces the reduction in H⁺ ions.

■

Line 60: Is it appropriate to consider that the reduction in partial pressure of CO₂ in seawater is the result of the reduction of CO₂ rather than being concurrent?

■

Line 65-67: Repetition; the authors state twice that OAE counters two aspects of OA.

▪

Line 70: The author is not specific about what is being constrained.

▪

Line 71: The author does not indicate why the reduction in carbonate ions increases the aqueous CO₂ concentration rather than following Le Chatelier's principle to replace the precipitated carbonate ions.

▪

Line 72: The author does not explain why the precipitation of native Ca²⁺ and CO₃²⁻ ions further reduces the effectiveness of the added alkalinity.

▪

Line 77: Does the indicated increase in TA to reach the described critical threshold hold if a non-lime alkalinity source such as olivine were used? If the estimate is specific to quick/ hydrated lime, can the author specify this when the estimate is stated.

▪

Line 78: Is the critical threshold given here calculated by the author or given in Marion et al. (2009)? The citation location and the previous postulation leaves this ambiguous.

▪

Line 91: Acronym "ESI" is used without prior definition; is this familiar to all relevant members of field?

▪

Line 94: The author describes how the seawater samples were stored to reduce bacterial metabolic activity before being sterilised; why were the samples not sterilised before storage?

▪

Line 102: The author describes the seawater as being "accurately weighed" without indicated the degree of accuracy used; please clarify.

▪

Line 102: The beakers are referred to as "high-quality"; what is the metric used to

determine the quality of the beakers?

■

Line 105: The term "floating lid" is unfamiliar and it was unclear how this minimised gas exchange. The discussion indicates that while this apparatus minimised gas exchange at the beginning of the experiment, a gaseous head space was created with subsequent sampling and therefore the lid did not float on the surface of the reduced fluid level. Can the apparatus and its function be described more clearly?

■

Line 106: Amounts is vague; were the alkaline minerals added in masses or a different metric? What masses were added?

■

Line 106: The author refers to "alkaline compounds" in general while the context of the paper indicates that only quick and hydrated lime were used, were other compounds used in the experiments?

■

Line 106: "...taken in increasing time intervals..."; can the author give the specific time intervals used in the experiments?

■

Line 108: The author does not indicate how the pH monitoring was performed such as if the monitoring was continuous or periodic; please clarify

■

Line 109: The contents of the beaker were "carefully transferred" but there is no indication of how care was taken or why the contents of the beaker were transferred; please clarify

■

Line 110: What was the "corresponding" volume? This is not specific

■

Line 114: It is not immediately clear that these are details of the descriptions of "alkalinity addition" given in the previous paragraph; make this clearer

■

Line 114: Why was this mesh size selected or why were these particles sizes selected for the described experiments?

■

Line 115: Why was it important to mass particles smaller than 63 μm when the experimental design was only for the addition of particles larger than 63 μm ?

■

Line 119: Why is it pertinent that the massing steps were performed in less than 5 minutes?

■

Line 119: Why were these addition concentrations used? What do these addition concentrations represent?

■

Line 121: The author has not prepared the reader for this set of experiments. There is no apparent preceding justification or explanation how these experiments tie into the three research questions posed in section 1

■

Line 123: The author describes the weighing as accurate without indicating the degree of accuracy

■

Line 124: "sonicated for 15 minutes with gentle shaking" is vague; unclear how ultrasonic vibrations can be applied gently, can the author give the frequency or some other metric?

■

Line 126: "almost twice the alkalinity increase" is vague; in the first, "almost" indicates a concentration less than double while Table 1 indicates the target was higher in both instances and theoretically lower in one instance, and in the second, it would be clearer if the specific concentration was given instead

■

Line 129: The author does not explain why quartz powder was added until section 4; please indicate the justification earlier

-

Line 130: "amount" is vague; was a specific mass of quartz particles added?

-

Line 131: "amount" is vague; context suggests area is an appropriate metric?

-

Line 132: This assumption seems unavoidable; is it possible to assess the errors from this assumption?

-

Line 135: Is "particle filtering experiment" a fixed term? Please give more detail

-

Line 135: "following the same setup as described above"; does the author refer to the setup described in 2.2.1 or 2.2.2 here?

-

Line 136: Why was a 4 hour period selected for the reaction? This contrasts with the 5 hours allocated in 2.2.1

-

Line 137: Here it is indicated that a "2L Schott beaker" was used but in 2.2 this is vaguely given that either 2L or 5L beakers were used; can the author describe which experiments used which volume and justify why different volumes were used?

-

Line 143: The acronym "NSW" is used without introduction; please clarify

-

Line 143: Is it necessary to tell the reader that something will be described rather than moving on to the description?

-

Line 145: "amounts"; was hydrated lime added in masses or another metric?

-

Line 147: The description of how the hydrated lime powder was added is given here after a vague "added" at line 145; why is a vague term used and detail given later when the detail could be given in the first instance?

-

Line 148: The author does not explain how the dilutions were performed? Was the solution transfer performed following the method given at line 109 or 137?

-

Line 151: Why were these time intervals selected?

-

Line 165: Why were samples collected at the end of some of the experiments? Which experiments were sampled and why?

-

Line 166: Is this a standard protocol? If so, please indicate reference, if not, please justify. Furthermore, why were TPC and POC samples treated differently?

-

Line 170: Please explain how standard deviations were calculated without replication

-

Line 174: Why was CaCO_3 precipitation "suspected", this indicates to the reader that there was insufficient evidence to support the conclusion

-

Line 175: Why were samples taken in a 10-15 mL range of volumes and not a more consistent and precise volume?

-

Line 180: "Most" indicates that not all parameters were calculated; which were/ were not calculated and why?

-

Line 182: Which parameters were calculated and why? What could not be calculated?

■

Line 189: "amount"; was alkalinity added in concentrations or another metric?

■

Line 203: The author states that a " $\sim 250 \mu\text{mol kg}^{-1}$ TA addition" was performed for the first quicklime experiment but Table 1 describes a Theoretical TA addition which exceeds this target by 9.6%; calling the experiments by the target TA addition is then misleading given that the Theoretical and Actual TA additions varied significantly from these targets; name the experiments in a clearer and more consistent manner.

■

Line 204: The author describes an increase in DIC concentration in the seawater incubations despite using a "floating lid" to minimise gas exchange (Section 2.2 and Fig. A2), this is not appropriately addressed in Section 4. Was there a port for gas exchange in the "floating lid"? If the volume of the gas phase in the beaker changed, what was the CO_2 mixing ratio of the introduced gas? How comparable is the CO_2 uptake with a system of free gas exchange?

■

Line 204: "slowly" is vague and subjective; please quantitatively indicate the rate of change of DIC concentration

■

Line 208: "...before slowly decreasing two days later..." indicates that there is no change between the first 4 hours until after 48 hours while Fig. 1 suggests a decline over the 48 hours and a greater rate of change 6th and 8th sampling points; can the author clarify this discrepancy between description and illustration?

■

Line 209: "eventually" is vague and subjective; specify when this ΔTA was recorded

■

Line 210: "slight" is vague and subjective

■

Line 210-211: "much more considerable" is vague and subjective

■

Line 211: The Δ DIC is given as approximately $465 \mu\text{mol kg}^{-1}$ but Fig. 1b indicates that this should be a negative value

■

Line 213: "rapidly" is vague and subjective

■

Line 217: The sentence beginning "The DIC concentration..." indicates a constant rate of increase to a total of $70 \mu\text{mol kg}^{-1}$ but Fig 2b indicates a greater rate of change between the 6th and 9th sampling intervals; please clarify

■

Line 221: "relatively steady" is vague and subjective; what was the rate of decrease?

■

Line 221: "decrease accelerated" is vague; by how much? From what to what?

■

Line 222: Why is the TA loss smaller with $\text{Ca}(\text{OH})_2$ than with CaO ?

■

Line 223: Does the DIC concentration drop "in a similar fashion as TA"? Fig. 2 indicates these were measured at different frequencies; what caused the difference in sampling frequency?

■

Line 227-229: The first two sentences of section 3.4 are the preparation that was missing from section 2.2.2

■

Line 228-229: "...designed to result... precipitated (Table 1)."; 1) Table 1 does not indicate the aragonite saturation state so the reader cannot confirm that these states are comparable; 2) After being presented with figures 1 and 2, the reader notes that the aragonite saturation states produced by this set of experiments significantly exceeds any of the previous sets of experiments, how can comparison be made?; 3) According to Table 1 the Na_2CO_3 solutions were precisely comparable to 0.02 mg kg^{-1} and $0.16 \mu\text{mol kg}^{-1}$ TA and yet produced significantly different aragonite saturation states illustrated in Fig. 3; can the author address this disparity?

-

Line 233-234: there is repetition of methods here

-

Line 235: "about a week later" is vague; what period does this include? 6-8 days? 5-9 days? Please clarify

-

Line 238: "pronounced decline" is vague and author does not indicate rates; Fig. 3 indicates the majority of the decline occurred in the first 20 days

-

Line 239: further repetition of method

-

Line 241: What is the variable with concentration of $470 \mu\text{mol kg}^{-1}$?

-

Line 245: 1) Section 2.2.3 description was insufficient to prepare the reader for Figure 4; it was not clear that each dilution was performed ONCE at different times and that five solutions were subsequently incubated. 2) Section 2.2.3 also does not address how the author ensured that the particles from the parent solution were proportionately included in the diluted solution. Was it assumed that the stirring was sufficient to maintain a homogeneous suspension? 3) It needs to be more clearly stated that it is not a single solution diluted multiple times but that it is a source solution diluted a single time at multiple instances.

-

Line 249: Is it intended that the reader should compare figure 4a with figure 4d?

-

Line 252: repetition of method

-

Line 255: "slightly lower" is vague; what was the difference?

-

Line 255-256: sentence starting "In all dilutions..."; while the direction of change appears independent of dilution timing, the magnitude appears to vary; replication may have provided more information

■

Line 260: This is the first instance where the author refers to the Theoretical TA addition from Table 1; it would be insightful for the reader to see both Actual and Theoretical TA additions in Table one for comparison

■

Line 262: sentence beginning "From then on..." is vague

■

Line 264-265: Can the author address the source of DIC increase given the use of the "floating lid" to minimise gas exchange?

■

Line 266: "up to ~16.7"; the author uses "up to" indicating a maximum and then gives an approximation, is the approximation a rounding of the maximum value? Why use an approximation?

■

Line 268: repetition of method

■

Line 273: repetition of lines 269-270

■

Line 274: "slowly increased over time" is vague

■

Line 274: "~5.0-5."; what value is being presented here? It reads as approximately 5.0 to 5.0, which would not appear to be correct. Please clarify

■

Line 275: states where the aragonite saturation state drops to but not where from; please expand

■

Line 275: " Ω_{ar} decreased slightly over time after dilutions"; both the 1 day and 1 week dilutions appear to exhibit an increase before the final sampling?

■

Line 278: Can the author address the difference between the estimated and measured PIC?

■

Line 280-281: Source of percentages unclear; reviewing Table 2 indicates a 14% and a 7% overestimation respectively in the 500 $\mu\text{mol kg}^{-1}$ cases and 67% overestimation in the the 2000 $\mu\text{mol kg}^{-1}$ case; can the author explain the source of the given percentages?

■

Line 284: "In some..." is vague; which experiments?

■

Line 285: "...TA additions equal to or higher than 500 $\mu\text{mol kg}^{-1}$ " seems misleading as these refer only to target or theoretical TA additions while the results indicate that the experiments with target TA additions of 500 $\mu\text{mol kg}^{-1}$ had lower measured TA changes; please clarify

■

Line 287: "...we simulated ocean mixing..."; how do the dilutions represent ocean mixing timescales? Please clarify

■

Line 290: Subsection 4.1 also addresses errors including CO_2 invasion from the headspace beyond precipitation which are not indicated by the title

■

Line 297: Section 2.5 state that only aragonite saturation states were reported and all Figures present aragonite saturation states. Is the author able to discuss calcium carbonate saturation state thresholds in general given the reported data?

■

Line 300: Reference to Eqn. 1; The statement is not disputed. However, the relevance of the citation to Eqn. 1 to explain how the change in carbonate ions accounts for a

change in TA is questioned. The equation expressed in Eqn. 1 does not describe the contributions to TA.

■

Line 301: sentence beginning "Hence, any loss..." is well established in literature; would a citation not be appropriate?

■

Line 302: "suspected" suggests a lack of evidence. If the author detected a 2:1 ratio of TA to DIC changes and then investigated to confirm if this was precipitation, then the authors' actions are based on empirical evidence rather than a feeling or thought. Please clarify.

■

Line 306: addresses comment at line 204: No information is given in section 2.2 regarding sample collection. What volume of samples were taken? Does this mean that the headspace volume increased with each sampling? What was the ambient CO₂ mixing ratio in the room during sampling? Was the floating lid removed completely for sampling? If so, how long for? Was a port in the floating lid used for sampling? Figure A2 does not indicate an additional port to allow gas into the vessel during sampling.

■

Line 309: "a significant fraction could be hydrated" is given as a reason for lower efficiency, could this have been prevented or quantified; please clarify.

■

Line 310: see comment at line 203: In section 3.2 the target TA values are given for the additions and compared with the measured TA increase. When comparing the TA target with the Theoretical TA addition being discussed here; there are significant deviations of 8-10% in the CaO experiments. The measured TA values are reported as approximates but comparing the Theoretical TA increase and the measured TA increase (as indicated at line 308) these deviations appear to be 35% and 33% and not the 25-27% reported at line 310. Where does this discrepancy come from?

■

Line 311: see previous; these percentages also appear to be underestimated.

■

Line 314: "same conclusions"; there is no apparent conclusion in the following statement to qualify this aside; please clarify.

■

Line 314: repetition of results

■

Line 315: "about $-970 \mu\text{mol kg}^{-1}$ " this value deviates from the $-950 \mu\text{mol kg}^{-1}$ given in the results; please clarify which is the correct concentration

■

Line 317: "Although a perfect match"; an absolutism and not technically accurate; for CaO with a reported TA loss of $-950 \mu\text{mol kg}^{-1}$ this would reflect a change in DIC loss of $475 \mu\text{mol kg}^{-1}$ rather than the reported $465 \mu\text{mol kg}^{-1}$; similarly, for the $\text{Ca}(\text{OH})_2$ this would be $430 \mu\text{mol kg}^{-1}$ rather than the reported $395 \mu\text{mol kg}^{-1}$; the absolute statements appear misleading

■

Line 320-323: sentence beginning "This discrepancy..." to "...dissolution rates."; not clear how the statement follows; The discrepancy described until this point is the invasion of CO_2 from the headspace which mitigated the DIC loss. It is not clear how this is explained by an inability to measure full dissolution in the presence of concurrent precipitation?

■

Line 323: "It also explains..."; what is the "it" in this context? Is the author arguing that the concurrent dissolution and precipitation prevents a true maximum ΔTA and therefore explains the lower PIC estimate?

■

Line 325: "This could be explained..."; this reads as speculation? Was there no way to verify this? Was there visible precipitation on the bottle walls? Why would this particular experiment be influenced by bottle wall-precipitation to such a significant degree compared to the other experiments?

■

Line 326: "In this sense"; in which sense? Please clarify

■

Line 327: "theoretical TA generation by dissolution"; why would dissolution generate theoretical TA? Please clarify

■

Line 329: "in between the other estimates"; which estimates are being referred to here?

▪

Line 329: "lies in the fact..."; appears to be a repetition of the speculation at line 326

▪

Line 333: "An interesting finding" is subjective

▪

Line 335-337: "in all these experiments... initiated in natural seawater"; this conclusion is supported by 4 incubations in different experimental conditions and is not convincing

▪

Line 327-329: sentence beginning "This is to be"; how does this follow? If precipitation occurs more readily at higher saturation states, why does a non-linear relationship between precipitation rate and saturation state explain the continued precipitation at saturation states where precipitation is not naturally initiated?

▪

Line 344: "modelling precipitation"; there is no explanation of modelling in the method section. Is this from Zhong and Mucci (1989)? It is set in the text and phrased as though it were part of this work. Please clarify

▪

Line 349: "...experiments were not replicated." This is a key concern of this Reviewer over the strength of the manuscript's conclusions and further justification to this lack of replication is required

▪

Line 353: "In contrast"; is this comparing the results of this manuscript with the results of Lioliou et al. (2007) or is this work from Lioliou et al. (2007); please clarify

▪

Line 354: "For that to occur"; what is the purpose of this statement? The next statement states that the experiments do increase the aragonite saturation state.

▪

Line 355: "above an Ω_{ar} of ~ 9.2 CaCO₃ precipitation did actually occur"; This statement is factually correct but incomplete as no tests were performed at lower aragonite saturation states to determine if precipitation occurred.

■

Line 357: "this idea" is vague; which idea? How does it support the idea? If anything it does not refute the hypothesis of heterogeneous precipitation e.g. an unfiltered sample which exhibits natural precipitation would be required to test the hypothesis

■

Line 360: first reference to Figure 5; if the figure is important enough to be included, why are they not described in Section 3?

■

Line 366: "Early CaCO₃ precipitation..."; Repetition. Early precipitation during dissolution was discussed at length in the previous subsection.

■

Line 370: "When CaCO₃ precipitates..." is repetition

■

Line 372: it is not explained how the changes described are "impacting OAE potential"; this should be stated explicitly

■

Line 375: sentence beginning "If fully..."; Why would a DIC increase further increase the pH? Are the numbers cumulative with the 8.61 and 8.45 reported on line 375? After reading Table 3, are these numbers after subsequent reductions in pH and aragonite saturation state during equilibration? If so, this should be clarified in text

■

Line 379: It was not clear how the author arrived at this value. Given that the author describes the use of quick and hydrated lime in the manuscript, these materials would sequester ~ 1.30 and ~ 0.99 tonnes of CO₂ respectively at the efficiencies described. The mixed use of "base material" and "source material" did not associate that the author intended this estimate to be based on CaCO₃ masses; please clarify in the text if this is the case. Additionally, does this estimate include the effects of mining, processing and transportation on the estimate of CO₂ sequestered per tonne of source material mined? If not, these estimates could be misleading

■

Line 381-382: Please explain the source of these sequestration estimates. Caserini et al. (2021) only indicate the given annual discharge rate of $\text{Ca}(\text{OH})_2$ but estimate a CO_2 uptake of 1.5-3.3 Gt a^{-1} . What is the source of the 1.2-2.8 Gt a^{-1} CO_2 given in the text? Please clarify the arrangement of the citation to indicate the correct sources of information

■

Line 382-384: c.f. comment for Line 40; the author addresses the Caserini et al. (2021) estimate but also includes the extreme input rate discussed by Feng et al. (2016) without acknowledging the extreme nature of the latter estimate

■

Line 385-392: "If as much... drop below 1."; This is difficult to follow and the Reviewer is not convinced there is sufficient evidence to support these conclusions. 1) It does not appear that the results shown indicate an experimental case where "as much CaCO_3 precipitates as TA was added". In cases where precipitation occurred, "runaway CaCO_3 precipitation" also appeared to occur? 2) If all of the TA added precipitates; how can the alkalinity of the ocean be enhanced at all, let alone by 10-60%? 3) This is the first time that "secondary CaCO_3 precipitation" has been mentioned; please define in more detail what is meant by this term. 4) How relatable is the DIC uptake of restricted gas exchange experiments with open ocean implementations?

■

Line 399: "While above considerations stress..." is vague, which "considerations" does the author mean? Further, how has this been stressed? 4.1 describes errors of unmeasured precipitation, 4.2 describes the influence of undissolved mineral phases on heterogeneous precipitation, 4.3 discusses the impact of precipitation on OAE. There has been no or very limited stress on the importance of monitoring; please clarify

■

Line 399: Please clarify what is meant by "natural" here; ships and their wakes are not natural phenomena?

■

Line 401: It is unclear why Feng et al. (2016), Caserini et al. (2021) and Mongin et al. (2021) are cited here as the statement made appears to be regarding the assumptions of the present work?

■

Line 401: "In our experiments..."; How does mixing after 10 minutes, 1 hour, 1 day or 1 week simulate dilution in wakes or natural mixing? Ship wakes would have a limited surface mixing effect in the short-term. Natural mixing would have a longer term and extremely regional impact c.f. Burt et al. (2021). Both approaches neglect continuous diffusion.

■

Line 403: "at a surprise"; why is this surprising? Following the arguments and work discussed in this manuscript; no significant precipitation was reported for the 250 $\mu\text{mol kg}^{-1}$ CaO or Ca(OH)₂ experiments (Fig. 1 and 2) and dilution of particles to that concentration could be expected to respond in a similar manner.

■

Line 403: Is dilution by half not the intention of the 1:1 dilution? Please clarify

■

Line 404: "precipitation rates..."; this is not apparent from Fig. A4. Fig. 4c indicates a change from aragonite saturation state of 7-8 between the 4 dilutions to approx. 4-5. Fig. A4 indicates an aragonite saturation state of 7-8 would have a precipitation rate of 800-1100 to a precipitation rate of 200-400?

■

Line 409: How is a comparison of Fig. 5c and Fig. 5d relevant here? The author appears to be discussing the precipitation response of Ca(OH)₂ dilutions from 500 $\mu\text{mol kg}^{-1}$ to comparing Na₂CO₃ experiments with quartz seed minerals rather than comparing precipitation between quartz seed minerals?

■

Line 410: "precipitation can be avoided"; there seems insufficient evidence to support that a 1:1 dilution is required as no other dilution ratios were tested at this concentration; please clarify

■

Line 411: "The quicker dilution..."; the similarity indicated appears limited; perhaps only to the aragonite saturation state achieved within the first day? Plot axes make comparison difficult for the reader

■

Line 415: "that verification... problematic"; it is unlikely that measurements of TA additions would be used to monitor CO₂ sequestration in the long term and "permanent" sequestration is difficult to define; please clarify what is meant by "TA measurements" and "permanent" in this context

■

Line 419: "Staying clearly..." is ambiguous; in what context is "clear" being used here?

■

Line 419-420: The author should address the fact that Feng et al. (2017) consider olivine rather than CaO or Ca(OH)₂ and that the drawdown the author indicates were modelled using less than 10 µm grain sizes compared to the greater than 63 µm grain sizes used in this study

■

Line 425: "Hence, even with..."; the Reviewer finds insufficient evidence to conclude that this is the optimum threshold

■

Line 431: "equilibrate it with air"; this reads that the mineral must first be equilibrated with air? Please clarify

■

Line 431: "CO₂ enriched flumes"; unclear what the author means by this term

■

Line 431: "This would allow... passive"; does the author mean that excessive aragonite saturations be permitted such that a rapid equilibration with air and CO₂ uptake would reduce the aragonite concentration again? What conditions could permit this in the ocean?

■

Line 433-438: From "In this case..." all reads as speculation and feels "tacked on" to the end of the manuscript. Is this what is referred to in the subsection title as "and other TA additional strategies"? How viable would these strategies be in terms of equilibration times? The author expresses that this extra step is time and cost intensive; is the trade-off worth discussing?

■

Line 440: "promising" is subjective and reader may not agree

■

Line 444-445: "suggesting... of about 5" reads as speculation and the breadth of the experiments described in this study appear insufficient to support this conclusion

■

Line 447: Is this intended to build upon the condition of the previous statement which

referred to coastal settings? i.e. expanding beyond coastal settings into the open ocean? Please clarify

▪

Line 448: What is meant by "major mixing", please clarify

▪

Line 448: "equilibrating the seawater... mineral dissolution"; unclear why this is stated. Surface water should always be in or approaching equilibrium with the surface atmosphere. What situations would this not occur?

Technical Corrections:

▪

Line 16: In what way does the sentence beginning "However, ..." contrast or oppose the preceding sentence?

▪

Line 16: "...dissolved..." could be used in the present tense here as the results should hold constant in time

▪

Line 20: the use of "...by which..." and the sentence structure of the statement makes the sentence feel unnecessarily complex and slows reading

▪

Line 25: Why is a comma used after the temperatures are given?

▪

Line 31: Capitalise "IPCC" in the given citation

■

Line 38: Why is "ocean alkalinity enhancement" italicised and the acronym given after a dash rather than in parenthesis?

■

Line 42: "...similar to what occurs..." is vague; what occurs during rock weathering? The sentence structure can be made more concise and less vague.

■

Line 51: The sentence structure here is complicated by the mixture of multiple comma-enclosed parentheses and comma-separated clauses. The sentence should be simplified

■

Line 58-59: "This" is vague, clarify what is increasing the pH i.e. the addition of quick/hydrated lime. Additionally, "lowering" and "increasing" are nouns describing the actions rather than the verb forms "lowers" and "increases" which are indicated contextually.

■

Line 60: "This in turn..." is vague, what it turn?

■

Line 70: Why is the symbol Ω introduced here in the second use of CaCO_3 saturation state rather than the first use at line 66?

■

Line 74-81: The paragraph structure here is meandering; the author begins on the topic of the critical threshold for precipitation and then go on to postulate about the TA change to achieve this threshold before returning to introduce the reader to other types of precipitation. The paragraph should be restructured to more clearly make the author's intended point.

■

Line 90: missing article before "Agilent"; grammar indicates plural but noun is used in the singular without article.

■

Line 115: the sentence beginning "The mesh..." is written in the past tense but the present tense "avoiding" is used, to be correct this should be changed to "to avoid"

▪

Line 122 and 123: the author repeats the CAS number information; is this necessary?

▪

Line 129: "identical" is an absolutism; Table 1 indicates that the experiments were similar but not identical

▪

Line 142: There is a mixture of singular and plural in the statement beginning "Ca(OH)₂ was added"; "final alkalinity enrichments" in the plural should be used

▪

Line 147: please add space between value and unit for "200 rpm"

▪

Line 154: Incomplete statement or missing word; "Samples for TA and DIC..." context suggests the author intended "analyses" or "measurements" etc.?

▪

Line 190: Unclear if " Δ TA" is given as an expression for the preceding text, then a comma separation is required, or intended to be included in the sentence but then more explanation is required for what is meant by the term "CaCO₃ precipitation Δ TA"

▪

Line 208: plural "decreases" suggests multiple negative change events; there is insufficient sampling to distinguish multiple independent decreases

▪

Line 290: "not measured" may be better phrased as "unmeasured"

▪

Line 299-331: Is this a single paragraph? Please restrict paragraphs to a single topic, it will benefit readers

-

Line 306: missing punctuation; should there be a period before "A"

-

Line 307: sentence beginning "This in turn..."; the use of "when" suggests that a cause and effect relationship is being introduced but only active forms are used; if a cause and effect relationship is intended, clarify grammatically

-

Line 312: missing punctuation; should there be a period before "In contrast..."

-

Line 317: "exactly"; an absolutism and not technically accurate

-

Line 317: "Although a perfect match"; an absolutism and not technically accurate

-

Line 325: "...similar results. However,..."; these sentences are linked and should not be separated by a period

-

Line 328: "Finally"; this appears to be a new topic and could be a new paragraph?

-

Line 357: "...this idea. Not yet dissolved..."; sentences are connected, join them with appropriate punctuation

-

Line 401: Sentence beginning "In our experiments..." is not clearly structured

-

Line 406: The use of "however" and "but" both indicate contrast but the first half of the sentence appears to agree with the previous linked statement; please clarify

-

Line 414: from context it is assumed this is a spelling error and "from" was intended?

■

Line 430: Is "mineral" intended to be singular?

■

Line 440: Why is "Ocean Alkalinity Enhancement" italicised?

■

Line 446: "as of potential" doesn't make grammatical sense

■

Line 447: Grammar of sentence beginning "Safely increasing..." is ambiguous