Dear referees,

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.

Comment 1. Line 160 – please specify units (PSU)

Response 1. In this instance, we measured salinity via the use of electrical conductivity measurements. As such, the salinity value here does not require units as per the international system of units (SI) in oceanography, pages 40 and 44 (UNESCO,1985).

Comment 2. Lines 470 and 563 – its suggested that CO2 and H+ are the most "physiologically-relevant carbonate chemistry parameters." I do not necessarily agree with this statement and other parameters are certainly relevant. For example, diatoms use SLC4 bicarbonate transporters (Nakajima et al. 2013). Iron uptake in diatoms can also depend on carbonate (McQuaid et al. 2018).

Response 2. We agree that other parameters are indeed relevant. In light of your comment, we will adjust both lines to better reflect this. line 470: "The lower peak chlorophyll *a* in the equilibrated treatment was unexpected as CO_2 and H⁺, two carbonate chemistry parameters believed to drive phytoplankton growth (Paul and Bach 2020) were relatively similar to the control and within natural ranges (Fig. 4d, e)."

Line 563: "The observation is also remarkable because the differences in BSi occur between the control and both alkalinity treatments even though differences in CO_2 and $[H^+]_F$ are much larger between the equilibrated and unequilibrated alkalinity treatments (Figs. 4d, 4e)."

Comment 3. Lines 523-525 – How is it known that the largest cells were roughly 50um? From the SEM? Please clarify.

Response 3. Yes, this was indeed determined from SEM images. We will adjust these lines to reflect this. Line 523-525: "The largest diatom cells in our experiment were roughly 50 μ m and we did not find any diatoms smaller than 3 μ m (determined from SEM). Thus, all diatom cells are most likely found in the nano- and microphytoplankton groups in flow cytometry data."

Comment 4. Lines 528-530 – Please clarify what you mean here by "variation in the diatom communities." To me, that means a community shift, but I do not agree that the differences in bSi and Si uptake translate to a shift in the diatom community. I think perhaps some other phrasing might be better here.

Response 4. We thank the reviewer for highlighting this. We also agree that the differences observed in our study may not necessarily translate to a shift within the diatom community. Our aim in this sentence was to elucidate that we believe observed differences between treatments (in BSi and Si) indicate some effect and change relating to the diatom community. What this is effect/change is we cannot say with our current data set, in light of your comment we will change this sentence to hopefully better reflect this. Line 529-531: "In addition, significant differences in the build-up of BSi and drawdown of Si(OH)₄ between the control and treatments strongly suggests that the alkalinity treatments influenced the diatom communities."

Comment 5. Line 571 – The authors suggest that there could be a shift in the diatom community towards nonsilicifying species. The only diatom I'm aware of that doesn't use silica is Phaeodactylum tricornutum and it is highly unlikely to have been in these incubations so I don't agree with this conclusion. Or if the authors mean a shift in the phytoplankton community away from diatoms they should clarify that.

Response 5. Thank you for highlighting this, we agree that it is highly unlikely that there was a shift in the community towards non-silicifying diatom species. As such we will adjust this sentence to reflect the reviewer's recommendation. Lines 570-571: "For example, there could be a shift in the diatom community towards smaller, less heavily silicified species and/or a higher fraction of non-silicifying phytoplankton."

Comment 6. Line 576 and Figure A5 – Rather than stating "data not shown," can the authors please include these data in appendix?





"Figure A5 Average diatom a) biovolume and b) abundance, during the peak bloom (day 6) within treatments determined via SEM. Data are presented as mean values \pm SD."



"Figure A6 Temporal variation in the molar ratios of TPC to BSi within microcosms. Coloured shading around the respective means represents the standard deviation."

Comment 7. Line 695 – A major conclusion is that diatoms were affected due to differences in bSi and Si uptake, but the authors have not considered silicious grazers such as certain Rhizaria. Could those not also influence these results?

Response 7. We thank the reviewer for highlighting this very important point. Based on our personal observations and the microscopy conducted during this study we believe that it is highly unlikely that Rhizaria or any other silicifying plankton caused the differences observed in our study. This is because we did not find any silicifiers (except diatoms) in this study, the pilot study we conducted before this study or subsequent sampling from the same location. We do highlight this in line 540 of the text: "Scanning electron microscopy investigations of samples taken before, during, and after the phytoplankton bloom revealed that diatoms were the only silicifiers detected in the plankton community.". We hope that this is sufficient to alleviate the reviewers concerns

References

UNESCO (1985) The international system of units (SI) in oceanography, UNESCO Technical Papers No. 45, IAPSO Pub. Sci. No. 32, Paris, France.