Response to the Reviewer 3

General Comment

Summary:
This study examines OAE's long-term efficiency and potential to remove CO2 from the atmosphere by simulating OAE in the GFDL ESM2M model from 2026 until the year 2500 under three different warming scenarios. The simulations show that atmospheric CO2 drops by 73-130ppm until 2500 with a larger reduction contribution occurring earlier in the simulation and under higher warming scenarios, while SAT tends to decrease nearly linearly over time and independently of the background warming level. This study also goes into detail about different metrics for OAE efficiency and how the chosen metric can influence the narrative and examines the ocean acidification mitigation potential of OAE, which can be traced back primarily to the reduction in atmospheric CO2 especially over centennial time-scales.
General comments:
The paper is overall of high quality, well structured with high quality figures, addresses relevant scientific questions related to the mCDR method OAE with a state of the art fully-coupled Earth system model and fits the scope of BG. However, the manuscript could be improved by primarily clarifying / adding details in the Methods and Results sections as listed below.

Response: We thank the reviewer for the positive assessment and the constructive suggestions to further improve the manuscript. Detailed replies to each specific comment follow below.

Specific Comments

L106-112: A more detailed description of the AERA would be beneficial here. It is without being familiar with the cited paper unclear how the emission pathways were chosen by the algorithm. Why / how does the algorithm e.g., lead to significantly different time windows for reaching the target temperature? Is a maximum positive / negative CO2 emission or target year specified?

Response: We have revised the text to clarify how AERA determines emissions pathways and associated timing of reaching the target temperature: " The AERA algorithm determines the emissions pathway by estimating anthropogenic warming, calculating the forcing-equivalent remaining emissions budget for the prescribed warming target using the transient climate response to cumulative CO_2-forcing-equivalent emissions (TCRE-fe), and then distributing the forcing-equivalent emissions budget over future years according to a third-order polynomial that leads to zero emissions. The timing of net-zero emissions, and thus the time window for reaching the target temperature, is an emergent property of the algorithm rather than an externally imposed constraint. If temperatures overshoot, the AERA responds by prescribing negative emissions. The pathway is recalculated every five years based on updated estimates of global mean temperature and the remaining CO_2-forcing-equivalent emission budget at the time of the stocktake (*Terhaar et al.*, 2022)."

L206: The results section provides lots of details via numbers, which partly compromises readability and dilutes the key findings. Maybe offloading details mentioned in the text to a new table could be a solution to maintain details and allow for quicker and easier comparison between simulations, while improving text readability.

Response: We appreciate the reviewer's suggestion and agree that the density of numbers can affect readability. Instead of introducing an additional table, we have revised the text to improve clarity by reducing the number of values reported within individual sentences and emphasizing the key findings more clearly. Specifically, we now focus on the most relevant quantitative results in the main text. We chose not to introduce a new table, as this would require frequent cross-referencing between text and tables, which can interrupt the narrative and make it more difficult to follow the interpretation of results. We believe that the revised presentation strikes a better balance between readability and providing sufficient quantitative detail.

L318-319: In the model description the authors mention the possibility of sedimentary calcite shell deposition (L94). In combination with the statement here, it is unclear how much of the added alkalinity via OAE is actually removed from the water column via the ecosystem and enters the sediment.
On that note, the authors mention in the Methods section that the model represents alkalinity well (L99), but don't show a Figure of the background ocean alkalinity inventory and no comparison of how much the inventory changes as a response to the yearly alkalinity addition over time.

Response: We thank the reviewer for highlighting this important point. We have revised the manuscript to clarify the fate of added alkalinity and to better document changes in the ocean alkalinity inventory.

To address the concern regarding alkalinity removal via sedimentary processes, we now explicitly describe how calcification responds to changes in saturation state in the model and how this affects the alkalinity budget. In particular, increased calcification under OAE conditions enhances alkalinity removal from the water column, which is reflected in changes to the internal alkalinity sources and sinks, including sediment interactions.

In addition, we have included a new figure in the Appendix (Fig. C3; also shown below) showing (i) the evolution of the global ocean alkalinity inventory over time, (ii) the additional alkalinity in the ocean in comparison to the amount of alkalinity enhancement and (iii) the combined internal alkalinity sources and sinks, including sedimentary processes. This allows for a direct assessment of how much of the added alkalinity remains in the ocean versus being removed.

We now refer to this figure at several points in the manuscript. At the beginning of Section 'Global carbon flux response', we describe the overall increase in alkalinity inventory due to OAE and its implications for carbon uptake: "Ocean alkalinity enhancement substantially increases the oceans' alkalinity inventory (Fig. C3). The added alkalinity in turn leads to additional ocean carbon uptake and lowers atmospheric CO_2, but part of this drawdown is offset by carbon release from the land biosphere (Fig. 5)." Furthermore, in the section on net ocean capture efficiency, we explicitly link enhanced calcification to a reduction in the effective alkalinity source (Fig. C3c): "In our Earth system model, calcification responds to changes in the calcite and aragonite saturation states and we find more calcification under OAE than in the reference scenarios, leading to an overall reduction in additional alkalinity within the ocean (Fig. C3c)." We also expand the discussion of biogeochemical feedbacks to clarify that both chemical adjustments and climate-driven responses contribute to reducing the net alkalinity gain, as illustrated by the differences between simulations in Fig. C3c: "Overall, these feedbacks substantially lower the total additional alkalinity in the ocean, where a quite equal part of the reduction comes from chemical changes due to OAE or is related to the reduction in atmospheric CO_2 following OAE (see the difference of OAE and Ref* to Ref in Fig. C3c)."

We believe these additions clarify how alkalinity is redistributed and partially removed within the Earth system, and directly address the reviewer's concern.

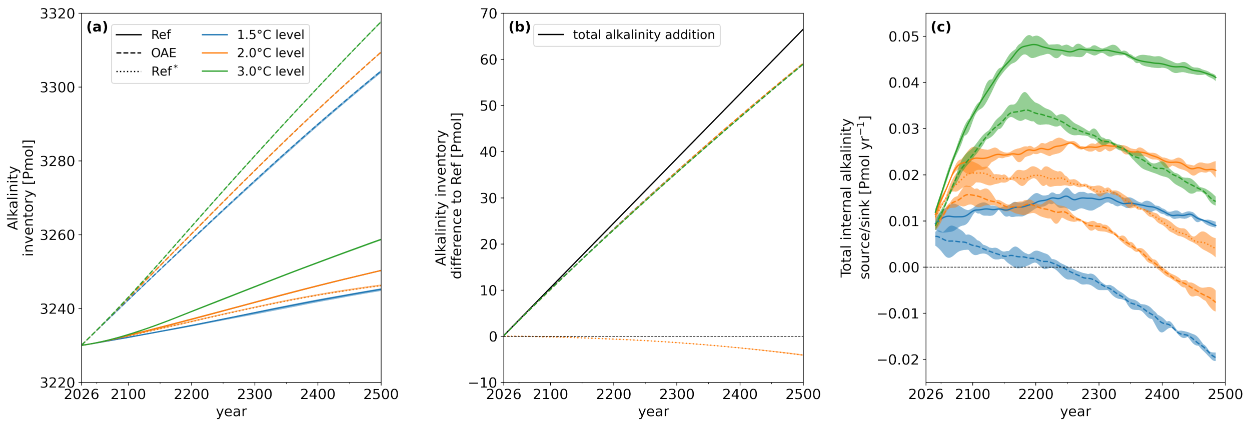

Figure C3. Simulated ocean alkalinity inventories (a), differences in the alkalinity inventory of the OAE and Ref* simulations to the Ref simulation (b) and the sum of all ocean internal alkalinity sources and sinks, including sediment processes (c) for all global warming scenarios with and without OAE. Lines are the five member ensemble mean and shading refers to the ensemble range. Lines in panel (c) are smoothed with a 31-year running mean.

> L339-341: This detail should be mentioned in the Methods section and be included in Figure1. Currently, Figure1 is inconsistent with Figures 8-10.

Response: We have adapted Figure 1 and moved the sentence to the end of the "Simulation setup" section, now saying: "To investigate the effect of OAE termination on both the efficiencies and the ocean acidification response, we extended one ensemble member of the 2°C OAE simulations to the year 3000, but terminating OAE in 2500, and extended both reference simulations as well."

Technical Corrections

> L30-31: Please add a citation to the statement or rewrite.

Response: We have added a citation here and rephrased it a bit: "Current efforts have largely focused on implementing land-based CDR methods, while ocean-based approaches remain comparatively understudied (*Smith et al.*, 2024; *Boyd et al.*, 2025)."

> L34: "alkaline materials" - not just materials, but also alkaline solutions, which would actually represent the model OAE implementation much better due to not modelled mineral dissolution processes in the surface ocean, but instantaneous dissolution.

Response: We have clarified: "OAE involves adding alkaline materials, such as olivine or quicklime, or alkaline solutions to seawater to enhance its natural ability to absorb atmospheric CO_2."

> L36: 'bicarbonate and carbonate ions ... stable ... for 10000 to 100000 years': This is a bit misleading formulation. First of all bicarbonate and carbonate ions are not conservative tracers, i.e. they are not even stable wrt mixing. More importantly, this quote ignores potential non-stability (or reduced durability) wrt to interactions with CaCO3 sediments and also wrt to modified terrestrial weathering. While this is often not considered in OAE studies (since they usually focus on centennial timescales) and also not by the paper under consideration, it has been studied by Köhler et al. 2020 (https://doi.org/10.3389/fclim.2020.575744) with a box model that can simulate such long time scales. I suggest to rephrase and refer to Köhler's work here.

Response: We thank the reviewer for this important clarification. We agree that the original wording was misleading, as bicarbonate and carbonate ions are not conservative tracers and their long-term persistence is influenced by processes such as $CaCO_3$ sediment interactions and changes in terrestrial weathering. We have therefore rephrased it to better reflect the timescale dependence and underlying assumptions, and we now explicitly acknowledge these longer-term processes and cite Köhler et al. (2020). The revised text reads: "Through this process, aqueous CO_2 is converted into bicarbonate and carbonate ions, which represent longer-lived forms of inorganic carbon in the ocean on centennial to millennial timescales (*Renforth and Henderson*, 2017; *Middelburg et al.*, 2020; *National Academies of Sciences, Engineering, and Medicine*, 2022; *Ho et al.*, 2023). On longer timescales, the persistence of the added alkalinity is limited by $CaCO_3$ sediment interactions and weathering feedbacks (*Köhler*, 2020)."

> L43: "CO2 sequestered" - additional ocean carbon uptake

Response: Changed.

> L52: "decrease" - reduce

Response: Changed.

> L56/64: 'most modelling studies' (56) vs. 'only a few ... modelling studies' (64), however, in both cases you cite six studies, each - I don't see your rational for 'most' vs. 'few' here

Response: We thank the reviewer for pointing out this inconsistency. Our original wording ("most" vs. "few") was not sufficiently precise given the similar number of cited studies in both cases and could therefore be misleading. We have revised the text to avoid this ambiguity and now use more neutral and consistent wording. In particular, we rephrased the latter sentence to: "Recently, more fully coupled modelling studies have become available, although the complexity of the models differs. They consistently show that interactions among the atmosphere, ocean, and land biosphere strongly reduce OAE efficiency compared to modelling experiments with prescribed atmospheric CO_2 (*Jeltsch-Thömmes et al.*, 2024; *Schwinger et al.*, 2024; *Tyka*, 2025; *Wey et al.*, 2025; *Sathyanadh et al.*, 2025; *Nagwekar et al.*, 2026)."

> L68:Unclear what is meant with "single" Earth system models. Maybe "other" or "previous"?

Response: Changed to "other". Thank you.

> L88: Please change to 'It simulates marine and terrestrial anthropogenic carbon uptake ...', to be explicit here. (I assume you refer to both marine and terrestrial uptake and storage.)

Response: Yes, this it correct. We have changed it to: "It simulates the uptake and storage of anthropogenic carbon by the marine and terrestrial carbon sinks close to observations (*Frölicher et al.*, 2015; *Bronselaer et al.*, 2017), and it represents alkalinity well by actively simulating both calcite and aragonite cycling in comparison to other climate models (*Planchat et al.*, 2023)."

> L126: Can you please clarify how/weather non-CO2 GHG forcing was applied in the Ref* simulations.

Response: We have added: "The non-CO_2 forcing is the same as in the OAE and Ref simulations."

> L175: 'to reduce atmospheric pCO2 and therefore global warming', I find this formulation a bit awkward, since in a transient climate warming is related to cumulative emissions (i.e. the TCRE), not the atmospheric pCO2

Response: We agree that, in a transient framework, global warming is more directly related to cumulative CO_2 emissions (via the TCRE) rather than instantaneous atmospheric CO_2 concentrations. However, we chose to retain the original formulation for clarity, as ocean alkalinity enhancement directly reduces atmospheric CO_2, which in turn affects radiative forcing and thus contributes to limiting warming.

> L190-4: I am not really convinced by the name of the term delpH-Hysteresis

Response: We thought about this again and came up with a new description. We now refer to the process as the "equilibrated pH response" and refer to it in the formulas and figures as "$\Delta pH_{equilibrated}$". We chose this to also refer to the analogy of equilibrated and un-equilibrated OAE often used in research about the ecosystem impact of OAE and added a description about this in the methods section: "Research about the ecosystem impact of OAE often distinguishes between equilibrated and un-equilibrated OAE (*Hartmann et al.*, 2023). Equilibrated OAE assumes that the alkaline solution added to the ocean has already re-equilibrated with the atmosphere and taken up CO_2 according to the maximum ocean capture efficiency, characterized by the equilibrated pH response. Un-equilibrated OAE assumes that pCO_2 is not yet back to equilibrium with the atmosphere, and the pH response is here characterized by the equilibrated pH response together with the pCO_2 disequilibrium effect (described below)."

> L212 / caption Fig. 3: 'air temperature anomalies'

Response: Changed.

> Caption Fig. 4: alike: indicate that you show anomalies hier

Response: We refrain from adding anomalies here, since it is just the difference between the two simulations, as stated.

L255: 'when global warming is stabilized': sounds as if the warming (carbon-climate feedbacks) is the central point here, but I guess that the decrease in marine CO2 uptake (rate) is primarely carbon-concentration driven, i.e. related to having negative emissions in Ref

Response: That is a good point, we have rephrased it to be more precise: "In the reference simulations, the total cumulative ocean carbon uptake since pre-industrial increases rapidly initially with increasing CO_2 emissions and slows markedly toward the end of the simulation when emissions reach near-zero or become negative (solid blue lines in Figure 5)."

Fig. 5: Interesting that in this model the land is a net source of carbon until about 2050 in OAE and Ref. Please add a comment on why this behaviour occurs (c-climate vs. c-concentration feedback?) and point to a reference, if this was discussed in a previous paper.

Response: We thank the reviewer for this interesting comment. The negative cumulative land carbon flux in the early period is primarily driven by historical land-use change emissions, which dominate over the land carbon uptake, as shown by *Silvy et al.* (2024). Until around 1970, the land is a net carbon source following land-use change, but a sink thereafter. It takes until 2050 for this initial loss to be compensated by additional land carbon uptake. We have clarified this in the manuscript: "The reduction in atmospheric CO_2 is modulated by anomalous carbon release from the land biosphere. Note that the historical carbon release from land is due to land-use change emissions, which dominate over the land carbon uptake due to increases in atmospheric CO_2 (*Silvy et al.*, 2024)".

L272: "signal(s)" typo

Response: Changed.

L417: "experimental setups" - such as carbon / temperature overshoot scenarios.

Response: Thank you. We now state: "Nevertheless, the robustness of this result remains to be tested across other Earth system models and experimental setups, such as carbon and/or temperature overshoot scenarios."

L436: Please add a comment if this difference is (in)-significant.

Response: We have highlighted the difference but we think a detailed statistical analysis about significance is not necessary here. We do not infer the significance of differences between lines or scenarios throughout the manuscript and we do not see an extra benefit of stating, if the two lines are significantly different or not at the specific time in 2500, further based on defining a significance level and the chosen statistical test. We now say: "In 2500, inventory-based and flux-based efficiency differ by 0.01 units with distinct ensemble ranges."

Figure C2: Please add temperature labels above the three columns similar as done in Figures 4-5.

Response: Good point. We have added it.

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