Reviewer comments – EGUsphere 2026-982

Reply to comments on EGUSPHERE-2026-982
Implemented Changes

The manuscript egusphere-2026-982 assesses the impact of varying alkalisers on the broader carbon pool. It focuses on three main alkaline feedstocks, NaOH, CaO and a mix of $NaHCO_3$ and $CaCl_2$, which are used to increase alkalinity between 500 and 1000 µmol kg^{-1}. It then explores the evolution of the various carbon pools, both dissolved and particulate, as well as dissolved O_2 and chlorophyll a. A last interesting aspect is the use of heavy carbon (^{13}C) to enhance the analysis of the carbon budget. The paper requires some revisions with emphasis on consistency before being ready for publication, but I strongly believe that once my general comments and suggestions, as well as the specific ones, have been addressed, the manuscript will be more ready for publication.

We thank you for your thorough and helpful comments and detailed review. We have tried to address all your comments.

General comments:

The experimental setup is rather straight forward and should be easily replicated, though some more information would be beneficial. First and foremost, I am questioning the surprisingly consistent salinity measurements in Table 1? Considering the study area and the inputs from both seawater (which one could assume to have a salinity of 35) and the Southern California Bight (which based on quick reading seems to have slightly lower salinity), I am doubtful the water collected at different points I time will have a salinity of exactly 35. This may need further research/analysis/discussion.

All salinities were measured with a refractometer with delineations of 1 ppt; hence we rounded to the nearest whole number. We did not previously report the uncertainty of this scale which is ± 0.5 ‰ (parts per thousand), which we have now added to the manuscript. Given the poor resolution for these measurements, we have removed these values from Table 1 but mention the rather coarse salinity measurements in the text.

Some more work is also needed in the "2.2 Alkalinity Types" and "2.3 Experimental Set Up" sections. First and foremost, the choice of alkaline source to simulate AWL is questionable, given that limestone, $CaCO_3$, would dissociate into Ca^{2+} and CO_3^{2-}. I am therefore wondering why using $NaHCO_3$ (which need to be added twice as much to match the TA increase from $CaCO_3$ dissolution) instead of Na_2CO_3, which would have provided CO_3^{2-} ions as when $CaCO_3$ dissolves.

In this study we were exploring two different styles of alkalinity enhancement – (1) a base addition method and (2) accelerated weathering of limestone (AWL). In the case of AWL, the CO_2 has already reacted with water forming carbonic acid, which, when encountering $CaCO_3$, results in byproducts Ca^{2+} and $2HCO_3^-$, hence we chose to use $CaCl_2$ and $NaHCO_3$ to simulate this scenario. We have also added equations (Eq. 2 & Eq. 3) to emphasize this distinction.

$$\boldsymbol{CO_2} + H_2O + Ca\boldsymbol{CO_3} \leftrightarrow Ca^{2+} + \boldsymbol{2HCO_3^-} \tag{2}$$

$$2Na\boldsymbol{HCO_3} + \boldsymbol{Ca}Cl_2 \leftrightarrow \boldsymbol{Ca^{2+}} + \boldsymbol{2HCO_3^-} + 2Na^+ + 2Cl^- \tag{3}$$

Furthermore, I fully agree with the statement made in lines 142-143 about deciding against introducing particles. However, some information on the stock solution preparation is missing, most importantly the targeted molarity of said solution. This leads to a rather abrupt end to the paragraph stating that somewhere between 100 and 160mL were added. While I agree that this only represent between 2 and 3.2% of the final volume (assuming 5L bottles, if I am not wrong?), the additions are significantly different.

We agree that specific stock solution molarity and information on volumes added was missing. This has now been added to the manuscript.

The handling of carbonate chemistry samples would also benefit from further details. For example, it is stated that a portion of the filtrate is used for DIC (lines 207-208) but without any further information. DIC sampling is very sensitive to exchanges with the atmosphere, particularly with alkaline samples. Some standard operating procedures SOPs (e.g., Dickson 2007) have been published to assist scientists with such issues. I recommend adding more information on the sampling procedures and, if different from such SOPs, argue the limitations and potential changes in samples concentrations.

Noted; references to SOPs have been added to the manuscript. In short, we are very careful that DIC sampling avoids the loss of CO_2.

The results section is significantly large and clearly shows the efforts and work put into this research. Overall, it is well written and there are no major gaps, though there are some points that require further justification. A very first comment is regarding the section "3.1.1 Observing Short Term Impacts at Extremely Low Alkalinity Additions". I believe that some information presented here would fit better in the Materials and Methods section (here named "Study Area & Methods" which may need to be edited, though this is to the discretion of the editor).

We thank the author for appreciating the research effort. In line with your suggestion, we have moved specific information about the experiments conducted on Catalina Island (in Section 3.1.1) to "Study Area & Methods"

Furthermore, I am also questioning the measured alkalinity. Following the description, one part of 5,000 µmol kg^{-1} water was mixed to 600, 300 and 150 parts for each dilution. Assuming the TA of the water used to dilute was the same as the control, a quick calculation reveals that expected final TAs should have been 2227.6, 2232.3 and 2241.5, respectively. Similarly, since the alkalinity addition was performed with bicarbonate (if I am correct, line 282), the changes in DIC should have been very close to the changes in TA which is not the case here. This needs to be verified and discussed. The remaining part of the Results section are good and mostly fine in my opinion.

Our manuscript included a rounded-up concentration of the stock solution that was used for the experiments in Section 3.1.1. We have corrected this value to the precise concentration of 4754 µmol kg^{-1} rather than the 5000 µmol kg^{-1} reported. Additionally, in this section we reported uncertainty values as the standard deviation of replicate measurements. We have updated this error to include the instrument error and the CRM correction used on these samples. In all data used in other sections the standard deviation of replicates surpasses the instrumental error; hence, instrumental errors have not been plotted but included in the datasheet – altasea-all-summary-datasheet-light.csv.

The differences between the expected final TA and the measured final TA are within these uncertainties, but we have included an additional column in Table 2 to reflect these differences.

A small suggestion would be to report ΔTA and ΔDIC instead of the values. This would make the reading of the graphs and especially small changes more easily visible. This would also help the reader see whether the targeted changes in TA in all experiments were reached.

We have considered this suggestion; however, we want to highlight the variability of TA and DIC in the replicate measurements—using ΔTA and ΔDIC values would compound uncertainties in the difference.

Another important point on the graphs is the mislabelling of the calcium ions, it should read Ca^{2+} rather than Ca^{+2}.

This change has been made throughout the text.

Finally, the discussion section is a little short but interesting as it combines several key OAE studies and puts the results of this research into context. Some edits may be suggested into this section, and a deeper interpretation of the data and implications for the OAE field should be done. In the paragraph "4.1 Secondary Precipitation of $CaCO_3$", the observed patterns are compared with other studies, but the section lacks discussion about time. While I agree that the risk for $CaCO_3$ precipitation is directly linked to surface area and ΩA, a third important factor is time. One could have expected that if the experiments had run for longer, $CaCO_3$ precipitation may have eventually occurred. This should be added to the paragraph and can done by comparing the duration of the experiments presented with the literature mentioned.

We agree with the reviewer that time is another important factor—we have another two manuscripts in preparation that deal with this topic. To help clarify this point we have rephrased Section 4.1 Secondary Precipitation of $CaCO_3$ and $Mg(OH)_2$. The restructuring of this section was done by summarizing the following tables as text. We do not include these tables, we provide them to the editor and review as evidence of our intent to summarize other works.

Summary of studies – Onset of $CaCO_3$ Precipitation

NO PRECIPITATION OBSERVED

Study	**Feedstock**	**Media**	**$\Omega_{aragonite}$**	**Temperature (°C)**	**Time (days)**
Hartman et al. 2023	$NaHCO_3$ + Na_2CO_3	0.2 µm filtered SW	< 15	24	4
		55 µm filtered SW	~ 6	24	4
Suitner et al. 2024	$NaHCO_3$	0.2 µm filtered SW	< 15	12-16	20

	+ Na_2CO_3	55 µm filtered SW	< 15	12-15	20
Moras et al. 2022	Na_2CO_3	0.2 µm filtered SW	~ 9	21	40
		0.2 µm filtered SW + Quartz crystals	~ 9	21	5
Hartman et al. 2023	NaOH	0.2 µm filtered SW	<20	24	4
		55 µm filtered SW	<20	24	4
Suitner et al. 2024	NaOH	0.2 µm filtered SW	< 15	11-13	25
		55 µm filtered SW	~19	10-11	25
Ringham et al. 2024	NaOH	Unfiltered SW	< 30	19-21	20-60
Moras et al. 2022	CaO	0.2 µm filtered SW; solid feedstock	5	21	40
	$Ca(OH)_2$		4	21	30

PRECIPITATION OBSERVED

Study	**Feedstock**	**Media**	**$\Omega_{aragonite}$**	**Temperature (°C)**	**Onset of Precipitation (days)**
Suitner et al. 2024	$NaHCO_3$ + Na_2CO_3	0.2 µm filtered SW	15-45	12-16	1-20
Moras et al. 2022	Na_2CO_3	0.2 µm filtered SW + Quartz crystals	~ 9	21	>5
Hartman et al. 2023	NaOH	0.2 µm filtered SW	~16-19	24	0-1
		55 µm filtered SW	~15-17	24	0-1
Suitner et al. 2024	NaOH	0.2 µm filtered SW	> 15-30	11-13	0-25
		55 µm filtered SW	> 19-30	10-11	1-15
Ringham et al. 2024	NaOH	Unfiltered SW	> 30	19-21	1
Moras et al. 2022	CaO	0.2 µm filtered SW; solid feedstock	~7	21	0.2
	$Ca(OH)_2$		~7	21	0.2

One other point which could help the reader follow better is referring to the experiments in a consistent manner. For example, the experiments are identified by the TA additions performed earlier on, i.e., +500 and +1000. In the discussion, the experiments are referred to as “pH<8.55” (line 542 for example). It would be easier to follow if each experiments labelling was constant throughout.

We have corrected references to our experiments throughout the document to be more consistent. In the discussion, in addition to the change in alkalinity we have also included the pH range of these experiments to improve comparison with other studies who focus on changes in pH.

Finally, the section would benefit from a paragraph discussing the limitations of this research (see comments above). This will reinforce the research outreach and will give interesting guidelines for other researchers that may be interested in running similar experiments.

We have added some more information on the limitations of assessing effects to the ecosystem with our measured parameters and experimental set-up. We have added more information in Section 2.3 Experimental Set-Up and Section 2.2 Alkalinity Types.

Specific comments:

Line 1: calcium ions should be written (conventionally) as Ca^{2+} instead of Ca^{+2}. Please edit throughout the text and graphs.
This has been corrected in the text and figures.

Lines 17-19: I believe as per Biogeosciences guidelines that one should limit the use of acronyms in the abstract.
All abbreviations have now been removed from the abstract.

Line 21: shouldn't this read "selected experiments"?
This line was rephrased for better flow.

Line 26: the acronym C for carbon is not introduced before
C now written out as carbon in the abstract.

Line 29: I believe the concluding section of the abstract is repeating lines 23-24. The abstract may need to be rephrased to enhance flow.
Repetition was reduced, and abstract was rephrased.

Line 32: consider "atmospheric carbon dioxide"
Added.

Line 33: the acronym IPCC needs to be introduced
Corrected.

Line 48: here the use of the acronym OAE would be suitable, please edit throughout
Corrected.

Line 49-51: this sentence is somewhat too broad? Maybe consider "understanding the impact of different alkaline feedstocks and concentrations on the ocean chemistry and ecosystems is still limited."
Rephrased.

Line 55: here the use of the acronym $CaCO_3$ would be suited as well, please edit throughout the text
Changes made throughout the text.

Line 69: I believe "when" should be "whether" instead
Suggestion accepted.

Line 73: here, Ω could be introduced, and also Ω_A with a quick explanation why choosing aragonite over calcite (aragonite is the morphotype that precipitates in seawater rather than calcite, see Morse 2007 for example)
Suggestion accepted and reference of (Morse et al., 2007) was added.

Line 79: here, the acronym DIC is used without introduction before. Consider "showing the loss of total alkalinity (TA) and dissolved inorganic carbon (DIC)…", this way both are introduced together and can be substituted later in the text
Added.

Line 82: I am not fully sure, but I believe that the agreed upon/conventional labelling is Ω_A instead of $\Omega_{aragonite}$, though this may need to be double checked. If so, consider changing
We have seen this value labelled as Ω_A / $\Omega_{aragonite}$ / Ω_{arag} in the literature. We would prefer to refer to this as $\Omega_{aragonite}$ for readers that may not be familiar with conventional terminology.

Line 83: I believe the sentences need to be reformulated. It is not "$NaHCO_3$/Na_2CO_3 addition" that is "stable up to $\Omega_{aragonite} \sim 20$" but rather the seawater alkalinity after addition of said alkaline feedstock. Please rephrase
Noted and corrected.

Line 89: I believe it was both CaO and $Ca(OH)_2$ though this may need to be double-checked
Agreed, a correction has been made.

Line 92: you could consider mentioning and discussing quickly Suitner et al., 2025 (https://doi.org/10.5194/egusphere-2025-381) that provides a first attempt at characterising the precipitation pattern as a function of ΔTA
We thank the reviewer for highlighting this interesting paper. We have added this reference as (Suitner et al., 2025 Preprint) in the text.

Line 93: one could argue that the research has grown exponentially rather than steadily
We have not conducted a literature survey quantifying publication vs. time so, to be safe, we edited our text to describe the growth as "rapid".

Line 95: consider mentioning the acronym LA Harbour used later in the text
Noted.

Line 100: here I would argue that both TA and DIC are increased, not only DIC
This statement has been changed to directly state that $[Ca^{2+}] + [HCO_3^-]$ are being added. Further details are included in Section 2.2 Alkalinity Types.

Line 134: here, “Sodium Hydroxide” is introduced while earlier, the acronym NaOH is already used (e.g., lines 81, 86, …). Please review for consistency.
Noted.

Line 135: CaO is commonly referred to as “quicklime”, please edit. Also, is there any information on the purity and potential traces of $CaCO_3$?
Suggestion noted, more information about the purity of chemicals has been included in Section 2.2 Alkalinity Types.

Line 136: I am not sure about the formulation “purposeful weathering” please elaborate
Here we were differentiating between natural carbonate (limestone) weathering and a geoengineering term labelled ‘accelerated weathering of limestone’ where the limestone is dissolved to neutralize pre-concentrated CO_2 dissolved in seawater. This section has been rephrased.

Line 138: this is not correct, when this occur, the water is enriched with Ca^{2+} and CO_3^{2-}. Though the latter reacts with CO_2 to give 2 HCO_3^-, the dissolution of $CaCO_3$ lead to Ca^{2+} and CO_3^{2-} additions. Please edit
See earlier comment about this…[This section describes the reaction of ‘accelerated weathering of limestone’ which is what we are trying to replicate as one of the alkalinity enhancement methods. We have added some equations to specify the reaction of AWL, where Ca^{2+} and HCO_3^- are added in a 1:2 ratio and how we mimic this ratio with the addition of calcium chloride + sodium bicarbonate.]

This section now reads as:

The other method considered here is accelerated weathering of limestone (AWL) (Dong et al., 2025; Kheshgi, 1995; Rau and Caldeira, 1999) where atmospheric CO2 and water are pumped through a limestone reactor to neutralize the CO2. The outflow water is enriched in the byproducts of this reaction (Eq. 2) – Ca2+ and HCO_3^- ions. In this method the CO2 is neutralized to alkalinity before it enters the seawater, as opposed to being neutralized in-situ during air-sea gas exchange with the base addition method. We mimic AWL with the addition of Calcium Chloride (CaCl2, Sigma-Aldrich, Missouri. Assay ≥ 97%, Batch No.: SLCL2754) and Sodium Bicarbonate (NaHCO3, Sigma-Aldrich, Missouri. ACS Grade, Batch No.: SLCM3421) with a molar ratio of 1:2 for Ca2+:HCO_3^- (Eq. 3).

$$\boldsymbol{CO_2} + H_2O + \boldsymbol{CaCO_3} \leftrightarrow Ca^{2+} + 2\boldsymbol{HCO_3^-} \qquad (2)$$

$$2\ Na\boldsymbol{HCO_3} + \boldsymbol{Ca}Cl_2 \leftrightarrow \boldsymbol{Ca^{2+}} + 2\boldsymbol{HCO_3^-} + \boldsymbol{2Na^+} + \boldsymbol{2Cl^-} \qquad (3)$$

Line 145: here the reader needs information on the targeted molarity for reproducibility
Molarity of stock solutions added.

Lines 184-186: here it would be interesting to know how the addition was performed. Was it with a pipette? Where the bottles mixed?
The following information was added:

Bottles were opened, 50-150 mL of seawater was removed; water was pulled out of the alkalinity stock solution foil bags into volumetric syringes and injected into the bottles. Any remaining headspace was once again filled with seawater; bottles were sealed and then inverted five times before being tied to the dock.

Line 187-189: how was this tracer added? Was it prepared as a stock solution? And though I understand the idea of increasing ^{13}C for detection, the statement “a couple hundred per mil” is not sufficient for publication, this should be precise. One should report the targeted enrichment to enable proper carbon budget analysis.
We added more information about the preparation of the tracer solution, volumes added, and the resultant change in 13C to the second paragraph of section 2.2 Alkalinity Types.

Lines 207-209: as mentioned above, the handling of carbonate chemistry samples may significantly alter the measurement of parameters. This section needs more details.
Sampling procedure was conducted in accordance with established SOPs (Dickson et al., 2007), and further details have been added in Section 2.3 Experimental Set-Up and 2.4 Sample Processing

Line 213: if I understood correctly, 5L bottles were used. About 250mL was removed for filtration and another litre for genomic analysis. It stated that 4.5L were remaining, shouldn’t it be 3.5L instead?
The vendor sells these bottles as “5 L Thermo Scientific Nalgene PETG bottles” but these bottles were filled with no headspace, so the total volume with no headspace was closer to 6L. We added this information to avoid further confusion.

Lines 231-232: was it a single point standard curve?
An eight-point standard curve is used for this analysis. Information has been added.

Line 285: I believe the “and” should be removed
Noted.

Line 294: as mentioned above, the results rise some questions. Assuming starting TA is 2223 µmol kg^{-1}, adding 1:600 of 5000 µmol kg^{-1} would lead to 599 parts of 2223 and 1 part of 5000. Hence, 2223/600*595 + 5000/600*1 = 8.3 + 2219.3 = 2227.6 µmol kg^{-1}. Though here it is reported to be 2232. This also true for the other dilution where I would have expected 2232.3 µmol kg^{-1} for 1:300 and 2241.5 µmol kg^{-1} for 1:150.

As mentioned above, we acknowledge that there are differences between the expected final TA and the measured final TA, so we have included additional columns in the table to reflect these differences.

Treatment	**Target ΔTA (µmol kg^{-1})**	**Measured ΔTA (µmol kg^{-1})**	**Target ΔDIC (µmol kg^{-1})**	**Measured ΔDIC (µmol kg^{-1})**	**Δ Ω$_{aragonite}$**	**ΔpCO$_2$ (µatm)**	**ΔPIC (µg L^{-1})**	**ΔPOC (µg L^{-1})**
1:600	12	9.3 ± 8	12	11.1 ± 20	-0.01 ± 0.02	9 ± 8	-0.3 ± 0.2	0.6 ± 9.7
1:300	16	11.5 ± 5	16	21.8 ± 17	-0.09 ± 0.02	38 ± 8	-0.4 ± 0	-1.5 ± 6.7
1:150	25	10.4 ± 5	25	24.9 ± 22	-0.12 ± 0.06	51 ± 21	-0.1 ± 0.3	12.2 ± 20.5

The control seawater had an alkalinity = 2223 ± 6 µmol kg-1, DIC = 2015 ± 21 µmol kg-1, Ωaragonite = 2.41 ± 0.02, pCO2 = 536 ± 4 µatm, PIC = 7.1 ± 0.2 µg L-1, and POC = 68.4 ± 7.7 µg L-1. The treatments are shown as dilution ratios - i.e., the 1:600 treatments received 1 part of a stock alkalinity (4754 µmol kg-1) per 600 parts of seawater. Expected TA and DIC have been calculated by only assuming [HCO$_3^-$] has increased. The reported error is standard deviations between triplicate bottles (n = 3).

Line 323: as mentioned above, presenting the data as ΔTA and ΔDIC might make it easier for the reader to identify changes overtime
Agreed, we have taken this comment into consideration but have decided to keep the measured values of TA and DIC to highlight the differences in the starting seawater that also links to variability in other measured parameters.

Line 328: the pH reported as pH^{STP} is not a standard pH scale. Consider introducing the meaning or to use a more traditional scale, e.g., total, free or seawater scale
All pH was calculated on the total scale. The figures have been edited to remove this confusing label.

Line 340: why writing "the Omega value" instead of "the Ω value" when the latter is used in the graphs?
Corrected.

Line 342: I believe this should read "µmol"
Corrected.

Line 356: the opening sentence is a bit awkward, consider rephrasing
Rephrased.

Lines 449-450: the formulation may be misleading. While the graph does show that DIC decrease is compensated by either POC or DOC, the graphs represent only proportions, i.e., %. Hence, a DIC decrease may not lead to an increase in POC or DOC, but rather in their contribution to the carbon pool. Please review
In Figure 7, we show changes in the total carbon pool in the experiments that have all PIC, POC, DOC and DIC measurements. We do this through a mass-balance calculation. Line rephrased to say increase observed in %POC or %DOC.

Line 464: here again, for consistency, use CaCO3. This applies all throughout the text and needs to be addressed
Corrections made.

Lines 471-474: the experiments are once referred to as "calcium + bicarbonate experiments" and then as "Ca^{+2} + HCO_3^- experiments". Please keep it consistent
Noted.

Lines 516-522: this is a heavy paragraph, consider rephrasing in 2-3 sentences
Rephrased.

Line 551: consider rephrasing as "addition, which they attributed to a possible…"
Rephrased.

Lines 556-562: this is a very interesting point and is sadly way too short. These hypotheses should be expanded to explain that naturally ecosystems are subject to such alkalinity changes and may therefore be able to handle to some extent OAE
Thank you for highlighting this point, we have revised this paragraph to include the reviewer's insights and added more suggestions for future studies.