Reviewer 2 (Devon Cole)

Hashim et al. investigate the rate of mineral formation during the addition of liquid alkalinity to unfiltered seawater in a shipboard experiment to gain insight on the potential outcomes of certain methods of OAE. This paper provides a new aragonite precipitation rate for 'unseeded' scenarios, although once precipitates are present they should act as seed for further precipitation. This manuscript is clearly written, well organized, and was a pleasure to read. With minor revisions I look forward to the publication of this work.

Thank you!

General Comments:

Very curious if the unfiltered seawater could have had suspended particles of any sort that might act as seeds in this experiment. From the omega threshold/induction period time, would it be possible to compare to other experiments of homogeneous precipitation and make a guess about potential impact of such particles? Although from a real-world standpoint, regardless of mechanism, this does make the most realistic approximation of the conditions that would be faced during an actual deployment of liquid alkalinity in the ocean.

We do believe that the particles present in our unfiltered seawater make a difference in the threshold and induction period of precipitation. The observation that the threshold for precipitation observed in our experiments is lower than that of Hartmann et al. (2023) for the filtered seawater experiments is likely because of nucleation on particles. Since we did not conduct any experiments with filtered seawater, an insightful discuss of the effect of particles is not possible. However, Hartmann et al. (2023) conducted identical experiments using filtered and unfiltered seawater (they refer to as abiotic and biotic) and demonstrate the effect of particles on the threshold of precipitation.

Did the authors by any chance filter the control and analyze to get a sense of what might have been suspended in the seawater to start?

We did filter the control. Visually, there was very little material on the filter, certainly not enough for any mineralogical work. We analyzed the sample for particulate inorganic carbon and found that it has 0.13 µmol/L seawater.

Would it be possible or useful to quantify the amount of precipitate and then use an estimate of BET surface area for the aragonite and work out rates which include surface area for the latter parts of the experiment once there is precipitate present? I am wondering if that could inform on the comparison of this rate to the Mucci rate more directly. Could this also allow a better understanding of the ease of precipitation on the inside of the bag (how much less efficient is it than particles) and confirmation of the normalization scheme used? It seems that if the answer to my very first comment is that omega values got high enough without precipitation that we are confident we generated homogenous precipitation, then I would think using the inside of the bag to normalize for surface area/with the Mucci rate is a sort of false value. That is, the initial surface area was effectively zero. So to convert to units w/ surface area, the authors would have *to rely on the accumulation of precipitate. This is quite a tricky problem and I very much look forward to seeing the authors thoughts on this!*

> Thank you for the interesting and thought-provoking question. The initial Ω was high enough, especially in the +1000 and +2000 alkalinity addition experiments, to induce homogenous nucleation, based on previous published work. As you mention, this means that the surface area was effectively zero and increased with time as these nuclei grew. Therefore, we are not sure if there is a way to constrain the evolving surface area without direct measurements and experiments that are specifically designed to test this.

Line-by-line:

Ln 40: should be "correlated"

> Noted. We will change that.

Ln 316-321: add ref to the figure here (fig 9?)

> Yes. We will refer to Figure 9 at the end of the sentence.

Ln 385: should be "may have prevented all of the brucite from redissolving,…"

> We will change the sentence according your suggestion.

Ln 391: define ACC up here

> Noted.

Ln 442: reaction order stated as 2.2 but shown as 2.16 on the figure, I think should make consistent throughout

> Noted. We will make sure to fix this. We also would like to mention that we discovered a small mistake in the code that performs the regression to calculate the reaction order and constant. It seems that the data from experiment D were mistakenly excluded which led to a small error in the values of n and k. The new value for the reaction order is 2.15 (compared to 2.16) and for the reaction constant is 0.20 (compared to 0.15). We will make sure to update the manuscript and figures accordingly.

Ln 444: Curious about the temp differences. I think this warrants a bit of discussion – either as to why the author has decided the slight differences don't matter, or if they do, how they ought to be handled to make all of these experiments comparable.

> We assume you are referring to the fact that our experiments were conducted at 27 ºC, Mucci et al. (1989) and Burton and Walter (1987) at 25 ºC, and Moras et al. (2022) at 21 ºC. We have thought about conducing a temperature correction based on available data

regarding the effect of temperature on kinetics, we decided that the correction is not necessary and could be counterproductive for two reasons. First, the temperature different between our experiments and those of Mucci et al. (1989) is only 2 ºC yet the difference in reaction order is 2.15 – 1.48 = 0.67 and the difference in reaction constant is 45 – 0.2 = 44.8. Based on the data of Burton and Walter (1987), the reaction order increases by ~ 0.066 per 1 ºC and the reaction constant by ~ 0.5 per 1 ºC. This suggests that differences in reaction order and constant would be within the reported uncertainties and are too small to make a difference.
Regarding the experiments of Moras et al. (2022), the difference is 4 ºC with our experiments. While recalculating their data to 25 ºC might bring them in closer alignment with our data (since they generally plot lower than ours in Figure 9 and a temperature increase leads to higher reaction order (steeper slope) and higher reaction constant (intercept), we think that a correction is not warranted because the data of Burton and Walter (1987) is based on seeded experiments where the reaction constant is much higher than in our and Moras et al. (2022) experiments, so it is unclear whether this extrapolation is justified given that it would only make a small difference. We will make sure to add a few lines in our manuscript so that the reader is aware of the temperature differences between experiments.

Ln 445: seems like the reaction orders are rounded here too

Noted.

Ln 457: this surface area comparison and normalization is very interesting to me, and I appreciate the detail. Given the bag material and the Subhas reference, it seems that precip on the bag should be retarded relative to if that 0.24m2 surface were carbonate. So if you convert this rate to a umol/m2/time unit, it is perhaps the case that the reaction constant should be even lower if the bag surface area is not very efficient? See the rest of my thoughts on this above

We agree that the bag inner walls, despite being non-ideal for nucleation, did serve as nucleation sites, which perhaps impacted the reaction kinetics to some degree.

Fig 10B – Should the Y axis be negative? Its making the point that more TA is removed by the end of the experiment than what it was to start, so the delta TA should be a negative number, right?

No, because it represents initial TA - final TA (after precipitation) which is always less than the initial.

SI Data:

The data copied over from Mucci et al 1989 do not appear to match that paper. I believe the data in the rate column are in fact omega-1 and I am not sure what the data in the log(omega-1) column are. I think these data should be from table III in that paper. I worked to calculate the surface area normalized data which are shown in Fig 9 of this manuscript for use in my own *work, and have come up with the same values that are plotted in Fig 9 working directly from the Mucci text, so I believe this is just a small copying error here.*

> Thank you very much for pointing this out! There was indeed a copying mistake. We will make sure to update the table with the correct data.

As a small gift to future close readers, the authors could add which table in Mucci these data come from to column A

> The data were taken from Table III. We will make sure to mention this in the spreadsheet.