

## Reply to Comments on bg-2022-126

Referee comments on "Stability of alkalinity in Ocean Alkalinity Enhancement (OAE) approaches – consequences for durability of CO<sub>2</sub> storage" by Jens Hartmann et al., Biogeosciences Discuss., <https://doi.org/10.5194/bg-2022-126-RC1>, 2022

### RC1: 'Comment on bg-2022-126', Olivier Sulpis, 26 Jun 2022

The manuscript presents results from a set of experiments assessing the fate of alkalinity added under various forms to natural seawater. Predicting alkalinity stability is key to quantify the potential of OAE to neutralize atmospheric CO<sub>2</sub>. Hence, this study is particularly timely.

The particularity of this study is that it is composed of many different experiments, providing a vast quantity of results, all quite convincing, and numerous take-home messages. The authors seem to have designed their series of experiments incrementally, guided by the preliminary results obtained along the way. As a result, they went very far into the search for answers to their initial research questions, and the diversity and quality of results they produced is impressive. The discussion section nicely summarizes and explains the key findings, analyzes them in the context of preexisting literature, and offers guidance for implementing OAE techniques. This study is overall a pleasure to read, and it will certainly have broad implications. That said, the fact that numerous experiments were carried on leads to some lack of clarity in the methods. A proper statistical treatment on the collected data is also missing. Finally, the absence of analyses on the recovered precipitates is frustrating, as knowing what these precipitates are and how they form would be capital to better understand how to deal with them.

REPLY: Thank you very much for your detailed and constructive evaluation. Following your suggestions, we implemented CO<sub>2</sub>sys\_v2.5. A comprehensive description of used constants and uncertainties and their resulting errors will be provided in the supplements. If feasible, uncertainties (/error bars) will be added to each plot. As inquired by both reviewers analyses of several precipitated materials were conducted using SEM/EDX. Information about the findings will be added to the manuscript. For detailed replies, please see below.

### General comments:

It is clear from the water chemistry data that carbonate minerals are precipitating. In abiotic experiment II, the authors observed immediate precipitation of centimetric needle shaped precipitates floating at the center of the bottles, suggesting spontaneous nucleation of aragonite. After 1 day, they observed precipitates on the walls of the reactors. They also observed these precipitates on the walls in experiment III. In all other cases, it seems that precipitation happened on preexisting particles, such as natural particles smaller than 55 microns, or minerals added as a TA source. In all those cases, precipitation occurred presumably without the need to nucleate in the first place, as growth on preexisting nuclei. What is precipitating exactly is unknown, but the results shown on Fig. 7 suggest that it may be a phase more soluble than aragonite.

REPLY: “Fig. 7 suggest that it may be a phase more soluble than aragonite” Yes this is correct, as the Omega for aragonite is larger 4. A discussion about ACC can be found in section 4.2.

The majority of CaCO<sub>3</sub> precipitates observed by Moras et al. (under review) were aragonite, and a little of vaterite. It would be enlightening to provide visuals of what the precipitates observed in the present study look like, and analyses of their mineralogy, as other similar studies have done using a combination of scanning electron microscope or energy dispersive X-Rays (Moras et al., under review; Fuhr et al., 2022). At minimum, it is needed to discuss the hypothesis of ACC precipitates in comparison with observations from the Moras et al. study.

REPLY: As suggested, SEM images of the precipitates for experiment IIa were prepared, including EDX measurements. Information will be provided.

There is a lack of proper statistical treatment. Results should be given with an associated uncertainty that should reflect uncertainties arising from (i) the experiments, from (ii) the instruments/analyses and from (iii) the computations (errors on thermodynamic constants, uncertainty propagation, etc.). (i) it seems that only experiment V was conducted in replicates, but any resulting uncertainty quantification is absent in Fig. 7. (ii) in the measurements section, no precision is provided for the instruments used, and even though it is stated that some samples were taken and measured in duplicates, the resulting uncertainties are not provided in the text or shown in figures (apart from Fig.4). (iii) the authors should use the new CO2SYS versions recently published, available in excel (Orr et al., 2018), that allow to include and propagate all uncertainties including those from the thermodynamic constants.

REPLY: As proposed, CO2sys\_v2.5 was implemented, constants: K1/K2 from Lueker et al. 2000; KHSO<sub>4</sub> Dickson 1990; KHF Dickson and Riley 1979, pH scale Total scale; [B]TValue Uppstrom 1974 were used. Like recommended uncertainties were calculated in CO2sys, based on following input conditions: Uncertainties for Salinity were constantly set to 0.1 and 0.020 units for pH, both based on the provided uncertainties of the used calibration standards. Depended on the experimental measuring series the input for TA and DIC uncertainties were individually calculated by the average deviations from the measured Dickson standards for each experiment. The resulting uncertainty outputs of CO2sys\_v2.5 for each parameter (TA, DIC, pH, pCO<sub>2</sub>, Ω(ar), Ω(cal) and Salinity) will be provided.

uncertainty quantification is absent in Fig. 7:

REPLY: Will be provided and implemented in Fig. 7.

some samples were taken and measured in duplicates, the resulting uncertainties are not provided:

REPLY: Double measurements for experiment I+II (only abiotic) are available for TA measurements and will be provided in the supplements. Comment: the average deviation is about +/-2.0  $\mu\text{mol/kgw}$ , just two pairs of measurements reached deviations of +/- 7.5 – which is basically not plottable by using error bars. For experiment V triplicates exist and statistics will be implemented. For all other experiments there are no duplicates or multiple measurements due the nature of the conducted experiment. However, we gain robustness in the interpretation using concentration gradients in the experimental design and by the coherent results. In general, the uncertainties in TA/DIC exist, but are of minor importance and almost neglectable. Using pH probes, as the ones used here, always produces uncertainties which are quite high with around +/- 0.02 units. However, we got constantly relative robust and reproducible measurements.

Reading through the methods section was challenging, due to the large number of experiments and to the lack of details provided, see list of specific comments. In particular, the motivations for the experimental design are not well explained. It would be helpful that the paper reflects the same succession of thoughts and actions that the authors went through when designing their study.

REPLY: Methods and experimental design will be adjusted to improve clarity in the section “introduction”.

#### Specific comments:

L19: precise which minerals and that it does not apply for the deep ocean

REPLY: Change to “Seawater is already supersaturated with respect to calcite and aragonite, ...”

L26: precise that the carbonates mentioned are carbonate *minerals*

REPLY: “... due to carbonate formation if added total alkalinity ( $\Delta\text{TA}$ )...” will be changed to “ ... due to the formation of carbonate minerals if added ...”

L28: precise what kind of slurry

REPLY: The term slurry will be changed to “alkaline slurries”.

L31-32: “unless alkalinity addition shifts the system beyond critical supersaturation levels”: this portion of the sentence is unclear, please rephrase

REPLY: This will be rephrased.

L45: field experiments are lacking but not laboratory studies, although more are needed, please precise that and add relevant references

REPLY: **Will be adjusted.**

L48: modeling studies have also focused on the OAE applied specifically in coastal environments and in oceanic regions of high CO<sub>2</sub> outgassing, e.g., Fakhraee et al., under review, please precise that

REPLY: **Will be adjusted.**

L52: here and after, please precise carbonate *minerals*

REPLY: **Will be adjusted.**

L54: adding one sentence here on nucleation itself being the greatest energy barrier to encompass before crystal growth can continue would be insightful, possibly referring to Sun et al. 2016.

REPLY: **Will be added.**

L60: please precise that the optimal conditions refer to natural, typical sea surface environments

REPLY: **We will change to: “Even under optimal natural conditions for air-sea gas exchange, in a typical sea surface environment, CO<sub>2</sub> equilibration.”**

Methods: there are a lot of experiments and variables, and the methods section describing them is sometimes hard to follow. Table 1 really helps. In addition, in the methods, it would be helpful to explain which research questions enumerated in the introduction relates to which experiment, to guide the reader and explain why all these different experiments were needed.

REPLY: **The research questions raised in the introduction will be better linked to the conducted experiments and the list given in table 1.**

Table 1: precisizing the type of seawater used (North Sea or Gran Canaria) in this table would be useful. In addition, please provide elsewhere eventual details on dissolved silica and phosphate concentrations for both seawaters, as well as the TA and S for the North Sea seawater.

REPLY: The type of seawater will be added to table 1 adding a new column. The available information on DSi and phosphate will be added.

L101: please provide further details, e.g., how much of  $\text{NaHCO}_3$  and  $\text{Na}_2\text{CO}_3^{2-}$  were added, what was the  $\text{pCO}_2$  of the solution and of the air?

REPLY: Added amount  $\text{NaHCO}_3$  and  $\text{Na}_2\text{CO}_3$  for each treatment was calculated separately for each treatment (adjusted to the target TA level and the volume of water in every reactor). Detailed information will be provided in the supplements. For the equilibrated experiments 420 ppm were set.

L110: is there a reason for not using the same Dickson bottles for all experiments?

REPLY: Yes, limited resources during field work (not enough bottles to conduct two experiments simultaneously – biotic/abiotic at the same time) and the volume of Dickson bottles is too small for the additional sampling during the experiment for biotic measurements.

L114: please precise why should airspace be avoided in the case of Ib, since this experiment is in principle equilibrated

REPLY: To prevent gas exchange, as organism might impact the carbonate system due ongoing life processes.

L115: why not keeping the same alkalinity addition steps than for the abiotic experiments?

REPLY: In parallel other research questions were answered during the biotic experiments, benefiting from smaller TA steps of 150.

L119: replace “gained” by “retrieved” or “collected”

REPLY: Replaced gained by collected.

L119: at this stage we have no information on what those precipitated carbonates are. What are the mineralogy, grain size, composition?

REPLY: SEM/EDX data will be provided, see above. Still, we do not have any information about the type of Ca-carbonate. Size distribution and chemical composition can be delivered.

L120: that the experiment took place in the harbor introduces temperature as a new and potentially important variable: what was the temperature and its difference between day and night, and how were the effects of temperature kept track of when interpreting the results and comparing them to experiments carried out in temperature-controlled environments?

REPLY: Experiment III tested the general concept of inducing precipitation, there was no tracking of temperature or any comparison to lab experiments. The average temperature range in the harbor area during the experiment was between 20-23°C.

L121: how much liquid was sampled?

REPLY: In general, 150 ml each sampling day (day 0-10), 40 ml day 46 and 90.

L121: replace “the 10 days of processing” by “10 days of experiment,”

REPLY: Will be done.

L138: was only the liquid sampled or some solid with it?

REPLY: Just liquid.

L142: how many Erlenmeyer-flasks were there and why?

REPLY: Three flasks per timestep (triplicates) plus one control flask – 12 timesteps => 48 flasks.

L142: it is a different type of seawater than for the previous experiments. What was the composition of that seawater? What was the temperature for those experiments?

REPLY: The composition and temperature of the used North Sea water: a) TA 2297  $\mu\text{mol/kgw}$ , b) pH 8.05, c) DSi 16  $\mu\text{mol/L}$ , d) temperature 23 °C (constant lab conditions).

L143: do “timesteps” here correspond to sampling times? Please clarify, and precise how much liquid was sampled each time.

REPLY: The timesteps describe the time material was in contact with water, so the duration between adding the mineral powder to the flasks and filtering it out again. We will

replace the word. Out of 300 g of seawater per flask roughly 200 ml were saved, stored and used for measurements.

L145: please precise why the need for a shaking table and why 125 rpm.

REPLY: 120 rpm, is the maximum frequency without risk of splashing. The idea was to simulate favorable conditions for gas exchange as possible.

L145: delete "value"

REPLY: Will be done.

L149: how many different solid masses were investigated? How much solid was added in each? Were the experiments conducted in triplicates?

REPLY: 15 masses + 1 control, per 300 g of seawater following masses [g] of Mg(OH)<sub>2</sub> were added 0, 0.001, 0.005, 0.01, 0.02, 0.03, 0.05, 0.125, 0.2, 0.500, 0.800, 1.000, 1.500, 1.750, 2.000 and 4.000 g. No triplicates, just one flask per treatment.

L152: replace "as" by "than"

REPLY: Will be done

L155: please provide more details on how airtight filtrations were done

REPLY: For experiment I, II, V and VI Nalgene filtration bottles were used (0.2 µm filter). Immediately after the end of each experiment the whole water volume was carefully purred atop the filter in the filtration vessel. For experiment III and IV a peristaltic pump and an attached 0.2 µm syringe filter was used. In general, unnecessary contact to the atmosphere of non-CO<sub>2</sub>-equilibrated samples have been avoided as much as possible.

L157: were dissolved silica and phosphate not measured in the two types of natural seawater used? If not, how were their contributions to total alkalinity removed in CO<sub>2</sub>SYs?

REPLY: DSi and phosphate were not measured during this experiment. Other working groups on Gran Canaria measured Silica and phosphate in the water of the Mesocosms placed in the harbor of Taliarte. North Sea water was measured for DSi (16 µmol/L) in another

project conducted with the same batch of water. Neither DSi nor phosphate were included in the CO2sys calculations.

L165: what scale was the pH expressed on and how was this conversion made?

REPLY: We used the log-scale with the basis 10 for the pH. The pH is the negative logarithm of the hydronium activity.

L183: the term “carbonate chemistry remained stable” is unclear. In fact, it seems that this last sentence does not bring anything new relative to what was said earlier in that paragraph and could be removed.

REPLY: Will be removed.

Fig.3 I suggest adding dashed lines in the background showing the 2:1 ratio, as a reference for the reader to assess the decline in TA:DIC ratio observed in the experiments

REPLY: We provided a grey “2:1” arrow for that purpose in Fig. 3. To ensure consistency it would be possible to transform it into a dashed line like in Fig. 1 & 2.

Fig.3: is “added TA” similar to “ $\Delta$ TA”? If so, precise or update to  $\Delta$ TA for consistency with the text and the other figures

REPLY: “added TA” occurs in Fig. 1, 2 and 5. This will be adjusted.

L243: were floating precipitates, as described in experiment II, also observed here?

REPLY: In comparison to experiment II (non-CO2 equilibrated) experiment III is CO2-equilibrated, accordingly Omega(aragonite) values are much lower and, in this case, even below the critical threshold for precipitation. As described, precipitation was only observed at the walls after considerable time.

L246: “due to other surfaces abundant”: this portion of the sentence is unclear, please rephrase

REPLY: Will be rephrased.



Fig.5: are the black/grey, and light blue/dark blue lines replicates? This is not indicated. If so, it would be clearer and make more sense to combine them and use the spread between them as a measure of uncertainty (which should also include and reflect the other uncertainty sources).

REPLY: No, each line represents an independent treatment. Description of Fig. 5 will be adjusted. Black (day 0), red (day 1), dark blue (day 4) are abiotic treatments and grey (day 0), brown (day 1) and light blue (day 4) are biotic treatments (same color scheme like Fig. 1 and 2).

L255: can more information on the dissolution rate of brucite be provided, e.g., approximately how much was left after 4 days?

REPLY: No additional data are available.

L260: the use of “aspired” seems odd, perhaps change to “targeted”

REPLY: Will be changed.

L273: what did the aggregates look like, and couldn't they be disaggregated with gently shaking the bottles?

REPLY: Aggregates looked like flakes in a snow globe (mm to cm sized) and could not be disaggregated, even with intense persistent shaking.

L299: the title of section 3.7 mentions CO<sub>2</sub> equilibration, but Table 1 says the opposite. If I understand correctly, there is an ambiguity here because Ca(OH)<sub>2</sub> dissolving induces a disequilibrium with the atmosphere which is then compensated by CO<sub>2</sub> uptake from the air due to the open flasks. Here and in general, it would be good to distinguish more clearly between “CO<sub>2</sub>-equilibrated” as in preparing a solution with amounts of TA and DIC targeted to produce a certain pCO<sub>2</sub> that would match that of the air and “CO<sub>2</sub>-equilibrated” as in actual exchanges between air and seawater until both are at equilibrium.

REPLY: “CO<sub>2</sub>-equilibrated” will be rephrased to avoid misunderstandings.

L327: please rephrase “wave line”

REPLY: “directly below the sea surface”

L342-343: “The reason is that the carbonate system (based on *W* of the bulk solution) stayed within boundaries to avoid mineral precipitation” and L345-346: “The reason is that, for the same DTA, perturbations of the carbonate system that affect mineral precipitation are much stronger in this non-equilibrated scenario”: because it is a very important issue for anyone implementing OAE to understand, I believe those two sentences should be explained in more details

REPLY: **Additional explanation will be provided.**

L360: the threshold observed by Morse and He is for nucleation, which is more specific than spontaneous carbonate formation. It should be made clear here and in the intro that nucleation and growth are two different things with different energy barriers

REPLY: **Nucleation and growth will be defined properly to avoid ambiguities.**

L367: “between adding carbonate precipitates and not” this portion of the sentence is understandable but could be better phrased

REPLY: **Will be rephrased.**

Data availability: the data should be made available at the time of submission for reviewers to be able to see it

REPLY: **All generated data were plotted. A supplement xls-file will be provided.**

## References:

Fakhraee et al., under review, <https://doi.org/10.21203/rs.3.rs-1475007/v1>

Fuhr et al., 2022, <https://doi.org/10.3389/fclim.2022.831587>

Moras et al., under review, <https://doi.org/10.5194/bg-2021-330>

Orr et al., 2018, <https://doi.org/10.1016/j.marchem.2018.10.006>

Sun et al., 2016, <https://doi.org/10.1073/pnas.1423898112>

**RC2: 'Comment on bg-2022-126', Scott C. Doney, 09 Jul 2022**

**RC3: 'Reply on RC2', Scott C. Doney, 10 Jul 2022**

Overall this is an important study presenting a series of small-scale experiments regarding the potential for mineral precipitation during ocean alkalinity addition, with the possibility of mineral

formation significantly counteracting the intended net change in total alkalinity. The authors do a good job framing the experimental results of the study in terms of the efficiency or efficacy of ocean alkalinity enhancement approaches. The study highlights a number of key nuances in terms of the amount and character of the added alkaline solutions or alkaline solid phases. I did not identify any significant conceptual issues with the study or with the main data analysis approaches and results presentations. The 6 sets of experiments involve a wide range of conditions, source waters, materials added, time durations, and underlying scientific questions. Interpreting these different experiments, therefore, requires some effort, and most of my comments below involve possible clarifications to the text that may help improve the readability of the study.

REPLY: Thank you very much for your supportive comments, which help to improve comprehensibility a lot. Especially the misleading use of "Delta TA" will be adjusted. As inquired by both reviewers' analyses of several precipitated materials were conducted using SEM/EDX. For detailed replies, please see below.

Abstract

Line 25-27:

"The application of CO<sub>2</sub>-equilibrated alkaline solution bears the lowest risk of losing alkalinity due to carbonate formation if added total alkalinity (Delta TA) is less than 2,400 micromol/kgw. The addition of reactive alkaline solids can cause a net loss of alkalinity if Delta TA > 600 μmol/kgw (e.g., for Mg(OH)<sub>2</sub>)."

If I am interpreting the text correctly, the notation "Delta TA" is used in three different, and somewhat confusing, ways:

- 1) initial amount of total alkalinity added to the seawater solution prior to mineral precipitation (e.g., Delta TA<sub>1200</sub>, etc.
- 2) net change in total alkalinity reflecting both the amount of alkalinity added as well as the loss of alkalinity due to mineral precipitation (e.g., Delta TA in Figure 8).
- 3) loss of alkalinity due to mineral precipitation following alkalinity addition (e.g., Figure 4)

The addition of an equation along the following lines may be useful:

net TA change = TA<sub>final</sub> - (TA<sub>initial</sub> + TA<sub>added</sub> + TA<sub>loss</sub>)

[RC3: 'Reply on RC2', Scott C. Doney, 10 Jul 2022] rephrased:  
Delta<sub>net</sub> TA = TA<sub>final</sub> - TA<sub>initial</sub> = Delta<sub>add</sub> TA + Delta<sub>loss</sub> TA

It would be very helpful to avoid multiple usages for Delta TA and be more specific about the quantity being presented.

REPLY: Notation "Delta TA" will be adjusted generally to avoid misunderstandings; equation will be implemented for clarification.

Also, as a general comment it would be helpful to clearly state at each use of the term what is meant by "loss of alkalinity". In some extreme cases in the experiments "loss" refers to a "net loss" relative to the natural background concentration. In other situations, "loss" appears to refer loss of alkalinity relative to the expected increase in alkalinity following the addition of alkaline solutions or solids.

REPLY: Will be rephrased.

Line 43

"(NAS,2021)" (also Line 655 in references)

Please update the citation and reference to "NASEM, 2022" for the final report:

National Academies of Sciences, Engineering, and Medicine, 2022: A Research Strategy for Ocean-based Carbon Dioxide Removal and Sequestration, Washington, DC, The National Academies Press, <https://doi.org/10.17226/26278>

REPLY: Will be done.

Line 49:

"negative TA could be a consequence"

I think this should read: "negative net change in TA could be a consequence"

REPLY: Will be adjusted.

Line 70:

The authors present a well-formulated set of research questions. Very helpful.

REPLY: Thank you.

Lines 74 to 85 and Table 1:

The study used quite a number of different experiments with different conditions for filtering, material added, storage, etc. The list of the six main experiments in the text and in Table 1 provide a reasonable description, but the details perhaps could be amplified. In particular, a better link early in the Methods

section to how the experiments address the science questions at the end of Section 1 (Line ~70) would be helpful. This is done well for filtering in Line 88-93 but could be done for other conditions within the experiment introduction section.

REPLY: For better understanding the research questions L66-72 are going to be connected to the list of experiments L76-82 by adding a short description to each point (I-VI) to clarify the thought process.

Line 96

For CO2SYS, a brief description should be added of any choices made in equilibrium coefficients, carbonate saturation coefficients, etc.

REPLY: A description of all used equilibrium constants will be added. (as also requested by [RC1: 'Comment on bg-2022-126', Olivier Sulpis, 26 Jun 2022]) version 2.5 has been implemented, including uncertainties for each constant, accordingly a much more detailed explanation will be given in the supplements)

Line 99-109

It may be beneficial for readers who are not seawater carbonate chemists to add a sentence or two on the difference chemically between "equilibrated" (Ia) and "non-equilibrated" (IIa) experiments using NaHCO<sub>3</sub>/Na<sub>2</sub>CO<sub>3</sub> versus NaOH. While perhaps obvious to chemists, the distinction may not be immediately obvious to the full range of potential readers or why the added alkaline solution is clearly equilibrated with the atmosphere CO<sub>2</sub>.

REPLY: For clarification the concept of "equilibrated" and "non-equilibrated" will be explained in more detail in the introduction.

Line 134

From the text, it appears that the particle experiments incorporated mixing only for 5 minutes at the beginning of the experiment. Some comment is needed on the affect of no mixing over several days in the tubes for particle dissolution kinetics and particle boundary layer dynamics relative to what would happen to particles added to the real ocean. I am not sure the kinetics are actually that comparable to ship disposal as mentioned in line 139 where most schemes assume a turbulent dispersion component during discharge plus natural background turbulence in the water column.

REPLY: A comment will be added stating that the dissolution kinetics are probably lower as compared to water being continuously mixed with untreated water. The term ships-disposal is referring to the amount of solid material per kg of water added.

Line 141

"This experiment focused on redissolution of alkalinity"

Perhaps consider an alternate phrasing:

"This experiment focused on addition of alkalinity due to the redissolution of alkaline precipitates"

REPLY: Will be rephrased.

Line 148-149:

"The experiment was set up to check if potential threshold values for TA loss/gain after 24 hours exist in case larger amounts of solids are dissolved than in the previous experiments, resulting in the loss of TA."

Perhaps add clarification "for net TA loss/gain with the combined effects of alkaline material addition plus mineral precipitation ..."

REPLY: Will be adjusted.

Line 154:

Are the particles captured on the filters for each experiment analyzed for the amount and character of the material? In particular, was anything done to analyze precipitates?

REPLY: Filters exist for experiment IIa (day 0, 1 and 4) and experiment V. SEM-images have been taken now (including EDX-analyses) for preparing the revision and will be implemented.

Line 169:

which coefficients are used for carbonate saturation state?

REPLY: A detailed description of all constants will be given. See above.

Line 172-175:

Perhaps worth noting that the notation Delta TA\_2400 (or Delta TA\_1200) is only approximate as the actual increases are not exactly 2400 or 1200 but are close, for example 2400 is actually 4750 - 2411 = 2339.

REPLY: Delta TA 2400 is a label to characterize the target change. The simplification of choosing the target TA level is supposed to make treatments of different experiments comparable. A short explanatory note will be added to clarify.

Line 178-179:

"Neither in the abiotic (0.2 micrometer filtered) nor in the biotic (55 micrometer filtered) treatment, including living planktonic organisms, a detectable loss in TA could be observed after 1 and 4 days."

Perhaps reorganize this sentence as:

"No decidable loss in TA was observed after 1 and 4 days following the initial TA addition at time 0 for either the abiotic (0.2 micrometer filtered) or in the biotic (55 micrometer filtered) treatment, including living planktonic organisms."

Also, this sentence helps better understand some of the earlier text in the Abstract and the Methods regarding the meaning of "alkalinity loss". Perhaps a sentence or two could be added along the lines of:

"The seawater TA was monitored after the initial alkalinity addition to determine if any alkalinity was lost subsequently do to mineral precipitation processes."

"Perhaps a sentence or two could be added along the lines of":

REPLY: Text will be adjusted, as suggested.

Line 180

"while carbonate chemistry remained stable in all TA treatments [after the initial alkalinity addition]."

From Figure 1 there does appear to be a noticeable, if relatively small, decline in pH and aragonite saturation state in the abiotic-equilibrated experiments. This should be mentioned briefly in the text.

REPLY: Part1: "after the initial alkalinity addition" will be added.

Part2: decline will be mentioned in the text.

Line 185, figure 1

The change in seawater carbonate system in the experimental treatments is over-determined with the measurements of 3 variables of the seawater carbonate system (pH, alkalinity, and DIC). Would be good to clarify for plots what is measured versus what is calculated from CO2SYS (only saturation state?) and if the measurement and CO2SYS calculated values are consistent.

REPLY: There is no over-determination: for Exp. Ia TA and pH, for Ib TA and DIC were used to calculate the other third parameters of the carbonate system. This will be clarified better in the caption like done in Fig. 2.

Minor point but in all of the captions it would be good to add the number and name of the experimental treatment to connect back to the notation on Table 1. So add Ia and Ib in the bolded text in the caption.

REPLY: Captions will be adjusted as proposed.

Line 221-226:

"Consequently, while precipitation was absent in the abiotic and biotic set-up in the equilibrated treatment, strong alkalinity loss through precipitation occurred in the non-equilibrated experiment ... The loss ratio in TA:DIC was about 2:1 (Fig. 3b, d) in all treatments, indicating the loss of alkalinity due to the precipitation of carbonates."

This is a good line and perhaps something like this would be helpful in the Abstract.

REPLY: Information about the 2:1 loss ratio will be added to abstract.

Line 250, Figure 4:

"development of alkalinity loss (Delta TA) over time"

Here is a third different usage of "Delta TA", here the "loss" due to mineral precipitation.

REPLY: As mentioned above, for clarification we will adjust the wording of Delta TA.

Line 330, Figure 8

A point about notation. Here Delta TA label on y-axis of both sub-panels indicates the net change in alkalinity rather than the use of the notation Delta TA in some previous figures for TA initially added via solution or solids. Perhaps change label to "net Delta TA".

REPLY: Correct, the label will be adjusted as suggested.

Line 335

Section 4.1 General discussion



This is a well written summary of the many of the key results.

REPLY: Thank you very much.

Line 377-378:

"Not only particle surface processes on added solid alkaline particles are to be considered."

Perhaps rephrase as, "In addition, particle surface processes ..."

REPLY: Will be rephrased.

Line 385:

"... partly be released again. Here probably facilitated ..."

I think these should be a single sentence (second sentence may be a sentence fragment), so

"... partly be released again, here probably facilitated ..."

partly be released again. Here probably facilitated

REPLY: Text will be adjusted.

Line 408:

"... lead not to a positive TA ..."

perhaps change to

"... did not lead to a positive change in TA ..."

REPLY: Will be done.

=====

[RC3: 'Reply on RC2', Scott C. Doney, 10 Jul 2022], included into the text above:

" I noticed an error in my post near the beginning. The base equation should be:

Delta\_net TA = TA\_final - TA\_initial = Delta\_add TA + Delta\_loss TA ”

See reply to “Abstract Line 25-27”