Review:

Schneider et al.: „Air-sea gas exchange measurements helped derive in-situ organic and inorganic carbon fixation in response to Ocean Alkalinity Enhancement in a temperate plankton community "

Key results

The authors investigated the effects of ocean alkalinity enhancement (OAE) on pelagic communities in a temperate fjord. In particular, they investigate carbon dioxide removal efficiencies as well as community responses to gradients of two different alkaline minerals (calcium- and silicon-based). Their results show that the amount of added alkalinity modifies the CO2 removal capacity, independent of the mineral. Furthermore, calcification is enhanced under medium alkalinity additions and net community production is stimulated by the addition of the silicon-based mineral, likely due to increasing diatom productivity.

The study is timely and important to improve the understanding of ecosystem effects of OAE, especially as knowledge on environmental impacts of OAE is still very limited. The authors made big efforts to analyse and understand the dynamics in their mesocosms, using a variety of methods. The investigation of the carbon removal efficiency as well as ecosystem impact of two different alkaline materials, inspired by the white vs. green ocean hypothesis of Bach et al. (2019), is a vital contribution to move OAE research closer to actual application. The findings of the study can be an inspiration for similar studies in other ocean regions, as well as for more detailed investigations of the separate processes.

Although I strongly encourage publication, I recommend major revisions prior to acceptance. In the following I add my main issues for each of the sections. Below you can find line-by-line comments. My main comments concern the presentation and discussion of the results and not the experimental setup in general. I have to add that I'm not working in the field or laboratory myself and cannot fully evaluate the methodologies in this study.

General comments

The title is very descriptive. It's a matter of taste, but I would recommend to include your main findings, e.g. the optimum functional response of coccolithophores to alkalinity addition.

The abstract is concise, although the description of the results could in parts be improved, see line-by-line comments.

The introduction successfully introduces the reader into the functioning of ocean alkalinity enhancement. The authors could improve the last part by removing some of the method description and adding more details on the motivation to conduct experiments with different alkaline material under nutrient addition.

The results could be restructured to highlight similarities and differences in the carbonate system, calcification rates, NCP etc. of the different experiments (i.e., Si vs Ca-based mineral and the effects of nutrient addition), sticking to the initial research questions of the study. In parts this is already done, but somewhat hidden in additional calculations and assumptions which could rather be moved to the result (and partly discussion) section. A clear structure and a consistent naming of the experiments would help the reader to understand the basic results of the study. I do like Table 1, which nicely summarizes the findings of the study.

The analysis of the carbonate system changes reads a bit like all over the place. I suggest to carefully disentangle the different measurements you have (TA, DIC, pCO2, FCO2) and what you can learn from these changes (e.g., are they biologically mediated or caused by physics and resulting changes in the carbonate system).

I couldn't find a comparison of CALC and CCPP as announced in L. 246-247 (apart from Fig. 3b) and an interpretation of this. I'm not sure if this is relevant for the manuscript, but it should at least be added to the supplement.

The discussion is very close to the results of the study, which is good, but could me moved from a description of the results to an interpretation of overall findings embedded into existing literature. In parts this is done, but it is hidden in a repetition of the result.

Conclusion and outlook are rather a summary of Table 1 than a real perspective on the study. I suggest to think about what we can learn from the findings of the study, what could be potential follow-up experiments, and which advices you would give for large-scale applications of OAE.

Finally, not being a native English speaker myself, I would recommend to do a thorough language check as there are some grammar and phrasing issues here and there.

Line-by-line comments

<u>Abstract</u>

L. 15-16: Reduced pCO2 and pH only until equilibrium is reached, right? Maybe it's worth to add this to the sentence.

L. 17: Better: "in *a* temperate fjord"

L. 23: "with cumulative coccolithophore calcification showing an optimum curve response to decreasing pCO2" – this sounds very complex. Can you simplify the message?

L. 24: "while no mineral-type effect was observed" – compare to first part of the sentence ("there were a number of mineral-type and pCO2/pH effects"); this seems to be contradicting. Perhaps splitting the sentence in two parts would help?

L27-28: “These complex findings suggest both direct and indirect effects of mineral type and OAE level and provide a valuable foundation for designing future OAE field trials” – can you be more specific about the message of your findings? For example, carbonate-minerals should be favored over silicate-minerals (smaller environmental impact) and the amount of added alkalinity must balance effectivity (CO2 flux into the ocean) and environmental impact (calcification).

Introduction

L. 44-46: “This variability…” – yes, but the variability that you mention in the previous sentence only refers to modelling studies. There are a number of reasons why modelling studies vary in their estimates of CO2 uptake potential, in the end they assume different alkalinization scenarios etc. I agree that the CO2 uptake potential of real-ocean applications is difficult to predict because we do not know enough, but then you need to rephrase the sentence.

L. 50: “Since gas exchange *can* take …”

L. 60: “all of which release soluble products of interest for marine organisms” – unclear; why are the alkaline minerals “of interest” for marine organisms?

L. 61-64: “For these materials …” – this sentence is very long and quite confusing, can you rephrase? For example, it is currently unclear what you mean with white and green ocean for readers who don’t know the study of Bach et al. You could also emphasize more that you aim to test the white vs green ocean hypothesis of Bach et al., this is currently somewhat hidden in your statements.

L. 67ff: I suggest to shorten, parts of this are methods.

L. 74: “These perturbations would be highest close to an OAE point source” – okay, but you don’t test OAE effects with distance to a point source in your study, right? I suggest to shorten here.

L. 77-78: “Processes that need to be better understood before large field deployments of OAE and for later monitoring and verification.” – Please check, this is not a proper sentence. And here you could add a few more details – which processes do you aim to understand? What is the goal of your studies, which parts of the ecosystem do you assess?

Material and Methods

L. 88: “surrounding” (without *s*)

L. 91: “to remove any heterogeneously distributed nekton” – what is this? Wouldn’t “remove nekton” be enough?

L. 92-94: “Additionally, a volume determination …” – this sentence is unclear; do you measure the volume change or the total volume, and why is this important? Please rephrase.

L. 99-101: As a reader who is not familiar with OAE experiments I would wonder why you don’t just add lime and olivine power?

L. 103: “*in* the olivine scenario”

L. 109-111: “(the unavoidable …)” – this sentence is unclear. Can you state more clearly that the addition of silicate leads to a shift in alkalinity that is not wanted in your experiments because you want to control TA only by the addition of NaOH? At least this is how I understand your description, but it took a while until I got your message.

L. 119-121: Why is the concentration of nutrients lower than in the surrounding water? And what is your motivation to add nutrients? This has not been explained in the introduction, has it? Furthermore, please check the sentence “Nitrate concentrations …” – is it possible to describe the amount of added nitrate and phosphate in a similar manner?

L. 122: “Ca treatments” – I recommend to use uniform terms for your experiments throughout the manuscript.

L. 123-124: “Nutrient concentrations …” – please check sentence structure and grammar.

L. 128: What exactly do you mean with “randomized sampling”?

L. 149-150: change “The filtration process resulted in the removal of biomass” to “The filtration process aimed at removing” or “The filtration process removed”

L. 151f: “Water for gas-sensitive parameters…” – please check the sentence structure.

L. 161: To compute average and standard deviation you would need triplicates, not duplicates. Is there a reason why you did not perform three measurements?

L. 171: Change “Said samples” to “Samples”

L. 196: “Throughout the experiment, …” – please check the wording. “Coupled” does not sound correct in this context.

L. 202: Please check the section title. The part “and community production estimates” appears also in the next section title in line 234, and this is where you actually describe community production.

L. 242: It would be helpful if you could give a unit for CALC.

L. 244: How do you know that coccolithophores are the main calcifiers in your mesocosms? I suppose that your experiment was performed jointly with other experiments in which the community composition was analysed, but if so it should be mentioned.

L. 244-245: Can you give an equation and a unit for CCPP? Which light conditions do you mean? Surface or at a specific depth?

L. 249: Maybe I missed this, but where do you have cCO2 from?

L. 254: "The following change in sign …" – I know what you mean, but the phrasing is a bit odd. Consider to rephrase to something like "We define NCP to be positive" or alike.

L. 255: The description of NCP from O2 measurements is missing.

L. 260: "for 5 additions of increasing dye addition" – rephrase to "for 5 additions of increasing dye concentration"

Results

L. 272: "After the manipulation on day 7" – but the manipulation took place on day 6? It would be more precise to say "After the manipulation on day 6" on "On day 7, one day after the alkalinity manipulation".

L. 273: "while pCO2 decreased to 61" – unit is missing.

L. 273: "In contrast, DIC was hardly affected." – Compare to Fig. 1d and L. 277: DIC is considerably affected by the alkalinity manipulation (and nutrient addition).

L. 274: "Overall, TA remained relatively stable throughout the experiment, regardless of the phase and the treatment" – this is only true for phase I and II, but not between phase 0 and I.

L. 276: "from day 35 onwards" – it would be easier for the reader if you would consistently refer to the phase and/or the respective manipulation (here: "after the nutrient addition in phase II").

L. 280: "DIC decreased in all mesocosms again, …, coinciding with average Chl a changes" – I suggest to mention that DIC is decreasing as soon as Chl a increases (i.e., different direction of change).

L. 289: "All treatments showed an immediate uptake of CO2" – where do you see this? In FCO2?

L. 290: "While FCO2 didn't change significantly for the control treatment right after the TA addition" – but you did not add TA to the control treatments, did you? TA should not change in these treatments anyway? Generally, I feel that you could use the control treatments a

bit more as such, i.e., you could analyse the changes in the other treatment relative to the control treatment to make sure that the only difference is the addition of the alkaline material and no other changes in the system that may play a role in the course of the experiment (being aware that experimental treatments are not 100% replicates of each other).

L. 290-291: “it increased with increasing deltaTA and corresponding lower seawater pCO2” – FCO2 is decreasing, right? Double-check sign convention. I also recommend to rephrase this sentence as it currently implies that deltaTA in the control treatment is increasing, but you mean the different TA-addition treatments.

L. 294: “Notably, no differences in FCO2 were observed between the different minerals” – This is not entirely true, I do see differences in Fig. 2. They may not be statistically significant, but you should not say “no differences”.

L. 299: “Except for the Ca-treatment deltaTA 150” – I like the idea of giving simple but specific names to the different treatments, but they should be introduced and consistently used throughout the manuscript. Indeed, this could simplify some phrases in the remaining manuscript.

L. 303: How about the comparison of CALC and CCPP as displayed in Fig. 3b?

L. 304: “at the beginning” – what exactly do you mean? Phase 0?

L. 308: Are you sure that you show cumulative changes? What is negative cumulative NCP then? More respiration than production?

L. 310-311: “Though such a statement assumes that …” – important to mention, but I suggest say a few more words on this and perhaps move to the discussion, possibly even to a a separate “limitation” section.

L. 314: “An ANOVA analysis showed no alkalinity effect, but a clear mineral effect on production” – if you tested NCP vs. pCO2 as described in the previous sentence, it would rather be a pCO2 effect and not an alkalinity effect.

Fig. 4c: Why triangles here? They should be diamonds, right?

<u>Discussion</u>

L. 333ff: Because it is an integral part of your experimental setup (although I miss a clear motivation for this as mentioned earlier) I suggest to add the nutrient addition to the sentence.

L. 335-336: “Manipulation of … which remained relatively stable over time” – please specify, what remained stable over time? DeltaTA? The treatment pairs?

L. 336: Hm yes, but as you mention correctly an omega aragonite of 11.1 may be the threshold under idealized conditions. However, the ocean is not particle-free (plus you have different temperature and salinity values compared to the idealized study of Marion et al. 2009), and these particles can serve as precipitation nuclei, decreasing the threshold of abiotic CaCO3 precipitation. It seems that this is not the case in your experiments, but you could add this argument in your discussion.

L. 339: “plus no significant change to DIC” – this is confusing, you do see quite some changes in DIC?

L. 341: “all manipulated treatments increased their DIC content” – I suggest to rephrase, the treatments did not actively increase the DIC content.

L. 343: “Changes in DIC ranged from about 23 to 49, in contrast to expected Redfield C:N of 20” – I cannot follow this argumentation, which DIC change would you have expected?

L. 351: “This is related to the fact that we arrived at the study site during in a post-bloom period” – it doesn’t really matter when you arrived ;-) Perhaps rephrase to “the experiment was started during a post-bloom period” (also note that you have a phrasing typo: “*during in* a post-bloom…”)

L. 353: “daily rates increased” – note sign convention, currently it would be “decreased”.

L. 354: “which is within the range of air-sea flux estimates from the region” – what do you mean, is the additional CO2 uptake after alkalinity addition not caused by ocean alkalinization but is rather caused by natural variations of the CO2 uptake?

L. 358-359: Temperature unit is missing (I guess it is °C).

L. 381-382: “In this regard, …” – this statement is unclear to me. Increasing ocean CO2 uptake is more likely explained by changes in temperature than by changes in wind speed over the time of the experiment because estimated wind speeds do not fit to observations?

L. 384: “Therefore, our results indicate that seasons should also be considered …” – but if the equilibration time takes more than 1000 years, passing through all seasons of almost 3 years, does the day of deployment really matter? Furthermore, I do not agree with the statement “at least in models”, because models should try to simulate what could be done in the real ocean.

L. 387-389: “Lastly, by accounting for the measured …” – this is what you will do in the following, right? If yes, you can add this to the sentence.

L. 391: “Because of the presence of the coccolithophore E. huxleyi …” – how do you know that Ehux is growing in your mesocosms? I assume that a community analysis was done in the framework of other studies on the same mesocosms, but this should be mentioned somewhere (here or in the methods). Otherwise, the certainty that Ehux is the main calcifier

in your experiments comes out of nowhere. You could, for example, give a short summary of the community analysis including relative contributions of all planktonic calcifiers.

L. 403 and L. 407: Right parenthesis missing

L. 390: Do you observe this bloom only in the experiment with alkalinity manipulation or also in the control experiment? I suggest to be more specific here, because it can be a bit confusing to understand which changes are caused by the alkalinity / nutrient manipulations and which changes are part of the natural change in the fjord.

L. 410: "Additionally, when cumulative TA-based calcification is compared to cumulative coccolithophore abundance …" – what you want to say is that the per cell calcification remains constant but the number of cells is increasing? Hence, the optimal conditions for calcification rather lead to proliferation than to heavily calcifying cells? This is an interesting finding and could be highlighted more. Although I got a bit confused by the following discussion on cellular rates.

L. 433: typo in "treatments"

L. 433: "nitrogen uptake was ~3.6 µM" – cumulative? Per unit time?

L.436-437: "… it was diatoms (if not in quantity, in frustule thickness) that was heavily silicified …" – in the second part of this sentence you imply that diatoms increase their frustule thickness and kind of exclude that the strong Si uptake can be caused by a proliferation of diatoms. If you don't know for sure I suggest to keep this open. But maybe you do have an idea as it seems like you have an impression of the community composition, given your previous statements on Ehux calcification?

L. 438: "Thus, this suggests that diatoms could allocate additional resources to growth and photosynthesis …" – doesn't this just show that diatoms were Si-limited previously?

L. 440: "… in turn, contributing to the mineral effect …" – is "contributing to" the correct word here? Shouldn't it rather be "causing"?

L. 444: "One reason could be low abundance of coccolithophores at the onset of the experiment …" – okay, but you do see an increase in coccolithophore abundance in the experiment under medium alkalinity additions (see for example L. 423-425)? The remaining sentence ("the other the fact that ….") is unclear to me, please rephrase.

L. 450: "The delay in bloom formation observed here" – this is a bit unspecific and as a reader I have to go back to the results to remember what you mean by "here". It would be easier if you would recap that you are talking about the delayed bloom peak in the high-alkalinity experiments. In the same sentence, you refer to the study of Martín-Samper et al. 2024. Is that study using the same experimental setup or even the same mesocosms? Or is that study completely independent from yours? This is currently unclear.

L. 450-456: This entire paragraph needs some more details. Why do you think the delay of the bloom could be caused by the substrate/inhibitor concept? What is a “knock-on effect”? What are the open questions and which additional considerations are required to be able to provide recommendations?

L. 458: As mentioned above, “NCP obtained from O2 measurements” was not described in the methods.

L. 458-466: Consider to move to the results and/or methods.

L. 467-468: “Interestingly, the treatments with lower NCPDIC had higher RZ, possibly indicating a top-down control on primary production.” – you can make it easier for the reader if you repeat which experiment resulted in lower NCPDIC (or higher NCP?). Also, you will always have a top-down control on primary production in a natural system, won’t you? Or do you want to say that you observe a “stronger” top-down control / more grazers in the system?

L. 468-472: “Reasons for this observation could be …” – here you mix the arguments for a weaker phytoplankton-zooplankton link due to increasing alkalinity addition and the effect on zooplankton metabolism (first part of the sentence) and due to mineral-mediated changes in the phytoplankton community composition (second part of the sentence), do you?

L. 475: But you do not investigate single species? How can you draw the conclusion that responses are species-specific? It is probably true, but I think the jump from your results to this conclusion is a bit too big.

L. 475: “ … and that a top-down control on primary productivity might have been exerted” – this sentence is unclear to me.

L. 475: Here again, how comes that Goldberg et al. 2024 knew what is going on in your experiments? If this study is based on the same mesocosm experiments than yours, it should be mentioned. Furthermore, the link between your findings and the findings on fish biomass vs abundance is not completely clear to me.

L. 479: “…. although respiration rates didn’t seem to be correlated (Bhaumik et al. 2025).” – add: “to be correlated to the amount of alkalinity added” or similar.

Table 1: A very nice summary table!

<u>Conclusion and outlook</u>

L. 496: “potentially due to enhanced Si(OH)4 concentrations.” – consider to add “and the concomitant proliferation of diatoms”

L. 498: “indicating species-specific responses and potential top-down control on primary production” – see comments above.