



EGUsphere, author comment AC2
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Reply on RC2

Jing He and Michael D. Tyka

Author comment on "Limits and CO₂ equilibration of near-coast alkalinity enhancement"
by Jing He and Michael D. Tyka, EGU sphere,
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We thank the reviewer for a thorough review and many thoughtful comments which we have incorporated into the manuscript.

- *The choice of pH and omega anomalies as opposed to absolute values when constraining OAE rates: Given the authors justification that the CO₂ system constraints they impose on potential OAE are to avoid calcite precipitation, why use pH and omega anomalies as opposed to absolute values? I find this choice problematic given both the log-scale of pH and the non-linear response of the CO₂ system (including omega) to alkalinity enhancement. For example, higher pH waters exhibit a smaller change in pH for the same change in [H⁺] just due to the log-scale.*

While lower omega waters will exhibit a smaller change in omega for the same increase in alkalinity (and the opposite is true of pH). I don't want to necessarily force the authors to rerun computationally expensive simulations but feel this decision to use anomaly thresholds needs to be better justified. Perhaps the authors could assess the extent to which permissible OAE is dependent on background omega and pH values. I also feel that they are missing a proper comparison between the use of the pH versus omega threshold.

We considered both absolute and relative thresholds as possible experiment designs, but settled on relative thresholds for a variety of reasons, detailed below. We use pH and Omega as two convenient proxies to evaluate the impact of OAE: we want to consider the impact on ecosystems and precipitation.

First let's consider impacts on ecosystems: Here relative changes made more sense to us than absolute ones, since the locally present organisms are likely adapted to the current environment. Imposing an absolute limit everywhere thus doesn't make sense in our opinion. Both pH and Omega affect organisms and thus the relative limit makes sense here.

Next let's consider the use of pH vs [H⁺] (or [OH⁻]). In general pH is used as a parameter to describe the ocean state rather than [H⁺], however the use of [H⁺] has been suggested before (e.g. Fassbender et al 2021). In our case, the choice of Δ[H⁺] vs ΔpH depends on whether the actual impact on an organism is proportional to the absolute or relative change in the activity of [H⁺] (or [OH⁻]). While this will inevitably depend on each

organism, it is reasonable to assume that a relative change is more appropriate. For example, the additional energy expenditure of an organism to maintain its intracellular pH is proportional to the logarithm of the concentration gradient. Thus, to a first approximation, the incurred metabolic cost given a ΔpH is the same for an organism adapted to a pH of 8 as for one adapted to a pH of 7.

As for Omega as a proxy for the avoidance of precipitation, we agree that an absolute limit would capture the true "limit" with respect to precipitation much better than a relative one. However, simulating OAE to such an extent that would raise Omega all the way to its absolute limit everywhere has some concerning drawbacks: The necessary perturbation of [Alk] would be so large in polar regions (where Omega is currently low) that the simulated carbonate system would be very far from where it is currently. It's not clear to us that the ocean state model can be expected to give realistic results since the perturbed state is now far outside the bounds of where the model was parameterized. Nor would we ever expect such a radical addition of Alk to be done in practice, on pH grounds alone (as that limit would be exceeded much sooner).

Therefore we opted to stick with a relative limit and we note that in our simulation the allowed small ΔOmega keeps the absolute Omega below the precipitation limits even in areas (equatorial) where the starting omega is already high. Thus the globally sustainable OAE limits obtained with respect to Omega can be seen as a lower bound on the limit rather than the actual limit itself, which is clearly much higher (from an Omega perspective alone).

We've expanded the methods section to elaborate more deeply on the choices made in our methodology in a new paragraph.

We also noted that we found no correlation between the background pH and the obtained limit (at constrained ΔpH), as the influence of local currents is too dominant a factor. When comparing the alkalinity flux obtained at ΔpH and ΔOmega we find their order of magnitude is highly correlated (consistent with the fact that the limits are largely influenced by current patterns). The correlation is not exact as the relative sensitivity of pH vs Omega wrt Alk (i.e. $\partial\text{pH}/\partial\text{Alk}$ vs $\partial\text{Omega}/\partial\text{Alk}$) differs, mostly meridionally. We've added sentences to the results section pointing out these properties of the carbonate system.

- *The dependence of results on current climatic conditions: The authors say little about how dependent their results are on current climatic conditions. I would be particularly interested to see how the permissible OAE rates and associated CDR differ under a much higher atmospheric CO2 concentration. Presumably permissible OAE rates are higher (at least when using the delta omega threshold) and the impact on pCO2 (and in turn CDR) will be far greater per unit alkalinity added. Such a simulation may be beyond their scope but I still think some discussion of how their results might change as the anthropogenic carbon content of the ocean increase is required.*

We agree this is a fascinating and important, albeit difficult question. We did not explicitly address this for two reasons. Firstly the ocean state model we use (ECCO) was specifically parameterized from contemporary data. Future oceans will potentially change significantly in flow patterns and stratification and thus the application of a contemporary flow field seems inappropriate. Secondly, the anthropogenic emission trajectories are highly uncertain - which future do we even simulate? Of course multiple IPCC scenarios could be played out however this would increase the scope of this paper very significantly. We feel that future work should address these issues but we believe this is well beyond the scope of the current work. We've added some prose to the discussion section that reflects on

these questions and future directions.

- *Figures: The current content and layout of many figures in the manuscript makes for quite painful reading. So much detail is often squeezed into the multi-panel figures that often nothing is legible/understandable without using a high zoom in a pdf viewer. I suggest the authors simplify their figures, reducing the number of panels and repetitive information. All figure text should conform to a minimum font size.*

We simplified each of the figures as suggested in the detailed comments below, increasing font size and reducing duplication by removing panels that are already shown.

Note also that the final double-column figures will utilize the full width of the page, rather than here in this preprint the LaTeX template limited us to 75% of the page width. Thus the fonts will be 35% larger in the final print.

Minor comments

- *L7-14 The first time I read this I thought you were confusing OAE and CDR. I think you need to introduce the concept of OAE thresholds (and why they may be important) before you mention the steady state OAE rate which most readers (myself included) will assume you are free to choose.*

As suggested, we've re-phrased that paragraph, starting the line of thought first with the chosen limitations and only then proceeding to the idea of steady state: "Choosing relatively conservative constraints on ΔpH or $\Delta\Omega$, we examine the limits of OAE using the LLC270 (0.3deg) ECCO global circulation model. We find that the sustainable OAE rate vari..."

- *L21 Isn't it the CDR potential that is lost not the OAE potential? Ie the transport of alkalinity to depth actually facilitates higher surface addition of alkalinity addition without exceeding a given threshold.*

Changed to "...as in such locations up to half of the CDR potential of OAE can be lost to bottom waters."

- *L32 CO2 doesn't dissolve as bicarbonate (although most ocean DIC is in the form of bicarbonate).*

Changed to "... and the excess atmospheric CO2 dissolves into the ocean, largely reacting to form (bi)carbonate ions".

- *L66 "moieties" is not the correct term to use here as it typically refers to part of a molecule. You just mean the sum of the aqueous forms of inorganic carbon.*

Changed "carbonate moieties" to "carbonate species"

- *L91 lat-lon grid?*

The grid of points used in the cited study was a latitude/longitude spherical coordinate system. Happy to rephrase this if there is a better way to express succinctly?

- *L104 avoid "moieties"*

Changed "of all chemical moieties involved in the carbonate system" to "all forms of CO₂ in the carbonate system"

- *L119-120 provide the units of DIC and Alk used otherwise it's unclear that this is a molar ratio.*

Added "ηCO₂ is a unitless molar ratio."

- *L121-122. Can the authors say more here about this 0.8 value and what effects it?*

"The exact value depends on the parameters of the carbonate system, i.e. Alk, DIC, temperature etc., with a typical range of 0.75-0.85". The local surface variation was explored in Tyka et al 2022."

- *L144-145 The movement of DIC is controlled by the ocean physics not these 5 biogeochemical tracers which are presumably used to compute the ocean CO₂ system.*

Changed "...uses 5 biogeochemical tracers [...] to simulate the movement of total dissolved inorganic carbon (DIC) within the ocean" to "...uses 5 biogeochemical tracers [...] to simulate the carbonate system."

- *L147 Is this freshwater flux from rivers?*

Yes, it is from rivers and from rainfall.

- *L150 uatm are units of partial pressure. Do you hold the concentration or partial pressure of atmospheric CO₂ constant ie can local changes in atmospheric pressure influence gas exchange?*

We hold the partial pressure constant, not the absolute concentration.

- *L154 More detail on how the imported wind speeds are used to calculate gas exchange would be useful here.*

We included a specific reference to the way gas-transfer is calculated: "Wind speeds, used to calculate the gas exchange, are imported from the LLC270 forcing data and the air-sea exchange of CO₂ is parameterized with a uniform gas transfer coefficient [Wanninkhof, 2018]."

- *L234 Any rationale for January pulses?*

No particular rationale. We only had enough computing time to run a handful of these pulse simulations so we had to pick a time point.

- *L240-241 Not sure this sentence should be a distinct paragraph.*

Merged with the following paragraph.

- *L265-277 Perhaps make it clear that you're only considering the financial and not carbon cost of transport.*

We changed the first sentence to "Alkalinity prepared on land must be transported out to sea, which adds to the total cost of the achieved negative emissions (in \$/tCO₂).". Of course reducing the shipping load also reduced CO₂ emissions associated with that

shipping.

- *L278 Results "and discussion" – there is a lot more than results in this section.*

Changed to "Results and Discussion"

- *Figure 1. A very busy figure. Fonts are too small to be legible. Axes labels are missing on panels e/f/g.*

Removed panels e/f/g as they are replicated in supplementary figures which increased the size of figure, especially fonts. Y Axes labels (histogram counts) were added in the supplementary figures.

- *Figure 2. Another very difficult figure to read. Suggest reducing the number of panels and enhancing font size. Some alkalinity flux axes labels for are missing in panels a/b.*

We split the future into two figures and fixed the axes labels.

- *Figure 2 legend, Line 2- I would call this CDR potential not OAE potential Given the different research questions this manuscript addresses I think this distinction needs to be clear.*

This sentence was removed from the figure legend and worked into the text as suggested by Reviewer 1. However, we agree that the distinction is critical and we have tried to make the distinction between OAE and CDR clearer throughout the text.

- *Figure 3. Panel labels b/c are incorrectly ordered. Panels e/f/g are not interpretable at their current size.*

Panel label order fixed. Increased the size of panels e/f/g and increased font sizes overall.

- *L326 Largest per unit area fluxes. The globally aggregated fluxes are shown to be higher for wider strips.*

My understanding is that the meaning of "flux" is already area normalized, i.e. a flux is "substance flow rate per unit area".

- *L331-332 This sentence is unclear, please clarify.*

Hopefully clarified by changing and expanding the sentence to "In general we find that the sensitivity of pH and Ω_{Arag} with respect to the local flux of alkalinity highly dependent on the surrounding pattern of injection. In other words the limit at which alkalinity can be added at a given location depends on the alkalinity addition rate at neighboring locations, up to some distance.

- *L335 Unclear where this 25% comes from. Fig 3e/f/g appear to show 8% and 30% declines in the 200km and 400 km simulations.*

"Placing injection patches 400 km apart instead of 200 km apart did not further increase the sustainable flux in each injection patch, and reduced the overall injection capacity by 25%". This sentence is comparing panels f and g, not e and f. In other words g is 25% lower than f. ($233 \text{ Tmol/yr} / 312 \text{ Tmol/yr} = 0.75$)

- *L341. Do the authors expect this length scale to vary considerably regionally?*

My sense is that yes, this length scale is non-isotropic and will vary from place to place.

Indeed Jones et al. 2016 reports "The global median spatial autocorrelation (e-folding) length is 400 ± 250 km, with large variability across different regions."

We amended the sentence at L341 to indicate the variability "... injection areas should be placed at most 200-400 km apart, however the optima will depend on the local current patterns."

- *L351-352 clarify these are molar ratios.*

Changed sentence beginning to "After 1 year the molar uptake fraction.."

- *Figure 4. Reiterate the units are mol/mol.*

Added "(molar ratio)" to figure legend.

- *Figure 5. Another busy figure. The "surface pCO₂" label is unnecessary and conflicts with the surface pCO₂ deficit label.*

We've removed the extraneous "surface pCO₂" label and will increase the font size to match that of the main text in the final layout of the paper.

We agree the figure is busy, but we believe this is justified as we're trying to visually show the interplay between spread of the deficit, the CO₂ uptake behavior and the depth-loss of alkalinity. We chose three scenarios to exemplify the breadth of behaviors possible.

This figure is intended to utilize the full width of the page in the final print, rather than 75% as in this preprint.

- *Figure 5. legend. I think some of this legend may be misleading. Presumably change in the surface pCO₂ over time is also influence by mixing/circulation/biology (and not only gas exchange).*

Reviewer 1 suggested moving interpretations and discussions from figure legends to the main text (to avoid duplication). Thus we have moved discussion of effects other than gas exchange into the main text.

- *L371 Some local deficits $>1\mu\text{atm}$ are shown in Figs 5bii and 5cii.*

We amended the paragraph to better reflect the intended point:

"We note that for all cases the alkalinity-induced CO₂ deficit spreads over a very large area within one year and a significant fraction of the CO₂ uptake occurs after the deficits have diluted to the sub μatm range. This makes direct monitoring and verification of OAE extremely challenging and will likely need to rely on modeling and indirect experimental verification."

- *L405 mol/kg?*

Changed to mol/kg.

- *L413 I would call this a high-resolution global ocean model (particularly compared to most global obgc models and papers that have simulated OAE previously).*

Changed "medium" to "high"

- *L421 Shouldn't the R in MRV be for Reporting?*

Changed to "reported".