This study investigates the mechanisms that control runaway calcium carbonate ($CaCO_3$) precipitation during ocean alkalinity enhancement (OAE). The researchers use previously published data to construct a process-based model for estimating total alkalinity (TA) losses due to runaway precipitation. The manuscript is well presented and makes significant contributions to our understanding of runaway calcium carbonate precipitation during OAE deployments.

General comments

The manuscript largely builds upon previously published data from Hartmann et al. (2023) and Suitner et al. (2024). This manuscript does build upon the earlier work; however, the authors frequently direct the reader to these articles throughout the manuscript. In several instances I believe it would be beneficial to simply provide the information within this manuscript rather than directing the reader to another article, while in other circumstances I do not feel it necessary to continuously cite these articles. Lines 160-161 direct the reader to the supplementary information of Suitner et al. (2024) while this information if important should be included in this manuscript or its supplementary information. Also, lines 252-259 cite the articles 5 times, in particular line 253 refers the reader to Suitner et al. (2024) for images of the aragonite precipitates which are also depicted in the current manuscript in figure 8.

To my knowledge increases in total alkalinity of 1600 – 2800 µmol/kg above current levels is not realistic in real world settings. The authors state themselves that the standard experimental setup (which used relatively high delta TA values) did not provide enough precipitates. This raises questions about the applicability of such extreme perturbation studies to real-world scenarios. Understanding such processes are undoubtedly important, however their relevance to actual OAE seems somewhat limited. I would welcome further discussion around this point expanding upon section 4.5. Additionally, the authors utilise mesocosm and small-scale bottle experiments, while the limitations of such experiments are well understood a short discussion of the limitations of such datasets would be beneficial.

In contrast to the discussion surrounding omega and precipitation under high alkalinity values I feel the manuscript could discuss the influence of suspended particles further. This factor appears to be much more likely to result in runaway precipitation than the intentional increase in omega aragonite above 20, particularly as researchers and companies look to rivers to transport alkalinity to the ocean.

Specific comments

Comment 1

Figure 4. This is an extremely busy figure, and I would recommend removing some of the text from within the actual figure and placing it outside. For example, the APP explanation, breaks up the plot to a point where it seems as if there are two distinct plots. A distinct legend would likely be beneficial, and I recommend the authors consider this as well as simplifying/removing some of the text within the plot.

It is unclear as to why the triangles for the TA1200 treatment are hollow and smaller in comparison to the other two treatment levels utilise filled triangles.

The secondary x axis is difficult to interpret as it appears to show start and end deltaTA values for CO2 equilibrated but only start values for the unfiltered and end values for filtered non-co2 equilibrated measurements. Proper axis labelling here would be beneficial or removal of this secondary x axis.

Comment 2

Both figures 4 and 5 depict TA on the x axis and time on the y axis, however all other figures in the manuscript appear to utilise the opposite axis labelling. This is confusing. I strongly recommend the authors use consistent labelling of the axis throughout the manuscript.

Comment 3

Lines 222 – 224. Suggest that the initiation of APP may be estimated via initial TA or omega aragonite values. However, figure 6 illustrates this to also be dependent on the approach and assumingly particle density. I appreciate this is highlighted in the figure caption but believe this should be explicitly stated in the text.

Comment 4

Line 299: Might help prevent

Comment 5

Line 390: it is unclear what cf. stands for and no black triangles are present in fig 3c

Comment 6

Line 389: description of the line/fits in figure 9 throughout this section are misleading please change wording from “graph” to “line/fit” throughout.

Comment 7

I appreciate section 4.5 and the discussion surrounding the context of this experiment in a real-world setting. However, lines 427 – 429 state “Nevertheless, since these projected APP induction times are also within the suggested residence times of treated water in the upper ocean layers, it is necessary to conduct studies lasting at least for the projected timespans, depending on the local environmental conditions”. I question whether a perturbed water parcel would remain in its perturbed state given the physical processes occurring in the surface ocean. Irrespective of its residence time, if the water parcel becomes diluted the omega values which the authors state as a controlling factor for determining precipitation would be significantly lower rendering any further precipitation highly unlikely.

Comment 8

I question the use of stokes law which as stated by the authors is used for solid spherical particles. Given the SEM images provided by the authors the particles appear to have significant cavities likely increasing the SA/V ratio and thus significantly influencing the sinking velocity. It is also unclear whether measured or calculated sinking velocities are used.

Comment 9

I question the relevance of using 10 m as the upper water layer given mixed layer depths often range from 10 – 100m. Understandably it is important that the perturbed water remains in contact with the overlying atmosphere for CO2 uptake to occur. However, if one is to consider the removal of particles from a system, they must consider it in the context of the MLD. Any particle flux above the MLD at the current point in time is not equivalent to the removal of the particle from the discussed water layer as it is entirely possible that the particle may re-enter the 10 m layer due to various mixing processes.

Further to this I question if the authors considered two important aspects of particle fluxes 1) attenuation with depth and 2) variable mixed layer depth and reinjection of particles into the surface layer. Understandably the particles described here are abiotic but a comment on the potential attenuation of these particles would be appreciated. Following this, have the authors considered reinjection of the particles via mixing processes? Although unlikely for the fast-sinking fraction I question if this may enable a delayed CO2 uptake or alter the OAE efficiency.

Comment 10

I appreciate the authors thoughts surrounding the transport of particles and their subsequent effect on runaway precipitation. However, given the context of this study I believe an estimation of the effect of particle export on OAE efficiency would be beneficial. Especially considering that the sinking velocities and abundance of particles have been calculated. I also again query how the authors differentiate between small particles capable of staying in the upper layer for months and those which sink “while still growing”. Could it be more realistic to assume that most particles continue to grow until they aggregate or reach a sinking threshold? And if so, how would this translate to OAE efficiency? Particularly given that the removal of growing or fully grown particles would likely begin to dissolve as they descend the vertical water column and have differing impacts on the alkalinity export.

Comment 11

Supplementary figure S1 could benefit from some slight adjustments so that the brightness of each image is similar. S1.B is very dark making it difficult to see any important details such as the branching shown in figure S1 A and B.

Comment 14

Image quality of the supplementary figures S2, S3, S7, S8, S9 and S10 is poor and should be improved prior to publication.

Comment 15

Figure S8 describes plots as the interplay between omega aragonite and surface area of particles. However, I would argue that it is the omega aragonite controlling the size of the precipitated particles and thus surface area.

Comment 16

Figure S10. I appreciate the lengthy explanation by the authors here however they fail to link such particle transformations back to the primary purpose of OAE, namely CDR. It is important to understand how such particle transport mechanisms would act upon the overall efficiency of the OAE deployment not just the particle surface area. To expand, an export of particles due to caco3 precipitation is still a loss of alkalinity and thus a reduction in efficiency. What would be interesting is to understand at what point this reduction in efficiency is beneficial if at all? Is there a point at which such extreme perturbations would be beneficial over smaller or medium sized perturbations. Otherwise, such a discussion may have limited relevance unless connected directly to implications for CDR efficiency.