

Dear referees,

We thank you for your comments on our manuscript. We appreciate the time and effort you have dedicated to providing valuable feedback on our manuscript. Here are our point-by-point responses to your comments.

General comment: I caution the authors about making too many broad, overly-generalized statements like in the abstract “In summary, our findings suggest limited genus-specific impacts of alkalinity on diatoms.” Statements like this need to be contextualized to prevent readers from making generalized conclusions that might not be applicable in other environments. This study looked at only one aspect of diatoms, that being silicification via PDMPO incorporation. Furthermore, it was done in an environment where there were very few diatoms to begin with.

Response to general comment: We thank the reviewer for highlighting this and agree that such a statement is far too generalised for this specific manuscript. We have adjusted this sentence and other similar instances throughout the manuscript.

Line 35- 38: “In summary, our findings illustrate that the enhancement of alkalinity via simulated silicate- and calcium-based methods has limited genus-specific impacts on the silicification of diatoms. This research underscores the importance of understanding the full breadth of different OAE approaches, their risks, co-benefits, and potential for interactive effects.

Comment 1: Lines 24-33 “ Silicification was significantly greater in the silicate-based mineral treatments, with all genera except *Cylindrotheca*, displaying an increase in silicification as a result of the increased concentration of silicate. *Pseudo-nitzschia* and *Nitzschia* were the only genera directly affected by alkalinity. The four other genera investigated here displayed no significant changes in silicification as a result of alkalinity increases between 0 and 600 $\mu\text{mol kg}^{-1}$ above natural levels.”

This is confusing. You have the first sentence saying all genera were affected except *Cylindrotheca*. Then it says only *Pseudo-nitzschia* and *Nitzschia* were affected. Then it says 4 genera were not affected. Please be more clear in the summary of your findings.

Response 1: We thank the reviewer for highlighting this and have adjusted this section in light of their comment Lines 29 – 35: “Silicification was significantly greater in the silicate-based mineral treatment, with all genera except *Cylindrotheca*, displaying an increase in silicification as a result of the increased concentration of dissolved silicate. In contrast to the effect of differences in dissolved silicate concentrations between the two mineral treatments, increases in alkalinity only influenced the silicification of two genera, *Pseudo-nitzschia* and *Nitzschia*. The four other genera investigated here (*Arcocellulus*, *Cylindrotheca*, *Skeletonema*, and *Thalassiosirra*) displayed no significant changes in silicification as a result of alkalinity increases between 0 and 600 $\mu\text{mol kg}^{-1}$ above natural levels.

Comment 2: Figures should be introduced in the order they appear in the text. (e.g. Results start with Fig. 2d as opposed to 2a).

Response 2: We thank the reviewer for highlighting this mistake and have adjusted the order which the figures are introduced within the text.

Line 291-300: “Concentrations of NO_3^- were below detection limit, thereby constraining phytoplankton growth during phase I of the experiment (mean NO_3^- day 7 – 25 = $0.004 \pm 0.035 \mu\text{mol L}^{-1}$) (Fig 2a). In contrast there was residual PO_4^{3-} ($0.021 \pm 0.022 \mu\text{mol}^{-1}$) and Si(OH)_4 (Ca-OAE treatment = $0.202 \pm 0.99 \mu\text{mol}^{-1}$, Si-OAE treatment = $67.929 \pm 1.04 \mu\text{mol}^{-1}$) which likely supported the phytoplankton community in utilising remineralised nitrogen until the addition of nutrients on day 26 and 28 (Fig 2b, 2c). Although $\sim 75 \mu\text{mol L}^{-1}$ of Na_2SiO_3 was added to the Si-OAE treatment, there was no discernible depletion of Si(OH)_4 during phase I (Fig 2c). The addition of macronutrients (NO_3^- , PO_4^{3-} , and Si(OH)_4) can be seen on day 26, with a secondary addition on day 28 to correct for unwanted differences in the stoichiometry between mesocosms (Fig 2). Chlorophyll *a* concentrations were relatively low at the beginning of the experiment with $1.01 \pm 0.17 \mu\text{g L}^{-1}$ (mean \pm SD) on day 3 (Fig 2d).”

Comment 3: Line 299-308 starting with “Chlorophyll *a* peaked between days 39 and 49 for all mesocosms with the Si-OAE treatment exhibiting a marginally higher mean peak in chlorophyll *a* (Si-OAE treatment: $3.78 \pm 1.7 \mu\text{g L}^{-1}$, Ca-OAE treatment: $2.97 \pm 0.3 \mu\text{g L}^{-1}$, Fig. 2d). From just the error alone, it doesn't not look like this difference is statistically significant so therefore saying that it is marginally higher is misleading. Either provide statistical support or remove such phrases. Same comment for subsequent statements about nitrate and silicic acid.

Response 3: We understand the reviewers concern and have removed the sections where comparisons between treatments with no statistical analysis were made. We have kept the discussion surrounding differences in silicic acid concentrations, with a modification so that only the highest and lowest alkalinity treatments are discussed. We feel that this is important to include as it relates directly to the observed differences in starting concentrations of dissolved silicate and BSi in the sediments.

Line 304 – 309: “There was no discernible relationship between total alkalinity and chlorophyll *a*, NO_3^- or PO_4^{3-} observed across the extent of the experimental period or in a particular phase (Fig 2). However, in the Si-OAE treatments initial concentrations of Si(OH)_4 were lowest in the high alkalinity mesocosm, with a difference of $2.45 \mu\text{mol L}^{-1}$ between the $\Delta 0$ and $\Delta 600 \mu\text{mol kg}^{-1}$ alkalinity treatments (Fig 2c). This trend appeared directly after the addition of the treatments but disappeared once nutrient uptake began (Fig 2c).”

Comment 4: Can the same symbols in Fig. 3 be used in Fig 2? This would improve the ability to differentiate the numerous lines shown in each graph which is currently very difficult to do. For example, it is stated that in Fig. 2c the silicic acid concentration in the higher alkalinity treatments is lower. Zooming it, it looks like one of the high alkalinity treatments has silicic acid concentrations similar to the low alkalinity, but it is difficult to match the exact color to the treatment.

Response 4: Yes, we agree with the reviewer and have adjusted figures 2 and 6 so that all graphs which utilise symbols to distinguish between alkalinity levels use the same symbols as figure 3.

Comment 5: Line 319, “small distances are observed between Pseudo-nitzschia and Nitzschia, likely due to their morphological similarities and therefore similar silica content”

Just because they have similar morphologies does not mean they have similar silica content. Furthermore, different Pseudo-nitzschia species can have different silica quotas in addition to the ability of diatoms to alter cellular silica quota.

Response 5: We agree that this statement is not well supported and would require further investigation. As such we have adjusted this sentence:

Line 328 – 330: “Nonmetric multidimensional scaling (NMDS) (Fig. 3) revealed distinct distances among treatments, including different alkalinity source minerals and total alkalinity, in relation to silicification of the various diatom genera.”

Comment 6: The inclusion of final counts for each genus provided in the methods is appreciated (1-176 cells per mesocosm per day), but all this says is that with respect to Figs. 4 and 5, these data could represent 1 cell or 176 cells. If the authors want to make the claim that Pseudo-nitzschia and Nitzschia have significantly higher silicification with OAE, knowing the number of cells this is based on would make the conclusion more robust.

Response 6: We agree with the reviewer that counts for these two genera specifically would be beneficial and have now included them within the text.

Line 362 – 364: “Exploration of this interaction revealed the silicification of cells in the genus *Pseudo-nitzschia* ($N = 3510$) to be significantly influenced by alkalinity in both the Ca and Si-based treatments, with silicification increasing with increasing alkalinity (Table 3). In contrast, the genus *Nitzschia* ($N = 677$) displayed...”

Comment 7: Do the authors have any thoughts as to how Pseudo-nitzschia was able to increase silicification in Ca treatment where there was very little Si?

Response 7: We are currently investigating this difference but believe that the increase in alkalinity and subsequent decrease in CO₂ may have slowed growth and therefore increased the time and uptake of DSi resulting in the observed differences in silicification.

Comment 8: Line 419, “Our results revealed silicate fertilisation associated with silicate-based OAE to significantly increase silicification in the diatom community and all genera with the exception of *Cylindrotheca*.”

This should be qualified by stating that this is relevant when dissolved silicon in the initial conditions are low as was the case in this study. If initial Si concentrations are high, it is possible the increase in silicification would not be seen in response to Si-OAE.

Response 8: We thank the reviewer for their comment and agree that if initial concentrations of dissolved silicate were high, further increases in dissolved silicate as a result of Si-OAE would most likely not increase rates of silicification. We have adjusted this section accordingly.

Lines 431 – 435: “It is important to note that low initial concentrations of dissolved silicate likely facilitated the observed increase in silicification. Under conditions where dissolved silicate is already replete, further increases due to silicate-based OAE may not result in similar increases in additional silicification. This increase in silicification was primarily a result of the difference in the dissolved silicate concentrations ($\Delta 75 \mu\text{mol kg}^{-1}$) between the silicate and calcium-based OAE treatments rather than an increase in alkalinity.”

Reviewer #2

Comment 1: In the text, check nitrate and phosphate: sometimes written NO-3 and PO-34?

Response 1: We thank the reviewing for highlighting this and will rectify this mistake throughout the manuscript.

Comment 2: L. 301-303: "A similar trend is observed in NO₃- uptake with mesocosms in the Si-OAE treatment, showing marginally faster NO₃ depletion ($-0.192 \mu\text{mol L}^{-1}$ per day) compared with the Ca-OAE treatment ($-0.158 \mu\text{mol L}^{-1}$ per day)"

Is there an error (SE) associated with these consumption rates?

Response 2: This section has been removed from the manuscript in line with recommendations made by reviewer 1.