

Authors point by point comment review

Referee #1

Referee #1 found that you have addressed most of his/her comments and that the manuscript has improved. He highlights that there remains a few minor issues. He provides several suggestions for further improvement (copied below) as well as an annotated pdf manuscript (link below).

Thank you for the several suggestions provided to improve the manuscript. The annotated PDF manuscript has been reviewed and the changes have been implemented in the updated manuscript.

- Lines 26-27: Unclear, awkward sentence structure! Please reformulate the sentence.

This sentence has been reformulated as: “Finally, OAE simulations suggest that for the same Ω_A threshold, the amount of TA that can be added to seawater would be more than three times higher at 5 °C than at 30 °C. The maximum TA addition could also be increased by equilibrating the seawater to atmospheric CO₂ levels (i.e., to a pCO₂ of ~416 μatm) during addition.”

- Lines 41-42: The authors should put these numbers in perspective with respect to current and projected global anthropogenic emissions.

Thank you for pointing this out. An estimate of the current CO₂ emissions has been added (line 43).

- Line 45: This is one of many OAE strategies, others included land-based mineral dissolution and discharge of the resulting fluids and discharge of alkaline by-product solutions from electrolytic green hydrogen production.

Thank you for pointing this out.

- Lines 51-52: necessary, not only “advisable”. The carbon signature of heating the limestone and its decarbonation must be mitigated to make the process viable.

Thank you for mentioning that. The authors agreed and changed “advisable” to “necessary”.

- Lines 72-74: I find this statement misleading. The increase in pH and TA, decreases the

pCO₂ (increased sink or decreased source, as indicated in the first sentence of this paragraph) and increases the carbonate ion concentration, an increase in CO₂ buffering or neutralizing capacity. The increase in [Ca] does increase the saturation state, but that only relates indirectly to mitigating OA.

Thank you for this comment, the sentence has been changed to: “Furthermore, dissolving CaO and Ca(OH)₂ can also counteract ocean acidification. During the dissolution of alkaline minerals, both pH and the CaCO₃ saturation state of seawater (Ω) increase through increasing Ca²⁺ and CO₃²⁻ concentrations.” (lines 75-77)

- Lines 89-90: This threshold value is for instantaneous precipitation. CaCO₃ will spontaneously nucleate/precipitate at lower saturation states after a resting or lag time. There is extensive literature on this subject.

Thank you for pointing this out. The authors took note of this.

- Line 96: "understanding" of what? Please be more specific.

The paragraph has been changed to: “To gain a better understanding on the consequences of CaO and Ca(OH)₂ dissolution for OAE, we conducted several dissolution experiments with CaO and Ca(OH)₂ to determine 1) how much alkaline material can be dissolved without inducing CaCO₃ precipitation, 2) what causes secondary CaCO₃ precipitation, and 3) how secondary precipitation can be avoided.” (lines 101-103)

- Line 120: Unclear what alkaline compounds the authors are referring to, CaO or Ca(OH)₂ (or Na₂CO₃ to which the authors have not yet referred to)? Hence, how do these experiments differ from those described in sections 2.2.1 and 2.2.2 or is it a preamble to the latter?

Thank you for this comment, here we stated that “calculated amounts of weighed-in calcium alkaline compounds were added” (line 125)

- Lines 141-142: Were precaution taken to avoid absorption of CO₂ from the atmosphere?

Yes, we changed the sentence, mentioning that the Na₂CO₃ solution was freshly prepared on the day to limit CO₂ ingassing (lines 144-145)

- Line 195: Per kg of seawater?

Thank you for noting this, it is per kg of seawater. This information has been added (line 202).

- Line 205: The B:S ratio of Lee et al. (2000) is preferred (see Woosley (Mar. Chem. 229, 103214, 2021), although given the other uncertainties, is it not likely to affect the results significantly.

Thank you for mentioning it. Indeed, it would only affect slightly the results, which is why the authors agreed on keeping the ratio from Uppstorm (1974).

- Line 282: 1.5 hours after the Ca(OH)₂ addition or filtration? Probably the former, but unclear as written.

Thank you for noting this, the sentence has been changed to “within the first 1.5 hours after Ca(OH)₂ addition” (line 291)

- Line 333: surfaces would be more appropriate than phases.

“Surfaces” has been preferred and changed throughout the manuscript.

- Lines 345: morphotypes or polymorphs?

“Polymorph” has been preferred and changed throughout the manuscript.

- Line 358: "head space" is written as a single word elsewhere in the manuscript (L. 349). The spelling should be consistent throughout the manuscript.

The spelling consistency has been checked, thank you for pointing this out.

- Lines 365-367: This would be true of the experiments where the dissolution of CaO or Ca(OH)₂ was incomplete before CaCO₃ started to precipitate (e.g., targeted delta TA of 2000 μmol/kg), as proposed on lines 394-395. Hence, this statement may be misleading and not pertinent.

Thank you for pointing this out, the authors agreed on removing the statement.

- Lines 380-381, 383-386, 394-395: The presence of undissolved CaO or Ca(OH)₂ particles, as proposed on lines 394-395, as well as the local supersaturation at their surfaces could readily trigger CaCO₃ precipitation.

Thank you for pointing this out. This information has been noted.

- Line 399: Upon the addition of quartz particles? Please be more specific.

Thank you for noting this, we changed the sentence to: “Here, we observed CaCO₃ precipitation on quartz particles at an Ω_A of ~9.2” (line 404)

- Line 422: That is a very critical assumption as the proposed OAE process is otherwise useless.

Thank you for mentioning this, it is indeed an important point.

- Line 426: What about the global anthropogenic CO₂ emissions?

This information has now been added at lines 431-432: “To put these model-derived numbers into perspective, the global cement industry currently produces about 4.1 Gt of cement per year (Statista, 2021)”

- Lines 428-429: Awkward sentence structure and poorly justified statement.

This sentence has been reformulated.

- Lines 432-433: Poorly formulated and confusing.

Thank you for mentioning it, this sentence has now been reformulated.

- Line 456: See previous comment about precipitation on the surfaces of CaO or Ca(OH)₂ where the local supersaturation could be higher than in the bulk solution.

Thank you for pointing this out.

- Lines 467-468: This statement required additional information. What is the rate of TA addition? Over what period? From present to 2100, I presume. I also presume that the authors assume that the reduction would be relative, as atmospheric CO₂ emissions will persist and atmospheric levels will likely keep increasing.

Thank you for this comment, more information from the research paper of Feng et al. (2017) has been added.

- Line 470: I presume that the authors mean additions of minerals other than CaO and Ca(OH)₂. The authors should write more complete sentences to avoid confusion.

Thank you for mentioning this. The authors added more information of the minerals of interest for OAE (line 474).

Referee #2

Reviewer #2 is also satisfied with the responses and revisions made to the manuscript by the authors. He suggests to provide information on the reproducibility and accuracy of the measurements. He also suggest to provide additional information to reproduce the simulations conducted by the authors such as the model formulation? I support this suggestion. This could be done in supplementary information and the code made available.

Thank you for your comments. The authors added some more information regarding the simulations, as well as the instruments uncertainty, lines 189-192: “The overall instruments uncertainty for TA and DIC was calculated as follows. For each measurement, a standard deviation was calculated, from duplicates of TA and triplicates of DIC. The samples and reference materials standard deviations were averaged, and an error propagation on these values were used to estimate average measurement uncertainty, i.e., $\pm 1.0 \mu\text{mol kg}^{-1}$ and DIC at $\pm 0.8 \mu\text{mol kg}^{-1}$, for TA and DIC, respectively.”

Editor comments

- 21: total alkalinity

Thank you for pointing this out, it has now been changed.

- 26-27: sentence very difficult to comprehend

This sentence has been reformulated as: “Finally, OAE simulations suggest that for the same Ω_A threshold, the amount of TA that can be added to seawater would be more than three times higher at 5 °C than at 30 °C. The maximum TA addition could also be increased by equilibrating the seawater to atmospheric CO₂ levels (i.e., to a pCO₂ of ~416 μatm) during addition.”

- 32: it is poorly informative to cite a full report that is thousands of pages long! Perhaps cite instead the Summary for Policymakers or a specific chapter

Thank you for pointing this out, the reference to the IPCC report from 2021 has now been changed to the IPCC summary for policymakers of 2021.

- 33: Not 30%. According to Friedlingstein et al. (2022): 26% over the period 1850-2021.

Thank you for this comment, the authors reviewed the paper from Friedlingstein, 2022. After reviews, and based on the Table 8 data, the authors estimated that oceans absorbed nearly 40% of the fossil CO₂ emissions between 1750 and 2020 ($180/460 = 0.391 \sim 39\%$).

- 42: use superscript for 10¹⁵

Thank you for pointing this out. It has been addressed.

- 72: I strongly discourage introducing the expression Ω_{CaCO_3} . Omega will do

The notation has been changed from Ω_{CaCO_3} to Ω throughout the document.

- 220-221: do you really need 4 citations for such an obvious point?

Thank you for the comment. Only the reference to Morse 1997 has been reported.

- add an unbreakable space between a number and its unit (4 h rather than 4h)

- unless one refers to aragonite or calcite, in which case it is standard practice to use Ω_a and Ω_c (rather than Ω_{Ar} and Ω_{Ca} as is done elsewhere in the manuscript)

- Despite earlier comments in-text citations are still not ordered chronologically and the list of references not always properly formatted. Please pay attention to that during revision.