RC2: ANONYMOUS REFEREE #2

Overall assessment:

This is a timely and significant contribution and I applaud the authors. This manuscript is ambitious, covering a range of relevant species for calcification rate responses to OAE, and will be of interest to biological oceanographers, ecologists and the wider carbon removal community and industry. The paper has a fantastic coverage of species and I really like the focus on functional groups; this is a sensible way to think about ecosystem response to OAE, in terms of what function each of these organisms play in the marine food web and carbon cycle more generally e.g. biological and inorganic carbon pumps. I really enjoyed reading the paper and thought it was generally well written and scoped. However, I have some major points raised below and also some more minor under "other comments". The conclusions of the study are solid - there should be more realistic manipulations of TA for responses of biological organisms in academic studies, and this is a fair interpretation of the data. Statistical analysis seems fair.

We would sincerely like to thank the reviewer for recognizing our objectives and efforts, and for the useful feedback. Comments are addressed below.

Major points for revision:

1. *Better elucidation of link between TA:DIC and Ω_{ar} needed. For example, in the text introducing the rationale, it is just described as TA:DIC being "essentially" related to Ωar, however this relationship is not explained in any depth and then in Figure 1, TA:DIC/Ωar is plotted on the x axis of the graphs, making an assumption that they are equivalent for those purposes. I think TA:DIC is a fair proxy for Ωar, but should not be plotted in a way that misleads to suggest they are functionally equivalent and could be 1:1 swapped out for one another in graphs such as those shown in Figure 1. A lot of this links back to methodology for this relationship being used which is shown in Figure 3; perhaps moving this graph to earlier in the flow of the manuscript could help to clear up some of this confusion.*

Response: We have covered the explanation of the link between TA:DIC and Ω_{ar} in our response to reviewer #1, point 1. We have expanded on this in the manuscript as well in section 1.2, and therefore believe leaving figure 3 in the start of the Results section is reasonable. We have removed Ω_{ar} from the figure 1 x-axis.

2. *It seems questionable that GLODAP should have been used, the data logically to me should have come from the individual studies relevant and used in this study; I am unclear of the reasoning here and it does not seem well justified to be using global datasets for mapping some of these responses, but I note this justification is not well detailed in the text and could be a lack of explanation rather than a fundamental flaw in the design of the study.*

Response: This is explained in our response to reviewer 1, point 3.

3. *Not enough examination of what the actual impact of increased/decreased calcification rate would be in the manuscript. This is generally discussed in the context of biological responses to OAE, insinuating that any change is a bad change - this is not true - we have to think about this in the context of some organisms having decreased calcification under acidification scenarios, so some increase in calcification rate for these organisms could actually represent a bounce back to pre-acidification conditions. Overall, important to understand that any intentional manipulation of the earth system like OAE will have an impact, but the actual "negativity" of the impact needs to be a nuanced and detailed discussion, which I think this manuscript could benefit from significant additional discussion on this in the Discussion section of the manuscript. Places where this is particularly relevant: lines 562-564 and onwards.*

Response: We appreciate this valuable comment by the reviewer that helped to elucidate more nuanced discussion about the categories-specific calcification while taking into account the anthropogenic changes in carbonate chemistry since the pre-industrial times. We have done additional analyses of the pre-industrial calcification now and have amended the text in the manuscript to be more precise in addressing the complexity in responses. Below is the short *background* on how we have tackled this in our edits of the manuscript.

Anthropogenic CO2 uptake in the ocean resulted in lower pH and Ω_{ar}, resulting in lowering of the calcification rates in the species, in particular the ones that are in our study were categorized as the positive responders (linear and threshold positive due to increase in TA-induced Ω_{ar}). With respect to the changes since the pre-industrial times, the aim was to examine the difference in calcification between current and pre-industrial times, and to what extent NaOH would be able to compensate for this difference. We have attached two figures as examples of linear positive and threshold positive responders, in which we have clearly indicated the current and pre-industrial level of calcification to help visualize the text we provided (see Figure 5).

To understand this, we have done the following: first, we inferred the pre-industrial TA:DIC ratio of 1.16 (Feely et al., 2004) vs. a current TA:DIC of 1.12 and use the regression lines of TA:DIC vs calcification rate to calculate the corresponding calcification rates. Then we calculated calcification due to the addition of NaOH and Na_2CO_3 from the species-specific baselines (see Method section 2.4 for detailed explanation) for the positive responders. This is done using the principles of mass balance approach for the carbonate system via CO2SYS, where the carbonate system is calculated for each increment of NaOH or Na_2CO_3 added. The difference between the pre-industrial calcification and current, increased by the NaOH was assessed and compared. We observe the following:

- The average calcification rate in the species with the *threshold positive* rate was similar for the pre-industrial and current conditions, or with generally small difference between the pre-industrial and current conditions. This is likely because they retain max calcification rate even at the current rate and the changes since the pre-industrial did not induce calcification challenges yet.
- However, a very different case was evident for the linear positive calcifiers, where the current calcification rates are substantially lower compared to the pre-industrial calcification rates,

indicating that the calcification rates have been substantially compromised already in these species. An increase using NaOH would compensate for the calcification loss due to lowering of pH and aragonite saturation state since the pre-industrial times. Interestingly, we find a rather uniform pattern for most calcifiers where the difference between the pre-industrial and current conditions would be compensated if an addition of 50 to 150 µmol/kg NaOH was added. This amount is species specific, but at least this represents a wider range of NaOH that would be needed to revert back to the pre-industrial calcification. We also emphasize that for a much smaller amount of species (e.g. coral *Duncanopsammia axifuga* from Bove et al., 2020), we would need less than 50 µmol/kg NaOH to achieve pe-industrial calcification.

- This represents a substantial increase, translated into the pH and Ω_{ar} of an average increase of 0.09 for pH, and 0.54 for the aragonite. For these species, we could then indeed conceptualize that this NaOH addition would then allow these species to *first bounce back to the pre-acidification conditions*, and as such not induce competitive challenges in the community up to the values of NaOH added of 50-100 µmol/kg. Achieving such long-lasting increases of NaOH is currently not envisioned and as such, we are not expecting this as it represents huge carbonate chemistry amendments, meaning we do not envision competitive challenges to be an issue for the linear responders. We have amended our text in the discussion in view of these new findings.

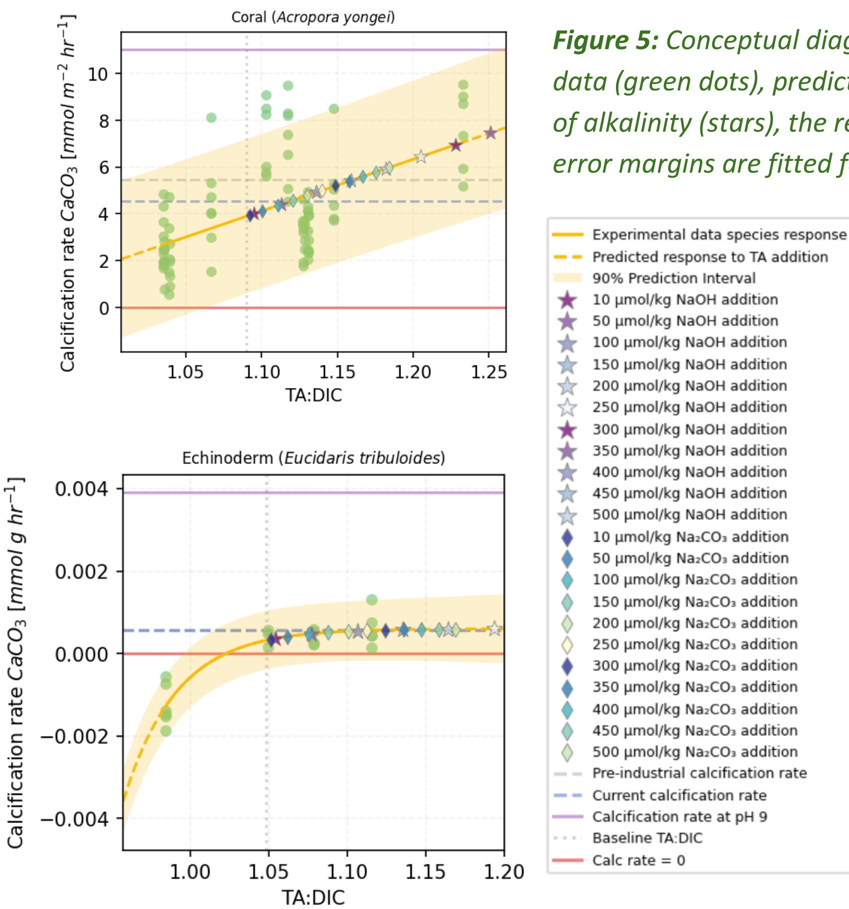

Figure 5: *Conceptual diagram to show how experimental data (green dots), predicted values at various additions of alkalinity (stars), the regression line and prediction error margins are fitted for a given species a) linear positive (top) and exponential for threshold positive (bottom). The uncertainty interval indicates four standard deviations. The red line indicates zero net dissolution (calcification rate is equal to 0; dissolution rate = calcification rate). The blue dotted horizontal line indicates current calcification rate (TA:DIC = 1.12) and the gray dotted horizontal line indicates pre-industrial calcification rate (TA:DIC = 1.16).*

Other comments:

Generally found the presentation of the different categories of response e.g. linear +, linear -, exponential +, exponential -, etc a bit confusing, particularly in Figure 6 and 7. I wonder if a box calling out exactly what fits into each of those categories and what they mean would be helpful for the reader to refer to.

The following box will be added alongside the bar chart, and the color coding (green for positive, gray for neutral and red for negative) will be used in the bar chart to make it clearer.

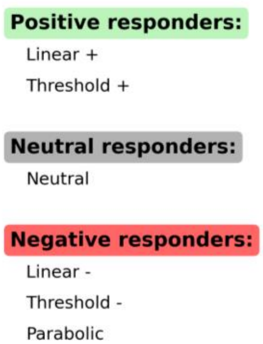

Figure 6: *Box with color coding to clarify which responses are positive, neutral and negative.*

line 483-485: not a sufficient explanation of the biological mechanisms involved in bicarbonate impact moderation in crustaceans

Calcification in crustaceans is enabled through the process of regulating pH at the site of the calcification, by converting HCO3- to CO32-, allowing the crustaceans to operate over large pH ranges, with such strategy being especially successful at low pH. While this gives the crabs competitive advantage at ocean acidification conditions (lower pH, less carbonate ion available) compared to the other species that are using bicarbonate ions directly, this might not be advantageous under high pH conditions where such mechanisms are needed. Studies are lacking on regulating calcification at higher pH, but there is evidence of the metabolic costs associated with the physiological regulation at higher pH (Cripps et al., 2013). Crustaceans show a disrupted acid–base regulation upon the alkaline compound addition. Individuals exposed to elevated $Ca(OH)_2$ concentrations showed an increase in hemolymph pH, K+ Na+ and osmolality and a decrease in extracellular PCO_2, TCO2 and HCO_3^-. These are essential physiological extracellular acid-based parameters, which were all significantly affected. Such response is according to the authors (Cripps et al., 2013) indicative of respiratory alkalosis (see also Truchot, 1984, 1986), which is often associated with hyperventilation. Hyperventilation is associated with increased respiration with the aim to increase the flow of water over the gills, flushing out the hemolymph CO2, increasing the affinity of oxygen uptake of the hemocyanin. While this creates physiological favorable conditions for crabs, it also represents the physiological costs and potential metabolic composition of other processes, such as calcification. We fully acknowledge that this remains a hypothesis right now, however this might nevertheless explain the potential mechanism of a decline due to alkalinity addition. We will additionally expand on this in the manuscript.

line 552: should read, such "a" guide

Fixed.

line 559: should be "better informing further experimental work"

Fixed.

line 585: when "the" grazing effects included - "the" should be removed

Fixed.

line 589, from "a" not the "the" biogeochemical perspective

Fixed.

line 600-601: NaOH is not a carbonate-based compound

We have removed this, it now reads: 'This study only considered the changes in carbonate chemistry due to the addition of NaOH and Na_2CO_3.'

line 605: "based" after OAE should be deleted

Fixed.

line 609: should be "were" not "was" used<<

Fixed.

line 618-619 - second part of clause not necessary, can be deleted

Has been deleted.

One of the major conclusions of the study is that the study is aiming to be used by the community to identify species that are at the largest risk of a "negative" response, or have the greatest uncertainties in their potential responses. However, nowhere in the paper is there a succinct summary of what those species are, in a box, or in a section, or a table. I think something like this would greatly improve the impact and applicability of the paper for the intended audience to use it, and the manuscript would be improved by this

We believe Table 2 provides a clear overview of the negative responders, along with their thresholds. We aim to improve this table by translating the thresholds to pH and Ω_{ar} and provide the respective errors (RMSE).

line 626: replace "the" with "further" experimental work

Fixed.

line 630: add "an" experimental framework

Fixed.