

RC3 Specific comments:

This study explores non-equilibrated Ocean Alkalinity Enhancement (OAE) using silicate and calcium-based Total Alkalinity (TA) gradients (0 to 600 $\mu\text{mol} \cdot \text{L}^{-1}$) under natural conditions. The manipulation increased pH and decreased pCO₂, impacting bloom formation after macro-nutrients were added. Overall, this study contributes to the current understanding of OAE field application. Here are my comments to help the authors refine this manuscript.

Major comments:

1. The authors proposed that the addition of TA higher than 150 $\mu\text{mol/L}$ had delayed the bloom of phytoplankton, but the results may not be strong evidence for this argument. The GP- ΔTA , Chl-a- ΔTA , and CR- ΔTA etc were analysed as shown in Figs.4,5,6 with a CI bar representing the Confidence Interval (CI). If I understand correctly, if CI bars overlap with the horizontal line ($y=0$), the GP or Chla and other parameters don't have a significant linear relationship with the ΔTA on that day. Considering the CI bars in many of the subplots in Figs. 4,5,6 are large (especially during the bloom time), so this argument about delayed bloom may be overinterpreted. In addition, in Fig.4 A and C, the peak of the curve seemed to occur earlier in the Si 150 treatment than in the Si 0 treatment, which conflicts with the argument. I would suggest the authors reconsider this argument throughout the manuscript.

We appreciate the reviewer's critical feedback and acknowledge the potential overinterpretation of our results, particularly when considering the Si control. This specific mesocosm displayed an atypical behaviour, differing notably even from the Ca control. It exhibited two small peaks in gross production (GP), net community production (NCP), and chlorophyll a (Chla), with a delayed response compared to the Si 150 treatment. We have discussed this anomaly in the discussion section, specifically on line 456 of the initial preprint, where we mentioned this may have been due to the mesocosm effect. In fact, it is worth noting that the average contribution during phase I of CR to GP in the silicate control was the highest at 63%. Thus, right before the nutrient addition that led to the described pattern, the community was likely much different in that treatment than the rest

The CI bars show that statistical significance was not found when these cross the y axis and, in most cases, are quite wide. This is mainly because of the variability observed on each individual day. The daily linear models employed to produce the effect size graphs have been included in the supplementary materials. As can be observed, the lower treatments do respond slightly earlier than the high TA ones, generating negative slopes that later invert, although most are not significant. We argue that this is because of the small sample size ($n = 5$ in the Si and Ca gradients separately). In fact, the variability that led to non-significant negative daily linear trends in terms of GP and Chla was due to just two treatments: The control in the silicate treatment set, and the $\Delta\text{TA} 150$ in the calcium one (which experienced a longer delay than the control).

While we acknowledge that these results are qualitative and not statistically significant (which has been highlighted further across the entire manuscript, and a paragraph addressing this has been added at the end of the results section), we believe they are still noteworthy. In fact, the delayed bloom formation in the high TA treatments has subsequently been observed in two follow-up mesocosm studies conducted a posterior (unpublished). Further, this trend, though subtle, emerged even after a nearly month-long acclimation period. Thus, the fact that when

closing the mesocosms, increased variability in community composition and structure amongst them is prompted, we believe further reinforces our argument. Additionally, we do not claim the delay to be proportional to the TA addition, but rather described it as a mild lag with lower TA treatments responding sooner. Despite being a qualitative observation in this specific experiment, we consider this a key result that warrants further study due to its potential implications.

2. The design of macronutrient fertilization is a good way to understand the field application in other seasons, but there is limited information about why authors choose these certain macronutrient levels. Is it close to the real nutrient levels in different seasons? Please explain more details about the design in lines 251-260.

We appreciate the reviewer's insightful comment. To address this, we have expanded the methods section "2.2 Carbonate chemistry manipulation and nutrient fertilization" to clarify our rationale behind the chosen macronutrient levels. The amount of NO_3^- of $4 \mu\text{mol/L}$ was added to simulate upwelling of deeper nutrient rich waters, creating a phytoplankton bloom comparable in biomass to natural occurrences in the area. The N:P ratio was targeted at 16:1, aligning with the Redfield ratio, which represents the standard nutrient composition of marine phytoplankton and is widely recognized in ecological studies. The Si:N ratio was set at approximately 1:4. This design choice was made to create a niche for coccolithophores so that they would not be outcompeted by diatoms, which would be silicate limited under these conditions (Gilpin et al., 2004; Schulz et al., 2017).

3. In discussion 4.1, I appreciate the authors trying to explain the reasons why a potential delayed bloom would occur. However, there is no sufficient information about local phytoplankton community composition in the experimental sites. Were there diatoms? Were there calcifying phytoplankton? In Fig.3, the peak of Chla was similar or even higher than the Fjord, is it possible some species benefited from the addition of TA? Considering the pH tolerance and CO_2 utilization are species-specific, the information about local phytoplankton will be useful.

Data on phytoplankton community composition and structure were not made available for this publication since its focus was on the trends observed in terms of microbial metabolic rates. A separate publication that is currently in preparation will report and discuss the observed changes to the community composition in relation to the trends detected here and the carbonate chemistry conditions.

There were diatoms present (Ferderer et al., 2023). Although indeed, information about the local phytoplankton community would be very useful (added to the manuscript at the end of the third paragraph in section 4.1). For now, plausible hypotheses, based on the relationship of pH and pCO_2 with growth rates being species-specific, are presented.

The difference between the fjord and the mesocosms is interesting. The phytoplanktonic communities were definitely different considering their GP:Chla, as well as the differences in Chla concentrations described by the reviewer. Leading to the conclusion that certain species benefited from the carbonate chemistry conditions inside the mesocosms. Although the mesocosm effect may have also played a role in this differentiation, this has been added at the end of the section 4.1's 4th paragraph, as well as in the results section.

Minor comments:

1. Line 76 “iron, which is a co-limiting micronutrient”: please add a reference.

Addressed

2. Line 130: five mesocosms?

Addressed

3. Line 162: please add a reference about how you correct the pH.

Addressed

4. Line 200: Where is the T in the equation?

Addressed

5. Line 230: The unit of TA is $\mu\text{mol/L}$ in previous paragraphs, and the delta TA unit is $\mu\text{Eq/L}$ in table 1. Please double-check and explain $\mu\text{Eq/L}$.

Addressed. We want to thank the reviewer for noticing. They were meant to be all in $\mu\text{mol/L}$.

6. Line 236: “The difference between ... was quite steep”, please rephrase this sentence.

Addressed

7. Line 265: “In the silicate ones, ...” should be “calcium ones”?.

“In the silicate ones...” here is correct. Initially, the average depletion of Si(OH)_4 , NO_3^- , and PO_4^{3-} in the calcium treatments are listed. Afterwards, the same is described for the silicate treatments. First the consumption of NO_3^- , and PO_4^{3-} in the latter, which were alike to those observed in the calcium treatments. The Si(OH)_4 consumption in the silicate treatments is stated afterwards separately because of the different concentrations of this nutrient between the two sets of treatments throughout.

8. Line 268-270: It looks like the drawdown of N and P in Ca treatments was just as much as in Si treatments. Therefore, they were both N and P limited.

We agree with the reviewer and this comment has been addressed by removing this sentence altogether.

9. Line 274: What’s the grey line? Please explain the greyline, the dash lines, the phases I and II here.

Addressed in all figure captions

10. Line 297: Please explain more about the top subplots in Fig. 4 and Fig. 5. Does the positive value mean the positive relationship between the parameter and delta TA?

Indeed, if the slope is positive, it means there may be a positive (although not necessarily significant) relationship between the parameter in question and the delta TA. The statistical strength is though lacking because of the small sample size (as explained above). These plots are just a representation of a trend that, with this experimental setup, cannot be statistically “verified”. This has been further clarified in a paragraph added at the end of the results section and in the conclusions.

11. Line 308-315: Please consider moving this paragraph to the method.

We want to thank the reviewer for this suggestion. We believe however that, since these correlations were carried out once the data were obtained and processed, that it belongs in the results section.

12. Line 358: Please explain more about what hydrated lime and forsterite are.

Addressed

13. Line 374 -376: Please add references.

Addressed

14. Line 399 “There was high variability in their tolerance to high pH”: what is the “high pH” range?

Addressed. Hansen (2002) reports some species stopped growing at pH 8.3-8.4, while other were still able to grow at pH 10. Thus, the range stipulated here: 8.4 to 10.

15. Line 415-419: Please add references.

Addressed

16. Line 436: “In the latter, the community”, please state the specific phase or day.

This sentence has been removed upon realizing that it was not pertinent to the argument being presented.

17. Line 441-443: The relationship between diatom silicifying and Chl-a concentration is not clearly explained here. Please rephrase the sentence.

This sentence has been extended and modified: “Diatoms in these treatments may thus have been able to allocate more resources to growth and photosynthesis (Inomura et al., 2023). Therefore, the excess availability of silicate could have aided the faster response to the mixing event simulation, as well as contributed to the higher absolute GP and Chl-a concentrations observed during phase II in the silicate treatments, compared to the calcium ones”.

References

- Ferderer, A., Schulz, K. G., Riebesell, U., Baker, K. G., Chase, Z., and Bach, L. T.: Investigating the effect of silicate and calcium based ocean alkalinity enhancement on diatom silicification, *Biogeosciences Discuss.*, 1–28, 2023.
- Gilpin, L. C., Davidson, K., and Roberts, E.: The influence of changes in nitrogen: silicon ratios on diatom growth dynamics, *J. Sea Res.*, 51, 21–35, <https://doi.org/https://doi.org/10.1016/j.seares.2003.05.005>, 2004.
- Hansen, P. J.: Effect of high pH on the growth and survival of marine phytoplankton: implications for species succession, 28, 279–288, 2002.
- Inomura, K., Pierella Karlusich, J. J., Dutkiewicz, S., Deutsch, C., Harrison, P. J., and Bowler, C.: High Growth Rate of Diatoms Explained by Reduced Carbon Requirement and Low Energy Cost of Silica Deposition, *Microbiol. Spectr.*, 11, <https://doi.org/10.1128/spectrum.03311-22>, 2023.
- Schulz, K. G., Bach, L. T., Bellerby, R. G. J., Bermúdez, R., Büdenbender, J., Boxhammer, T., Czerny, J., Engel, A., Ludwig, A., Meyerhöfer, M., Larsen, A., Paul, A. J., Sswat, M., and Riebesell, U.: Phytoplankton blooms at increasing levels of atmospheric carbon dioxide: Experimental evidence for negative effects on prymnesiophytes and positive on small picoeukaryotes, *Front. Mar. Sci.*, 4,

<https://doi.org/10.3389/fmars.2017.00064>, 2017.