

First, I want to highlight, that I came into this review process after the first iteration. This implies that I tried to find a balance between looking at the manuscript with an unbiased opinion, but at the same time trying to acknowledge the already past review process.

#### Summary:

The study aims to understand the impact of advection in the water column on CO<sub>2</sub> uptake and the subsequent change in mineral dissolution and alkalinity stability during ocean alkalinity enhancement. The major results are that faster equilibration leads to higher amounts of dissolved alkalising material (in the case of solid feed stock) and in a lower loss to brucite and subsequent CaCO<sub>3</sub> formation (in the case of liquid fed stock).

#### General comments:

Given that this is the second iteration of the script, the state it is in is still poor. The method section is still very messy and difficult to follow. Whilst at some point unmercenary details are presented, major methodologies are not well explained.

The result part needs severe streamlining and reduction to results. Very often the authors mix in discussion-like snippets that do not belong here.

At the same time the authors readily state that their data supports certain ideas without explaining in what way that is the case. This coincides with at least loose if no uninformed statements about diffusive processes around grains.

The discussion needs a clearer line of thought and less attempts to invent a new concept. Additionally, the discussion would greatly benefit from a more comprehensive comparison of this studies new findings with other studies to emphasise the impact and the relevance of the new results. Overall, I want to urge the authors to do a thorough literature research before revising the manuscript. The last iteration was, as far as I saw it definitely not thorough enough.

Overall, though, the results are relevant and certainly worth being published in this journal. Hence, I recommend an absolute major revision of the script and will gladly review the result.

Here the specific comments as occurred upon reading:

#### Line-by-line comments:

Line 10/11: "...using solid..." please rephrase with "...using sea water amended with solid..."

Line 13a: Compared to expected values?

Line 13b: Bottles don't lead alkalinity generation. Please rephrase this.

Line 20: Grammatically incorrect: precipitation does not redissolve.

Line 22: Data does not serve as a threshold. Data can support an argument for lower thresholds. Or sth. in these lines.

Line 27: I thought that this was already in the 2021 IPCC report. Please check citation

Line 28/29: "...the highest carbon capture potential...". Please provide a citation for this statement, and a rough number.

Line 32: Some issues is really not what you want to say in a scientific paper. Secondary effects... or „Several processes “. Yes, let's do that.

Line 33a: The potential is not decreased. The efficiency is. Please rephrase.

Line 33b: "itself" is redundant here. Just say the ocean or alkalised seawater.

Line 34: Not sure, whether the formula needs to be introduced since it's been used in the abstract before.

Line 36: Despite the fact that these authors used olivine or dunite respectively, Fuhr et al. 2022 and Rigopoulos et al. 2018 were the first to report on CaCO<sub>3</sub> precipitation on grain surfaces during OAE. Please add references.

Line 39: Or you add the mentioned references here after aragonite.

Line 41: "...where the precipitation speed increases...". I would rephrase to sth. like "with increasing precipitation rates".

Line 44: "Hence, one could suggest": Then why does one not simply suggest this, when they could? ;)

Jokes aside. Please rephrase this.

Line 47: "...loss to CaCO<sub>3</sub>": Please rephrase to "...loss to CaCO<sub>3</sub> formation"

Line 48a: If it's microenvironments around added feed stock particles, how much of a biological risk can that be? Clarify or cite a respective study, please.

Line 48b: "This period of high alkalinity is therefore critical to the overall safe and effective implementation of OAE to limit negative impacts of OAE and maintain high CDR potential, which is later referred to as the critical alkalinity period (CAP).": This sentence at this point suggests that CAP refers to the time when microenvironments exist. Is that the idea? Or should it also refer to times when the entire water column is critically alkalised? Please clarify and rephrase if necessary.

Line 51: If you say "First", you must say second, which you do not. Please rephrase.

Line 52: "...weeks.": Here you need to cite the most relevant lab-based OAE studies that fulfil your description. Keep in mind that you were talking about "OAE-focussed experiments", which encompasses a broad range of experiments. So, either the list of references here will be very long or you want to rephrase this.

Line 53: "One can...": One should really do sth. rather than stating what one could do. please rephrase.

Line 54a: "these experiments": Which ones? Please provide references.

Line 54b: In what way is a stir bar problematic? Please provide a short explanation and again: references.

Line 57: "...what would occur in a natural setting": How high would that be? Please provide respective references.

Line 58: "One could..." Please rephrase.

Line 61: How do you define open ocean conditions? What parameters are considered, how are these defined? Is it just the mixing? In the abstract you say about 3 ms<sup>-1</sup> wind speed, but what does that mean? airflow? turbulence? an expected gas-exchange velocity calculated via the Schmidt number?

Line 64: "and gas velocities for CO<sub>2</sub> and equivalent open ocean wind speeds were derived": This does not really make sense. What gas velocities? What equivalent? Please explain and rephrase.

Line 75: "more appropriate to resuspend": Compared to what? Please clarify.

Line 76: "without breaking water tension": What do you mean by that?

Line 77: How did you determine the Reynolds number?

Line 80ff: "The bottles were cleaned... ": This is oddly detailed. Not bad per se, but also not necessary. Additionally: If you want to keep this sentence, place it after the section with the

sea water. That is logically easier to follow: 1: Collect water, 2: Prepare bottles. 3: Set up experiment.

Line 84/85: Sea water was collected and filtered: When? Directly after collection? How long was it stored before the experiment? What depth was it collected from?

Line 89: "One major issue when it comes to open bottles is evaporation. To compensate for such loss...": Shorten to "To minimize evaporation,"

Line 89/90: "...was provided in all bottles using a similar process to the one described above for the seawater.": That sounds like a lot of redundant words: Why not just say „In order to avoid evaporation the headspace of the open bottles was continuously replaced with water-saturated air. For this purpose....“. And then a brief explanation of what the setup looked like.

Line 94/95: "allowing for easier comparison of results from bottles as they were all exposed to the same conditions.": This would also have been true (within error) if the headspace had not been modified.

Also: Have you measured relative humidity in the air? And: Have you measured the flow rate at each outlet? Experience tells me that it is not easy to construct a gas distributor that provides the same flow rate at every outlet. A photo of the device would be very helpful.

Line 96: "to ensure that evaporation was not too important": This is not an appropriate wording. Please rephrase this to sth. like "...2-3 days). Evaporation-induced weight loss was compensated for by..."

Line 98/99: "Another issue that was raised before the start of the experiments was the risk of organisms growing in the bottles. Hence, experiments were mostly conducted in the dark": Same pattern as before: Many unnecessary words. „To reduce the potential growth of autotrophic organisms, the experiments were conducted..."

Lines 110-112: I thought this had happened already? Please reorganize the methods thoroughly so that there is a chronological order. Right now, it's quite a lot of jumping back and forth.

How many of each treatment and controls?

How much powder? How much solution?

Line 113/114: "+100  $\mu\text{mol kg}^{-1}$  to +600  $\mu\text{mol kg}^{-1}$  with 100  $\mu\text{mol kg}^{-1}$  TA increments for both alkaline powders, and only one set of +600  $\mu\text{mol kg}^{-1}$  using NaOH.": After the powder had dissolved? why the 125 rpm then?

Line 114 – 118: This is very confusing. please provide a table or a figure to visualize this.

Lines 123 – 125: Please provide an overview of the amounts added to the different treatments.

Lines 126 – 134: This section needs thorough revision: Too much unnecessary information (14-gauge needle), and a lot is missing: How was pH measured? What scale are values reported on? What total amount of sample was collected? Was the volume replaced?

Lines 136 – 138: Ahhh here it comes. Why the paragraph above?

Line 138: "as per Badocco": Quickly describe how that is done. A seawater buffer?

Line 139: What do you mean by "estimated". Please describe measurements with appropriate references.

Line 159: Expectations do not go into the method section. Please just describe what you did.

Line 160: "Scanning electron microscopy (SEM) analyses were conducted on a tabletop Hitachi TM4000 II, connected to a

Bruker QUANTAX energy-dispersive x-ray spectroscopy system (EDS)": This sentence is somewhat misplaced. Please report the methods in a chronological order. What sample was taken in what way and then what was measured on which device.

Line 162: What do you mean by remaining?

Line 163: Rinsed for how long? Was the MQ pH neutral? Usually, it's corrosive.

Line 165: What does "higher treatment" mean? Please rephrase.

Line 166a: "could have been used for CaCO<sub>3</sub> precipitation.": Do you mean that CaCO<sub>3</sub> might have formed around Mg(OH)<sub>2</sub> particles? If so, say that, please.

Line 166b ff.: „the case where CaCO<sub>3</sub> would have precipitated on yet to be dissolved Mg(OH)<sub>2</sub> particles, a clear signal of Mg<sup>2+</sup> would be observed. Otherwise, if CaCO<sub>3</sub> precipitated from the water

column only, the Mg<sup>2+</sup> signal would be absent. ": This sentence belongs to the discussion section.

Line 172a: Why HNO<sub>3</sub> and not HCl?

Line 172b: "try and". Delete that.

Line 173c: Rather remove *from* the wall?

Lines 174 – 176: Please clarify what you mean by this, what TA loss is referred to and please provide a formula that was used to carry out the calculations including an explanation of each variable used in the formula.

Line 179/180: "Throughout the experiments, quality control of the carbonate chemistry measurements was conducted using the TA, DIC and pH<sub>T</sub> data from each set to prevent loss of data to unexpected instrument misbehaviour.": This sentence is redundant. Please delete.

Line 180/181: "The DIC measurements from months 1 and 2 were likely underestimated, while the DIC measurements from month 6 were likely overestimated (Fig. 2).": This sentence belongs to the discussion along with an explanation why the authors come to this conclusion.

Lines 180 – 190: This entire section belongs to the discussion. The method section should only describe what has been done.

Figure 2 Belongs to the results section.

Line 197: This section along with the next one should be clearly marked as only referring to the solid phase experiments.

Line 199/200: "Within these first 24 hours, stark differences emerged between the material used and the advection regime.": In what way can there be a difference between a material and a reaction regime. Please rephrase this sentence.

Line 199/200: "The %TA<sub>realised</sub>, i.e., net measured ΔTA in μmol kg<sup>-1</sup> divided by the targeted ΔTA in μmol kg<sup>-1</sup>...": This term needs explanation and the formula for its calculation needs to be presented and explained.

Lines 202 – 203: "Advected bottles led to...": How much of the added material has hence dissolved? How did the saturation state with respect to the materials change? After what time is TA<sub>realised</sub> calculated? Is incomplete dissolution or already precipitation?

In addition: Bottles do not lead to anything. In bottles with advection higher values were observed.

Lines 205/207: "It appears that the measured TA...": Either it is lower or not. It does not appear to be something. Such formulations can be used in the discussion section, certainly not in the results part.

Line 214: Figure caption: "...after 1 day.": This explanation needs to appear in the main text.

Figure 4: Please add the control here. That makes it easier to understand  $p\text{CO}_2$  values.

Lines 272/273: “While both setups are meant to be identical...”: They are clearly not. Recent research that some of the authors were involved with have demonstrated that liquid addition differs significantly from solid phase addition.

Line 273: “clear difference in  $\Delta\text{TA}$  emerges”: Difference between what and what? The two solid state experiments were already different. Please rephrase for clarity.

Lines 291/292: “particles were used as surface area for  $\text{CaCO}_3$  precipitation”: Why not simply say whether  $\text{CaCO}_3$  had formed on  $\text{Mg}(\text{OH})_2$  particles? „Used“ sounds odd here.

Line 292/293: “The sample for the third replicate of the 0 RPM  $\text{Mg}(\text{OH})_2$  treatment was lost during analysis, and not all interpretations could be computed in Table 1 for this sample in particular”: This sentence appears out of nowhere. transfer it to the table caption or a point in the main text after Table 1 has first been mentioned for the data it contains.

Line 293: “The data”: Which data?

Lines 293 – 297: This is all interpretation. Only report results in the result part, please. Move it to the discussion or delete this section.

Line 298: “the 0RPM”: The 0 RPM what? a revolution per minute cannot really have  $\text{Mg}^{2+}$ . Please use precise scientific language.

Line 301: Table1: What does this table have to do with the section title? This should go to a respective discussion part. And please explain every parameter listed in the table and provide formulas how they were calculated in the method section.

Line 323: “Considering real-world applications for OAE is of high interest but is yet to be fully addressed.”: This sentence as it is, is completely redundant. Please delete or fill with meaning. Addressing a consideration does not make much sense.

Lines 324/325: “more classical experimental setup available in the literature”: What on earth is “classic” supposed to mean here? Likewise “available in the literature” is a weird wording. On top of that the authors should know that there are very well long-term experiments on OAE published in peer reviewed journals.

Lines 325/326: “intentional atmospheric  $\text{CO}_2$  invasion.”: Was the unnaturally high  $p\text{CO}_2$  in the atmosphere as indicated by the control  $p\text{CO}_2$  intended?

Line 329: Expectations do not go into the results part. Delete this sentence.

Lines 329 – 330: Clarify this statement: How does this affect the diffusion speed? To my knowledge the actual diffusion speed is always the same. The flux is just higher if  $\Delta C$  is higher or the diffusive distance is shorter if the dbf is thinner.

Lines 331/332: “Hence, TA generation was up to 50% lower at 0 RPM for the lower additions, due to the slower dissolution and higher saturation in the...”: A thicker dbf explains a time lack in alkalisiation, not a lower final alkalinity. Please rephrase this. Add: Depending on the conceptual model of a dbf, the saturation state in the dbf is always the same. The gradient is just steeper in this dbf.

Line 332: The concept of a diffusive boundary layer needs to be explained and backed up with an appropriate citation.

Line 333a: What disturbance to you refer to?

Line 333b: Flipkens 2023 proved that grain to grain interaction can foster olivine dissolution, but the conceptual idea is older. Meysman & Montserrat 2017 should be cited here.

Lines 338/339: Strictly speaking this is not an open ocean scenario. To argue in that direction, you need to refer to the  $\Omega$  values in your experiments.

Line 342: "...likely led to...": Without evidence in the form of particles or at least a drop in Mg concentrations, I would rephrase this to something like „Mg(OH)<sub>2</sub> formation and subsequent redissolution would therefore explain..."

Line 343: Which implications would that be?

Lines 347 & 348: Repeating "runaway fashion". Please rephrase.

Line 350: "Like in the NaOH experiments, CaCO<sub>3</sub> precipitation was suspected in the experiments with solid feedstocks": This sentence is redundant. Expectations make our lives easier, but are not an argument for or against anything. Please delete.

Lines 350/351: "The creation of highly alkaline microenvironments led to inevitable secondary CaCO<sub>3</sub> precipitation.": As long as you cannot measure or visualize these micro-environments you should use a more careful wording here.

Lines 351/352: "Furthermore, the low to inexistent advection led to longer CAP, allowing for more CaCO<sub>3</sub> to be formed": Why exactly? Please explain this in more detail.

Line 352/354: „The presence..." : This sentence just repeats what has just been said.

Lines 354/355: "In all experiments, CaCO<sub>3</sub> formation was suspected, with a clear decrease in DIC after addition in the 0 RPM experiments, and a loss in DIC for all levels above 300 µmol kg<sup>-1</sup> at 125 RPM.": What do you mean by "was suspected"? Either there is evidence supporting it or not.

Line 355/356: „This was confirmed by SEM imaging (Fig. 7) and particulate analysis (Table 1).": Please explain in what way that was supported by these data.

Line 357: Which data support that and in what way?

Line 358: What do you mean by the signal of Mg? Please clarify.

Line 359: If it was not confirmed via SEM it is rather possible than likely.

Line 363: Which external factor? Just name it, please.

Lines 363 – 367: These sentences are stating the absolute obvious. In addition, the citation does not really make sense here. Furthermore, the ratio of 2:1 would be realistic in a scenario without a source for TA. The loss term is not clearly defined as it consists of two terms: inhibited dissolution and precipitation. This ratio is therefore not to be expected in these experiments anyways, except from the liquid addition.

Line 375/376: Please explain that further. Why would the CAP be responsible for processes in the dbl?

Lines 377/378: This study has not looked at dbl processes, which are relevant here. Please provide more detail on your ideas on what has happened in the experiments.

Line 380/381: "Under such considerations...": Under what consideration? and why? Please be more specific.

Line 389a: "...in all experiments.": Did you find precipitates in your NaOH experiments?

Line 389b: What DIC fluxes? Into the bottles? Figure 8 shows concentrations, not fluxes. Can the actual fluxes even be calculated?

Lines 393/394: Why is that expected in Seawater? Please provide a respective reference.

Lines 416/417: Again. in the one hand side you talk about dbl processes, on the other here it's about impacts on organisms. Microbes? What effects are those? Please restructure this, so that the reader can better follow your thoughts.

Line 418: What changes in the carbonate system? You can shorten sections like this one, by getting to the point directly.

Line 420a: Which effects? Please explain this.

Line 420b: "Hence, the duration..." : This sentence contains the same information as the second sentence of this section, but is at the same time just as non-informative. Please

restructure this section to "Observed effects, where and why do they occur and then how your new (!) findings shed a new light on the current state of research".

Line 444: "see below": Where below? It is always a pain to have to jump back and forth in a script to understand it.

Line 446: "One could argue that...": I'm not going to highlight this anymore. Replace throughout the manuscript. Please. Especially, because it does semantically not make any sense. "One could argue that... , but at the same time this and that speaks against that argument".... that would be a valid construction.

Line 463: What does usually mean here? in which studies was that done?

Lines 450 – 468: For this section: I am missing the comparison to real world applications.

Planetary in Halifax is already using  $\text{Mg}(\text{OH})_2$  for alkalising the water column. How do they manage these questions? There should be at least some citable studies out there.

Line 470: Again: Name them. Unwanted changes can be anything.

Line 478: Here you would also need to cite Fuhr et al. 2023 and 2024 where these authors actually showed that added calcium carbonate dissolves under various geochemical conditions.

Line 480: In which bottle experiments. In general: The citations in this script are very scarce. The authors should definitely step back and redo their literature research on the various aspects discussed here. And despite the one answer to a previous reviewer: There are studies out there that have investigated OAE and e.g. effects on advection, grain interaction or secondary minerals. Despite the fact that some of them used materials other than  $\text{Mg}(\text{OH})_2$  and  $\text{Ca}(\text{OH})_2$  I am lacking a comprehensive comparison with their results.

Line 485a: Which consideration? How can one actually use a consideration?

Line 485b: "one could" see above.

Line 487: "However such needles may...": Based on what is this assumption made? Please provide a reference.

Line 489: "...allows for a higher CAO in the dbl": This is still contradictive. How do you define CAP in a dbl? At the grain surface, the A would always be C for long P (as long as the grain exists). Please clarify this at the respective point in the manuscript.

Line 490: "This was identified by...": In what way do these results support that? Please explain.

Line 494: "These statements are only hypotheses...": Please rephrase this to something like "Understanding the precise behaviour of the different particles, though, will need dedicated experimental work."

Line 496: "Current understanding of the global CDR potential...": Anything to back up this statement? Otherwise, I wonder where the authors take this information from.

Line 498: "DIC fluxes": I haven't seen a single flux calculation so far, let alone a resulting value.

Line 499: "ours": Our what? results? Please rephrase this sentence.

Line 499b: "first steps after": if you need to, at least say time steps, please.

Lines 502 – 505: Please provide an informed guess on how much  $\text{CaCO}_3$  has formed in each experiment and account for that in your flux calculations. I will therefore skip the rest of this section, until this has been performed.

Line 540: Likewise, I will skip the conclusion part for now. There are too many issues in the script that may or may not change the conclusion. Hence, I will gladly have closer look at it, once the other issues reported above have been fixed.