Referee #3

General comments

The manuscript investigates the potential for atmospheric CO₂ removal via ocean alkalinity enhancement using waste products from cement and lime kilns. The study is based on laboratory dissolution experiments and evaluates the dissolution kinetics, CO₂ sequestration potential, and ecological risks associated with cement kiln dust (CKD) and lime kiln dust (LKD). In my opinion, the manuscript offers a valuable contribution to scientific progress within the scope of Biogeosciences, presenting new concepts and ideas for CO₂ removal that worth further testing and investigation.

Overall, the manuscript is well structured and clearly written. The language is fluent, the figures are clear and easy to interpret, and the amount and quality of the supplementary material are appropriate.

The conclusions are generally supported by the experimental results. However, I have reservations about the representativeness of the reported values, given that the experiments were conducted over relatively short timescales (8 hours and 15 days) in 200-mL polystyrene vials filled with filtered seawater under controlled laboratory conditions. These constraints limit the extent to which the findings can be extrapolated to real-world applications. In my view, the discussion, conclusions, and abstract should adopt a more cautious tone regarding the scalability and environmental impacts of the results.

The manuscript also addresses the potential ecological impacts of kiln dust dissolution. However, the assessment focuses primarily on turbidity and trace metal concentrations, without sufficiently considering biological responses or broader ecological consequences of kiln dust deployment in marine environments.

Finally, the Methods section could be strengthened by providing additional detail to facilitate reproducibility by other researchers.

I hope the comments below help the authors improve the clarity, rigor, and reproducibility of the study.

We sincerely thank the reviewer for their thoughtful and supportive comments on our manuscript. The feedback has significantly improved the clarity, organization, and overall quality of the paper. In the sections below, we address each of their points and outline the corresponding revisions. Our responses are presented in regular black text, while the revised manuscript text appears in blue with changes highlighted in yellow.

We agree with the reviewer's general comments and have incorporated their suggestions throughout the manuscript, as detailed in the specific comments section below. In response to the recommendation that the discussion, conclusions, and abstract should adopt a more cautious tone, given that the study was conducted under controlled laboratory conditions, we have revised the abstract accordingly. Revisions to the discussion and conclusions are described under the relevant specific comments. The revisions made to the abstract are as follows:

Based on current industrial production rates, this translates into global CDR potentials of up to 8.7 ± 0.6 Mt CO_2 yr^{-1} for LKD and 25 ± 2 Mt CO_2 yr^{-1} for CKD. These estimates suggest that both materials could be viable OAE feedstocks, although further testing under conditions that more closely mimic natural coastal conditions is needed.

Specific comments

Material and methods

- Please specify the source of all materials used in the experiments (e.g., supplier, kiln type, facility, geographical origin). This information is essential for assessing the broader applicability of the results and ensuring reproducibility.

Due to a non-disclosure agreement, we are unable to provide the name of the kiln dust supplier or specific details regarding its production process. However, we respectfully disagree that this specific information is essential for evaluating broader applicability or ensuring reproducibility. The comprehensive physicochemical characterization included in our study provides all necessary information for comparing our results with future work using kiln dust from other sources, thereby enabling meaningful assessment and replication.

- Please clarify which analytical methods were applied in each of the experiments. For Experiment II, indicate how total alkalinity (AT) was measured.

The same analytical methods were used for both experiments, which are described in section 2.3 to avoid repetition. To improve clarity, we have now added a reference to this section within the methods description of Experiment II. The revised text now reads as follows:

Duplicate samples for dissolved inorganic carbon (DIC), dissolved metals, turbidity, and A_T analysis were collected on both sampling days by drawing water with a syringe after opening the vials (analytical procedures are described in Sect. 2.3).

We have also added a reference to the section describing the solid-phase analyses to clarify that the same SEM–EDX procedure was applied to the recovered samples from Experiment II as to the fresh kiln dust.

The remaining suspension in the incubation vials was filtered through a 0.2 µm polycarbonate membrane filter to collect solids, which were rinsed with deionized water and then oven dried at 40°C in preparation for SEM-EDX analysis (see Sect. 2.1).

- It appears that only two sampling times (after 1 day and after 15 days) were analyzed for Experiment II. Please clarify in the manuscript.

For Experiment II, two sampling times were indeed selected. To improve clarity, this information, which was originally placed mid-way through the Experiment II description, has now been moved to the beginning. Furthermore, the term “longer-term” has been removed from the text in accordance with a later comment.

To assess the alkalinity generation potential and the possibility of secondary mineral formation, we conducted a second dissolution experiment with incubation periods of one and 15 days. The one-day (i.e. 24 h) incubation ensured complete dissolution of the reactive phases in the kiln dusts, while the 15-day incubation allowed for the verification of secondary mineral precipitation, in case this would occur.

Lines 90-92 – In the sentence “Based on preliminary tests, three different masses of CKD and LKD were added targeting a specific aragonite saturation state (ΩArg) at the end …” To facilitate reproducibility, please indicate the correspondence masses values (in grams) added to the 200 mL of FSW.

This information was already provided in Table 1, but has now also been added to the text to clarify the kiln dust concentrations and their corresponding targeted aragonite saturation states:

Based on preliminary tests, three concentrations of CKD (30, 130, and 309 mg kg^{-1}) and LKD (11, 48, and 113 mg kg^{-1}) were selected to target different aragonite saturation states (Ω_{Arg} = 3.6, 5.7 and 9.7) (Table 1).

Lines 103- 104- Referring to the 15-day experiment as a "long-term dissolution experiment" may be misleading in the context of OAE. I suggest referring simply to a 15-day experiment, unless additional justification is provided.

We agree, the wording "longer-term" has been removed from the text (see revised text in previous comments).

Lines 105-109 – For reproducibility, please provide details on the number of replicates used and the number of vials per treatment.

Thank you for noting that this crucial info was missing. The number of replicates has now been added to the text.

Vials were closed tightly and had minimal headspace to minimize gas exchange with the atmosphere. Experiment II was conducted in triplicate at ambient room temperature (17.5–22.7 °C).

Line 104 – Are 200 mL plastic vials representatives of sea water collum? Please explain the rationale for choosing this container size.

We agree with the reviewer's later comments that larger-scale experiments, which better mimic natural conditions, are ultimately needed to fully assess the CDR potential and ecological risks of kiln-dust-based OAE. However, the use of 200 mL vials in this study was intentional: small-scale incubations allow rapid, cost-effective screening of multiple treatments simultaneously, which is appropriate for a first assessment of whether a material is suitable for OAE. This has been clarified in the text as follows:

Kiln dusts were weighed in small aluminium (Al) foil cups using a micro balance (XP26 Excellence Plus, Mettler Toledo) and then transferred to 200 mL polystyrene vials with polyethylene screw caps containing approximately 200 mL of FSW. These small-scale laboratory experiments provide a high-throughput, cost-effective first assessment of a material's suitability for OAE.

Line 109 – Please clarify why different rotation speeds were used in the two experiments (700 rpm in Experiment I vs. 14 rpm in Experiment II). What was the intended effect of this difference?

Different rotation speeds were used due to differences in the experimental set-ups. In Experiment I, vials were kept upright on a magnetic stirrer to allow continuous pH monitoring. In Experiment II, bottle rollers were employed since pH monitoring was not required and this setup allowed simultaneous incubation of a larger number of vials. In both experiments, rotation speeds were selected to ensure effective mixing and to maintain particles in suspension, creating ideal conditions for dissolution and enabling assessment of the maximum CDR potential. The rationale for these rotation speeds is further clarified in the text, which now reads as follows:

Seawater temperature was kept constant at 20°C during the incubation by means of a water bath (T100, Grant). Magnetic stirring was applied at a rate of 700 rotation per minute (RPM) to ensure good mixing of the suspension and to create optimal dissolution conditions.

Experiment II was conducted in triplicate at ambient room temperature (17.5–22.7 °C). Vials were subsequently incubated on bottle rollers (ThermoFisher Scientific) for 1 or 15 days at 14 RPM, a speed sufficient to keep particles suspended and ensure optimal dissolution conditions.

Lines 109-110 – In the sentence "The one-day incubation reflects the time needed for complete dissolution of the reactive phases in the kiln dusts.", please specify the exact duration in hours from the start of the experiment until dissolution was considered complete.

The exact duration of complete reactive phase dissolution is not known, but Experiment I suggests it occurs within 24 hours, a finding confirmed by the 15-day incubation, which showed no additional alkalinity release compared to the 1-day incubation of Experiment II. The sentence referred to by the reviewer has been rephrased to:

The one-day (i.e. 24 h) incubation ensured complete dissolution of the reactive phases in the kiln dusts, while the 15-day incubation allowed for the verification of secondary mineral precipitation, in case this would occur.

Lines 237-238 – In the sentence "…while for LKD, the ΔAT curve showed a maximum at higher concentrations (Fig. 2B)" does not seem to match the figure: the maximum appears to occur before the highest concentration. Please revise accordingly.

You are correct; the sentence has been revised as follows:

After one day, ΔA_T showed a monotonous increase with the CKD concentration, while for LKD, the ΔA_T curve reached a maximum at 69 mg kg^{-1} and decreased at higher application concentrations (Fig. 2B).

Lines 318-319 – Regarding this sentence "The compositional complexity of kiln dusts underscores the need for detailed mineralogical and chemical characterization to properly assess the CDR potential and environmental risks in OAE applications.", more details about the provenance of the kiln dust should be added to the methods section.

As mentioned previously, we are unable to disclose the origin of the kiln dusts; however, we believe this information is not critical given the detailed physicochemical characterization provided.

Line 363 – In the sentence "… leading to a net AT loss, as seen at the highest LKD concentration after 15 days (Fig. 2A)". the reference appears to be to Figure 2B rather than Figure 2A, please check.

You are correct, we changed the reference from Figure 2A to 2B.

Line 371 – In the sentence "… several minutes during ship-based ocean liming Caserini et al. (2021), which may have an impact on marine life if pH exceeds". It seems that the reference should be placed between brackets, please check.

The reference should indeed be between brackets, this has now been corrected.

Line 373-374 – "To avoid temporary exceedances of pH 9, CKD concentrations should stay below 343–502 mg kg-1, and LKD below 102–149 mg kg-1, depending on local seawater conditions (AT = 2350 µmol kg-1, DIC = 2100 µmol kg-1, salinity = 35, temperature = 10–25 °C)."

Your experiment was conducted at a salinity level of 32.3 ± 0.5 and a temperature level of 17.5–22.7 °C. Are these values comparable to a salinity of 35 and a temperature of 10–25 °C? Would these variations in salinity and temperature influence the concentrations of CKD and LKD? What impact would they have on the pH? These points need to be clarified in the manuscript.

Temperature, salinity, hydrostatic pressure, and the background seawater total alkalinity and DIC all influence seawater pH, and thus the amount of kiln dust that can be added before reaching pH 9. The values reported in the manuscript correspond to average surface seawater conditions under temperate and tropical temperatures, calculated from the maximum specific alkalinity release measured for LKD (8.0 mmol g^{-1}) and CKD (2.4 mmol g^{-1}). Using these specific alkalinity release rates, local maximum application concentrations can be readily calculated once the seawater conditions at a specific deployment site are known. This clarification has now been added to the text as follows:

To avoid temporary exceedances of pH 9, CKD concentrations should stay below 343–502 mg kg^{-1}, and LKD below 102–149 mg kg^{-1} under average surface seawater conditions (A_T = 2350 µmol kg^{-1}, DIC = 2100 µmol kg^{-1}, salinity = 35, temperature = 10–25 °C). Application concentrations must be further tailored to local seawater geochemistry at the deployment site to prevent exceeding the pH 9 threshold.

Line 385-387 – "In real applications, kiln dust will be rapidly mixed into much larger volumes of surface water, …."

How representative is your experiment, that used 200 ml sea-water bottle, to be extrapolated for real application conditions? How would be guarantee a concentration below the referenced concentrations at the discharge point? In my opinion, without stronger justification, the scalability of the results remains uncertain.

Our results demonstrate a clear linear relationship between suspended particle concentrations and seawater turbidity for the kiln dust studied. To ensure concentrations remain below turbidity guidelines, numerical modelling is required that incorporates local hydrodynamic conditions and kiln dust particle behaviour to determine appropriate discharge rates from the ship. We have elaborated on this in the text and cited two studies addressing modelling considerations for OAE and the effective settling of mineral particles in the ocean relevant to mCDR applications.

In real applications, kiln dust will be rapidly mixed into much larger volumes of surface water, meaning that the allowable concentration in the input stream will depend on the discharge rate, intensity of local turbulence, and kiln dust settling time (which is primarily determined by initial particle size). Accurate numerical modelling to determine suitable discharge rates therefore requires detailed knowledge of the environmental conditions at the deployment site and the behaviour of kiln dust particles under varying hydrodynamic conditions (Fennel et al., 2023; Yang and Timmermans, 2024). This information is essential to extrapolate small-scale laboratory results to realistic field scenarios and ensure that concentrations remain below guideline levels.

Line 407-409 – "Assuming full mixing in the top 10 cm of the sediment, up to 1.4 kg CKD or 74.8 kg LKD per m^2 could be applied ….."

Please clarify what would be the resulting thickness of kiln dust deposited on the seafloor? Even if the 10-cm surface layer is assumed to be fully mixed over time, the initial deposition could create a substantial layer of fine material. Benthic infauna and epifauna cannot survive rapid burial under more than ~1–2 cm of sediment, and a thick layer of fine particles would also strongly reduce oxygen exchange at the sediment–water interface. This physical disturbance should also be addressed in the discussion, as compliance with chemical SQGs alone does not ensure ecological safety.

We agree that physical disturbance is an important aspect that should be incorporated into the discussion. The resulting thickness of a deposited kiln-dust layer depends on its bulk density and solid-phase density, which vary with grain morphology, mineralogy, and local grain packing. Assuming a kiln dust bulk density of 0.62 g cm^{-3} based on the study by Nikolov et al. (2025), the maximum application rates derived from trace-metal SQGs would produce surface layers of approximately 0.2 cm for 1.4 kg m^{-2} CKD and 12 cm for 74.8 kg m^{-2} LKD, assuming uniform spreading.

These estimates indicate that physical burial is of low risk during CKD spreading, whereas for LKD the potential for burial impacts is considerable. A layer of this thickness would be expected to cause mortality of low-mobility benthic fauna and strongly reduce oxygen exchange at the sediment–water interface, thereby altering sediment geochemical processes. However, from a CDR perspective, applying such a thick layer of fine-grained LKD would also be inadvisable for other reasons, including the reduced efficiency of $CaCO_3$ dissolution at high application rates (Dale et al., 2024) and potential changes in habitat suitability for benthic organisms (Flipkens et al., 2024).

We have now incorporated the estimated kiln-dust layer thicknesses and associated physical impacts into the revised manuscript text as suggested.

Assuming full mixing in the top 10 cm of the sediment, up to 1.4 kg CKD or 74.8 kg LKD per m² could be applied to pristine sediments without exceeding the strictest marine SQG of 30.2 mg kg^{-1} for Pb (Appendix C). Using a kiln dust bulk density of 0.62 g cm^{-3} (Nikolov et al., 2025), this corresponds to an applied layer of approximately 0.2 cm for CKD and 12 cm for LKD. The exact layer thickness would of course be dependent on the specific kiln dust properties and local grain packing. These estimates indicate that burial risk for benthic organisms is small for CKD, but could be considerable for LKD, since the deposition of a cm-thick layer could substantially impact the resident benthic infauna and epifauna. Moreover, applying LKD at this scale is also not advisable because it may lead to changes in habitat suitability (e.g., grain size, permeability, organic carbon content) (Speybroeck et al., 2006; Flipkens et al., 2024) and alter geochemical sediment processes (see Sect. 4.4). Overall, these findings underscore the need for ecotoxicological testing and cautious application of kiln dust to avoid ecological harm.

Line 420-424 – The manuscript notes that large-scale fining of permeable sediments by kiln dust could reduce oxygen penetration depth and thereby limit the zone of metabolic $CaCO_3$ dissolution. However, the biological implications of such physical and geochemical changes are not discussed. What would be the expected impact on benthic organisms living within these sediments, particularly those that produce or maintain calcium carbonate shells (e.g., molluscs, foraminifera, small crustaceans)? Reduced permeability, shallower oxic layers, and burial by fine material could harm calcifiers through smothering, reduced oxygen availability, and altered porewater chemistry. I recommend addressing these ecological consequences alongside the geochemical considerations

We have expanded the discussion to address additional geochemical and physical changes beyond reduced oxygen penetration depth. We also added a reference to Section 4.3, where potential ecological impacts, including burial and changes in habitat suitability arising from both physical and chemical alterations, are now discussed as mentioned in the previous comment. The revised text reflects these broader considerations.

However, large-scale fining of sediment with kiln dust could reduce sediment properties, such as the permeability, solute exchange rates, and oxygen penetration depth (Speybroeck et al., 2006; Ahmerkamp et al., 2017). The latter would limit the zone in which metabolic $CaCO_3$ dissolution can occur. Relatively high $CaCO_3$ concentrations may further reduce the dissolution efficiency (i.e. dissolution rate per amount of $CaCO_3$ added) (Dale et al., 2024). Additional ecological impacts may arise through changes in the physical texture of the solid sediment matrix and modifications of porewater conditions (see Sect. 4.3). The potential for enhanced sedimentary alkalinity generation via residual kiln dust addition to organic-rich, carbonate-poor marine sediments therefore warrants further experimental investigation.

Line 470-471 – "Overall, LKD and, to a lesser extent, CKD show promise for OAE, with a CDR potential of up to 13.4 Mt year-1 for LKD and 57 Mt year-1 for CKD at current production levels"

In my opinion, given the significant uncertainties surrounding ecological impacts, including water-column responses, sediment interactions, and potential biological effects, the Conclusions should acknowledge these uncertainties and reflect them in the overall assessment of kiln dust as a viable CDR strategy.

We agree that the conclusions could be more carefully worded, given that this is the first study assessing the potential of kiln dust for OAE and that our experiments were conducted on a small laboratory scale. Accordingly, we have revised the text to reflect these uncertainties and included recommendations for future research to address water column responses, sediment interactions, and potential biological impacts under more realistic environmental conditions.

Overall, LKD and, to a lesser extent, CKD show promise for OAE, with a CDR potential of up to 13.4 Mt year^{-1} for LKD and 57 Mt year^{-1} for CKD at current production levels, based on our small-scale laboratory experiments. However, additional experiments that more closely mimic natural conditions are warranted to further constrain particle behaviour in the water column, interactions with sediments, and potential biological impacts of kiln-dust–based OAE.

References:

Ahmerkamp, S., Winter, C., Krämer, K., Beer, D. d., Janssen, F., Friedrich, J., Kuypers, M. M., Holtappels, M.: Regulation of benthic oxygen fluxes in permeable sediments of the coastal ocean, Limnol. Oceanogr., 62, 1935-1954, https://doi.org/10.1002/lno.10544, 2017.

Dale, A. W., Geilert, S., Diercks, I., Fuhr, M., Perner, M., Scholz, F., Wallmann, K.: Seafloor alkalinity enhancement as a carbon dioxide removal strategy in the Baltic Sea, Commun. Earth Environ., 5, 452, https://doi.org/10.1038/s43247-024-01569-3, 2024.

Fennel, K., Long, M. C., Algar, C., Carter, B., Keller, D., Laurent, A., Mattern, J. P., Musgrave, R., Oschlies, A., Ostiguy, J.: Modeling considerations for research on Ocean Alkalinity Enhancement (OAE), State of the Planet Discussions, 2023, 1-47, https://doi.org/10.5194/sp-2-oae2023-9-2023, 2023.

Flipkens, G., Dujardin, V., Salden, J., T'Jollyn, K., Town, R. M., Blust, R.: Olivine avoidance behaviour by marine gastropods (Littorina littorea L.) and amphipods (Gammarus locusta L.) within the context of ocean alkalinity enhancement, Ecotoxicol. Environ. Saf., 270, 115840, https://doi.org/10.1016/j.ecoenv.2023.115840, 2024.

Nikolov, A., Kostov-Kytin, V., Tarassov, M., Tsvetanova, L., Jordanov, N. B., Karamanova, E., Rostovsky, I.: Characterization of cement kiln dust from Bulgarian cement plants, J. Chem. Technol. Metall., 60, 455-463, https://doi.org/10.59957/jctm.v60.i3.2025.11, 2025.

Speybroeck, J., Bonte, D., Courtens, W., Gheskiere, T., Grootaert, P., Maelfait, J. P., Mathys, M., Provoost, S., Sabbe, K., Stienen, E. W.: Beach nourishment: an ecologically sound coastal defence alternative? A review, Aquat. Conserv.: Mar. Freshw. Ecosyst., 16, 419-435, https://doi.org/10.1002/aqc.733, 2006.

Yang, A. J., Timmermans, M.-L.: Assessing the effective settling of mineral particles in the ocean with application to ocean-based carbon-dioxide removal, Environ. Res. Lett., 19, 024035, https://doi.org/10.1088/1748-9326/ad2236, 2024.