To the reviewers and editors:

We thank you for your second review of “An assessment of ocean alkalinity enhancement using aqueous hydroxides: kinetics, efficiency, and precipitation thresholds.” The initial round of comments led to major improvements in the manuscript, and the latest review has continued the trend towards a stronger and more efficient description of our work.

Below in our Response to Reviewers, we provide a point-by-point response to reviewer comments and explain the changes that were made to the manuscript, provided in track-changed and clean formats.

On behalf of all the co-authors,
Sincerely,
Dr. Mallory Ringham
Adjunct, Stony Brook University
Lead Oceanographer, Ebb Carbon, Inc.

Responses to reviewer comments
In the responses, “[Line N]” indicates that the location is line number N in the track-changed version of the latest reviewed version of the manuscript.

Report #1: Anonymous referee #2

Comment 1: The authors have addressed all comments. Their argument against “unclear novelty” is that tank scale experiments are a necessary transition from benchtop-scale to field studies. I partially agree, it is indeed desirable to show results like the ones presented here to confirm approximate kinetics of gas exchange and secondary precipitation derived from other studies and different scales. I only “partially” agree because I think their argument that not much known is for CO2 exchange in the context of OAE is not a good one. It is unclear why knowledge on air-sea CO2 exchange could not equally well be derived from totally different research fields. Obviously, such research exists and models are parameterized accordingly, which is why the timescales estimated with them are broadly consistent with the ones measured here. Nevertheless, the first argument (there is value of different lab scale experiments) may outweigh the latter, especially when it comes to permitting. The responses to the other comments are also satisfactory, although I was at times surprised by the authors’ defensiveness. For example, the authors argued against the comment that there was only one reference in the whole results/discussion/summary sections. I would have thought that there was not much to argue against. But in any case, the authors have enriched these parts and tried to add further context so ultimately considered the recommendation.

Response 1: We thank the referee for their comments on the novelty of this topic. We agree—prior research on air-sea CO2 exchange in other fields lends significant confidence to OAE research. The need for demonstration of this process at intermediate scales is driven by an abundance of caution in moving mCDR research from the lab to field trials and is critical to social license and scientific reputation in testing OAE approaches. We appreciate the valuable commentary throughout the discussion section, which we improved through comparison with relevant publications in the previous revision.

Comment 2: One remaining (minor) comment is that the summary still sounds more like a discussion. E.g. the repetition of methodological details seems somewhat out of place. Furthermore, the summary contains new discussion points. This is not critical but may be addressed.

Response 2: This was a helpful note. In finalizing this manuscript, we shifted several sentences from the Results and Discussion to the Methods, including lines 353-358 (discussion of changes in steps to homogenize control and experiment seawater before beginning an experiment), 452 – 455 (highlighting again the use of smaller aquaria to enable additional experiment cases on shorter timelines, and 474-480 (noting handling of evaporation in aquaria experiments. We have renamed the Summary section to Summary and Future Work, to clarify that discussion points in this section are specifically relevant to improvements and new directions that could be taken from this work.

Report #2: Referee #3, Sijia Dong, sijiadong@nju.edu.cn

Comment 1: Ringham et al. present original experimental work on the chemical consequences of ocean alkalinity enhancement. The authors exclude biological responses by pre-treating seawater with bleach and UV, and monitor the water chemistry and solid phase precipitation within the water column. The authors include, in their manuscript, a good incorporation of up-to-date studies, as related researches in ocean alkalinity enhancement have been emerging rapidly. I think this work provides important new data and information for the field, and should definitely be seen by the science and policy-making community soon. Overall, this work provides new experimental data and information to better understand the consequences of ocean alkalinity enhancement, a widely interested topic. I would recommend publishing the manuscript after minor revision.

A small concern I have about the research is:

In Section 3.3, the runaway precipitation section, the authors state that Mg(OH)2 immediately precipitate after NaOH addition, as thick, slurry-like precipitants. Then, within 24 hours of NaOH addition, most Mg(OH)2 signals disappear (from the XRD spectrum of filtered particle samples), and aragonite start to precipitate. TA decrease as the two minerals precipitate, and drop below that of the initial seawater within 24 hours of the NaOH addition (Line 543-544). In Figure 6, there seems to be another really intense and crystalline solid phase in addition to brucite at time zero. The authors do not specify what this phase is. And there seems to be aragonite peak as well (around 2-theta = 28 degrees) at t0. Brucite XRD peaks should be quite narrow and intense too (in Figure 6, at 0 hr and 3 hr, the peaks seem quite broad). But when there are “sharper” peaks, other peaks might be hard to see. In this case, “other peaks” would be brucite and/or aragonite. I would recommend the authors identify what the intense mystery peaks are (for example, it could be bleach that decompose with time?), and see whether removing these “noise signals” affects the interpretation of their results. If the authors still have spare samples, they can analyze the samples again after rinsing. I would also recommend a more quantitative analysis of Mg(OH)2 / aragonite ratio if possible.

Response 1: We thank the referee for their comments on this work. In addressing the questions on the XRD data presented in Figure 6, we have revised and updated the figure to include the peak angles characteristic of brucite, calcite, aragonite, vaterite, and NaCl. We have highlighted a few additional peaks showing the progression from brucite-dominated precipitation to aragonite-dominated precipitation over time. We note that the noise in this Figure in the 0 and 3 hour time points is likely due to the difficulty in removing salt from the thick slurry of these samples (see lines 564-566). The suggestion that bleach could be decomposing is an interesting one, but was ruled out through a series of Hach tests of free chlorine and monitoring of pH that suggested the effective decomposition of bleach before the beginning of each experiment. In regards to the 2θ peaks ~27°, we note that there is a single peak could be associated with NaCl at the 0 hour timestamp, and there are two peaks associated with aragonite at time = 3+ hours. We have added two figures to the Supplementary Materials (S7 and S8) from duplicates of this experiment at aquaria scale. While a t = 0 hour sample is only available in one of the two aquaria experiments, we note a similar distribution of peaks that indicate NaCl trending to aragonite over time. While there are no spare samples available for reanalysis, we agree that future studies would benefit from improved slurry collection and cleaning prior to XRD followed by additional analyses of the brucite/ aragonite ratio.