Referee #1

The authors present experimental results on the reactivity of kiln dust in seawater. Kiln dust is a waste product of the cement and lime industry that is either recycled or disposed in e.g. landfills. The authors show that reactive phases in kiln dust (CaO, Ca(OH)2) are rapidly dissolved in seawater such that most of these phases are consumed within a few minutes after exposure to seawater. Kiln dust added to marine surface waters would, hence, add alkalinity and promote CO2 uptake in the surface ocean. The remaining material (mostly calcite) may settle to the seabed where it might produce additional alkalinity if sediment porewaters are undersaturated with respect to calcite.

The experimental results are very solid and clearly show the potential of kiln dust for ocean alkalinity enhancement (OAE). The paper is well written and I would suggest to publish the paper after a few revisions that are outlined below:

We sincerely thank the reviewer for the thorough and thoughtful evaluation of our manuscript. We appreciate the time and effort dedicated to carefully reading our work and providing constructive feedback which helped us improve the quality and clarity of the paper.

The authors present the mineral composition of kiln dusts in Table 2. It would be good to add data on Ca-Si-phases that are formed during cement production and should be present in cement kiln dust (CKD). These cement phases should have a high reactivity in seawater. They could explain the release of excess alkalinity observed in CKD experiments (line 331). The authors should consider these Ca-Si-phases in the discussion of their experimental results and add information on these phases in Table 2 if possible.

Calcium silicates are indeed relevant when discussing the alkalinization potential of CKD. However, as noted in the review by Adekunle (2024), these phases are typically present in smaller quantities compared to lime and portlandite. In our study, we conducted quantitative XRD analyses in duplicate. No Ca–Si phases were detected in the first measurement, while the second measurement indicated the possible presence of small amounts of beta-dicalcium silicate (β-C_2S 2.1%), and also of the iron oxides hematite (0.68%), and maghemite (0.56%). The identification of these phases was uncertain, which is why they were not included in Table 2 summarizing the mineralogical composition. Still, we follow the valuable suggestion of the referee, and we now additionally discuss how calcium silicates can contribute to alkalinity generation upon CKD dissolution in seawater. So, we now discuss their potential role in the text (shown in blue below, with changes highlighted in yellow) and we also indicate in the header of Table 2 that these phases were detected with low confidence.

Line 184: Furthermore, minor phases including 2.1% larnite/β-C_2S (Ca_2SiO_4), 0.68% hematite (Fe_2O_3), and 0.56% maghemite (γ-Fe_2O_3) were identified with low confidence in one of the duplicate CKD samples analyzed by XRD.

Line 197: Furthermore, the hematite and maghemite present in the CKD are essentially insoluble under oxic conditions in natural seawater, and filtration would remove Fe-reducing bacteria capable of enhancing their dissolution (Canfield, 1989). In contrast, the dissolution of portlandite and lime each produces two moles of A_T per mole (Eq. 3), while larnite (potentially present in the CKD) would yield four moles of A_T per mole upon dissolution (Brand et al., 2019). Overall, complete dissolution of these phases corresponds to maximum alkalinity contributions of 8.8 mmol g^{-1} for the LKD and 1.7 mmol g^{-1} for the CKD, respectively.

Line 198: Physicochemical properties of the experimental cement kiln dust (CKD) and lime kiln dust (LKD). ND indicates that the phases were not detectable. The minor phases larnite/beta-calcium

disilicate (β-C_2S; 2.1%), hematite (Fe_2O_3; 0.68%), and maghemite (γ-Fe_2O_3; 0.56%) were detected with low confidence in one of the duplicate samples and are therefore mentioned here but not included in the table. The complete measured elemental composition is provided in Appendix A Table A1.

Table 2: theoretical alkalinization potential of CKD was changed to 1.7 mmol g^{-1} to account for A_T production by larnite dissolution.

Line 324: CKD is more compositionally complex, typically containing calcite along with various sulfates, chlorides, silicates, and aluminates, including belite, aphthitalite, spurite, ettringite, arcanite, and ferrite (Ayman et al., 2004; Siddique and Rajor, 2012; Beltagui et al., 2017; Adekunle, 2024; Lee and Choi, 2024; Nikolov et al., 2025).

Line 338: Calcium silicates (e.g. larnite Ca_2SiO_4) are also alkalinity-generating phases that occur in minor amounts in CKD. They originate form the raw materials used in cement production (e.g. iron ore, clay, or shale) and exhibit a relatively high reactivity in water (Brand et al., 2019; Adekunle, 2024). Dissolution of larnite present in our CKD sample (~ 2.1 %) could therefore further account for 17 ± 1% of the observed A_T release. The remaining ~29% of the alkalinity released from CKD likely originated from dissolution of amorphous phases, including (partially dehydrated) clay minerals, reactive amorphous silica, and kiln-derived materials such as fly ash or slag (Khanna, 2010; Pavía and Regan, 2010)

Line 572: Figure B2. Saturation state (Ω) of (A) portlandite, (B) lime, (C) aphthitalite, (D) anhydrite, (E) syngenite, (F) sylvite, (G) calcite, and (H) quartz, and (I) larnite as a function of the kiln dust application concentration (mg kg^{-1}) during experiment II.

The authors propose that kiln dust should be applied to the surface ocean in shelf regions where the seabed is covered by permeable (sandy) sediments to allow for calcite dissolution in sediments that would add further alkalinity to the ocean (line 420). It is, however, likely that porewaters of these permeable sediments have a composition that is close seawater due to the rapid exchange with ambient bottom waters driven by fast tidal currents. Since shelf waters are usually oversaturated with respect to calcite, calcite dissolution may not proceed in these permeable deposits. Muddy sediments with restricted advective porewater exchange might offer a better environment for respiration-driven calcite dissolution as discussed in Dale et al., 2024 and Fuhr et al., 2025. I would suggest to update the text accordingly considering that muddy deposits are at least as favorable for calcite dissolution as permeable (sandy) sediments.

We agree that the statement regarding preferred sediment types for kiln dust application should be more carefully formulated. The suitability of a given sediment strongly depends on the local degree of calcium carbonate undersaturation in the porewater, which in turn is controlled by multiple factors, including the rates of aerobic mineralization, oxidation of reduced compounds, and pre-existing $CaCO_3$ dissolution, as well as temperature and salinity. Because these parameters can vary substantially across spatial scales, local sediment characteristics should ideally be assessed when selecting kiln dust application sites.

Nevertheless, it is evident that organic-rich, carbonate-poor sediments represent the most favorable environments for enhanced carbonate dissolution. Furthermore, coastal upwelling zones have been proposed by Fuhr et al. (2025) as promising application sites due to their high-pCO_2 seawater and high mineralization rates. This clarification has been incorporated into the revised text as follows:

Line 429: If kiln dusts would be applied to continental shelf waters overlying sediments with potential for enhanced carbonate dissolution, including organic-rich, carbonate-poor marine sediments

(Lunstrum and Berelson, 2022; Bice et al., 2024; Dale et al., 2024; Fuhr et al., 2025) or coastal upwelling zones (Harris et al., 2013; Fuhr et al., 2025), weathering of all calcite in the residual kiln dust could additionally produce a maximum of 10.8 mmol A_T g^{-1} LKD and 7.4 mmol A_T g^{-1} CKD.

Line 435: 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 484: Therefore, application should focus on continental shelf seas, especially those with organic-rich, carbonate-poor sediments (Lunstrum and Berelson, 2022; Dale et al., 2024), to promote metabolic $CaCO_3$ dissolution and maximize the CDR potential.

Kiln dust disposed in landfills reacts with CO2-bearing rain waters which may lead to a substantial uptake of atmospheric CO2. It is not clear to me whether the total CO2 uptake is enhanced when this material is added to the ocean instead of being disposed on land. The authors should add a paragraph on cement and kiln dust weathering under terrestrial conditions which has been intensively studied over the past decades. They should also try to compare the net CO2 balance of their approach (using kiln dust for OAE) with alternative kiln dust uses (disposal on land, recycling).

We fully agree with this suggestion. The CO_2 uptake from using kiln dust for ocean alkalinity enhancement is now briefly compared to terrestrial applications, such as landfill disposal, enhanced weathering, and recycling, in the discussion section on carbon dioxide removal potential. Additionally, the potential benefits and drawbacks of using kiln dust use in a reactor type environment, such as in accelerated weathering of limestone or ship-based ocean deployment, are now addressed to respond to the community comment by Ken Caldeira.

Line 442: Achieving the Paris Agreement targets will require rapid and deep CO_2 emission reductions, complemented by 12–15 Gt CO_2 $year^{-1}$ of carbon removal by 2100 (Rockström et al., 2017; Minx et al., 2018). Kiln dusts could potentially contribute to this CDR portfolio. Currently, most kiln dust is landfilled (El-Attar et al., 2017), while the remainder is recycled for applications such as soil stabilization, concrete mix, chemical treatment, ceramics, and brick manufacturing (Al-Bakri et al., 2022). CKD can replace 5-10% of cement, or up to 20% when combined with pozzolanic materials (fly ash, slag), reducing waste, lowering raw material and energy consumption, and cutting CO_2 emissions by a similar percentage (Huntzinger and Eatmon, 2009; Al-Bakri et al., 2022). While this represents the ideal use of CKD in terms of CO_2 mitigation, high levels of alkalis, sulfate, and chloride limit the extent to which CKD can be recycled in cement manufacturing (Al-Bakri et al., 2022). Carbonation of kiln dusts, involving the reaction of metal oxides with CO_2 to form solid carbonates, has been proposed as an alternative CO_2 sequestration method (Huntzinger et al., 2009; Adekunle, 2024):

$$CaO + CO_2 \rightarrow CaCO_3 \quad (7)$$

This process captures 1 mol CO_2 per mol metal oxide, which is less than what can be achieved via CaO hydration and subsequent $Ca(OH)_2$ dissociation in seawater (~1.68 mol CO_2 mol^{-1} metal oxide). In landfills, both processes naturally occur when kiln dust is exposed to rainwater (Sreekrishnavilasam et al., 2006). However, limited water availability in large kiln dust piles promotes secondary precipitation of carbonates or clay minerals, reducing the effective CO_2 sequestration. As such, the ad hoc CDR effect that occurs during landfill disposal of LKD and CKD remains uncertain. Similarly, application of kiln dust to agricultural soils for enhanced weathering purposes (as an alternative to primary mined rocks such as basalt or dunite) could contribute to CO_2 removal, though restricted water availability may again increase the risk of secondary mineral formation (Buckingham and Henderson, 2024; Xu and Reinhard, 2025). The focus of this study is the usage of kiln dusts via OAE in natural marine environments.

Alternatively, kiln dusts could also be used in reactor-based OAE approaches, such as accelerated weathering of limestone (Rau and Caldeira, 1999; review in Huysmans et al., 2025). These methods allow fast, controlled, and easily monitored alkalinity addition, but require higher energy inputs and dedicated infrastructure compared to ship-based distribution in natural environments (Rau and Caldeira, 1999; Rau et al., 2007; Huysmans et al., 2025).