Reply to referee 1

We would like to thank the reviewer for their constructive and positive feedback on our manuscript. Their recommendations have significantly improved the structure and content of the text. Below we provide a response to all their comments and suggestions, and indicate how we have altered the manuscript in response; our responses are in blue, altered text is in shaded in grey.

1. Line 35 Use of CCS as the overarching term for point-source CO2 mitigation is inappropriate because CCS has come to mean a very specific form of that mitigation https://en.wikipedia.org/wiki/Carbon_capture_and_storage

CCS is replaced by CO_2 emission mitigation throughout the manuscript.

2. Line 79-81 "The concept of AWL was first proposed by Rau and Caldeira more than two decades ago (Rau and Caldeira, 1999). It provides a geochemistry-based method for CCS in which the dissolution of carbonate minerals is artificially enhanced (Rau and Caldeira, 1999)."

   You mean -

   The concept of AWL was first proposed more than two decades ago by Rau and Caldeira (1999). It provides a geochemistry-based method for CO2 emissions mitigation in which the aqueous reaction of carbonate minerals with CO2 is enhanced due to the elevation of CO2 in typical waste combustion gases (Rau and Caldeira, 1999). ?

The sentences are adjusted according to the recommendations of the reviewer.

3. Line 90 Cite Caserini et al (2021) in initially introducing/describing BAWL.

The citation is added.

4. Table 1

   Row 1 - The initial values here are very uncharacteristic of low latitude, surface SW. Chou et al. et al 2015 are referenced as the source, and the values appear to be taken from their Table 1 (representing offshore and probably deep water samples) although I don't see the specific At and DIC values used by the present authors. In any case, it is clear from Chou et al. et al Table 1 that the starting solutions were not air equilibrated, pCO2>700 uatms, thus DIC is elevated and pH and Omega are depressed. The more realistic starting conditions are listed in Chou et al. eta Table 2 where pH>8 and esp Omega(c) >4.5. The choice of starting conditions will have a very significant effect on the modeling outcomes of the present study, so I ask the authors to carefully justify their initial choice of values here.

   Row 2 The amount of DIC rise in equilibrium with 0.15atm CO2 will very much depend on the chemistry of the starting solution that I question above.

Row 3 Ditto. Why does Omega(c) only rise to 0.203? In a perfect world under full CO2 and CaCO3 equilibrium OmegaC = 1. Granted, the kinetics for reaching this equilibrium are too slow to be reached in a practical application, but why is CaCO3 dissolution stopped at OmegaC=0.203 when the solution is still significantly carbonate undersaturated? The ratio of DeltaDICseq/DeltaDICcarb = 0.83/0.19 = 4.4. Shouldn't this be closer to 1? Or is there a huge amount of excess, unreacted CO2aq in solution?

Rows 4 and 5 Values are highly dependent on the accuracy of the preceding conditions/modeling.

The reviewer is correct in his assertion that the outcomes of our thermodynamic modelling are dependent on the initial conditions. Therefore, we did not use hypothetical 'ideal' starting conditions – as these 'ideal' starting conditions are also location dependent, and would thus vary for each potential AWL reactor. Instead, we used data from published pilot studies. The aim of Table 1 was thus to give an example of the different states for a representative real life (bench-top) reactor setup. All the values calculated in Table 1 are based on the measured inlet and outlet A_T and DIC from the two-step bench-top reactor from Chou et al. (2015).

Note that the initial solution values from Table 2 from Chou et al. (2015) cannot be used for calculations as there are no measured values at the outlet for these starting conditions.

To avoid this confusion, we clarified the purpose of Table 1 upfront at Line 113-119:

"Table 1 shows the values for pCO_2, A_T, DIC, pH, and Ω_{calc} in each of the four states for a representative case, which is based on data reported from a two-step bench-top reactor consisting of a separate gas-liquid and liquid-solid reactor (Chou et al., 2015, reactor design as further discussed below). The CO_2 concentration of the gas stream was 15%, while the pCO_2 of the atmosphere is fixed at 420 ppm. The A_T and DIC values at the inlet and outlet of the reactor are based on measured values (Table 1 in Chou et al., 2015). The remaining variables are calculated using the CRAN:AquaEnv package for the thermodynamic equilibria of acid-base systems in seawater (Hofmann et al., 2010)."

And expanded the caption of Table 1:

*"Theoretical values for alkalinity (A_T), dissolved inorganic carbon (DIC), pH, and calcite saturation state (Ω_{calc}) in the four consecutive states of the example AWL reactor: (1) the process water that is used as intake (the process water was collected from an offshore station near the Hoping power plant and the inlet and outlet of the cooling water drainage of the Hoping power plant (Chou et al., 2015)) , (2) the process water with elevated DIC after CO_2 uptake, (3) the process water enriched in A_T and DIC after $CaCO_3$ dissolution, (4a) the unbuffered or (4b) buffered process water upon discharge. ΔDICseq is the DIC that is added to the process water due to dissolution from the gas stream and ΔDICcarb is the DIC added through the dissolution of $CaCO_3$ in the reactor. The pCO_2, A_T and DIC values (indicated by #) are based on values measured in a two-step AWL bench-top reactor (Chou et al., 2015). The values of A_T, DIC, pH, and Ωcalc (indicated with *) are calculated using CRAN:AquaEnv (Hofmann et al., 2010) for seawater at a temperature of 15 °C and salinity of 35."*

The reason Omega only rises to 0.203 at the reactor outlet, is because the dissolution reaction was too slow to completely buffer the saturation state drop. The omega is calculated for the

values at the outlet given by Chou et al. and are before re-equilibration. We clarified this observation at Line 149 – 152:

"Note that the effluent at state 3 in the example two-step reactor is not in equilibrium with respect to $CaCO_3$ dissolution ($\Omega_{calc} < 1$, Table 1). This indicates that the effectiveness of $CaCO_3$ dissolution in the reactor design of Chou et al. (2015) could still be improved (e.g. by implementing a longer residence time)."

5. Equ 1 Only valid at low pH (<7). The stoichiometry changes as pH rises so as to accommodate the spontaneous formation of (alkalinity hog) CO3-- ; ACO2 + BH2O + CaCO3 ---> Ca++ CHCO3- + DCO3-- + ….. such that the total moles carbon added is A+1=C+D (<=2) and A<=1 (see eq 1 here https://bg.copernicus.org/articles/20/27/2023/)

This is correct, in a sense that the carbonate system re-equilibrates after the reaction, which will set the eventual stoichiometry of the overall reaction. However, we think this leads to a confusing way of writing the equations, as A, B, C, … are dependent on conditions and obscure the fact that the dissolution of calcium carbonate always creates two alkalinity. We have chosen to explain the re-equilibration step (see Line 133 of the original manuscript), and if one would combine Eq. (1) and Eq. (2) for a given pH, you would get the equation the reviewer refers to. We prefer to keep our approach, as we think this makes the overall process clearer for non-specialist readers.

6. Line 143-4 "However, one can easily show that equilibration followed by mixing, provides the same CO2 transfer as mixing followed by equilibration." This assumes that discharing a supersaturated CO2 solution into seawater will in fact equilibrate with air (on human-relevant timescales). That is unlikely to happen due the slow kinetics of air/sea gas exchange coupled with vertical SW mixing that will remove some of the supersaturated solution out of contact with air prior to equilibration. Gorey details here: https://www.nature.com/articles/s41558-024-02179-9

   Bottom line: Assuming air equilibration underestimates C storage because some excess CO2aq added in unbuffered AWL will not have a chance to degas to air.

Full equilibration with the atmospheric pCO_2 will indeed be prevented when surface residence times of the discharged process water is shorter than the air-sea equilibration timescale. So, when the process water is discharged below a strong stratification layer or in locations where the discharged water quickly reaches the deeper oceans, assuming full equilibration would underestimate the CO_2 storage potential (He & Tyka, 2023; Jones et al , 2014). However, as most AWL plants, like the AWL pilot plant in Wilhelmshaven (Germany; Kirchner et al., 2020), will be located near the coastal ocean with shallow mixed layers with relatively efficient air-sea CO_2 exchange, equilibration will take place on timescales of months up to a year (Jones et al, 2014; Geerts et al, 2025).

We added the caveat about air-sea CO_2 exchange variation and the possibility at non-equilibrium degassing at Line 119-128:

"We assume full re-equilibration with the atmosphere (unbuffered AWL) or full buffering with slaked lime ($Ca(OH)_2$) upon discharge into the sea (buffered AWL). This condition of full re-equilibration requires consideration. In the well-mixed coastal zone, air-sea CO_2 exchange takes place on a time-scale of several weeks up to a year (Jones et al., 2014; He and Tyka,

2023; Geerts et al., 2025). When the surface residence time of the discharged process water is shorter than the air-sea CO_2 equilibration timescale, some of the dissolved CO_2 unbuffered by the A_T increase in the AWL reactor can move to deeper layers and so full re-equilibration will not be reached (Jones et al., 2014; He and Tyka, 2023). Likewise, when the process water is discharged below the stratification layer or directly in the deeper ocean, full re-equilibration will also be prevented (Jones et al., 2014; He and Tyka, 2023). In both the cases, the CO_2 sequestration is increased. Therefore, assuming full re-equilibration represents a conservative lower bound for the CO_2 sequestration during AWL."

7. Fig 2 Should be modified depending on the (new) outcomes listed in Table 1.

See our response to comment 4

8. Line 178-80. "In a similar fashion, the final alkalinity value is the result of alkalinity addition during carbonate dissolution and possibly some extra addition during lime buffering"

   Unclear. If you are adding lime you are adding alkalinity, no "possibly" about it. Or are you saying that adding lime is a possibility? In this region of the text the discussion seems to move from AWL with an option to lime to one where liming is now assumed/required. Please be clear from the start about how you are treating AWL +/-liming.

This phrasing is indeed a bit confusing. We adjusted the sentence to make it clear that we mean that the final alkalinity is the result from carbonate dissolution, with extra alkalinity added by $Ca(OH)_2$ addition in BAWL at Line 201-202

"In a similar fashion, the final A_T value is the result of A_T addition during $CaCO_3$ dissolution and the A_T that is added during buffering with $Ca(OH)_2$ in the case of buffered AWL."

9. Line193-4 Full air equilibration after discharge is unlikely (https://www.nature.com/articles/s41558-024-02179-9)

See our response to comment 6.

10. Equ 8 Missing an operator between the 2nd the 3rd right hand terms?

A multiplication sign is added between the two terms for clarification.

11. Line 196-204. Assumes full air/sea CO2 equilibration, unlikely (https://www.nature.com/articles/s41558-024-02179-9)

See response to comment 6.

12. Line 219-225 Revise depending on outcomes in (revised) Table 1?

See our response to comment 4

13. Line 297-and after Flows and efficiencies are calculated from data in Table 2 with the implication that these values will be characteristic of AWL at scale, yet what is the evidence that the data in Table 2 represent optimized systems?

The idea of Table 2 is to calculate efficiency values for different existing/conceptual reactor designs. Since we are reviewing the existing literature, it is not our goal to represent optimized systems at scale. The operational stage of each specific example reactor is specified in row 1 of the operational conditions. To prevent misunderstanding, we clarified that these are values for prototype/conceptual reactors and that the efficiencies are calculated based on the inlet an outlet A_T and DIC, and the given water/gas flow rate at Line 328 – 332:

> *"The operational conditions and process efficiencies of these reactor designs are summarized in Table 2. The presented operational conditions are given for specific example reactor setups (bench-top (Chou et al., 2015) or pilot plant (Kirchner et al., 2020b)) or conceptual designs (Caserini et al., 2021) and the process efficiencies are calculated based on published data for a specific operational condition. Changes in reactor design or operational conditions will change these calculated efficiencies."*

14. Line 258 You mean 150,000 m^3, yet eq 20 is in units of tonnes/tonne and the assumes that 1L SW = 1kg?

Correct, the exponent should have been 3 (150 000 m^3) instead of 2 (150 000 m^2). This has been changed.

The units for eq. 20 at $m^3_{seawater}$/tonne of CO_2, as is stated in the text: *'The volume of process water (m^3) that is used to capture one tonne of CO_2'*. We did notice a typo, 10^6 has to be 10^{-6}.

ΔDIC_{seq} is expressed in mol per unit of volume: $$mM = \frac{10^{-3}\,mol}{L} = \frac{10^{-3}\,mol}{dm^3} = \frac{10^{-3}\,mol}{10^{-3}\,m^3} = \frac{mol}{m^3}$$

The units for Eq. 20 are: $$\frac{1}{mol_{CO_2} / m^3_{seawater}} \frac{10^{-6}}{g_{CO_2} / mol_{CO_2}} = \frac{10^{-6}}{g_{CO_2} / m^3_{seawater}} = 10^{-6} \frac{m^3_{seawater}}{g_{CO_2}} = \frac{m^3_{seawater}}{10^6\, g_{CO_2}} = \frac{m^3_{seawater}}{tonnes_{CO_2}}$$

15. Line 293-6 What is the evidence that the efficiencies stated are representative of optimized systems?

See response to comments 4 and 13.

16. Line 301-2 You likely mean Rau (2011) rather than Caldeira and Rau (2000)? The former pub offers numerous results/data for a one step reactor. Compare/contrast with Chou et al. et al 2015 and you subsequent calcs?

The citation should indeed be Rau (2011). Results from Rau (2011) and Chou (2015) are compared in section 3.1 but comparing specific calculations is not possible as specific values for DIC, A_T, water/gas flow rate are not specified in Rau (2011).

17. Line 324-5 This does not jibe with Rau (2011) which states "Comparing resulting DIC and alkalinity to that of the original solutions and to ambient seawater demonstrates that 61-85% of the carbon originally added to the seawater remained in solution (Figure 2c), with

little change in alkalinity and with no visual evidence of carbonate precipitation after aeration."

The section in Rau (2011) that the reviewer refers to discusses the modified reactor in which seawater in equilibrium with the CO_2/air mixture was allowed to reside in the reactor for 1-2 weeks. This in essence becomes a two-step reactor with long residence time in the second reactor and is thus not applicable on the one-step reactor.

18. Line 327-8 "Consequently, the overall CO2 sequestration efficiency of a one-step reactor remains low due the lack of conversion from hydrated CO2 to HCO3-." Hydrated CO2 is HCO3- + H2CO3. What is apparently meant here is lack of conversion of hydrated CO2 balanced by Ca++ rather than by H+? Or do you mean lack of conversion of CO2 to carbonic acid? Anyway, how does this square with the 61-85% of the initially captured C shown to be air stable by Rau (2011)?

For the comparison with Rau (2011) see response to comment 17.

We agree with the reviewer that this formulation does not clearly represent the limiting step of $CaCO_3$ dissolution and the production of alkalinity. We have changed the sentence to better convey that we mean the buffering of the dissolved CO_2 by the increase in A_T at Line 355 – 356:

"Consequently, the overall CO_2 sequestration efficiency of a one-step reactor remains low due to a lack of $CaCO_3$ dissolution. A large fraction of the dissolved CO_2 remains unbuffered by the increase in A_T."

19. Line 417-19 If the now alkalized and carbonated SW is discharged at the same pH as ambient SW the pCO2 must be higher than ambient? Don't you need to discharge at higher pH to avoid this? And wouldn't higher discharge pH beneficially help counter ongoing ocean acidification?

The reviewer raises a good point. Based on the alkalinity and DIC values at the end of the buffering reactor (BR) from Table 1 in Caserini et al. (2021), and given a seawater temperature of 10 °C, the pH and fCO_2 of the process water can be modelled using CRAN:AquEnv.

Under these conditions, the pH is 8 (as in Table 1 in Caserini et al. (2021)) and the fCO_2 is 0.00483 atm or 4830 µatm. Thus, under conditions presented by Caserini et al. (2021) the fCO_2 is indeed higher than ambient if the pH is at the same level as the surrounding seawater.

20. Line 435-332 Check out Langer et al for further discussion of limestone sources (in the US): https://www.researchgate.net/publication/283868780_Accelerated_weathering_of_limestone_for_CO2_mitigation_Opportunities_for_the_stone_and_cement_industries

We thank the reviewer for this resource, we have now included it in our reference list.

21. Line 443-6 Here and elsewhere "high water demand" is implied to be an AWL showstopper, yet the global supply of seawater seems rather limitless. What is apparently meant here is that the pumping costs of seawater can become prohibitive, yet so far no discussion of exactly what these costs are, especially relative to the (high) cost of the industry darling, CCS – capturing, concentrating and storing molecular CO2 underground.

We agree with the reviewer that the supply of seawater should not be seen as a limiting factor, we have adjusted the text accordingly at Lines 469 – 480:

"Significant volumes of water are needed to dissolve the CO_2 and dilute the resulting bicarbonate in the original reactor designs (10^4 - 10^5 tonnes of water per tonne of CO_2; Table 2) (Rau et al., 2007; Rau and Caldeira, 1999), although more recent designs have reduced the water demand by a few orders of magnitude (~ 10^3 tonnes of water per tonne of CO_2; Table 2). The high water demand and the accompanying pumping cost could limit the feasibility of the overall AWL process. Therefore, a low-cost water source such as cooling water from a power plant or other sources of recycled water should be used preferably (Rau and Caldeira, 1999). Due the required quantities of process water, the favored locations for (B)AWL reactors would be coastal regions as seawater is a virtually limitless source and the bicarbonate-containing effluent could be directly dumped and diluted in the ocean after degassing or buffering and removal of potential contaminants (Rau and Caldeira, 1999; Rau et al., 2001). Pumping costs could further be reduced by reusing the large volumes of seawater already pumped and used as power plant cooling water (Rau et al., 2007; Kirchner et al., 2021). However, the elevated temperature of the seawater during the cooling of the power plants would reduce the CO_2 dissolution into the seawater (Kirchner et al., 2021)."

22. Line 447-8 Who has proposed the use of anything but seawater for AWL? The only places AWL will work are near the ocean, eps powerplants that use SW for cooling(?)

The different possible water resources were suggested in Rau and Caldeira (1999) on page 1807 in section 4: Water considerations.

AWL will indeed only be possibly economically feasible near the ocean and indeed especially when seawater used for cooling in power plants can be reused, limiting pumping costs. We have elaborated this paragraph to make it clear that seawater is the only viable option and that pumping costs could further be reduced by reuse of power plant cooling water, as outlined in our response to comment 21.

23. Line 458-9 "The BAWL reactor setup proposed by Caserini et al. consumes 0.4 tons of Ca(OH)2 to store 1 ton of CO2." Or 1/0.4 = 2.5 t CO2/t Ca(OH)2(?) Yet the delta DIC/deltaAlk in the surface ocean is about 0.85. Since 1 mole of Ca(OH)2=2 moles Alk, then the mole CO2 captured and stored per mol Ca(OH)2 should be 2x0.85/1 = 1.7 moles/mole. CO2= 44g/mol, Ca(OH)2= 74g/mol, Thus ,1 tonne of Ca(OH)2 is able to capture and store about 1.7x44/74 = 1 t CO2/tCa(OH)2 in seawater @pCO2= 420 uatms? Or does 2.5 t/t only apply to deep ocean, high pressures?

This sentence could lead to misunderstanding. We meant that 0.4 tons of $Ca(OH)_2$ was used on top of the 1.31 tonnes of $CaCO_3$ that was fully dissolved, as stated by Caserini et al. (2021). We decided to remove this sentence to avoid confusion.

24. Line 466-8 Seems pretty obvious from the previously published lit. Why even hint at the use of other water sources?

See response to comment 22.

25. Line 499-500 "..the increased alkalinity and pH could potentially limit ocean acidification.." You mean "…the increased alkalinity and pH would help counter ocean acidification and

its effect on marine biota, see for example Albright et al (2016)" https://www.nature.com/articles/nature17155

The sentence is adjusted according to the comment of the reviewer.

26. Line 514 How about inserting "All of the preceding argue for the use of relatively clean waste gas streams (such as from the combustion of natural gas) in (B)AWL applications." ?

We have added the sentence.

27. Line 515-20 Bach (2024) specifically discusses the application of alkaline solids to marine sediments and the effect of alkalinity generation there. Discharge of dissolved alkalinity into surface waters some distance from sediments and with rapid dilution, as characteristic of (B)AWL, would seem to pose much less risk to benthic/sediment processes.

The discharge of dissolved alkalinity pose less risk than addition of alkaline solids to the sediment. We have highlighted this in this section and further discussed the potential negative feedback in the water column based on the recently published manuscript by Lehman & Bach (2025) at Line 546 - 554:

"The disposal of large volumes of process water in the surface water of the coastal zone can locally increase pH and mitigate the adverse effect of ocean acidification on calcifying phytoplankton. However, this implies a reduction of the efficiency of the CO_2 sequestration via AWL, as part of the produced A_T will be consumed and lead to CO_2 degassing (Lehmann and Bach, 2025). Additionally, mixing of this A_T enriched coastal water within the coastal sediment through porewater flushing or diffusion could potentially inhibit natural $CaCO_3$ dissolution (Lunstrum and Berelson, 2022; Bach, 2024). If this would occur, the efficiency of the (B)AWL process would be reduced as the CO_2 sequestration by AWL would be partially compensated by a loss of natural CO_2 sequestration. However, this is less likely to occur with (B)AWL than with mineral-based OAE where alkaline minerals are directly added to the coastal sediment and A_T can build-up in the porewater (Hartmann et al., 2023)."

References

He & Tyka, 2023: https://doi.org/10.5194/bg-20-27-2023

Jones et al, 2014: https://doi.org/10.1002/2014GB004813

Kirchner et al., 2020: https://doi.org/10.1021/acs.est.9b07009

Geerts et al., 2025: https://doi.org/10.5194/bg-22-355-2025

Lehman & Bach, 2025: https://doi.org/10.1038/s41561-025-01644-0

Reply to referee 2

The paper is a useful summary of the chemistry and the applicability of accelerated weathering of limestone or buffered accelerated weathering of limestone, and it deserves publication. Minor comments below.

We would like to thank the reviewer for the positive feedback and the constructive comments. Below we provide a response to all their comments and suggestions, and indicate how we have altered the manuscript in response; our responses are in blue, altered text is in shaded in grey.

1. Lines 39-59. Please revise this section because it could lead to confusion among "enhanced weathering", "enhanced rock weathering" , "mineralization", and "carbonation" (in the case the mineral obtained is a carbonate mineral). The studies by Rau and Caldeira, 1999, Renforth and Kruger, 2013, Caserini et al., 2021, cited as "enhanced rock weathering" processes, could be better identified as accelerated weathering of limestone, to avoid confusion with enhanced weathering (that is a CDR approach that removes atmospheric carbon).

The start of this paragraph is rewritten to prevent confusion with enhanced weathering as CDR technology, Line 39 – 60*:*

"Industrial point-source CO_2 emissions from waste gas streams can be mitigated by geochemical-based processes in which CO_2 is reacted with solid carbonate or silicate rocks in the presence of water, which aims to enhance the natural weathering process of carbonate and silicate rocks (Rau and Caldeira, 1999; Renforth and Kruger, 2013; Caserini et al., 2021). This targeted weathering process can take place in situ, in which CO_2 is first captured from the flue gas and then injected into suitable silicate rock formations (basalts and ultramafic rocks). The CO_2 is then trapped by a carbonation reaction with the ambient silicate rock, thus ensuring a permanent, geological storage (Matter and Kelemen, 2009; Romanov et al., 2015; Gadikota, 2021; Cao et al., 2024). However, there are certain geomechanical risks associated with geological storage of CO_2, such as CO_2 leakage, induced seismicity, the loss of well integrity, and surface uplift (Song et al., 2023). Moreover, suitable rock formations for storage are not always in close proximity to the CO_2-emitting installations, thus requiring compression/liquefaction and transport of CO_2.

Alternatively, the chemical weathering can also be executed under controlled conditions in a land-based reactor, close to the industrial point source. Mitigation of CO_2 emissions via such reactor-based methods can follow two main approaches, depending on whether silicates are used as feedstock material (usually referred to a "ex-situ mineral carbonation" technologies; Romanov et al., 2015; Gadikota, 2021, or "mineralization"; Campbell et al., 2022) or whether carbonates are used as weathering substrates (referred to a as "accelerated weathering of limestone"; Rau and Caldeira, 1999). In ex-situ mineral carbonation (ESMC), a finely-ground silicate mineral (e.g. olivine Mg_2SiO_4) is fed into a reactor, where it reacts at elevated temperature and pressure with CO_2 from a flue gas to eventually form stable carbonates (e.g. magnesite Mg_2SiO_4) - see recent reviews (Snæbjörnsdóttir et al., 2020; Veetil and Hitch, 2020; Thonemann et al., 2022). Alternatively, during the accelerated weathering of limestone (AWL), CO_2 is stripped from the flue gas using a mixture of seawater and limestone (Rau and Caldeira, 1999; Renforth and Henderson, 2017), and the resulting effluent is discharged into the sea. "

2. Line 52-55 Please specify that what is called "ex situ mineral carbonation" (methods where alkaline minerals react with CO2, producing solid carbonate minerals) is also called "mineralization", as in Campbell et al (2022) https://doi.org/10.3389/fclim.2022.879133.

Mineralization is added as an alternative name for ex-situ mineral carbonation at Line 51 - 54:

"Mitigation of CO_2 emissions via such reactor-based methods can follow two main approaches, depending on whether silicates are used as feedstock material (usually referred to a "ex-situ mineral carbonation" technologies; Romanov et al., 2015; Gadikota, 2021, or "mineralization"; Campbell et al., 2022), …"

3. line 62: please specify that the CO2 removed by ocean alkalinization is atmospheric CO2

Adapted.

4. line 63. I don't see the need to add "chemical" between natural and weathering, since all the weathering processes are chemical processes.

'chemical' is removed.

5. Lines 91, 93, 99, and others: It is not clear what "upon discharge" means: just before the discharge of the process water or after the discharge? Sometimes, it seems just before (i.e.: … buffering with Ca(OH)2 upon discharge into the sea), in other cases, just after the discharge in seawater (upon re-exposure to atmospheric conditions, aqueous CO2 which is not stabilized by the increased AT will degas back to the atmosphere)

Clarified.

"After discharge into the surface ocean, there is no longer any CO_2 transfer to the atmosphere."

"The process water is discharged into the sea without any further treatment after which it re-equilibrates with the atmosphere at the lower pCO_2 ($pCO_2 \approx 0.00042$ atm) and the excess CO_2 (i.e., the part of DIC not stabilized by the increased A_T) will degas back to the atmosphere. "

"… (4a) the unbuffered or (4b) buffered process water after discharge into the surface ocean."

6. Line 99 "(4a-b) the unbuffered or buffered". Please clarify that 4a is unbuffered and 4b is buffered.

Adjusted according to the suggestion of the reviewer.

7. Lines 119-125 (table 1). It should be stated in the title what (1) (2) (3) (4a) and (4b) in the first column means. Since just before figure 1 there is (i) (ii) (iii) and (iv), there could be some misunderstanding.

The different states with number and explanation are now explicitly stated in the caption of Table 1.

"Theoretical values for alkalinity (A_T), dissolved inorganic carbon (DIC), pH, and calcite saturation state (Ω_{calc}) in the four consecutive states of the example AWL reactor: (1) the process water that is used as intake (the process water was collected from an offshore station near the Hoping power plant and the inlet and outlet of the cooling water drainage of the Hoping power plant (Chou et al., 2015)) , (2) the process water with elevated DIC after CO_2 uptake, (3) the process water enriched in A_T and DIC after $CaCO_3$ dissolution, (4a) the unbuffered or (4b) buffered process water upon discharge. ΔDICseq is the DIC that is added to the process water due to dissolution from the gas stream and ΔDICcarb is the DIC added through the dissolution of $CaCO_3$ in the reactor. The pCO_2, A_T and DIC values (indicated by #) are based on values measured in a two-step AWL bench-top reactor (Chou et al., 2015). The values of A_T, DIC, pH, and Ωcalc (indicated with *) are calculated using CRAN:AquaEnv (Hofmann et al., 2010) for seawater at a temperature of 15 °C and salinity of 35."

8. Line 124: the pH for 4a, unbuffered process water upon discharge, is 8.16, quite high, very close to the 8.27 for the buffered case. The pH is quite higher than in Caldeira and Rau 2000 https://doi.org/10.1029/1999GL002364. Please add some comments on this point.

The pH in Table 1 (4a) is calculated in R using the CRAN:AquaEnv package. The pH in state 4a is calculated based on the alkalinity content at the outlet of the reactor and the DIC content after full re-equilibration with atmospheric pCO_2.

In Caldeira and Rau (2000), the pH for the "degassed to seawater Ω_{calc}" is based on the alkalinity at the outlet of the reactor and the Ω_{calc} of 4.14. In this case, the seawater is not fully equilibrated with the atmospheric pCO_2 (0.000402 atm) and the fCO_2 of the seawater is still at 0.014808 atm. If we calculate the pH for Caldeira and Rau (2000) using an outlet alkalinity of 14808 µmol kg^{-1} and full equilibration with the atmospheric pCO_2, we get a pH of 8.5 due to the higher alkalinity compared to our example.

9. Lines145-149. Add more recent experimental studies:

Hartmann, J., Suitner, N., Lim, C., Schneider, J., Marín-Samper, L., Arístegui, J., Renforth, P., Taucher, J., & Riebesell, U. (2023). Stability of alkalinity in ocean alkalinity enhancement (OAE) approaches – consequences for durability of CO_2 storage. Biogeosciences, 20(4), 781–802. https://doi.org/10.5194/bg-20-781-2023

Moras, C. A., Bach, L. T., Cyronak, T., Joannes-Boyau, R., & Schulz, K. G. (2022). Ocean alkalinity enhancement – avoiding runaway $CaCO_3$ precipitation during quick and hydrated lime dissolution. Biogeosciences, 19(15), 3537–3557. https://doi.org/10.5194/bg-19-3537-2022

References included.

10. Lines 221-224. I would further clarify the reason behind the additional CO2 removal through liming. This represents a novelty of this study that was not addressed in Caserini et al. (2021), because buffered AWL is a carbon dioxide storage process. In contrast, ocean liming is a carbon dioxide removal process.

In this context, liming is meant as the addition of $Ca(OH)_2$ in the buffering reactor before discharge of the process water to the sea. The use of "liming" could indeed cause misunderstanding. It is changed to "buffering with $Ca(OH)_2$".

11. Line 258. I think the exponent of the unit of measurement is 3, not 2.

The exponent is changed to 3.

12. Line 427-428. I would provide more details about this calcination-free process as a method for $Ca(OH)_2$ recovery, since $Ca(OH)_2$ recovered from steel slag is obtained through calcination, then used in the steel industry, and ultimately ends up in the steel slag. Furthermore, I would elaborate on whether this process has other potential environmental side effects and provide more insights into its availability, as it depends on the residuals of an industrial process.

We do not think expanding on the process of forming $Ca(OH)_2$ fits within the scope of our paper, and would distract from the overall message.

13. Lines 461-462. Please provide a reference for the value of 1 ton of CO2 produced per ton of Ca(OH)2.

The value is changed to 1 – 1.8 tonnes of CO_2 per tonne of $Ca(OH)_2$., and we included two new references:

Oates, 2008: ISBN: 978-3-527-61201-7

Simoni et al., 2022: https://doi.org/10.1016/j.rser.2022.112765

14. Lines 504-514. It's worth adding that the problems of trace metals or other pollutants are much lower if AWL or BAWL are used just for the storage of the CO2 produced by calcination, i.e. in the case of electric calcination

Included at Line 544 -546:

"The potential negative effects from trace elements and other pollutants can be further mitigated by using of relatively clean waste gas streams (such as from the combustion of natural gas or calcination of $CaCO_3$) in (un)buffered AWL applications."

15. Lines 515-521. Regarding potential impacts on marine biota, I would also cite the recent study by Sánchez et al. (2024).

Sánchez, N., Goldenberg, S. U., Brüggemann, D., Jaspers, C., Taucher, J., & Riebesell, U. (2024). Plankton food web structure and productivity under ocean alkalinity enhancement. Science Advances, 10(49), eado0264. https://doi.org/10.1126/sciadv.ado0264

Included.