*Comments Reviewer 1*

*Minor Comments*

*Abstract: L19-21: Be specific, what changes are seen? In what parameter?*

Added "in alkalinity"

*L21-22: Not quite sure what that means.*

The sentence now reads: *Globally, while we see that under RCP2.6 the carbon uptake associated with AOA is only ~60% of the total under RCP8.5, the relative changes in temperature are larger, as are the changes in pH (140%) and aragonite saturation state (170%).*

*L22-23 The change in saturation state is ambiguously describe, refer specifically to changes in omega.*

With respect the actual values are listed in the body of paper, and to list all of the values here would make the abstract too long.

*L28 It's left a little open ended here, you could be more specific with the regional response. It is one of the more important findings from the experiment.*

We would love to but given the length of the abstract we feel that we are somewhat limited in terms of space. But we have tried to be a little clearer, the last sentence of the abstract now states:

*Finally, our simulated AOA for 2020-2100 in the RCP2.6 scenario is capable of offsetting warming and ameliorating ocean acidification increases at the global scale, but with highly variable regional responses.*

*Introduction: Good introduction. Clearly explains why we need CDR and more specifically AOA. Also gives a description of ocean acidification and how AOA works.*

Thanks

*L50: 'Including through coral bleaching' a little clunky, maybe remove 'through'*

We have removed this statement, it now states: *While warming represents an imminent global threat which is already significantly impacting the natural environment (Hughes et al., 2017), ocean acidification poses an additional and equally significant threat to the marine environment.*

*L54: Could you say something about the changing Revelle Factor, and the potential for AOA to impact this?*

We have now added the statement to the text: *As $CO_2$ is taken up by the ocean it changes its chemical equilibrium, reducing the carbonate ion concentration and decreasing pH, collectively known as ocean acidification.*

We have also added statement later in the introduction to say:

*Artificial Ocean Alkalization (AOA), through altering the chemistry of seawater, both enhances ocean carbon uptake (thereby reducing atmospheric $CO_2$), while at the same time reversing ocean acidification and increasing the buffering capacity of the ocean.*

*L148-150: simply states "impact" which could be a bit vague. Could go in further and state that they will be investigating the impact on the "carbon cycle, global surface warming (2m surface air temperature), and response and ocean acidification response to the 4 different AOA experiments under the high (RCP8.5) and low (RCP2.6) emissions scenarios." Which is stated in lines 207-210. Regarding my comment above, it is worth exploring the potential experiment space, magnitude of alkalinity addition, lo cation, emission scenario, and the resulting impacts site specific/regional/global/ open ocean/coastal etc. What parts of this picture does your model/this paper deal with, what has already been done by others, and what is left to do? are other models needed?*

The goals of this study of this focus on the global response to regional and seasonal AOA, therefore we don't explore are best ways to ameliorate local conditions through AOA. Regarding question of whether to explore experimental space further – the answer is yes there is a lot of work that needs to be done (please see the review by Renforth and Henderson (2017)) and these results are put into this context in the Results and Discussion section. AOA will also have addressed as part of the Carbon Dioxide Removal Model Intercomparison Project (CDR-MIP), which we are involved in.

We have now modified the paragraph to now say:

*In this work, we use a fully coupled ESM (CSIRO-Mk3L-COAL), which includes climate and carbon feedbacks, to investigate the impact of AOA on the carbon cycle, global surface warming (2m surface air temperature), and ocean acidification response to the global and regional AOA experiments under the high (RCP8.5) and low (RCP2.6) emissions scenarios*

*Methods:*

*Model seems appropriate for the scope of this paper. Clear description of the experimental design which seems appropriate to answer the research question proposed in the introduction. Could explain what the model outputs are? Also should mention the testing for seasonality? (mentioned in lines 538-554)*

This model has been assessed in a number of studies already cited here and the outputs are consistent with standard Earth System Model outputs. As stated above the key outputs we consider are surface air and ocean temperature, and changes in the surface pH and aragonite saturation state. We also describe in the methods sections a number of other model prognostic variables (lines: 153-168) and provide references to individual model papers that describes components of the CSIRO Mk3L-COAL earth system model.

*L162-164: Do you expect this assumption to hold up under elevated alkalinity? Could the rain ratio change?*

Probably, this is already addressed in the lines 445-460, and studies suggest a small feedback.

*L204: Fair assumption, but it is worth pointing out that alkalinity manipulation could be from carbonate dissolution or NaOH addition which would not induce and impact from iron and silicate. You are testing the fundamental impact intrinsic to all of these methods of C sequestration.*

We agree, the sentence now reads:

*We do not consider the biogeochemical response to other minerals and elements that can be associated with the sourcing of alkalinity from the application of finely ground ultra-mafic* *rocks such as olivine and forsterite, nor dissolution processes required to increase alkalinity (e.g. Montserrat et al., 2017).*

*Results and discussion:*

*L208 – 209: the sentence doesn't make sense, a typo somewhere?*

Yes, we have now removed *and response*

*L215: Why have you chose this addition rate? Also, should alkalinity not be expressed in equivalents rather than moles (and throughout)?*

As stated, this value is very close to that used by Keller et al (2014) following Kohler (2014), who estimated a value of AOA based on globally shipping. While this allows a comparison of simulated values and a quasi-physical value, our work is more focussed on comparing and contrasting the responses to AOA for low and high emissions to regional and seasonal AOA. Regards units we have followed the convention used by Zeebe and Wolf-Gladrow (2005) and followed Keller et al (2014) and to ensure consistency with previous work.

*L218-221: Fascinating, but why was the response different?*

This is now addressed in the discussion

*L232: "at" is missing*

Addressed

*L239: I think 'an overall' is missing before 525 ppm in the brackets*

Added

*L241: could you also give this as a % similar to how you did for RCP8.5*

This is a good comment – however. This doesn't really make sense as the atmospheric value at end is less than at the beginning.

*L251-254: This is really important...why was there an increase in export?... The 1% is an important outcome because it is the 'efficiency reduction' on the overall engineering system design.*

Firstly, this is a really small number (<1%) and as stated occur in the Arabian Sea. In section 3.2.3 it now reads:

*The very small changes in export production in RCP2.6 were located in the Arabian Sea (not shown), likely driven by enhanced mixing in this region.*

*Could tables 1-3 be summarised in one table? I think it would make things a little clearer.*

This is a good and we have now combined these three into a single table

*Line 331: could be explained more clearly rather than just "due to the Revelle factor" (see previous comment)*

We have now rewritten this section to be clearer, it now states:

*In the 2020-2100 period, AOA under RCP2.6 led to much larger increases in surface pH and aragonite saturation state, more than 1.3 times, and more than 1.7 times that of RCP8.5 respectively (Table 4). These changes reflect the differences in the mean state associated with high and low emissions, specifically the difference between Alkalinity and Dissolved Inorganic Carbon (ALK-DIC), a proxy for ocean acidification (Lovenduski et al, 2015). As the values of DIC in the upper ocean are larger under RCP8.5 than RCP2.6, the difference between ALK and DIC (ALK-DIC) is smaller and the chemical buffering capacity of $CO_2$ or Revelle Factor (Revelle and Suess, 1957) is less. This means that, for a given addition of ALK the increase in the upper ocean DIC will always be greater under RCP8.5 due to its reduced buffering capacity. Consequently, the changes in ALK-DIC with AOA are greater under RCP2.6 than RCP8.5, which translates to greater increases in pH and aragonite saturation state.*

*L374: 'This reflects the fact that' should be rewritten 'This is caused by the subduction processes..' or something similar.*

We have now added *through subduction* to this statement

*L396: 'Quite low', how low?*

We have now removed *Quite*

*L445-447: But could you speculate as you have in the previous sentence? How much would export have to change to make a material difference?*

We would not like to speculate to do this as many of the processes are not well understood, instead we reference Matear and Lenton (2014) for a discussion of these processes and feedbacks.

*L507: This doesn't quite ring with your abstract, which suggests that ocean acidification would be mitigated. Would it not partially ameliorate the impacts?*

The last line of the abstract refers to low emissions, so they are consistent. Yes, it would ameliorate some of the impacts under RCP8.5, as shown, but its impacts would much less than under RCP2.6.

*L599: 'Interestingly' is used a bit too often, it gets a bit jarring.*

Removed

*Figures:*

*All Figures are clear. Slightly too many for this type of manuscript. Could some be moved to the supporting information? Figures 11-12 are not referred to in the text.*

We feel all figures are warranted, and we have added references to figures in the text.

*Reviewer 2: Major comments:*

*Major comment 1: The changes in the land carbon uptake (table 2) in the AOA simulations based on the RCP2.6 are around 4 times higher than those of the simulations based on the RCP8.5. This is an important aspect because the variations in these carbon fluxes determine the final state of the climate. That is why I think that these results should be discussed properly and the cause of this differential behaviour should be explained.*

These differences are due to 2 main factors: (i) the temperature differences between RCP2.6 and RCP8.5. The mean SAT cooling over land under RCP2.6 is much larger (2x) this means that the decrease in carbon uptake would be larger RCP2.6 than RCP8.5; and (ii) as seen Zhang et al (2014a), the climate sensitivity of the land carbon-climate feedback under higher emissions is lower than other models' due to nutrient (N&P) limitation. This sensitivity in part explains why the response of the land carbon cycle is about half that reported in Keller et al (2014).

The text now states:

*…On the land, in the RCP8.5 simulation there was a smaller reduction in carbon uptake than in RCP2.6 (Table 1), due to larger decreases in surface air temperature (SAT) over land in RCP2.6 than RCP8.5 (~2x; see Section 3.1.2). The land carbon cycle response was also smaller under high than low emissions due to nutrient limitation being reached, thereby limiting the effect of $CO_2$ fertilization (Zhang et al, 2014a).*

*Major comment 2: The statements given between the line 285 and 289 are really confusing. On the one hand, it reads as the temperature change in the RCP8.5 experiment is higher than the one associated with the RCP2.6, which is not what I see in the numbers. And on the other hand, making reference to "potentially reflecting feedbacks" in order to explain this cooling signal does not help to understand the signal. Instead, it confuses the reader. Please explain properly how these feedbacks affect the results.*

This was a mistake and has now been corrected. We have now clarified the text and removed the reference to feedbacks which was not correct, please see the comment below showing that the disparate responses are primarily due to differences in atmospheric $CO_2$ growth rate, please the response to Major Comment 3 (next) for more detail.

*Major comment 3: The reduction in atmospheric CO2 concentrations by 2100 associated with the AOA scenarios under RCP8.5 emissions (app. 84 ppm) is higher than the one associated with the AOA scenarios conducted under the RCP2.6 (app. 40 ppm). Yet, the mitigated warming in the AOA simulations under RCP2.6 is higher than those conducted under the RCP8.5. This is one of the main findings of this publication, however, there is not any discussion/explanation of this result. Only stating what the model delivers is not enough, since it could be a model artefact, the signal might not be caused by AOA, etc. The RCP8.5 and 2.6 scenarios have atmospheres with quite different levels of CO2, which might lead to differences in the CO2-forcing response to changes in CO2 levels. Not only that, but also the RCP8.5 and 2.6 scenarios differ in the assumed land use and the sea ice extent by the end of this century. This might also cause changes in albedo and therefore in the cooling response due to changes in forcing.*

There are a number of mechanisms that may explain the differential response of the cooling which is larger under RCP2.6 than RCP8.5, these including the $CO_2$ vs outgoing long wave radiation (OLR) log relationship, and land and ocean albedo changes. We find that while these may play a minor role, the major driver of these differences are due to differences in atmospheric $CO_2$ growth rate between RCP2.6 and RCP8.5.

We have now added the following statement to the text:

*…In the period 2081-2100 we see larger mean changes in SAT under RCP2.6 than RCP8.5 primarily due to differences in atmospheric $CO_2$ growth rate. Krasting et al. (2014) showed that the slower rate of emissions, the lower the radiative forcing response. This occurs in response to the timescales associated with the uptake of heat and carbon. Consequently, under RCP8.5 the atmospheric $CO_2$ growth rate is much faster than RCP2.6, leading to a strong radiative forcing response. This explains why, despite a larger reduction in atmospheric $CO_2$ concentration under RCP8.5, the biggest reduction in global mean SAT occur under RCP2.6…*

*Ref:*

*Krasting, J. P., Dunne, J. P., Shevliakova, E., and Stouffer, R. J.: Trajectory sensitivity of the transient climate response to cumulative carbon emissions, Geophys Res Lett, 41, 2520-2527, 10.1002/2013gl059141, 2014.*

*Major comment 4: Between the lines 325 and 334 an explanation to the differential pH and aragonite saturation state responses between simulations is given. This explanation seems confusing and it refers to the other main finding of this publication. Because of this I think that it requires some supporting figures (which could be added into the supplementary information) and some extra work in order to clarify the message. I suggest to look at the buffer factors and the effects of AOA under the two different DIC/ALK regimes associated with the RCP8.5 and 2.6 scenarios. More information can be found in the paper by Egleston et.al. (2010) (http://onlinelibrary.wiley.com/doi/10.1029/2008GB003407/abstract).*

We have rewritten this paragraph to better capture our message make it more accessible. It now states:

*In the 2020-2100 period, AOA under RCP2.6 led to much larger increases in surface pH and aragonite saturation state, more than 1.3 times, and more than 1.7 times that of RCP8.5 respectively (Table 4). These changes reflect the differences in the mean state associated with high and low emissions, specifically the difference between Alkalinity and Dissolved Inorganic Carbon (ALK-DIC), a proxy for ocean acidification (Lovenduski et al, 2015). As the values of DIC in the upper ocean are larger under RCP8.5 than RCP2.6, the difference between ALK and DIC (ALK-DIC) is smaller and the chemical buffering capacity of* $CO_2$ *or Revelle Factor (Revelle and Suess, 1957) is less. This means that, for a given addition of ALK the increase in the upper ocean DIC will always be greater under RCP8.5 due to its reduced buffering capacity. Consequently, the changes in ALK-DIC with AOA are greater under RCP2.6 than RCP8.5, which translates to greater increases in pH and aragonite saturation state.*

*Minor comments:*

*L16, L27 and L561: "is capable of" gives the impression that AOA has not real big limitations to be implemented which is not the case, please modify the wording*

This wording is correct, AOA is capable and is analogous to alkalinity addition that occurs over geological timescales and this has been hypothesised to play a role in glacial-interglacial timescales.

*L18, L19: there are acronyms which the reader might have never seen in the abstract, please spell them out or remove*

We have now spelt out $CO_2$ and RCP

*L25: "lower" and "higher" emissions than what? I think that you meant "low" and "high"*

This is correct, we wanted to be more generic than just RCP 2.6 and 8.5, particularly as Shared Socioeconomic Pathways (SSPs) will be used CMIP6.

*L26: our simulations show that AOA during the period ... ; in any case I do not think that this very last sentence in the abstract is needed*

With respect, we think that this is an important statement to make.

*L46: ... could help to ...*

Corrected

*L53-54: CO2 that enters the ocean does not react with seawater to reduce the carbon- ate ion concentration, please reconsider this statement and use correct grammar*

This has now been changed to say:

*As $CO_2$ is taken up by the ocean it changes its chemical equilibrium, reducing the carbonate ion concentration and decreasing pH, collectively known as ocean acidification.*

*L59: ...changes in calcification...*

Corrected

*L60: are you sure that ocean acidification alters nutrient availability*

Yes e.g. Shi et al (2010)

Shi et al (2010) Effect of Ocean Acidification on Iron Availability to Marine Phytoplankton, *Science* 327, 676 (2010);

L62: please change order of publications

They are already in name order

L69: semicolon needed?

Removed

L77: weathering of minerals play a crucial role in modulating the state of the climate in geological timescales, please write an assertive statement

Removed *may*

L91: reviewed

This should be present tense, I believe

L92-95: way too long sentence, please simplify and split it

This has now been split

L98: Did Kohler used one or several models?

Changed to singular

L110: ocean only without the hyphen

Corrected

L110: and they showed

Corrected

L111: high CO2 emission

Corrected

L114: also concluded that

This should be present tense, I believe

L115, L118, L131, ...: impacts of

Changed

L124: from a high

Changed

L126: it would be required

Changed

L127: and it would come

Changed

L134: 78 ppm between brackets might look better

Changed

L134: a net atmospheric cooling

Changed to *surface air temperature*

L139: "to be very large" - (very) large in what respect? please clarify

The sentence now reads: *Capturing these feedbacks is critical as they have the potential to significantly increase atmospheric* $CO_2$ *concentrations (Jones et al., 2016).*

L141: currently assume (instead of "utilize")

Changed

L141 to L145: way too long sentence, please simplify and split it

Changed to: *Furthermore, the feasibility of these approaches which are increasingly questioned due in part to limited land (Smith et al., 2016), whereas the potential CDR capacity of the oceans is orders of magnitude greater (Scott et al., 2015).*

L149: and surface warming

L149: questions

L147 to L152: I think that the novelty of this study could be better emphasise. In any case, this last paragraph is crucial and therefore it should be improved since it does not read well.

The above three comments have been addressed in response to Reviewer 1 and the paragraph now reads:

*In this work, we use a fully coupled ESM (CSIRO-Mk3L-COAL), which includes climate and carbon feedbacks, to investigate the impact of AOA on the carbon cycle, global surface warming (2m surface air temperature), and ocean acidification response to the global and regional AOA experiments under the high (RCP8.5) and low (RCP2.6) emissions scenarios.*

L158: extra dot after citations?

Changed

L160, L163, ...: please remove the brackets in those citations which are subjects of the sentences, this occurs several times in this manuscript

Changed

L164 to L166: does this sentences really add any relevant information? Such a feature of the model is basic to conduct this study

Yes, we agree but it does provide confidence in the tool we are employing in this study – as seen by Review 1's comments

L171 to L172: the land carbon cycle currently has too many uncertainties to state something in such an assertive manner, please consider to modify this or even remove it

We have removed *realistically*

L185: from 2006 onwards, ...

Corrected

L186: corresponding to the Representative ...

Corrected

L218: Subpolar addition

Corrected

L232: is seen in 2100

Corrected

L233 to L235: if you mention this feature of the modelling tool, please explain the associated consequences for the simulations of AOA

We have discussed these results in the manuscript and identify why the sensitivity of the land carbon uptake, particularly under high emissions is less than other studies.

L242 and L256: "more than compensates" and "more than offset" are really confusing ways of describing the obtained values, please clarify

We have removed *more than* in both of these instances

L247: 50% instead of 1.5 maybe?

Corrected

L253: total ocean uptake ...

Corrected

L254 to L258: this explanation reads really confusing, please clarify

This has now been changed to say:

*The simulated cooling drove both a reduced net primary production, leading to reduced carbon uptake, and an increase in carbon retention associated with a reduction in heterotrophic respiration. However, overall, the net decrease in land carbon uptake means that in the response to AOA globally the reduced net primary production dominated.*

L266: addition studies such as Ilyina ... which demonstrated ...

Corrected

L270: 181 PgC is in ... (instead of was)

Corrected

L277: I think the authors meant "positive denotes enhanced uptake" (instead of "negative")

Yes – this is corrected thank you

L288: "large" twice in the sentence

Corrected

L294: "projected" instead of "anticipated"

Corrected

L295: why is this publication here cited?

Removed

L298: standard deviations with respect to what? what is this (1 - sigma)? Please clarify

The caption has now been improved, and now reads:

*Table 1 The differences in global mean surface air temperature in the period 2081-2100 (2090) and associated standard deviation (1-σ) (K; SAT; 2m) for the four different AOA experiments for each emission scenario, relative to the same emission scenario with no AOA.*

. L302 to L304: please consider to reformulate these sentences since "variability" might refer to many different things (e.g. inter annual, inter model, model internal, ...). In any case I think that "variability" is not really the term to use since what is described here are differences between simulations.

The sentence now reads:

*Within each of the scenarios, there are some differences in the magnitude of the cooling within the four different AOA experiments; however, these are smaller than the interannual variability over the last two decades of the simulations.*

L308: mean surface cooling

Corrected

L318: What is the point of this statement and citation? The pH and aragonite saturation state correlate really well as I can see in the figures.

Yes but the impacts are different and this motivates why we are interested looking at both aragonite saturation state and pH.

L321: despite the return

Corrected

L344 and L377: the citation here to Groeskamp et.al. seems unfounded

Removed

L348 and L349: Please elaborate on this so that the reader understand the context, e.g. discuss how this change in pH might (or not) matter, ...

We have modified this sentence to now say:

*To put these changes into context, the estimated decrease in pH since the preindustrial period is 0.1 units (Raven et al., 2005), and is responsible for already detectable changes in the marine environment (Albright et al., 2016).*

L380: How can one of the experiments (AOA_ST) reflect the timescales of the circulation of the subtropical gyres? Please explain this.

This sentence now reads:

*In the case of AOA_ST, this reflects the timescales associated with the longer residence time of upper ocean waters in the subtropical gyres.*

L382: ice covered (instead of "non-ice-free")

Changed

L386: by 2100 (instead of "in")

Corrected

L387 and L388: please clarify this, is not understandable

This is now clarified to say:

*Specifically, for AOA_G we see 31% remains in the upper ocean and for AOA_T and AOA_ST: 34%, while for AOA_SP: 22-24% remains in the upper ocean which (as anticipated) is lower than in other regions.*

L404: ...seen in the...

Corrected

L421 to L425: please work on the grammar of these sentences

Corrected

L442: ... in the ratio ...

Corrected

L444: Dot missing

Added

L445: ... remain poorly...

Corrected

L457 to L459: why do you obtain this result?

We have added: *likely driven by enhanced mixing in this region.*

L463: remove (SAT)

Removed

L468 to L470: why do you obtain this result?

This very much reflects the period over which the mean changes were calculated, and the simulated large variability in SAT in this region, which now stated in the text.

L538 to L554: why no figures are shown in this section on seasonality to support this discussion? Also, only AOA is implemented in the summer season under RCP8.5 emissions, which does not seem to me enough to explore the effects of seasonality.

This is a little confusing as the Reviewer wishes us to remove figures and now requests more. We don’t present results here, as it is a sensitivity experiment, rather than a major result.

L560: please remove (COP21)

Removed

L593 to L595: What do you mean? Please clarify this.

It now states:

*However, AOA under the RCP2.6 emissions scenario changes the roles played by the ocean and land in carbon uptake as compared with the scenario of RCP2.6 with no AOA, resulting in a reduced uptake in the terrestrial biosphere and increased uptake in the ocean.*

L605: double "that"

Corrected

L620: mention "preindustrial period" and remove (1850) reads better

Changed

L621: cases (subject?) leads ...

Corrected

L633: for the role

Corrected

L638: ...therefore it needs ...

Added

L642 and L645: Earth system (instead of earth system)

Corrected

L649: please put "e.g. mesocosm experiments" between brackets

Added

Please keep an eye on the format in which the references are given and be consistent with it.

Done

*Comments Reviewer 3*

*The Lenton et al., study investigates the impacts from adding artificial alkalinity to the oceans using the model CSIRO under 2 different emission pathways - RCP2.6 and RCP8.5. It was a really well done and interesting study to read technically, however my main comment is that the way the paper is currently structured makes it confusing to read. For example, each paragraph jumps back and forth between RCP26 and RCP85 making the story line hard to follow. I suggest setting up the story for one of the emissions pathways and then comparing to that one for the other pathway. It would also be useful to set up the chemistry in a little more detail or reiterate the paragraph in the intro. This would be useful when explaining why adding alk under a 2.6 scenario is more effective.*

At this stage, we do not feel that major rewrite or reordering of the paper is warranted. It is clear from other studies is that AOA will reduce OA and global warming; what is more interesting is whether the response to the same amount of AOA differs between emissions scenarios. This is the main focus of the study hence it does not make sense to restructure the paper as suggested. Instead we have gone through the paper to ensure that it is clearer and easier to follow.

*It would also be useful to set up the chemistry in a little more detail or reiterate the paragraph in the intro. This would be useful when explaining why adding alk under a 2.6 scenario is more effective.*

Please see the response to Reviewer 2.

*Lastly, section 3.1 was confusing (you may want to expand on the methods section to make this section clearer). For each run you added 0.25Pmol/yr of alkalinity but then I read in ln216 that the magnitude of the increase in alkalinity is dependent on where it was added. Is the 0.25Pmol/yr added to all the boxes? or is it divided up between the boxes for a total of 0.25Pmol/yr? Can you put everything in the same units to be constant?*

We apologise for any ambiguity and have now clarified this section, it now states:

*For each emissions scenario, we simulated four different AOA experiments, which all had the same 0.25 Pmol/yr of alkalinity added. In the case of the regional experiments the per surface values were larger than the case of global addition.*

*Minor comments: ln50: "including through coral bleaching" - not clear what this means*

We have now removed this statement.

*ln79-80: This sentence seems out of place.*

We have now removed this sentence.

*ln149: what do you mean by impact?*

We have now been more explicit and the sentence now says:

*In this work, we use a fully coupled ESM (CSIRO-Mk3L-COAL), which includes climate and carbon feedbacks, to investigate the impact of AOA on the carbon cycle, global surface warming (2m surface air temperature), and ocean acidification response to the global and regional AOA experiments under the high (RCP8.5) and low (RCP2.6) emissions scenarios.*

*ln158: extra period between feedbacks and references*

Corrected

*ln230: the first sentence does not make sense.*

Rewritten it now states:

*The large atmospheric $CO_2$ concentration at 2100 under RCP8.5 reflects the large projected increase in emissions during this century, while under RCP2.6 a similar atmospheric concentration of $CO_2$ is seen in 2100 as at the beginning of the simulation (2020) (Figure 2a).*

*ln250: why is there a difference in export?*

The text now reads:

…Consistent with Keller et al. (2014) and Hauck et al. (2016) the simulated changes in ocean export production were very small (~0.2 PgC) under RCP8.5 and due to small changes in ocean state, e.g. stratification. Under RCP2.6, it was slightly larger at 1.2 PgC, but still less than 1% percent of the total ocean uptake increase simulated under AOA, due to small changes in ocean state in a more stratified ocean…

*Section 3.1.2: I don't understand how soil-moisture feedbacks are influencing temperature in this case. Temperature is more variable on land than over the ocean, could it be simply that?*

We apologise for the confusion we have removed this section and attribute these changes to the differences different in atmospheric CO2 growth rate.

The section now states:

*…In the period 2081-2100 we see larger mean changes in SAT under RCP2.6 than RCP8.5 primarily due to differences in atmospheric $CO_2$ growth rate. Krasting et al. (2014) showed that the slower rate of emissions, the lower the radiative forcing response. This occurs in response to the timescales associated with the uptake of heat and carbon. Consequently, under RCP8.5 the atmospheric $CO_2$ growth rate is much faster than RCP2.6, leading to a strong radiative forcing response. This explains why, despite a larger reduction in atmospheric $CO_2$ concentration under RCP8.5, the biggest reduction in global mean SAT occur under RCP2.6…*

*Ref:*

*Krasting, J. P., Dunne, J. P., Shevliakova, E., and Stouffer, R. J.: Trajectory sensitivity of the transient climate response to cumulative carbon emissions, Geophys Res Lett, 41, 2520-2527, 10.1002/2013gl059141, 2014.*

*Table4: relative to what?*

It now reads:

*Table 4 The differences in surface value of aragonite saturation state and pH between the AOA experiments for each emission scenarios in 2100 relative to the emissions scenario with no AOA.*

*Figures: relative to what?*

We have added text to each of the captions to clarify

*Figures: There are a lot of figures which I'm not sure add much to main text. You could simply the story in the text and only look at a few and toss the rest into the supplement. Or could you combine the 2.6 and 8.5 figures into 1? Could you do a difference between them? That would visually show the reader where the differences between scenarios lie....*

While this seems attractive, we think that there is value in keeping these figures. Furthermore, we do not see a simply way of combining these into 8 panel figures, nor does doing the differences make much sense, as differences of differences is quite confusing.

*Section 3.2.5: I think the figures referenced are not the correct figures. Plus I think a description of figures 11 and 12 are missing.*

Thank you for this – we have now ensured that the figures are referenced correctly, and switched the order to better reflect the order they are appear in the text.

*I see that the previous reviewers picked up a bunch of typos*

Corrected.

# Assessing Carbon Dioxide Removal Through Global and Regional Ocean Alkalization under High and Low Emission Pathways.

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## 1. Abstract

Atmospheric Carbon Dioxide ($CO_2$) levels continue to rise, increasing the risk of severe impacts on the Earth system, and on the ecosystem services that it provides. Artificial Ocean Alkalization (AOA) is capable of reducing atmospheric $CO_2$ concentrations~~,~~ and surface warming and addressing ocean acidification. Here, we simulate global and regional responses to alkalinity (ALK) addition (0.25 ~~PmolAlk~~PmolALK/year) over the period 2020-2100 using the CSIRO-Mk3L-COAL Earth System Model~~ in the period 2020-2100~~, under high (Representative Concentration Pathway 8.5; RCP8.5) and low (RCP2.6) emissions. While regionally there are large changes in alkalinity associated with locations of AOA, globally we see only a very weak dependence on where and when AOA is applied. ~~We~~Globally, while we see that under RCP2.6~~, while~~ the carbon uptake associated with AOA is only ~60% of the total under RCP8.5, the relative changes in temperature are larger, as are the changes in pH (~~1.4x)~~140%) and aragonite saturation ~~(1.7x).~~state (170%). The ~~results of this modelling study are significant as they demonstrate that~~simulations reveal AOA is more effective under lower emissions, ~~and~~therefore the higher the emissions the more AOA is required to achieve the same reduction in global warming and ocean acidification. ~~-~~Finally, our ~~simulations show~~simulated AOA ~~in the period~~for 2020-2100 in the RCP2.6 scenario is capable of offsetting ~~global~~ warming and ameliorating ocean acidification increases ~~due to low emissions~~at the global scale, but ~~regionally the response is more~~with highly variable~~.~~ regional responses.

## 1. Introduction

Atmospheric carbon dioxide ($CO_2$) levels continue to rise ~~primarily~~ as a result of human activities. Recent studies have suggested that even deep cuts in emissions may not be sufficient to avoid severe impacts on the Earth system, and the ecosystem services that it provides (Gasser et al., 2015). Recent international negotiations (UNFCCC, 2015) agreed to limit global warming to well below 2~~°.~~ °C. The application of Carbon Dioxide Removal (CDR), sometimes referred to as “Negative Emissions”, appears to be required to achieve this goal, as emission reductions alone are likely to be insufficient (Rogelj et al., 2016). In this context, there is an urgent need to assess how ~~Carbon Dioxide Removal (CDR)~~CDR could help either mitigate climate change or even reverse it, and to understand the potential risks and benefits of different options.

While warming represents ~~a major~~an imminent global threat~~, including through coral bleaching~~ which is already significantly impacting the natural environment (Hughes et al., 2017), ocean acidification poses an additional and equally significant threat to the marine environment. ~~Ocean acidification occurs as $CO_2$ taken up by the ocean reacts with the seawater to reduce the carbonate ion concentration and decrease the pH. Annually~~At present the oceans take up about 28% of anthropogenic $CO_2$ emitted annually (Le Quéré et al., 2015). As $CO_2$ is taken up by the ocean it changes its chemical equilibrium, reducing the carbonate ion concentration and decreasing pH, collectively known as ocean acidification. Furthermore, as the ocean continues to take up carbon the buffering capacity or Revelle Factor (Revelle and Suess, 1957) of the seawater decreases, thereby accelerating the rate of ocean acidification.

Ocean acidification is the unavoidable consequence of rising atmospheric $CO_2$ levels and will impact the entire marine ecosystem ~~–~~– from plankton at the base~~,~~ through to ~~fish~~higher-trophic species at the top. Potential impacts include changes ~~to~~in calcification, fecundity, organism growth and physiology, species composition and distributions, food web structure and nutrient availability (Doney et al., 2012~~;Dore et al., 2009;Fabry et al., 2008;Iglesias-Rodriguez et al., 2008;Munday et al., 2010;Munday et al., 2009)~~; Fabry et al., 2008; Iglesias-Rodriguez et al., 2008; Munday et al., 2010; Munday et al., 2009). Within this century, the impacts of ocean acidification will increase in proportion to emissions (Gattuso et al., 2015).

Furthermore, these changes will be long~~-~~-lasting, persisting for centuries or longer even if emissions ~~were~~are halted (Frolicher and Joos, 2010).

To date, many different CDR techniques have been proposed~~ both on the land and in the ocean~~ (Royal Society, 2009; National Research Council, 2015). Their primary purpose is to reduce atmospheric $CO_2$ levels~~;~~, and thus most CDR methods will ~~ameliorate~~also reduce the impacts of ocean acidification~~,~~ although some proposed techniques such as ocean pipes (Lovelock and Rapley, 2007) and micro-nutrient addition (Keller et al., 2014) may actually lead to ~~an~~a regional acceleration of ocean acidification in surface waters.

Artificial Ocean Alkalization (AOA~~)~~), through altering the chemistry of seawater, both enhances ocean carbon uptake~~,~~ (thereby reducing atmospheric $CO_2$~~,~~), while at the same time ~~directly~~ reversing ocean acidification~~.~~ and increasing the buffering capacity of the ocean. AOA can be thought of as a massive acceleration of the natural processes of chemical weathering of minerals that ~~may~~ have played a role in modulating the climate on geological timescales (Zeebe, 2012~~;~~; Colbourn et al., 2015~~)~~; Sigman and Boyle, 2000)~~. Alkalinity changes may also have played an important role in controlling glacial-interglacial cycles of atmospheric $CO_2$ e.g. Sigman and Boyle (2000).~~.

Specifically, as alkalinity enters the ocean, the pH increases leading to an elevated carbonate ion concentration, a reduction in the hydrogen ion concentration and a decrease in the concentration of aqueous $CO_2$ (or $pCO_2$). ~~-~~This in turn enhances the disequilibrium of $CO_2$ between the ocean and atmosphere (or $\Delta pCO_2 = pCO_2^{ocean}$ - ~~$pCO_2^{atmosphere}$~~$pCO_2^{atmosphere}$) leading to increased ocean carbon uptake, and a reduction in the atmospheric $CO_2$ concentration. These increases in pH and carbonate ion concentration thus reverse the ocean acidification due to uptake of anthropogenic $CO_2$.

Kheshgi (1995) first proposed AOA as a method of CDR. Renforth and Henderson (2017) review the early experimental, engineering and modelling work undertaken to investigate AOA. From the observational perspective, we draw particular attention to the experimental work of Albright et al. (2016) which provided an in situ demonstration of localised AOA to offset the observed changes in ocean acidification on the Great Barrier Reef that have occurred since the ~~pre-industrial~~preindustrial period.

Several modelling studies have explored the impacts of AOA both on carbon sequestration and ocean acidification. Using ocean~~-~~-only biogeochemical models, Kohler et al. (2013) explored AOA via olivine addition. Olivine, in addition to increasing alkalinity also adds iron and silicic acid, both of which can enhance ocean productivity (Jickells et al., 2005~~;~~; Ragueneau et al., 2000). Kohler et al. (2013) estimated the response of atmospheric $CO_2$ levels and pH to different levels of olivine addition over the period 2000-2010, and ~~later~~ this was later extended to 2100 by Hauck et al. (2016). These studies demonstrate a global impact~~,~~ that ~~appeared~~appears to scale with the amount of olivine added. Importantly, Kohler et al. (2013) showed that the global effect of alkalinity added along shipping routes (as an analogue for practical implementation) was not significantly different from that of alkalinity added in a highly idealized uniform manner.

Ilyina et al. (2013) explored the potential of AOA to mitigate rising atmospheric $CO_2$ levels and ocean acidification in ocean-only biogeochemical simulations, and they showed that AOA has the potential to ameliorate future changes due to high $CO_2$ emissions. They did not limit the amount of AOA, as their goal was to offset the projected future changes~~;~~, and showed that the amount of AOA required to do this would drive the carbonate system to levels well above ~~pre-industrial~~preindustrial levels. Ilyina et al. (2013) also conclude that local AOA could potentially be used to offset the ~~impact~~impacts of ocean acidification, with enhanced $CO_2$ uptake being only a side benefit. This regional approach was explored further by Feng et al. (2016) who suggested that local AOA in the tropical ocean, in areas of high coral calcification, has the potential to offset the ~~impact~~impacts of future rising atmospheric $CO_2$ levels under a high emissions scenario (RCP8.5). This study also revealed strong regional sensitivities in the response of ocean acidification related to the locations in which it was applied.

~~To date several~~Several other studies ~~estimate~~have estimated the response of the Earth system to AOA. Gonzalez and Ilyina (2016) used an Earth System Model (ESM) to estimate the AOA required to reduce atmospheric concentrations from a high emissions scenario (RCP8.5) to the medium emissions scenario (RCP4.5). They estimated that to mitigate the associated 1.5K warming difference, via reducing atmospheric $CO_2$ concentrations by ~400 ppm, ~~would require~~ an addition of 114 Pmol of alkalinity (between 2018-2100~~),~~) would be required, and it would come at the cost of very large (unprecedented) changes in ocean chemistry.

Keller et al. (2014) used an Earth System Model of Intermediate Complexity (EMIC) to explore the ~~impact~~impacts of AOA over the period 2020-2100~~, to~~ arising from a globally uniform addition of alkalinity (0.25 PmolALK/yr), an amount based on the estimated carrying capacity of global shipping following Kohler et al. (2013). Keller et al. (2014) showed that AOA led to a reduction in atmospheric $CO_2$ of 166 PgC (or ~~~78ppm,~~78 ppm), a net surface air temperature cooling of 0.26K and a global increase in ocean pH of 0.06 in the period 2020-2100.

To date, not all modelling studies have been emissions driven, and this is important as potential climate and carbon cycle feedbacks may not have been accounted for. Capturing these feedbacks is critical as they have the potential to ~~be very large~~significantly increase atmospheric $CO_2$ concentrations (Jones et al., 2016). Further, no studies have explored the impact of AOA under low emissions scenarios such as RCP2.6. This is important because scenarios that limit warming to 2° C or less, currently ~~utilize~~assume considerable land~~-~~-based CDR via afforestation and/or ~~Bio-~~Biomass Energy with Carbon Capture and Storage (BECCS~~)~~). Furthermore, the feasibility of ~~which are~~these approaches is increasingly questioned due in part to limited land (Smith et al., 2016), whereas the potential CDR capacity of the oceans is orders of magnitude greater (Scott et al., 2015).

In this work, ~~using~~we use a fully coupled ~~Earth System Model~~ESM (CSIRO-Mk3L-COAL), which includes climate and carbon feedbacks, ~~we~~to investigate the impact of AOA on the carbon cycle, global surface warming (2m surface air temperature), and the ocean acidification~~, land and ocean carbon uptake and warming. Specifically, the question this study tackles is: What is the impact of global~~ response to the global and regional AOA ~~on the Earth System, and how efficient it is at mitigating global warming and ocean acidification~~experiments under the high (RCP8.5) and low (RCP2.6) emissions ~~trajectories?~~scenarios.

## 2. Methods

### *~~2.1~~ 2.1 Model Description*

The model simulations were performed using the CSIRO-Mk3L-COAL (Carbon, Ocean, Atmosphere, Land) ~~Earth System Model~~ESM which includes climate-carbon interactions and feedbacks~~.~~ (Matear and Lenton, 2014~~;~~; Zhang et al., 2014a). The ocean component of the ~~Earth System Model~~ESM has a resolution of 2.8° by 1.6° with 21 vertical levels. The ocean biogeochemistry is based on ~~(~~Lenton and Matear~~,~~ (2007~~;~~) and Matear and Hirst~~,~~ (2003) simulating the distributions of phosphate, oxygen, dissolved inorganic carbon and alkalinity in the ocean. The model simulates particulate inorganic carbon (PIC) production as a function of particulate organic carbon (POC) production via the rain ratio (9%) following ~~(~~Yamanaka and Tajika~~,~~ (1996). This ocean biogeochemical model was shown to simulate the observed distributions of total carbon and alkalinity in the ocean (Matear and Lenton, 2014) and phosphate (Duteil et al., 2012).

The atmosphere resolution is 5.6° x 3.2° with 18 vertical layers. The land surface scheme uses CABLE (Best et al., 2015) coupled to CASA-CNP (Wang et al., 2010~~;~~; Mao et al., 2011) which simulates biogeochemical cycles of carbon, nitrogen and phosphorus in plants and soils. The response of the land carbon cycle was shown to~~ realistically~~ simulate the observed biogeochemical fluxes and pools on the land surface (Wang et al., 2010).

To quantify the changes in ocean acidification, we calculate pH changes on the total scale following the recommendation of Riebesell et al. (2010). To calculate the changes of carbonate saturation state, we use the equation of Mucci (1983).

### *2.2 Model Experimental Design*

Our ESM was spun-up under a preindustrial atmospheric $CO_2$ concentration of 284.7 ppm, until the simulated climate was stable (> 2000 years) (Phipps et al., 2012). From the spun-up initial climate state, the historical simulation (1850 - 2005) was performed using the historical atmospheric $CO_2$ concentrations as prescribed by the CMIP5 simulation protocol (Taylor et al., 2012).

Following the historical concentration pathway from 2006 onward, two different future projections to 2100 were made using the atmospheric $CO_2$ emissions corresponding to the Representative Concentration Pathways of low emissions (RCP2.6) and high emissions (RCP8.5 or 'business as usual') (Taylor et al., 2012). All simulations include the forcing due to non-$CO_2$ greenhouse gas concentrations (Taylor et al., 2012). We define RCP8.5 and RCP2.6 as our control cases for the corresponding experiments below.

In the period 2020-2100, we undertook a number of AOA experiments using a fixed quantity of 0.25 Pmol/yr of alkalinity, ~~the same~~a similar amount used by Keller et al. (2014). Consistent with this study, we applied AOA in the surface ocean all year-round in ice-free regions, set to be between 60°S and 70°N~~.~~ (note that this ignores the presence of seasonal sea-ice in some small regions). For each of the two emissions scenarios, we considered four different regional applications of AOA, shown in Figure 1. These are: (i) AOA globally (AOA_G) between ~~60S~~60°S and ~~70N~~70°N; (ii) the higher latitudes comprising the subpolar northern hemisphere oceans (~~40N -70N~~40-70°N) and the (ice-free) Southern Ocean (~~40S-60S~~40-60°S) (AOA_SP); (iii) the subtropical oceans (15-~~40N~~40°N and ~~15S and 40S;~~ 15-40°S) (AOA_ST); and (iv) in the equatorial regions (~~15N-15S;~~ 15°N-15°S) (AOA_T). In this study, we only look at the response of the Earth system to alkalinity injection. We do not consider the biogeochemical response to other minerals and elements that can be associated with the ~~proposed~~ sourcing of alkalinity from the application of finely ground ultra-mafic rocks such as olivine and ~~fosterite~~forsterite, nor dissolution processes required to increase alkalinity (e.g. Montserrat et al~~. (~~., 2017).

## 3. Results and Discussion

To aid in presenting our results and to compare these with previous studies, we first discuss the carbon cycle, global surface warming (2m surface air temperature), and ~~response and~~ ocean acidification response to the ~~4~~four different AOA experiments under the high (RCP8.5) and low (RCP2.6) emissions scenarios. We then look at the regional behaviour of the simulations in the different AOA experiments.

### *3.1 Global Response*

For each ~~emission~~emissions scenario, we simulated four different AOA experiments, which all had the same 0.25 Pmol/yr of alkalinity added. In the case of the regional experiments the per surface values were larger than the case of global addition. As anticipated, by 2100~~,~~ AOA increased the global mean surface ocean alkalinity relative to the corresponding scenario control case, with the magnitude of the increase in alkalinity being dependent on where it was added (Table 1). ~~Polar~~Sub-polar addition (AOA_SP) led to the smallest net increase in surface alkalinity, while tropical addition (AOA_T) produced the greatest increase. As expected, the global mean changes in surface alkalinity between emissions scenarios are very small (less than 3 µmol/kg difference). The slightly greater increase in surface values in alkalinity under RCP8.5 likely reflects enhanced ocean stratification under higher emissions (Yool et al., 2015).

| | | ~~RCP8.5~~ | ~~RCP2.6~~ | | |
|---|---|---|---|---|---|
| | | **AOA_G~~-RCP~~** | **AOA_SP**~~108.3~~ | **AOA_ST**~~105.1~~ | **AOA_T** |
| *(a) Relative increase in global mean ocean surface alkalinity (µmol/kg) in 2100* | | | | | |
| RCP8.5~~AOA_SP-RCP~~ | | 108.3 | 79.7 | 115.1~~74.4~~ | 129.8 |
| RCP2.6~~AOA_ST-RCP~~ | | ~~115~~105.1 | 74.4 | 112.9 | 127.1 |
| *(b) Total integrated additional carbon uptake (in PgC) in the period 2020-2100* | | | | | |
| RCP8.5 | *Total* | 178.6 | 183.3 | 180.7 | 174.5 |
| | Ocean | 184.4 | 188.1 | 185.1 | 177.2 |
| | Land | -5.8 | -4.8 | -4.4 | -2.7 |
| RCP2.6 | *Total* | 121.1 | 122.1 | 122.0 | 116.0 |
| | Ocean | 143.1 | 145.2 | 143.1 | 139.2 |
| ~~AOA_T-RCP~~ | Land~~129.8~~ | ~~127~~-22.1 | -24.1 | -21.2 | -23.1 |
| *(c) Differences in global mean surface air temperature in the period 2081-2100 (2090) and associated standard deviation (1-σ) (K; SAT; 2m)* | | | | | |
| RCP8.5 | *Total* | -0.16±0.08 | -0.13±0.10 | -0.08±0.05 | -0.14±0.06 |
| | Ocean | -0.14±0.07 | -0.11±0.07 | -0.06+0.03 | -0.12±0.05 |
| | Land | -0.22±0.15 | -0.18±0.20 | -0.13±0.14 | -0.19±0.11 |
| RCP2.6 | *Total* | -0.25±0.08 | -0.23±0.08 | -0.20±0.09 | -0.16±0.06 |
| | Ocean | -0.19±0.05 | -0.18±0.05 | -0.15±0.06 | -0.13±0.05 |
| | Land | -0.39±0.22 | -0.35±0.22 | -0.30±0.20 | -0.24±0.16 |

*Table 1 For the two RCP scenarios, (a) the relative increase in global mean ocean surface alkalinity (µmol/kg) between each AOA experiment and control experiment in 2100. (b) The total integrated additional carbon uptake (in PgC) in the period 2020-2100 in different experiment and emissions scenarios, positive denotes enhanced uptake. (c) The differences in global mean surface air temperature in the period 2081-2100 (2090) and associated standard deviation (1-σ) (K; SAT; 2m) for the four different AOA experiments for each emission scenario, relative to the same emission scenario with no AOA.*

### *3.1.1 Carbon Cycle*

The large atmospheric $CO_2$ concentration at ~~the end of the century~~ 2100 under RCP8.5 reflects the large projected increase in emissions ~~projected (under RCP8.5),~~during this century, while under RCP2.6 a similar atmospheric concentration of $CO_2$ is seen in 2100 as at the beginning of the simulation (2020) (~~Figure2a).~~ Figure 2a). We note that atmospheric $CO_2$ levels in our CSIRO-MK3L-COAL for the control cases are greater than for their respective concentration driven RCPs due to nutrient limitation in the land, leading to reduced carbon uptake (Zhang et al., 2014a)~~.~~.

Under all emissions scenarios and experiments, AOA leads to reduced atmospheric $CO_2$ levels (Figure 2a). Under RCP8.5, AOA reduces atmospheric concentration by 82-86 ppm~~,~~ ~~this represents~~; representing a ~16% decrease in atmospheric concentration ~~(525 ppm increase over the period 2020-2100).~~. In contrast to RCP8.5, AOA under RCP2.6 leads to a smaller reduction in atmospheric concentration (53-58 ppm). Figure 2a shows that, by the end of the century, AOA ~~more than~~ compensates for the projected increase in atmospheric $CO_2$ due to RCP2.6.

Over the 2020-2100 period, the reduction in atmospheric $CO_2$ levels associated with AOA is primarily due to increased ocean carbon uptake, offset by small decreases in the land surface carbon uptake (Table ~~2~~1). In the ocean, RCP8.5 ~~has~~leads to much greater net uptake than RCP2.6, about ~~1.5 times~~50% more, due to the larger (and growing) disequilibrium between the atmosphere and ocean.

In the ocean, the relative increase in carbon uptake in response to AOA is primarily abiotic in nature. Consistent with Keller et al. (2014) and Hauck et al. (2016) the simulated changes in ocean export production were very small (~0.2 PgC) under RCP8.5~~. While under~~ and due to small changes in ocean state, e.g. stratification. Under RCP2.6, it was slightly larger at 1.2 PgC, but still less than 1% percent of the total ocean uptake increase simulated under AOA~~.~~ and due to small changes in ocean state in a more stratified ocean. In contrast, the relative decreases in land carbon uptake were biotic in nature. The simulated cooling ~~over land~~ drove both a ~~reduction in~~reduced net primary production ~~that more than offset the decrease in~~, leading to reduced carbon ~~flux due in~~ uptake, and an increase in carbon retention associated with a reduction in heterotrophic respiration. However, overall, the net decrease in land carbon uptake means that in the response to AOA globally the reduced net primary production dominated. On the land, in the RCP8.5 ~~simulated~~simulation there was a smaller reduction in carbon uptake than in RCP2.6~~, potentially due to a~~ (Table 1), due to larger decreases in surface air temperature (SAT) over land in RCP2.6 than RCP8.5 (~2x; see Section 3.1.2). The land carbon cycle response was also smaller ~~relative cooling (Table 3~~under high than low emissions due to nutrient limitation being reached, thereby limiting the effect of $CO_2$ fertilization (Zhang et al, 2014a).

~~-~~For both emissions scenarios, the ~~4~~four AOA experiments all produced similar reductions in atmospheric $CO_2$ concentrations (Figure 2) with less than a 5% difference in the total land and ocean carbon uptake. The global changes in land and ocean carbon uptake ~~do~~are not ~~appear to be~~ very sensitive to where we add the alkalinity to the surface ocean. This is consistent with Kohler et al. (2013) who saw little difference in adding olivine along existing shipping tracks, versus uniformly adding it to the surface ocean. It is also consistent with regional addition studies of Ilyina et al. (2013)~~ and~~, Feng et al. (2016) ~~who~~and Feng et al (2017) which demonstrated a global impact.

Our simulated total increased carbon uptake under AOA_G with RCP8.5 (179 PgC) is comparable to the 166 PgC reported by ~~(~~Keller et al., (2014). Their cumulative increase in ocean carbon uptake by 2100 of 181 PgC ~~was~~is in very good agreement with our value of 184 PgC. However, they simulated a reduction in land uptake nearly twice the -5.8 PgC reduction in our AOA_G simulation. These differences ~~may~~ reflect both the lower sensitivity of the simulated climate feedbacks in our ESM, and differences in land surface models.

|  | ~~Total RCP8.5~~ | ~~Ocean RCP8.5~~ | ~~Land RCP8.5~~ | ~~Total RCP 2.6~~ | ~~Ocean RCP 2.6~~ | ~~Land RCP 2.6~~ |
|---|---|---|---|---|---|---|
| ~~AOA_G-RCP~~ | ~~178.6~~ | ~~184.4~~ | ~~-5.8~~ | ~~121.1~~ | ~~143.1~~ | ~~-22.1~~ |
| ~~AOA_SP-RCP~~ | ~~183.3~~ | ~~188.1~~ | ~~-4.8~~ | ~~122.1~~ | ~~145.2~~ | ~~-24.1~~ |
| ~~AOA_ST-RCP~~ | ~~180.7~~ | ~~185.1~~ | ~~-4.4~~ | ~~122.0~~ | ~~143.1~~ | ~~-21.2~~ |
| ~~AOA_T-RCP~~ | ~~174.5~~ | ~~177.2~~ | ~~-2.7~~ | ~~116.0~~ | ~~139.2~~ | ~~-23.1~~ |

~~*Table 2 The total integrated additional carbon uptake (in PgC) in the period 2020-2100 in different experiment and emissions scenarios, negative denotes enhanced uptake.*~~

### *3.1.2 Surface Air Temperature*

In the control simulations, the global mean surface air temperature (SAT; 2m) increased in the period 2020-2100 with RCP2.6 simulating a net warming of 0.4±0.1K while RCP8.5 warmed by 2.7±0.1K (2081-2100). ~~All AOA experiments simulated a reduction in global mean SAT relative to their corresponding control simulation (Figure 2b). Within each emissions scenario the global mean SAT decline associated with AOA is always greater and more variable over the land than ocean (Table 3). While the mean cooling, in the period 2081-2100, is also greater over the land under RCP8.5 than RCP2.6, potentially reflecting feedbacks such as soil-moisture (Seneviratne et al., 2010) snow and ice cover changes. However, these changes are associated with large interannual large variability, and therefore not significantly different~~AOA experiments simulated a reduction in global mean SAT relative to their corresponding control simulation (Figure 2b). Within each emissions scenario the global mean SAT decline associated with AOA is always greater and more variable over the land than ocean (Table 1). In the period 2081-2100 we see larger mean changes in SAT under RCP2.6 than RCP8.5 primarily due to differences in atmospheric CO$_2$ growth rate. Krasting et al. (2014) showed that the slower rate of emissions, the lower the radiative forcing response. This occurs in response to the timescales associated with the uptake of heat and carbon. Consequently, under RCP8.5 the atmospheric CO$_2$ growth rate is much faster than RCP2.6, leading to a strong radiative forcing response. This explains why, despite a larger reduction in atmospheric CO$_2$ concentration under RCP8.5, the biggest reduction in global mean SAT occur under RCP2.6. These mean changes are also associated with large interannual variability.

~~Under RCP2.6, all the AOA experiments keep warming levels much close to values in 2020 than RCP2.6 by the end of this century (2100; Figure 2b). In contrast, under the RCP8.5 scenario, none of the AOA experiments have a significant impact on the projected warming by the end of this century (less than 10%) reflecting the large warming anticipated under high emissions (Rogelj et al., 2012).~~

|  | ~~Total RCP8.5~~ | ~~Ocean RCP8.5~~ | ~~Land RCP8.5~~ | ~~Total RCP 2.6~~ | ~~Ocean RCP 2.6~~ | ~~Land RCP 2.6~~ |
|---|---|---|---|---|---|---|
| ~~AOA_G-RCP~~ | ~~-0.16±0.08~~ | ~~-0.14±0.07~~ | ~~-0.22±0.15~~ | ~~-0.25±0.08~~ | ~~-0.19±0.05~~ | ~~-0.39±0.22~~ |
| ~~AOA_SP-RCP~~ | ~~-0.13±0.10~~ | ~~-0.11±0.07~~ | ~~-0.18±0.20~~ | ~~-0.23±0.08~~ | ~~-0.18±0.05~~ | ~~-0.35±0.22~~ |
| ~~AOA_ST-RCP~~ | ~~-0.08±0.05~~ | ~~-0.06+0.03~~ | ~~-0.13±0.14~~ | ~~-0.20±0.09~~ | ~~-0.15±0.06~~ | ~~-0.30±0.20~~ |
| ~~AOA_T-RCP~~ | ~~-0.14±0.06~~ | ~~-0.12±0.05~~ | ~~-0.19±0.11~~ | ~~-0.16±0.06~~ | ~~-0.13±0.05~~ | ~~-0.24±0.16~~ |

~~*Table 3 The differences in global mean surface air temperature and their standard deviations (1-σ) (K; SAT; 2m) for 2090 (in the period 2081-2100) for different AOA experiments and emissions scenarios relative to the emissions scenarios with no AOA.*~~

Under RCP2.6, all the AOA experiments keep global warming levels much closer to values in 2020 than RCP2.6 by the end of this century (2100; Figure 2b). In contrast, under the RCP8.5 scenario, none of the AOA experiments have a significant impact on the projected warming by the end of this century (less than 10%) reflecting the large warming projected under high emissions.

Within each of the scenarios, there ~~is~~are some ~~variability~~differences in the magnitude of the cooling within the ~~4~~four different AOA experiments~~,~~; however, these ~~differences~~ are smaller than the interannual variability over the last ~~2~~two decades of the simulations. Therefore, it appears that the global mean SAT decline with AOA is not very sensitive to where the alkalinity is added under either emission scenario.

The global mean cooling associated with AOA_G under RCP8.5 (-0.16±0.08K; 2081~~:~~-2100) is close to the mean surface air temperature ~~decrease~~cooling of -0.26K reported by ~~(~~Keller et al., (2014) for ~~the same~~similar levels of AOA. These differences may reflect the simplified atmospheric representation of the UVIC Intermediate Complexity Model and different climate sensitivities.

### *3.1.3 Ocean Acidification*

Here, we quantify changes in ocean acidification in terms of pH and aragonite saturation state changes. –We consider these two diagnostics because they are associated with different biological impacts and are not necessarily well correlated (Lenton et al., 2016). In the future, the global mean changes in pH and aragonite saturation state will be proportional to the emissions trajectories following Gattuso et al. (2015), with the largest changes associated with the higher emissions (RCP8.5) (Figure 2c-d). By 2100, despite ~~a~~the return to 2020 values of atmospheric $CO_2$ concentration under RCP2.6 (Figure 2), neither pH ~~or~~nor aragonite saturation state return to 2020 values, consistent with Mathesius et al. (2015).

In the 2020-2100 period, AOA under RCP2.6 led to much larger increases in surface pH and aragonite saturation state, more than 1.3 times, and more than 1.7 times that of RCP8.5 respectively (Table ~~4~~2). These changes reflect the differences in the mean state associated with high and low emissions, specifically the difference between Alkalinity and ~~DIC. The~~Dissolved Inorganic Carbon (ALK-DIC), a proxy for ocean acidification (Lovenduski et al, 2015). As the values of DIC in the upper ocean are larger under RCP8.5 than RCP2.6, ~~and therefore~~ the difference between ALK- and DIC (ALK-DIC) is ~~higher. For~~smaller and the chemical buffering capacity of $CO_2$ or Revelle Factor (Revelle and Suess, 1957) is less. This means that, for a given addition of ~~alkalinity,~~ALK the increase in the upper ocean DIC will always be greater ~~in the high emission case than the low emission case~~under RCP8.5 due to ~~the Revelle Factor (Revelle and Suess, 1957).~~ its reduced buffering capacity. Consequently, the ~~difference between Alkalinity and~~ changes in ALK-DIC with AOA ~~increases less in the high emission scenario than the low scenario~~are greater under RCP2.6 than RCP8.5, which translates ~~into smaller~~to greater increases in pH and aragonite saturation state~~ in the high scenario.~~ .

While there was a significant difference in pH and aragonite saturation state changes with AOA between high and low emissions cases, the global mean changes for different AOA experiments within each scenario ~~is~~are quite similar (Table ~~4). The~~2), the exception being the AOA_SP experiment, where ~~its~~the pH and aragonite saturation state changes are only ~75% of the change in the other AOA experiments. This reduced change in the polar region is consistent with the smaller changes in the surface ocean alkalinity values associated with AOA_SP (Table 1). These differences at higher latitudes reflect the enhanced subduction of alkalinity away from the surface ocean into the ocean interior that occurs in the high latitude oceans (Groeskamp et al., 2016).

AOA_G under RCP8.5 leads to a relative increase in pH of 0.06, which is consistent with ~~(~~Keller et al~~.,~~. (2014), while ~~our~~the relative increase in aragonite saturation state (0.28) is also very close to their simulated value (0.31). To put these changes into context, the estimated decrease in pH since the preindustrial period is 0.1 units (Raven et al., 2005)~~.~~ , and is responsible for already detectable changes in the marine environment (Albright et al., 2016).

| | Aragonite RCP8.5 | pH RCP8.5 | Aragonite RCP2.6 | pH ~~RCP 2~~RCP2.6 |
|---|---|---|---|---|
| AOA_G~~-RCP~~ | 0.28 | 0.06 | 0.50 | 0.07 |
| AOA_SP~~-RCP~~ | 0.20 | 0.05 | 0.39 | 0.07 |
| AOA_ST~~-RCP~~ | 0.30 | 0.06 | 0.54 | 0.08 |
| AOA_T~~-RCP~~ | 0.28 | 0.06 | 0.5 | 0.07 |

*Table ~~4~~2 The~~ relative~~ differences in surface value of aragonite saturation state and pH between the AOA experiments ~~and the high and low emissions ~~for each emission scenarios in 2100 relative to the emissions scenario with no AOA.*

*3.2 Regional Responses*

For both RCP scenarios, there are large regional differences in the relative surface changes in alkalinity, temperature, and ocean acidification associated with the different AOA experiments. The regional nature of these changes is closely associated with where alkalinity addition is applied, and the two different emissions scenarios considered here do not differ significantly in their behaviour. This implies that any differences in stratification and overturning circulation between the two scenarios ~~are insufficient to~~do not significantly ~~modulate~~alter the response to AOA.

### *3.2.1. Surface Alkalinity*

For both scenarios, the greatest surface alkalinity changes occur where the alkalinity is added (Figure 3). Spatially, under either emission scenario, the relative differences in 2090 are very similar~~,~~; consequently, we only show the changes under RCP2.6 (Figure 3). The only significant differences occur in the Arctic, reflecting larger longer-term changes in alkalinity projected under higher emissions (Yamamoto et al., 2012).

Overall, the greatest increases are seen in the tropical ocean (AOA_T) suggesting this is the most efficient region in retaining the added alkalinity in the upper ocean. This reflects the fact that subduction processes in the tropical ocean are less efficient than in other regions such as the higher latitudes. ~~In the~~The (ice-free) subpolar oceans (AOA_SP) produced the smallest relative increase in alkalinity, and this reflects the strong and efficient surface to interior connections through subduction occurring at higher latitudes (Groeskamp et al., 2016). The global mean relative increase associated with AOA in the subtropical gyres (AOA_ST) and globally (AOA_G) fall between the tropical (AOA_T) and higher latitude (AOA_SP) values. In the case of AOA_ST, this reflects the ~~time-scales~~timescales associated with ~~circulation~~the longer residence time of upper ocean waters in the subtropical gyres.

The most modest relative increase in alkalinity occurs in the ~~non-~~ice-~~free~~covered regions where alkalinity is not explicitly added. Interestingly, even when alkalinity is added in the very high latitude Southern Ocean, it is carried northward by the Ekman current ~~explaining~~which explains the very modest increase in the region where AOA occurs between 50S to 60S. In terms of the total alkalinity added to the surface ocean, about one-third remains in the upper 200m ~~in~~by 2100~~.~~ (Figure 4). Specifically, for AOA_G we see 31% remains~~,~~ in the upper ocean, and for AOA_T and AOA_ST~~:~~ 34~~%,~~% remains in the upper ocean, while for AOA_SP~~:~~ the figure is 22-24%~~ remains,~~ which (as anticipated) is lower than in other regions.

Spatially, AOA in the higher latitude regions (AOA_SP) leads to very large relative increases in alkalinity (> 1000 µmol/kg; 2090) occurring along the northern most boundary of the Northern Subpolar Gyres, particularly the North Pacific. Clearly, in this region the rate of AOA exceeds the rate of subduction allowing alkalinity to build up. Large relative increases in alkalinity also occur in the Southern Ocean under AOA_SP, particularly along Western

Boundary Currents. However, in contrast to northern high latitudes the values still remain ~~quite~~ low suggesting that the rate of addition does not exceed the rate of subduction even under the highest emission scenario.

AOA_ST shows a large relative increase of ~300 μmol/kg (2081-2100) in the ~~subpolar~~subtropical gyre regions. Overall, we find that these relative increases are quite homogenous across the entire subtropical gyres, with strong mixing with tropical waters leading to significant relative increases in tropical Atlantic, Western Pacific and Indian Oceans. ~~-~~Within the tropical ocean, under AOA_T the largest relative changes are found across the entire tropical Indian Ocean (~~~-~~400 μmol/kg) with large relative increases also seen in the Indonesian seas (~280 μmol/kg; 2081-2100). Away from the tropical Indian Ocean, we find that relatively homogenous increases occur in the Western Pacific and the Atlantic, with much more modest relative increases in the Eastern Pacific reflecting the dominant East to West upper ocean circulation. ~~Consistent with the response of AOA_ST,~~ AOA_T leads to relative increases in surface alkalinity that are consistent with the response to AOA_ST – in the ~~AOA_ST~~ region of ~130 μmol/kg (2081-2100).

In the case of AOA_G, a relatively uniform net increase in alkalinity occurs in all regions with the exception of the upwelling regions such as the tropical Pacific, which showed a more modest relative increase. In AOA_G there is little evidence of any of the very large increases in alkalinity seen in the more regional AOA experiments. This spatial pattern of relative increase is broadly consistent with the pattern of global alkalinity increase simulated by Ilyina et al. (2013) and Keller et al. (2014) for AOA in the (ice-free) global ocean.

### *3.2.2 Changes in the interior distribution of alkalinity in the global ocean*

As only about 30% of the total AOA remains in the upper 200m, we explore the fate of this alkalinity in the interior ocean in the zonal sections of alkalinity (Figure 4). As the pattern is very similar~~,~~ between RCP2.6 and RCP8.5, we only show RCP2.6, noting that in the North Atlantic ~~where~~ the projected ocean stratification is stronger under higher emissions (not shown) leading to slightly decreased subsurface values. This increased stratification is consistent with other studies (e.g. ~~(Yool et al., 2015)~~Yool et al., 2015).

Unlike the surface plots of AOA, the relative increases in subsurface alkalinity due to AOA are very similar across all experiments. This heterogeneous spatial pattern of alkalinity increase is associated with water entering the interior ocean along specific surface to interior pathways ~~(Groeskamp et al., 2016). Specifically, we see alkalinity moving~~. Alkalinity also moves into the interior ocean along the poleward boundaries of the subtropical gyres, associated with the formation and subduction of mode waters, and an increase in the subtropical gyres associated with large-scale downwelling~~,~~ and deep mixing in the North Atlantic. The changes in alkalinity are mainly found in the upper ocean (<~~1000 m~~1000m) which reflects the relatively short period of alkalinity addition. Given the short period, this is analogous to present-day observed distributions of anthropogenic carbon (Sabine et al., 2004).

As the changes in export production are very small, the large changes in the interior alkalinity concentrations primarily reflect the physical transport, rather than the sinking and remineralization of calcium carbonate. Clearly other biological processes, not represented in our model, have the potential to impact the surface and interior values of alkalinity (Matear and Lenton, 2014). One such process is the reduction in the (rain) ratio of PIC:POC under higher emissions (Riebesell et al., 2000) ~~however~~. However, it has been shown that even a very large reduction in PIC production (50%) would not significantly impact our results (Heinze, 2004). Unfortunately, at present~~,~~ the magnitude and sign of many of these other feedbacks ~~remains~~remain poorly known (Matear and Lenton, 2014); consequently, quantifying their impact on our results is very difficult, and beyond the scope of this study.

*3.2.3 Ocean Carbon Cycle Response*

The similarity in global ocean carbon uptake associated with all AOA experiments for a given emission scenario hides the large spatial differences between simulations. Given that the largest carbon cycle response occurs in the ocean (Table ~~2~~1), we focus on this response for RCP8.5 and RCP2.6 (Figures 5 and 6). As expected, ocean carbon uptake is strongly enhanced in the regions of AOA. Away from regions of AOA, there is a reduction in carbon uptake, associated with the weakening of the gradient in $CO_2$ between the atmosphere and ocean due to AOA. Interestingly, the largest increase spatially occurs in the Southern Ocean under AOA_SP for RCP2.6, while in contrast the largest changes under RCP8.5 ~~occurs~~occur in the tropical ocean under AOA_~~SP~~T. The very small changes in export production in

RCP2.6 were located in the Arabian Sea (not shown), ~~and while~~likely driven by enhanced mixing in this region. While these changes are <~~-~~1% of the total change in carbon uptake, they may nevertheless ~~they maybe~~be important regionally.

### *3.2.4 Temperature ~~(SAT)~~*

The decrease in global mean SAT associated with all AOA experiments for a given emission scenario again hides the large spatial differences between the simulations. ~~-~~The response of surface temperature is spatially very heterogeneous (Figures 7 and 8~~),~~) and~~ while~~ the regional surface temperature changes are very similar between the two emissions scenarios. The exception to this is the Arctic which did not show a consistent response across the different AOA experiments~~.~~, reflecting the period over which the mean changes were calculated, and the simulated large variability in SAT in this region. Under both emission scenarios, the largest cooling associated with AOA occurs over Northern Russia and Canada, and Antarctica (greater than a -1.5K cooling) with a larger cooling in these regions under RCP2.6.

~~In the surface ocean,~~ AOA in the RCP2.6 scenario ~~shows~~brings about a net cooling ~~over~~of the surface ocean~~,~~ with the exception of the North Atlantic, east of New Zealand, and off the southern coast of Alaska, which show a very ~~a~~ modest warming. A similar pattern is evident in RCP8.5; however, there is a greater cooling in the high latitudes, and less cooling in the lower latitudes than under RCP2.6.

### *3.2.5 Ocean Acidification Response*

Globally, the response of pH and aragonite saturation state associated with AOA are similar~~,~~; however, large spatial and regional differences are present~~.~~ (Figures 9-14). To aid in the interpretation of changes in aragonite saturation state, overlain on the aragonite saturation state maps ~~(Figures 9 and 10)~~ are the contours corresponding to the value of 3, the approximate threshold for suitable coral habitat (Hoegh-Guldberg et al., 2007). On these surface maps and subsequent section plots ~~(Figures 13  and 14)~~ we plot the saturation horizon, i.e. the contour corresponding to the transition from chemically stable to unstable (or corrosive), i.e. aragonite saturation state is equal to 1 (Orr et al., 2005).

The largest relative changes in pH and aragonite saturation state were associated with regions of AOA~~,~~ (Figures 9-12), reflecting increases in the surface values of alkalinity~~.~~ (Figure 3). All simulations increase pH and aragonite saturation state in the Arctic despite no direct addition in this region, with the largest changes here associated with AOA_G and AOA_SP. Interestingly, all simulations show little to no increase in the high latitude Southern Ocean, consistent with more efficient transport of the added alkalinity into the ocean interior.

The changes in pH associated with AOA experiments under RCP8.5, while spatially very different~~,~~ particularly when added in the subpolar ocean, are still much less than the decreases associated with RCP8.5 with no AOA~~.~~ (Figure 9). In terms of aragonite saturation state (Figure 10), the conditions for coral growth in the tropical ocean remain very ~~unfavorable~~unfavourable by the end of century (i.e. aragonite saturation state <~~-~~3) under all regional and global experiments, with the exception of AOA_T, where~~ only~~ a very small region in the Central Pacific Ocean exhibits suitable conditions.

Consistent with Feng et al. (2016), we find that this level of AOA under RCP8.5 is insufficient to ameliorate or significantly alter the large-scale changes in ocean acidification. More positively, at the higher latitudes the saturation horizon is moved poleward with the largest shift associated with AOA_SP, and the smallest shift at the high latitudes occurring under AOA_T. Consistent with these changes, we see a deepening of the saturation horizon everywhere~~ (Figure 13),~~, and little difference spatially between AOA experiments, consistent with zonal mean changes in alkalinity for the ~~4~~four AOA experiments~~.~~ (Figure 11).

The spatial pattern of changes associated with AOA under RCP2.6 ~~are~~is broadly consistent with ~~those~~that seen under higher emissions~~,~~; however, the magnitude of the response is much larger – again, due to the larger differences between Alkalinity and DIC with AOA under RCP2.6~~.~~ (Figures 12 and 13). In terms of aragonite saturation state, the area of tropical ocean favourable for corals is considerably expanded~~ suggesting that conditions for tropical coral growth are improved under AOA.~~. As anticipated the largest changes in the area favourable for tropical corals is associated with AOA_T, closely followed by AOA_ST. As the saturation horizon does not reach the surface under RCP2.6, we can only look at the changes in the interior ocean. Here, there is a deepening in the saturation horizon ~~in all experiments ~~of a very similar magnitude in all experiments (Figure 14), with the exception of the Arctic.

Here, the response of the saturation horizon is more sensitive to the location of the AOA, varying between ~100m under AOA_T and ~~~280 m~~280m under AOA_SP~~.~~ (Figure 14).

Spatially, the large changes in ocean acidification in response to AOA under RCP2.6 more than compensate for the changes in ocean chemistry due to low emissions in the period 2020-2100. Globally, ~~in~~ the changes in the period 2020-2100 are sufficient to ~~reversed~~reverse or compensate for the changes since the preindustrial period (1850). However, spatially in some regions such as equatorial upwelling, an important area of global fisheries (Chavez et al., 2003), AOA in fact leads to higher values of aragonite saturation state and pH than the ocean experienced in the preindustrial period (Feely et al., 2009). We can only speculate on the potential impact on marine biota ~~through~~of a reduction in aqueous $CO_2$ and elevated pH levels in these regions. For a recent review of the potential impact of rising pH and ~~Aragonite~~aragonite saturation state on marine organisms, we direct the reader to Renforth and Henderson (2017).

### *3.2.6 Importance of Seasonality*

In this paper, while we have focused on year-round AOA, as a sensitivity experiment we also explored whether AOA added in summer or winter was more efficient. To do this, we focused on the higher latitudes regions where the largest seasonal changes in mixing are found (de Boyer Montegut et al., 2004~~;~~; Trull et al., 2001). Here, we tested whether AOA in either summer or winter was more effective than year-round addition. To test this for RCP8.5, we add alkalinity only during the summer at half of the annual rate (or 0.125PmolALK/year) in the AOA_SP region.

Our results showed that the response to AOA in summer was very close to 50% of the response of the year-round addition associated with AOA_SP (or 0.25PmolALK/year). This suggests that the response of AOA appears invariant with regard to when the alkalinity is added. This also suggests, consistent with published studies (e.g. ~~(~~Keller et al., 2014~~;~~; Feng et al., 2016~~;~~; Kohler et al., 2013), that the response of the ocean to different quantities of AOA is scalable under the same emissions scenario. Whether this is true under very much larger additions of alkalinity, as simulated by ~~(~~Gonzalez and Ilyina~~,~~ (2016), is less clear.

## 4. Summary and Concluding Remarks

Integrated Assessment Modelling for the Intergovernmental Panel on Climate Change shows that $CO_2$ removal (CDR) may be required to achieve the goal of limiting warming to well below 2° ~~(COP21)~~C (Fuss et al., 2014). Of the many schemes that have been proposed to limit warming, only Artificial Ocean Alkalization (AOA) is capable of both reducing the rate and magnitude of global warming through reducing atmospheric $CO_2$ concentrations, while simultaneously directly addressing ocean acidification. Ocean acidification, while often receiving~~ often~~ less attention, is likely to have very long lasting and damaging impacts on the entire marine ecosystem, and the ecosystem services it provides.

Here, for the first time, we investigate the response of a fully coupled climate ESM~~, ~~(i.e. one that accounts for climate-carbon feedbacks~~,~~) to a fixed addition of alkalinity (0.25PmolALK/year) under high (RCP8.5) and low (RCP2.6) emissions scenarios~~ to a fixed addition of alkalinity (0.25PmolALK/year).~~. We explore the effect of global and regional application of AOA focusing on the subpolar gyres, the subtropical gyres and the tropical ocean. To assess AOA, we look at changes in surface air temperature, carbon cycling and ocean acidification (aragonite saturation state and pH) in the period 2020-2100.

Consistent with other published studies, we see that AOA leads to reduced atmospheric $CO_2$ concentrations, cooler global mean surface temperatures, and reduced levels of ocean acidification. Globally, for these metrics we observed that they do not vary significantly between the various AOA experiments under each emissions scenario. This implies that at ~~this~~the global scale there is little sensitivity of the global responses to the region where AOA is applied. We also investigate as a sensitivity experiment adding alkalinity in different seasons and see little difference in response to when AOA was undertaken.

We see under AOA that the increased carbon uptake is dominated by the ocean. Under RCP8.5, the changes due to AOA are only capable of reducing atmospheric concentrations by ~~a maximum of 86 ppm versus the projected change of 560ppm,~~16 % and, as such, the response of the climate system remains strongly dominated by warming. This is consistent with published studies of the response of the climate system under RCP8.5, and studies that have estimated the amount of AOA required to counteract a high emissions trajectory.

In contrast, AOA under RCP2.6 – while only capable of reducing atmospheric $CO_2$ levels by 58 ppm~~,~~ – is sufficient to reduce atmospheric $CO_2$ concentrations and warming to close to 2020 levels at the end of the century. This is significant as it suggests that, in combination with a rapid reduction in emissions, AOA could make an important contribution to the goal ~~to keep~~of keeping the rise in global mean temperatures below 2°. However, AOA under the RCP2.6 ~~does not ameliorate spatial~~emissions scenario changes ~~of~~the roles played by the ocean and land in carbon uptake ~~associated~~as compared with the scenario of RCP2.6 with no AOA, resulting in a reduced uptake in the terrestrial biosphere and increased uptake in the ocean. This highlights that, while the atmospheric $CO_2$ and warming may be reversible, the response of individual components of the Earth ~~System~~system to different CDR may not be (Lenton et al., 2017).

~~Interestingly despite~~Despite the impact of AOA on the atmospheric $CO_2$ concentration under RCP2.6 being only ~60% of the ~~value the~~impact under RCP8.5, we see much larger changes in ocean acidification associated with RCP2.6 than RCP8.5~~,~~ – more than 1.3 times in pH and more than 1.7 times in aragonite saturation state. This ~~reflect~~reflects the larger reductions ~~of~~in the difference between ALK and DIC that occurs under RCP2.6. We also see larger relative decreases in global temperature associated with RCP2.6. These results are very important as they demonstrate that ~~that~~ AOA is more effective ~~under lower emissions~~ in reducing ocean acidification and global warming under lower emissions.

While there is little sensitivity in the global responses to the region in which AOA is applied, spatially the largest changes in ocean acidification (and ocean carbon uptake) were seen in the regions where AOA was applied. Despite large changes regionally, these cannot compensate for the large changes associated with RCP8.5. Even targeted AOA in the tropical ocean can preserve only a tiny area of the ocean conducive to healthy coral growth; and even then the concomitant large warming is likely to be a stronger influence on coral growth than ocean chemistry (D'Olivo and McCulloch, 2017).

In contrast, AOA under RCP2.6 is more than capable of ameliorating the projected ocean acidification changes in the period 2020-2100. We see that, in all cases, the area of the tropical ocean suitable for healthy coral growth expands, with the largest changes ~~are~~

associated with tropical addition (AOA_T). In some areas, such as the equatorial Pacific, the changes that have occurred since the preindustrial ~~(1850)~~period are also completely ~~compensated~~reversed, and in some cases, leads to higher values ~~that are higher~~of aragonite saturation state and pH than were experienced in the preindustrial period.

While the amount of alkalinity added in this study is small in comparison to other published studies, the challenge of achieving even this level of AOA should not be underestimated. Indeed, it is not clear whether such an effort is even feasible given the cost~~,~~, and the logistical, political and engineering challenges of producing and distributing such large quantities of alkaline material (Renforth and Henderson, 2017). In the case of RCP8.5, it is unlikely that this level of AOA could be justified given our results. If emissions can be reduced along an RCP2.6 type trajectory, this study suggests that AOA is much more effective and may provide a method to remove atmospheric $CO_2$ to complement mitigation, albeit with some side-effects, and may be an alternative to reliance on land~~-~~-based CDR.

In this work, and other published studies to date, we have not accounted for the role of the mesoscale in AOA. In the real ocean (mesoscale~~)~~), eddies are ubiquitous~~,~~ and associated with strong convergent and divergent flows, and mixing~~ that~~ plays an important role in ocean transport (Zhang et al., 2014b). It is plausible that the mesoscale, and indeed fine~~-~~-scale circulation in the coastal environment (e.g. ~~(~~Mongin et al., 2016a~~;~~; Mongin et al., 2016b) ~~would~~, may modulate the local response to AOA and this therefore needs to be considered in future studies.

Furthermore, this is a single model study, and the results of this work need to be tested and compared in other models. The Carbon Dioxide Removal Model Intercomparison Project (CDR-MIP) was created to coordinate and advance the understanding of CDR in the ~~earth~~Earth system (Lenton et al., 2017). CDR-MIP brings together Earth ~~System~~system models of varying complexity in a series of coordinated multi-model experiments, one of which is a global AOA experiment (C4) (Keller et al., ~~2017~~accepted). This will allow the response of the ~~earth~~Earth system to AOA to be further explored and quantified in a robust multi-model framework, and will examine important further questions such as including cessation effects of alkalinity addition, and the long-term fate of additional alkalinity in the ocean. In parallel, more process and observational studies (e.g. mesocosm experiments~~,~~) are needed to better understand the implications of AOA.

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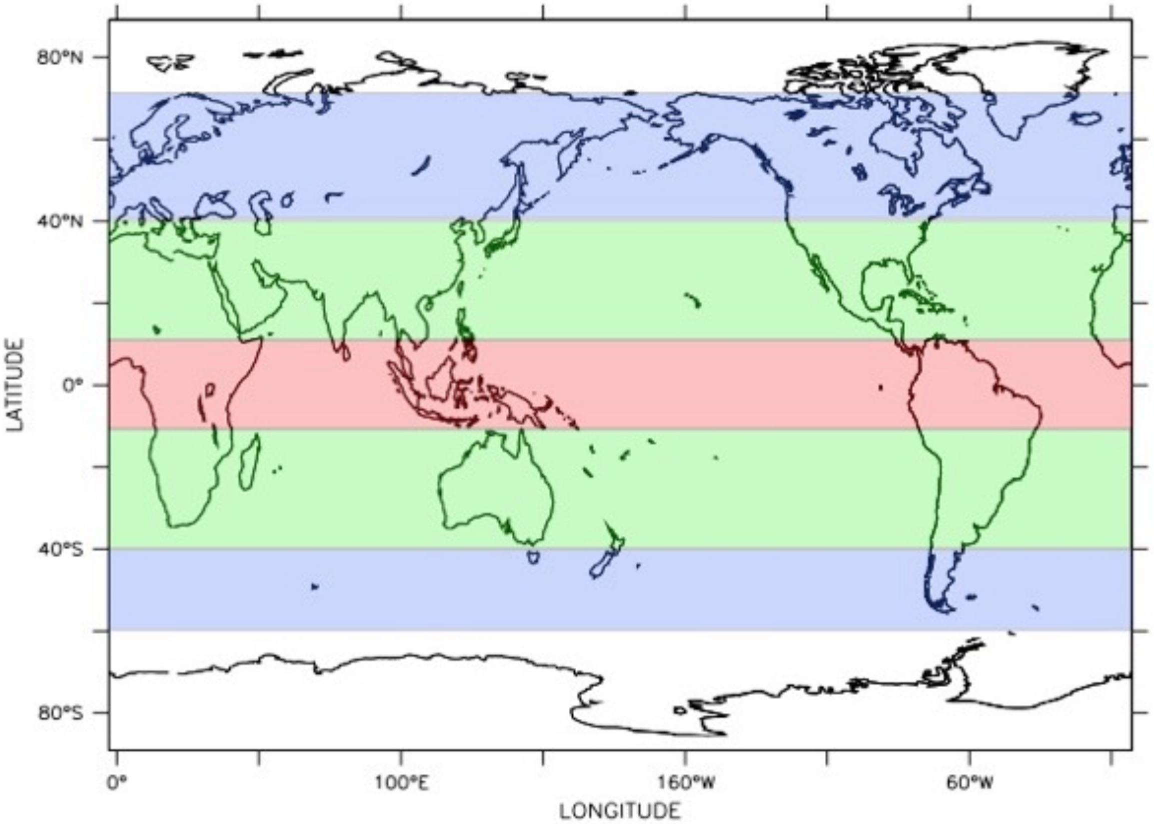


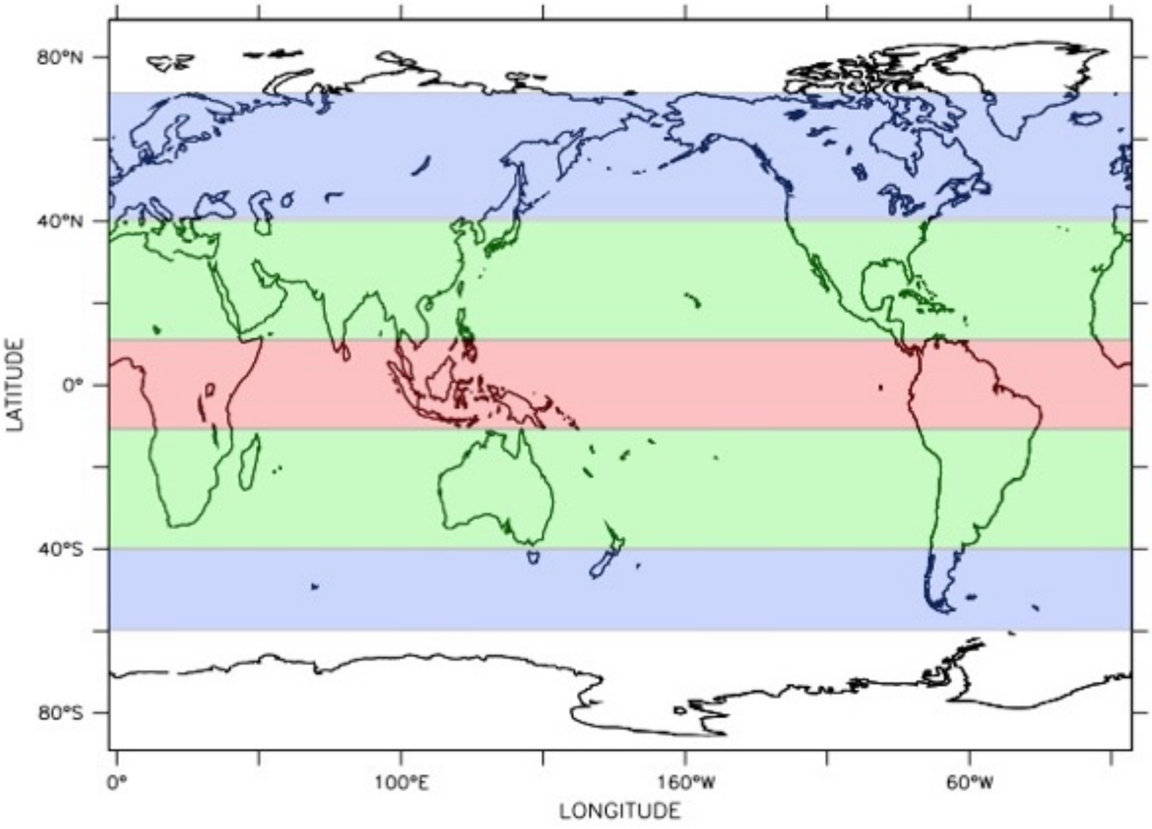
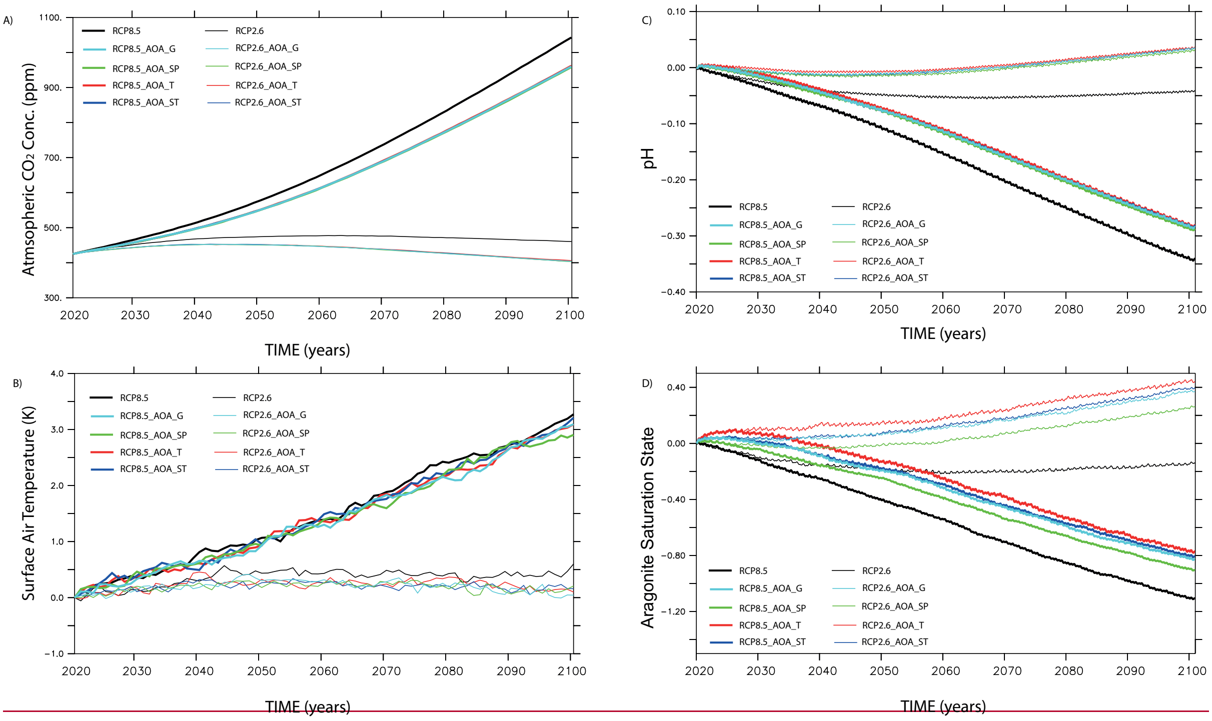


Figure 1 Ocean regions used for Alkalinity Injection in the period 2020-2100, the blue denotes the subpolar regions (AOA_SP), the green regions represent the subtropical gyres (AOA_ST), red the tropical ocean (AOA_T), and all ~~colored~~coloured regions combined the global alkalinity injection (AOA_G). Note that the ocean regions not ~~colored~~coloured represent the seasonal sea-ice, where no alkalinity was added in the simulation.



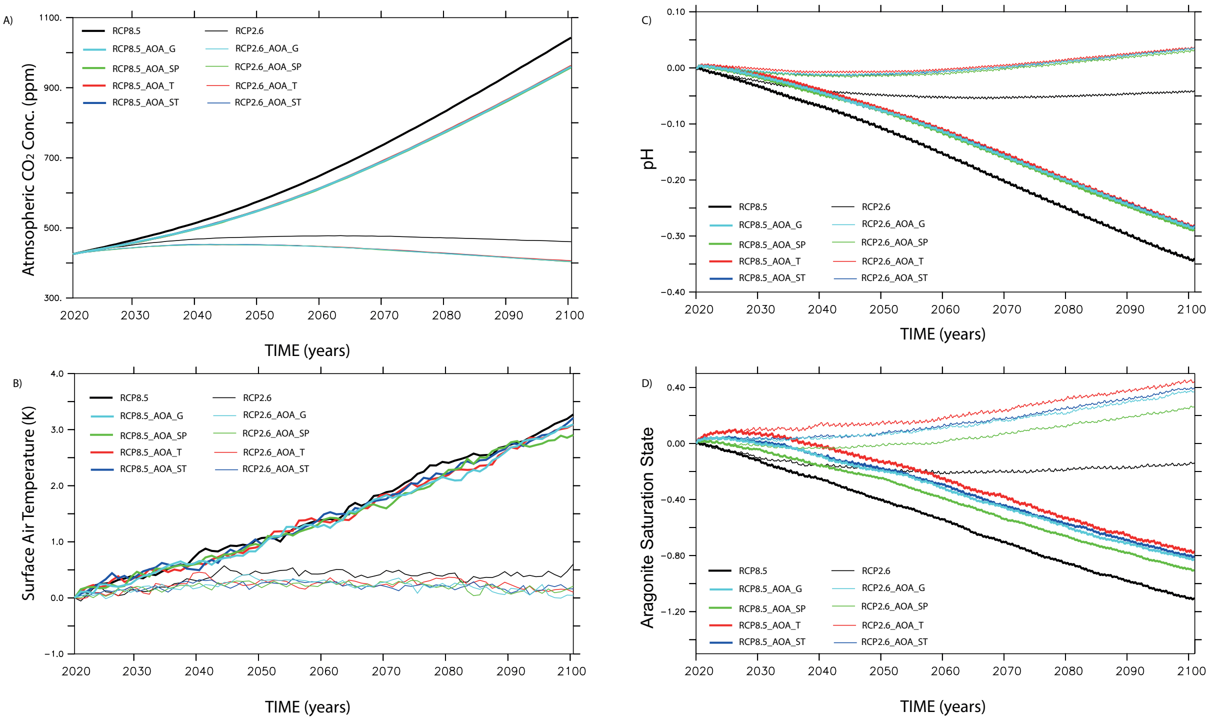
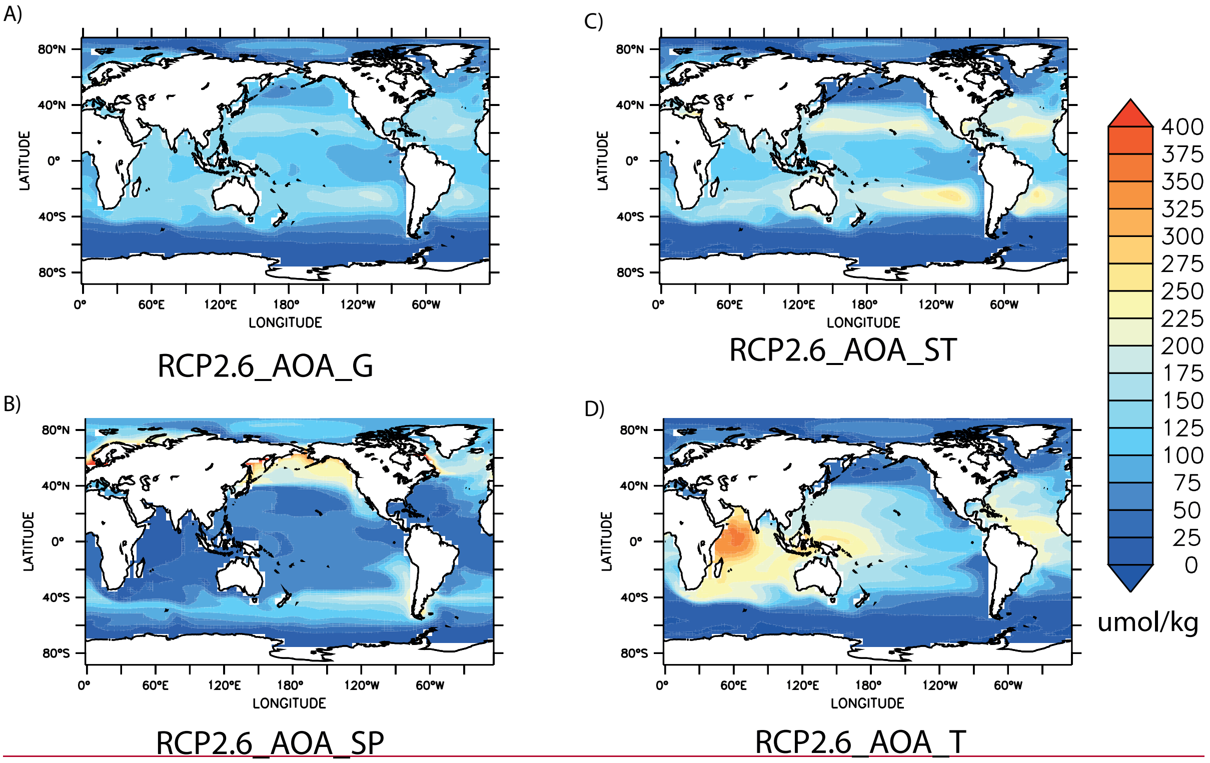
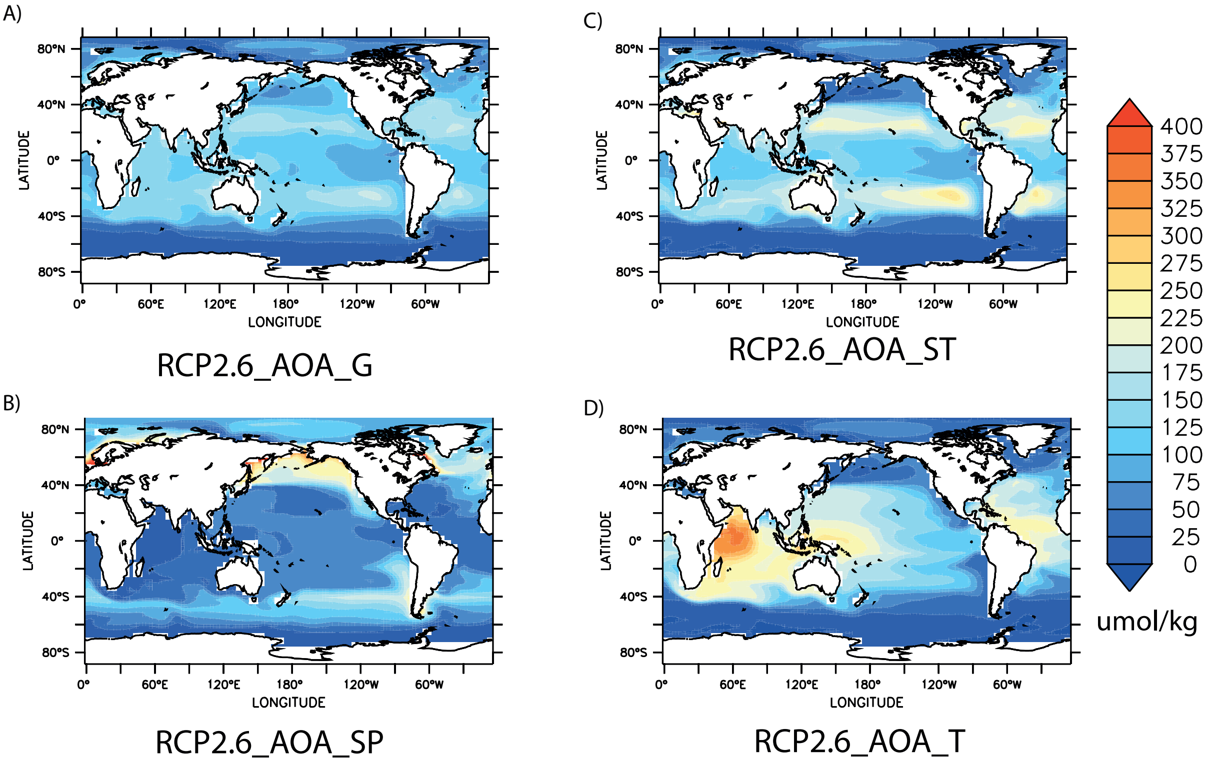


Figure 2 The global mean changes in: Atmospheric CO2 concentration (~~A~~a), Surface Air Temperature (SAT; ~~B~~b), surface ocean pH (~~C~~c) and Aragonite Saturation State (~~D~~d) for high (RCP8.5) and low emissions (RCP2.6) with global and regional AOA in the period 2020-2100.


A)
B)
C)
D)
80°N
40°N
0°
40°S
80°S
LATITUDE
0°
60°E
120°E
180°
120°W
60°W
LONGITUDE
RCP2.6_AOA_G
RCP2.6_AOA_SP
RCP2.6_AOA_ST
RCP2.6_AOA_T
400
375
350
325
300
275
250
225
200
175
150
125
100
75
50
25
0
umol/kg


A)
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80°N
40°N
0°
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80°S
LATITUDE
0°
60°E
120°E
180°
120°W
60°W
LONGITUDE
RCP2.6_AOA_G
RCP2.6_AOA_SP
RCP2.6_AOA_ST
RCP2.6_AOA_T
400
375
350
325
300
275
250
225
200
175
150
125
100
75
50
25
0
umol/kg

Figure 3 The spatial map of ~~relative~~the increase in surface alkalinity in 2090 (mean; 2081-2100) associated with global and regional AOA under RCP2.6~~.~~ relative to RCP2.6 with no AOA. Units are µmol/kg.

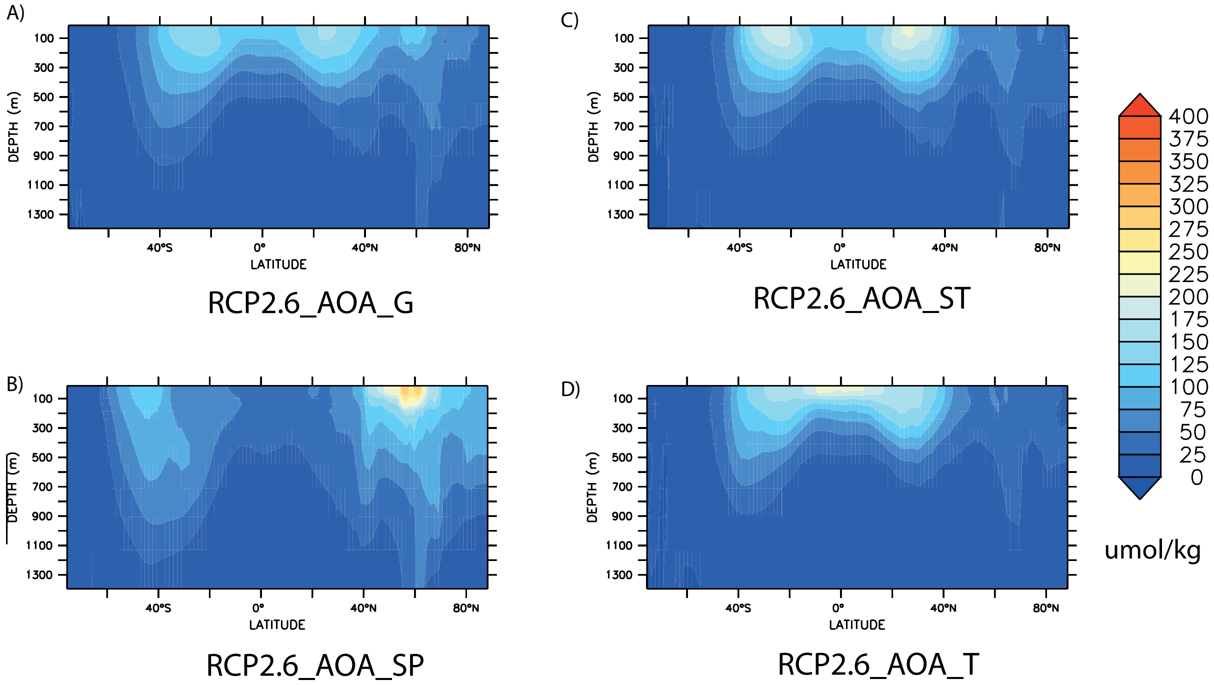


Figure 4 The zonal mean ~~relative~~ changes in alkalinity in the interior ocean associated with global and regional AOA under RCP8.5 in 2090 (mean~~:~~; 2081-2100~~).~~) relative to RCP8.5 with no AOA. Units are µmol/kg.

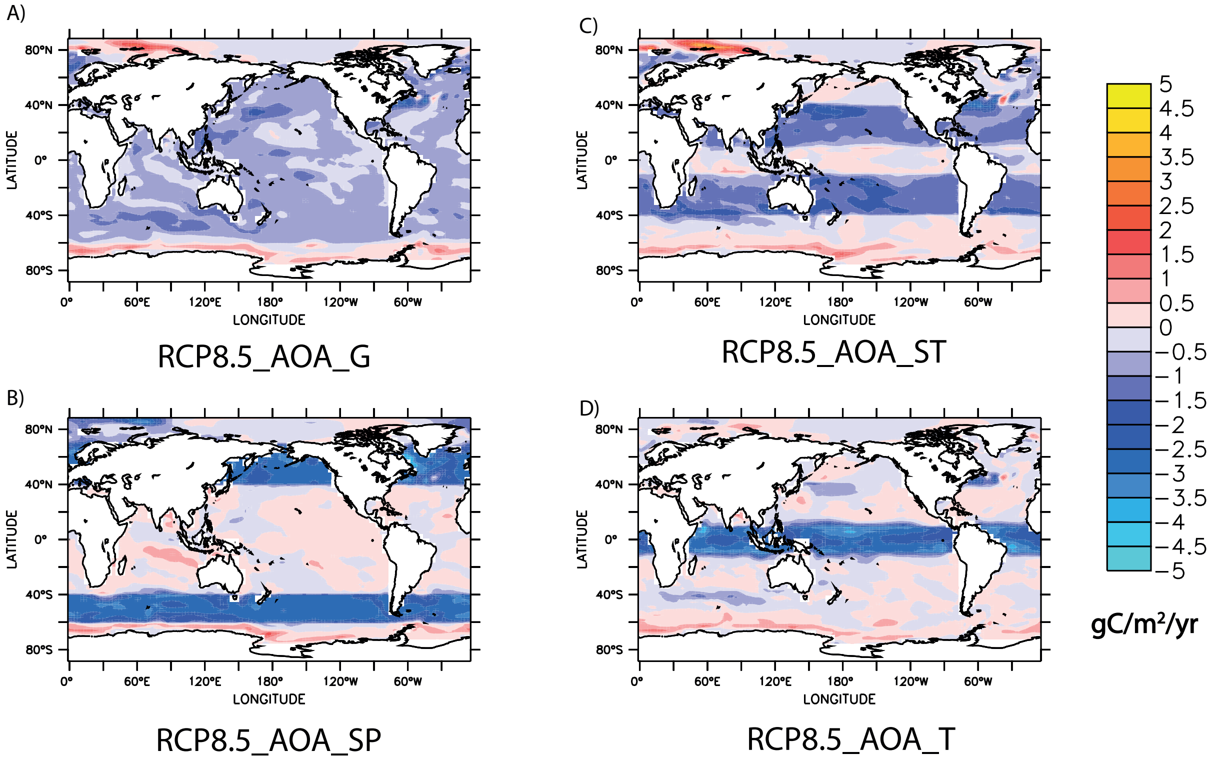
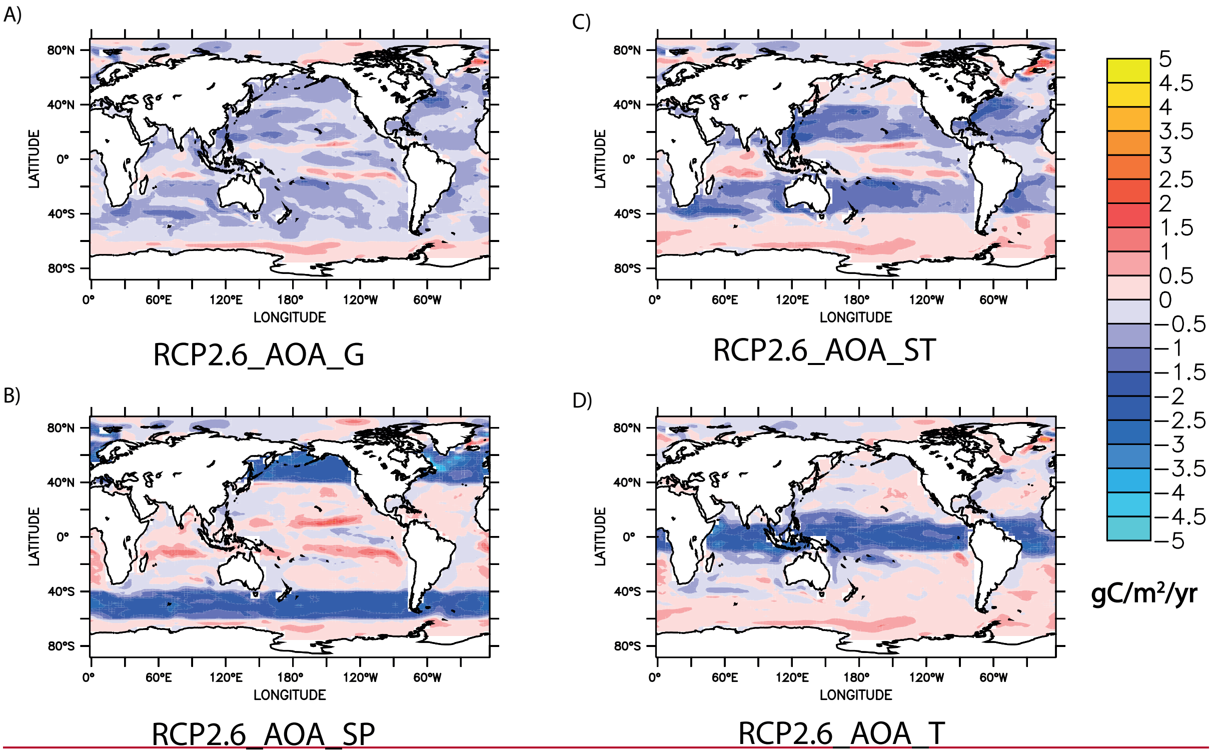
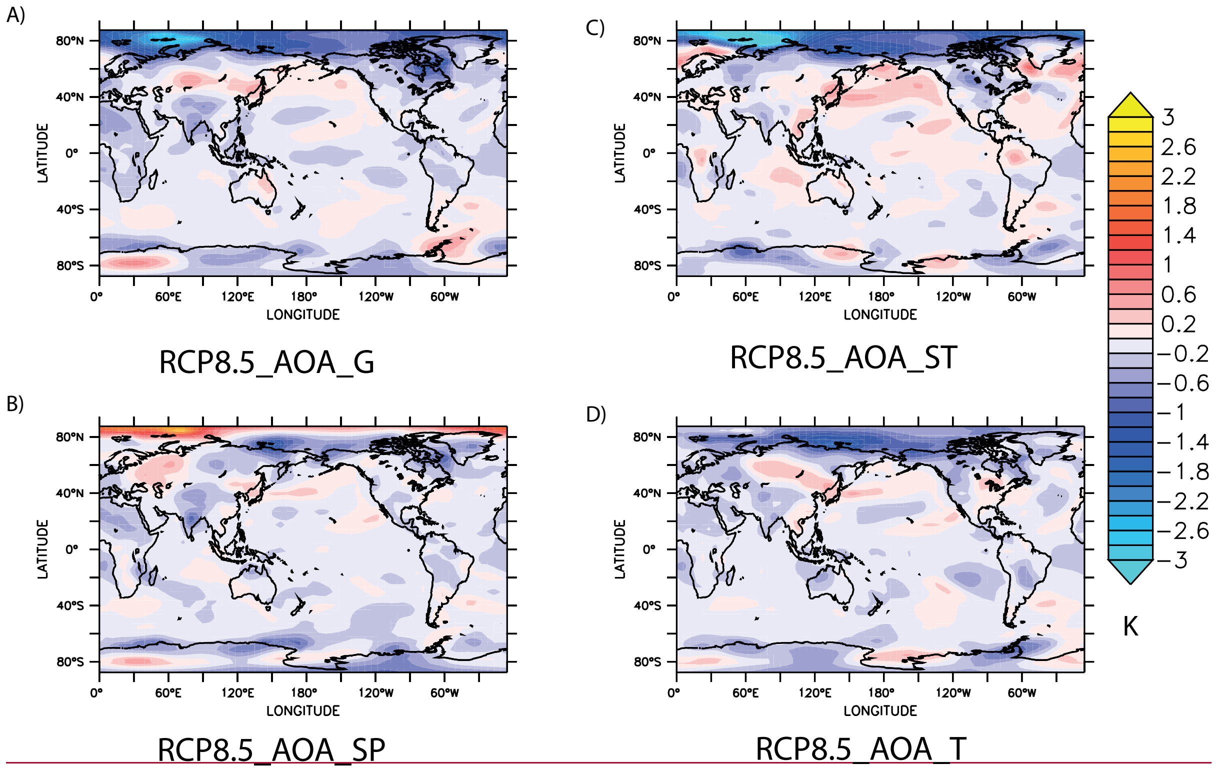


Figure 5 The spatial map of ~~relative changes in ocean carbon uptake in 2090 (mean; 2081-2100) associated with global and regional AOA under RCP8.5. Units are gC/m$^2$/yr~~



~~Figure 6 The spatial map of relative~~the changes in ocean carbon uptake in 2090 (mean; 2081-2100) associated with global and regional AOA under ~~RCP2.6.~~RCP8.5, relative to RCP8.5 with no AOA. Units are gC/m$^2$/yr.

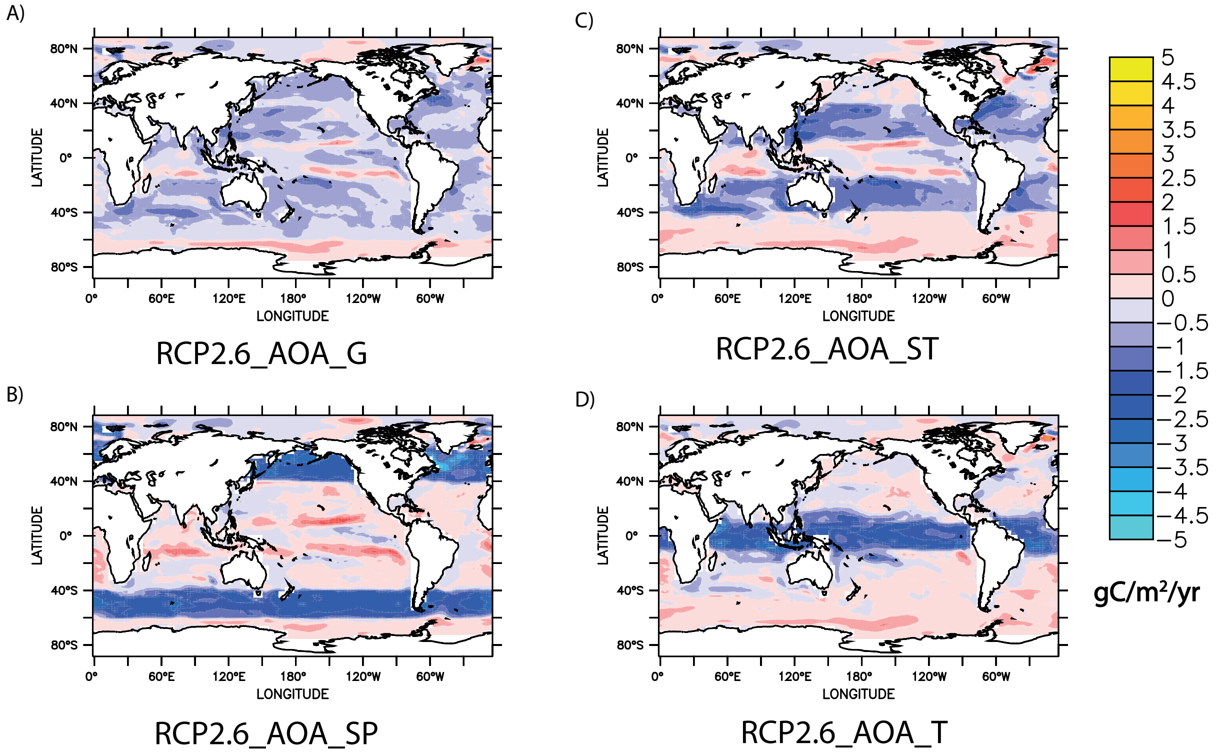


RCP8.5_AOA_G

RCP8.5_AOA_ST

RCP8.5_AOA_SP

RCP8.5_AOA_T



RCP2.6_AOA_G

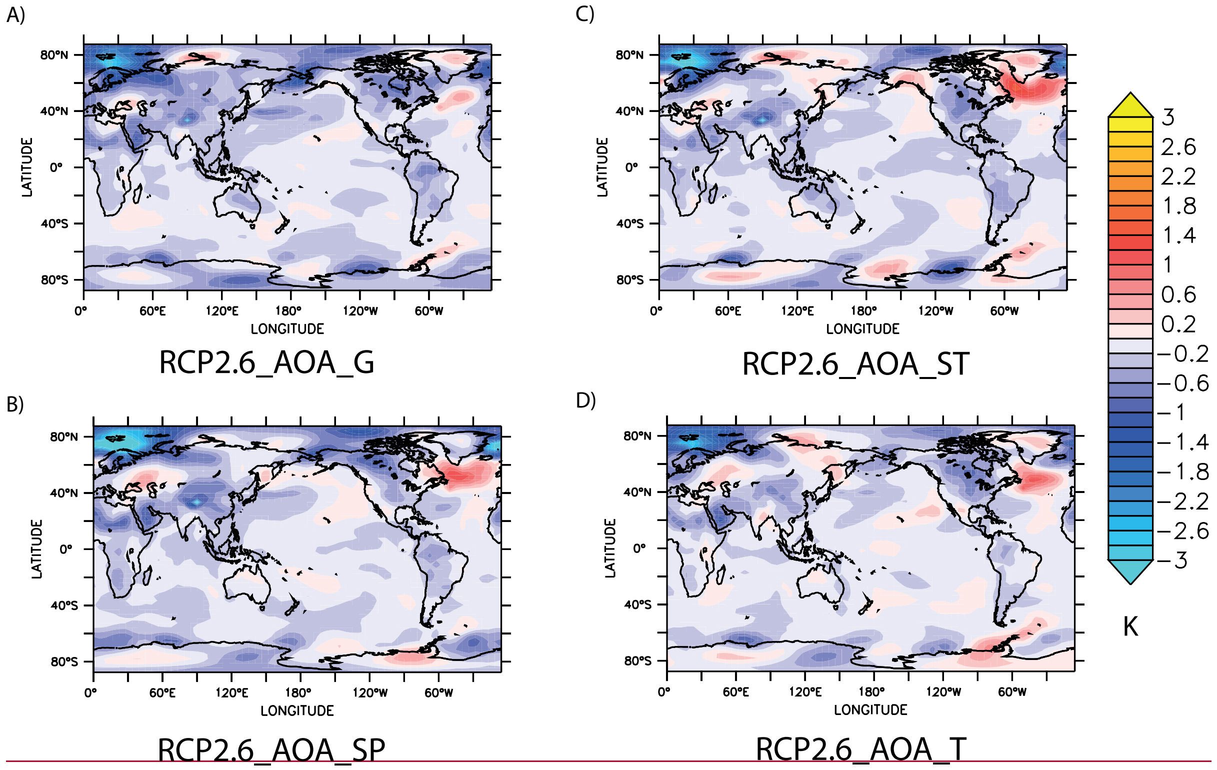
RCP2.6_AOA_ST

RCP2.6_AOA_SP

RCP2.6_AOA_T

Figure ~~7~~6 The spatial map of ~~relative~~the changes in ~~surface air temperature~~ocean carbon uptake in 2090 (mean; 2081-2100) associated with global and regional AOA under ~~RCP8.5.~~RCP2.6, relative to the RCP2.6 with no AOA. Units are ~~K~~gC/m$^2$/yr.

1043



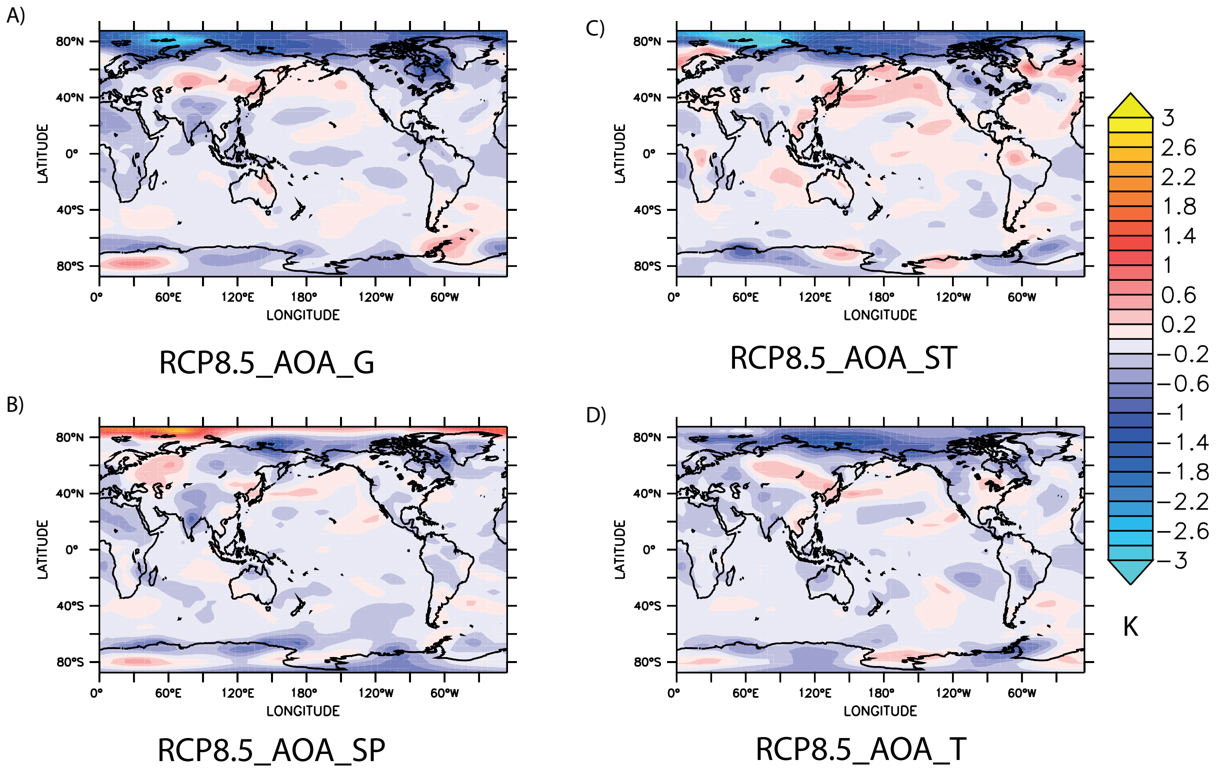
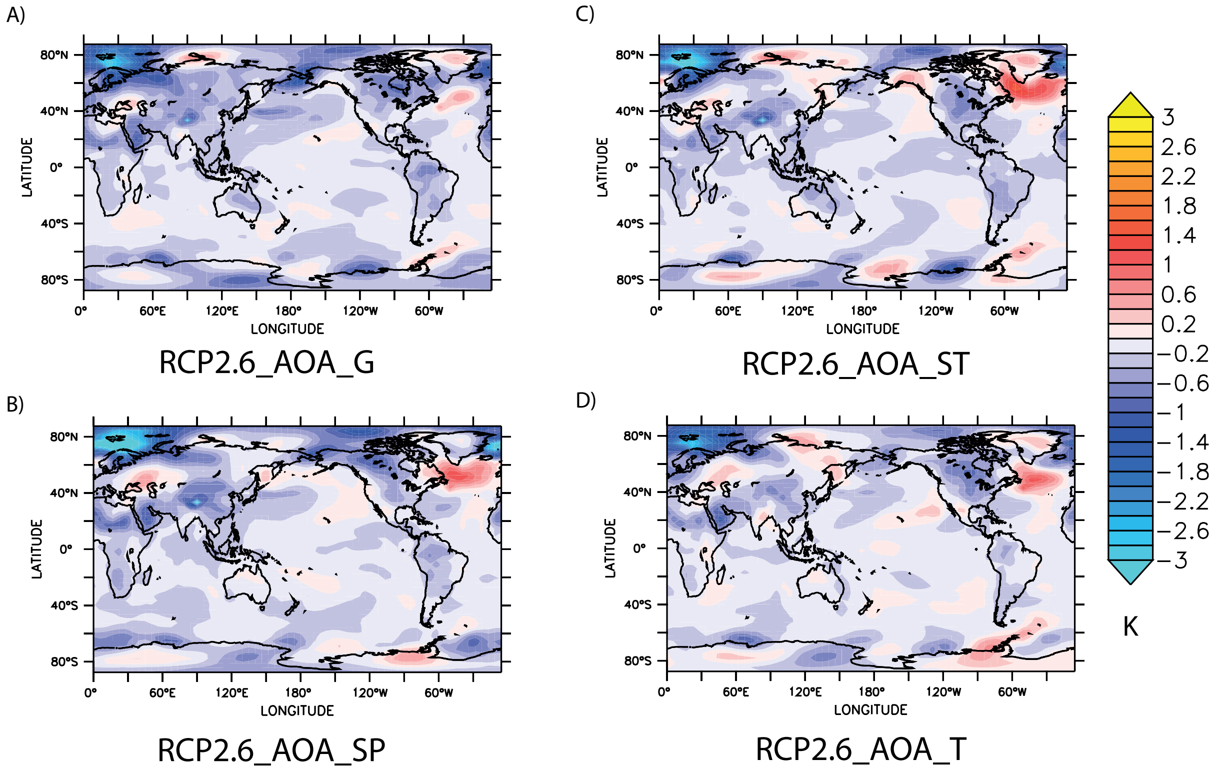


Figure ~~8~~7 The spatial map of ~~relative~~the changes in surface air temperature 2090 (mean; 2081-2100) associated with global and regional AOA under ~~RCP2.6.~~RCP8.5, relative to RCP8.5 with no AOA. Units are K.

1051



RCP2.6_AOA_G

RCP2.6_AOA_ST

RCP2.6_AOA_SP

RCP2.6_AOA_T

Figure 8 The spatial map of the changes in surface air temperature 2090 (mean; 2081-2100) associated with global and regional AOA under RCP2.6, relative to the RCP2.6 with no AOA. Units are K.

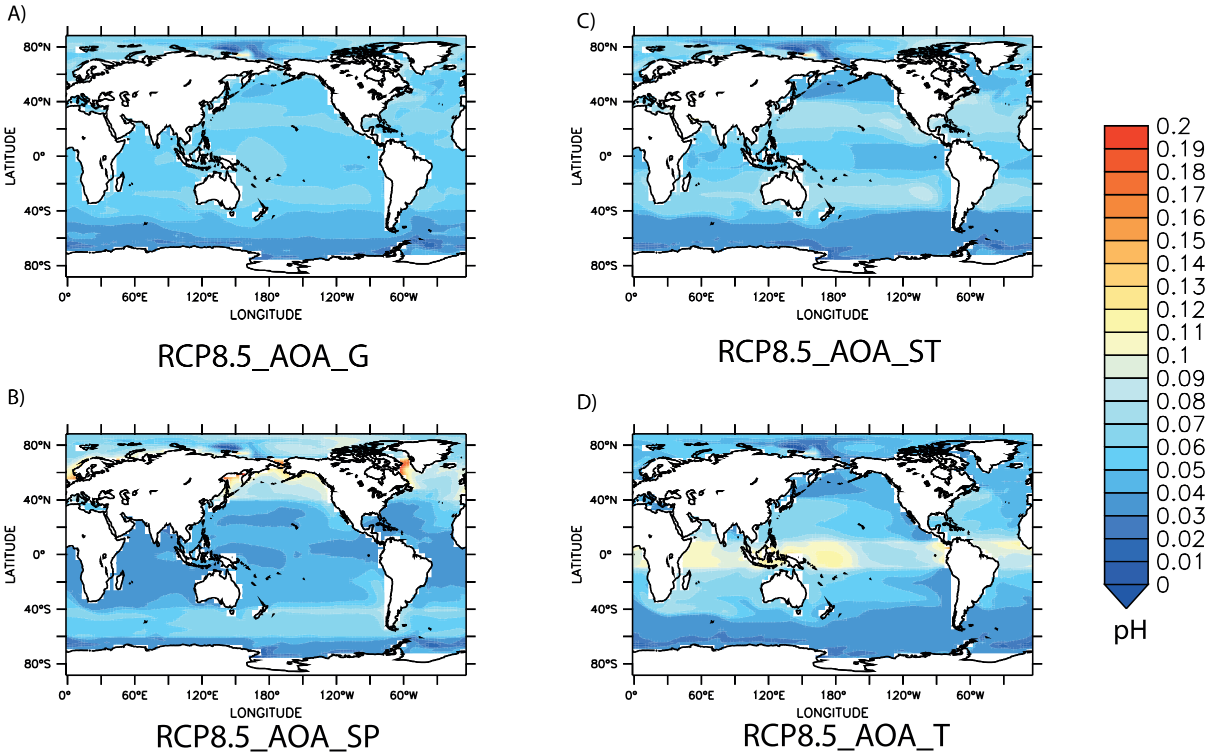
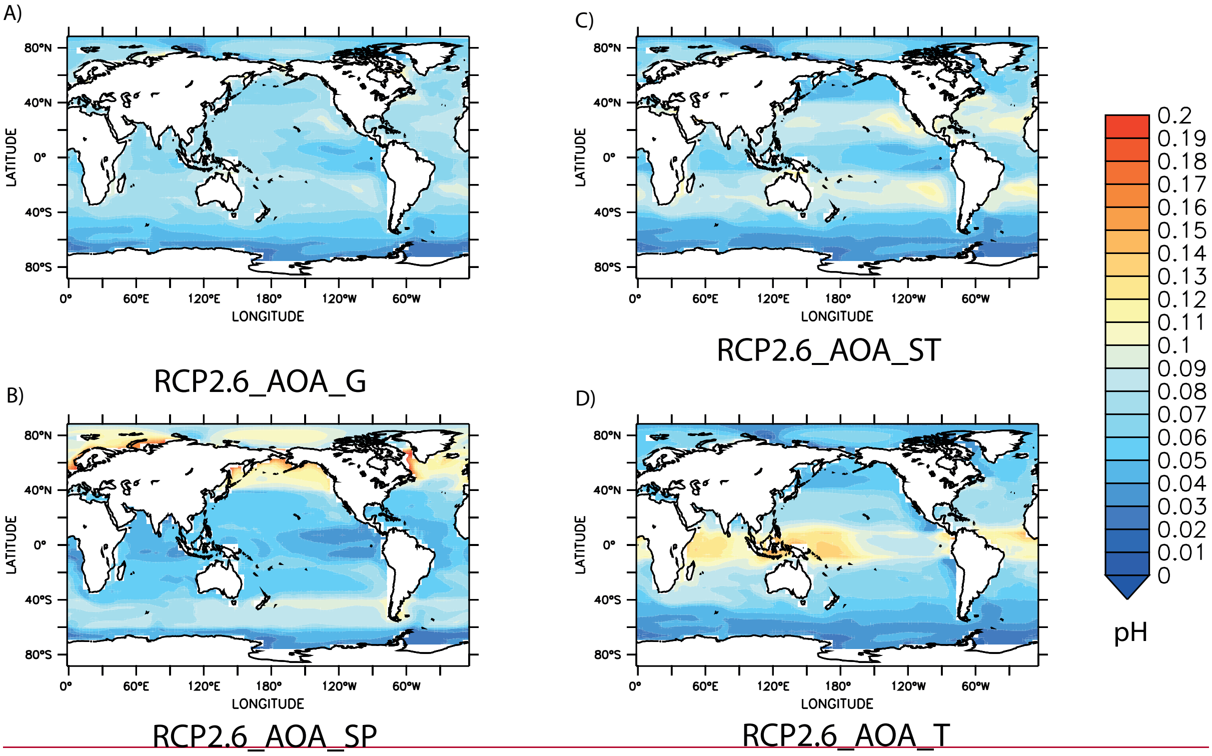


Figure 9 The spatial map of the ~~relative~~ changes in pH in 2090 (mean; 2081-2100) associated with global and regional AOA under RCP8.5, relative to RCP8.5 with no AOA.



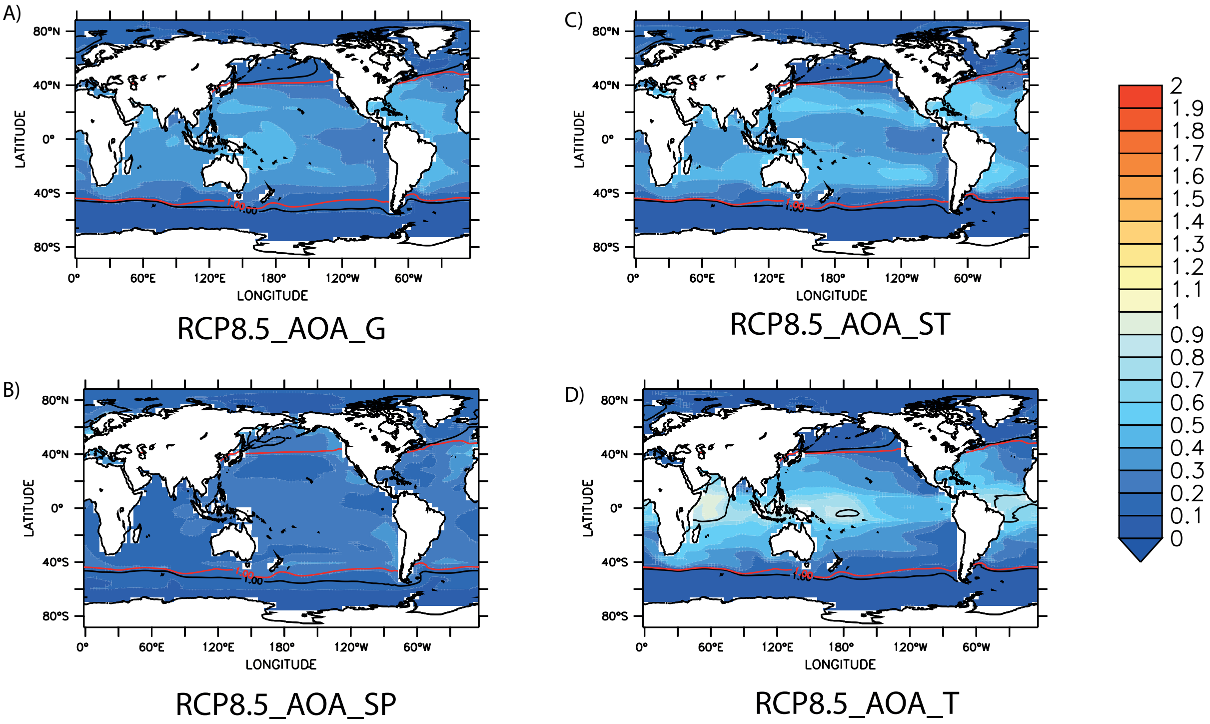
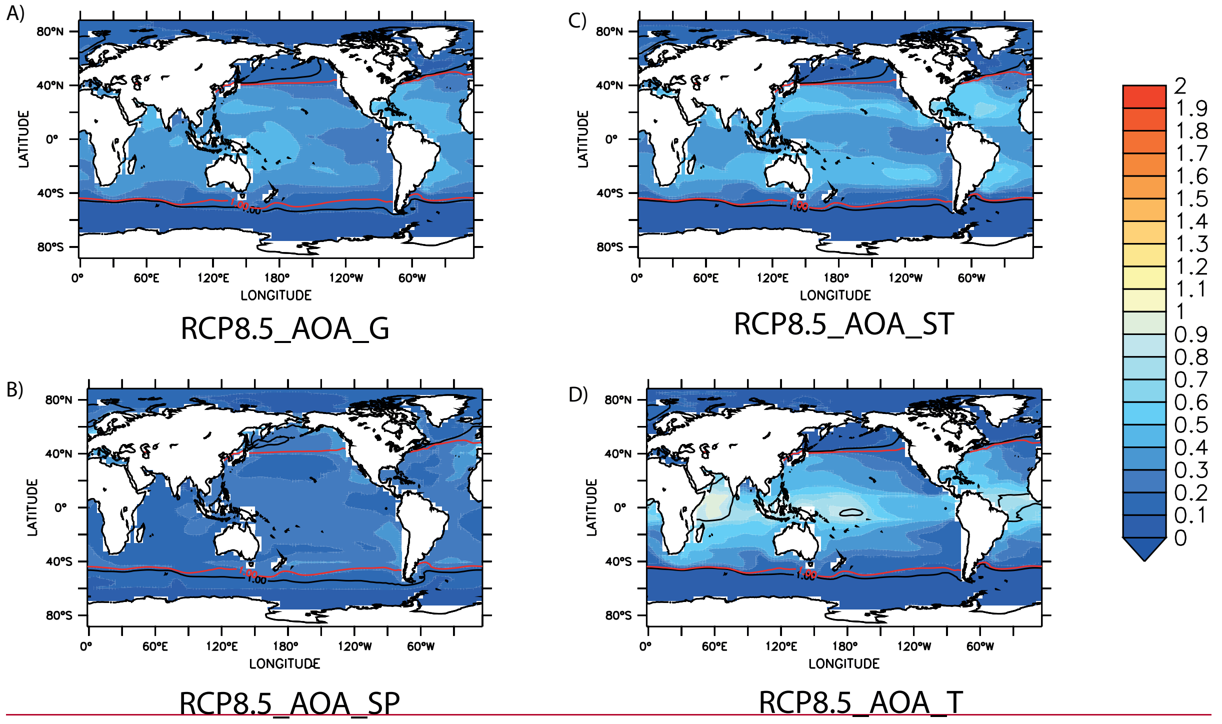
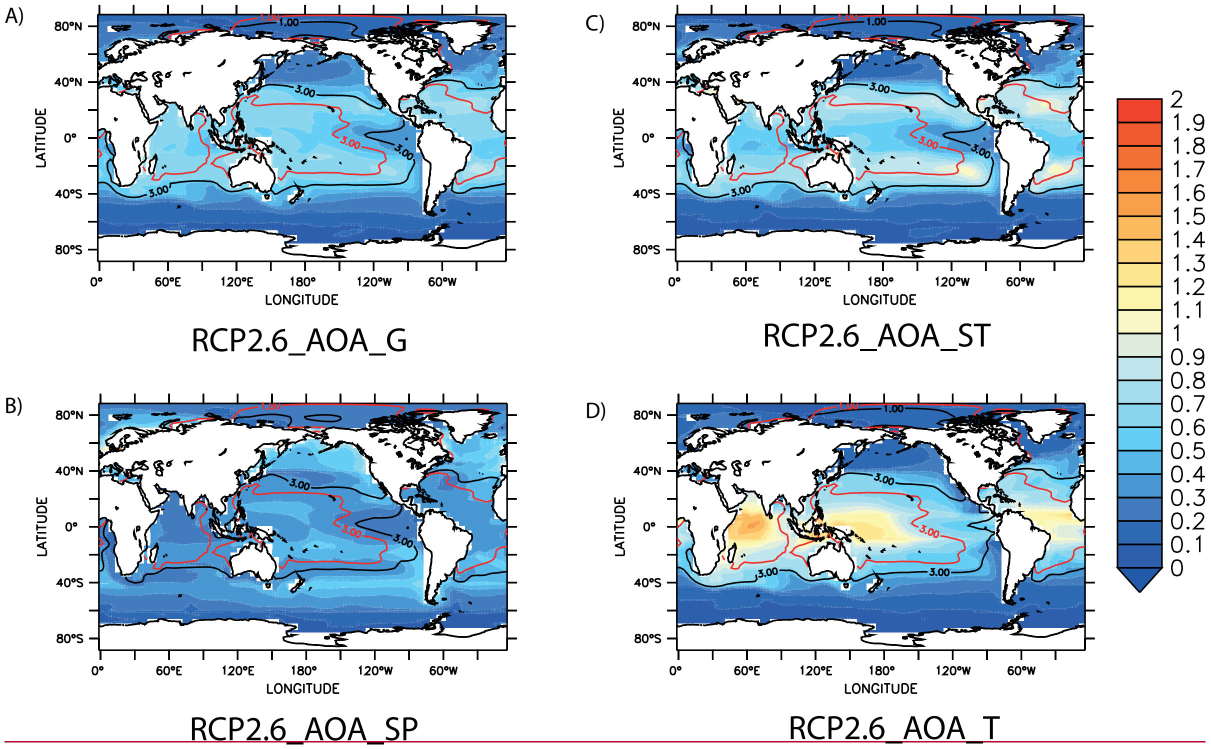


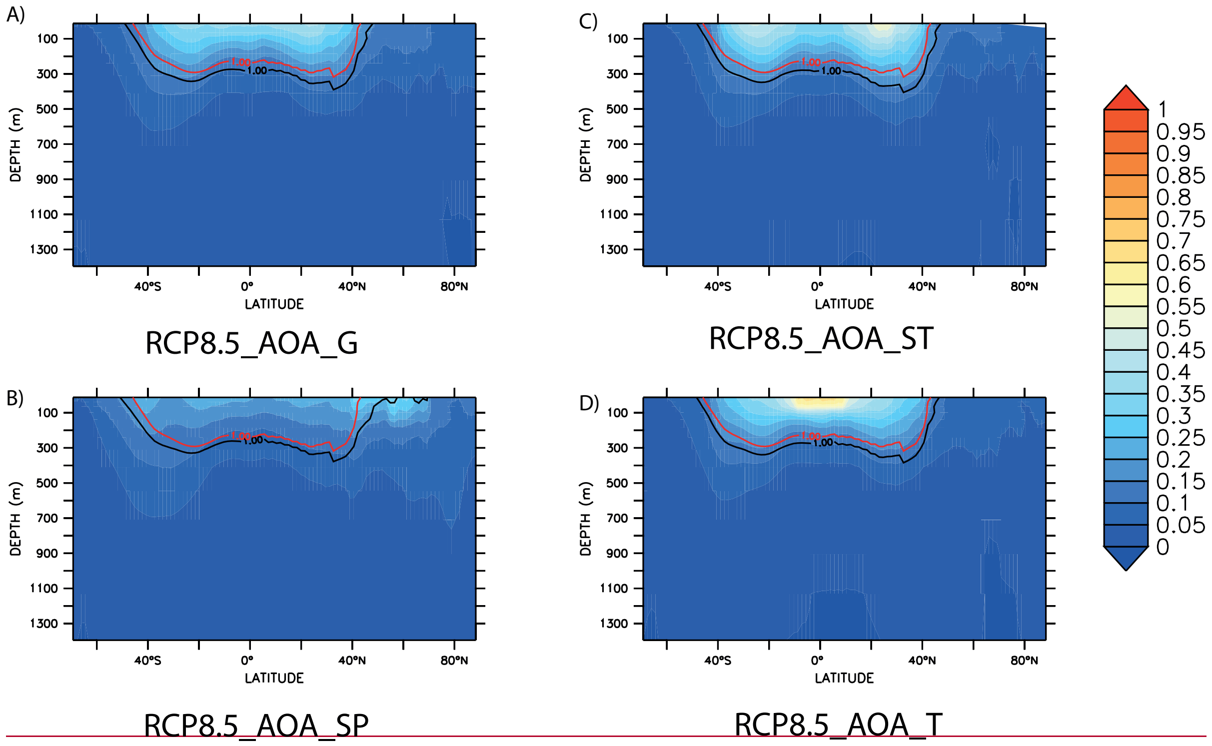
Figure 10 The spatial map of the ~~relative changes in pH in 2090 (mean; 2081-2100) associated with global and regional AOA under RCP2.6.~~



~~Figure 11 The spatial map of the relative differences in surface aragonite saturation state in 2090 (mean 2081-2100), associated with global and regional AOA under RCP8.5. Contoured on each map are the values of aragonite saturation state of 1 and 3, please see the text for more explanation. The red contours represent RCP8.5 and black AOA for each experiment~~



~~Figure 12 The spatial map of the relative~~ differences in surface aragonite saturation state in 2090 (mean~~2081-2100), associated with global and regional AOA under RCP2.6. Contoured on each map are the values of aragonite saturation state of 1 and 3, please see the text for more explanation. The red contours represent RCP2.6 and black AOA for each experiment~~



~~Figure 13 The relative zonal mean differences in aragonite saturation state in 2090 (mean;~~ 2081-2100), associated with global and regional AOA under RCP8.5~~.~~, relative to RCP8.5 with no AOA. Contoured on each map are the values of aragonite saturation state of 1 and 3; please see the text for more explanation. The red contours represent RCP8.5 without AOA and the black contours represent RCP8.5 with AOA for each experiment.

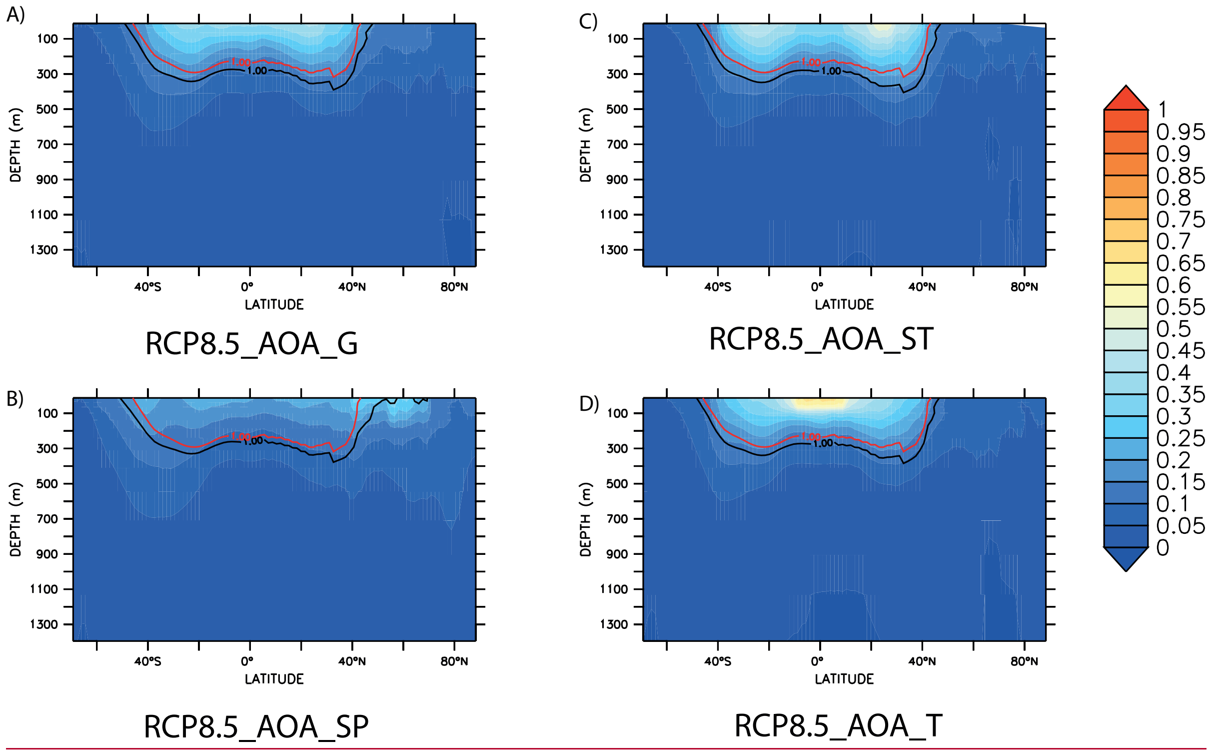


Figure 11 The zonal mean differences in aragonite saturation state in 2090 (mean; 2081-2100), associated with global and regional AOA under RCP8.5, relative to RCP8.5 with no AOA. Contoured on each map are the values of aragonite saturation state of 1~~,~~; please see the text for more explanation. The red contours represent RCP8.5 without AOA and the black contours represent RCP8.5 with AOA for each experiment~~.~~

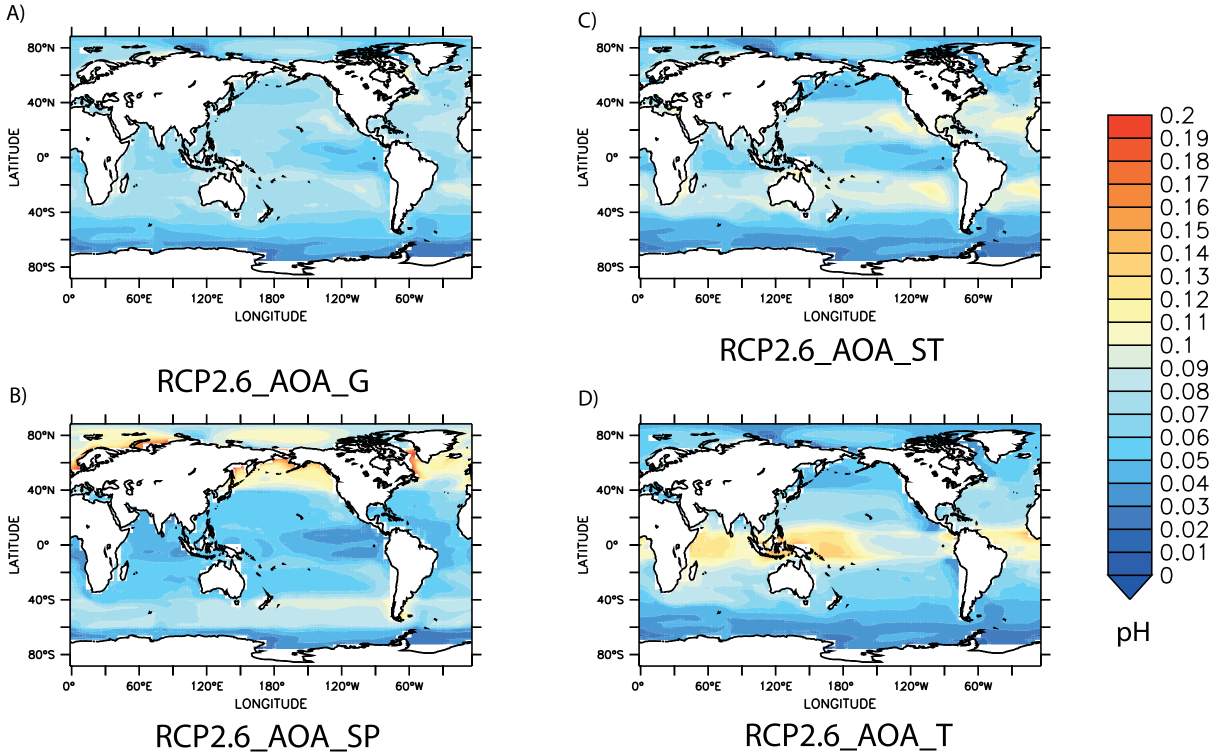


Figure 12 The spatial map of the changes in pH in 2090 (mean; 2081-2100) associated with global and regional AOA under RCP2.6, relative to RCP2.6 with no AOA.

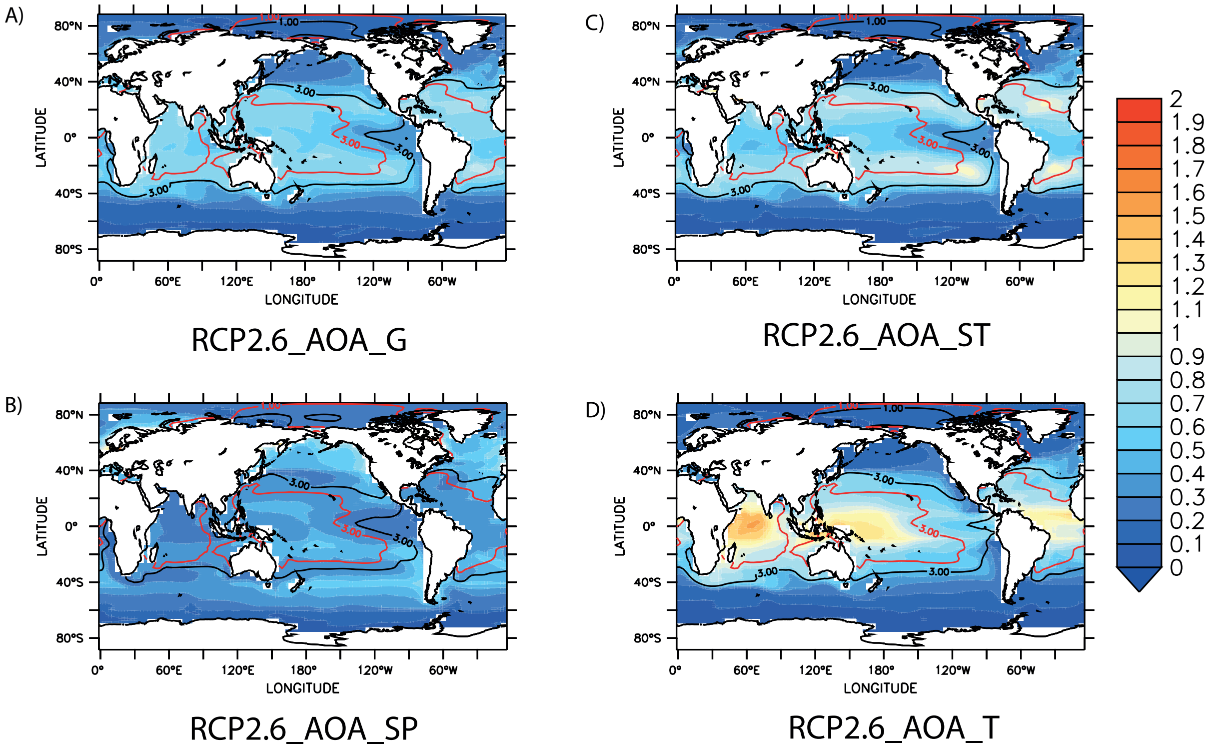


Figure 13 The spatial map of the differences in surface aragonite saturation state in 2090 (mean; 2081-2100), associated with global and regional AOA under RCP2.6, relative to RCP2.6 with no AOA. Contoured on each map are the values of aragonite saturation state of 1 and 3; please see the text for more explanation. The red contours represent RCP2.6 without AOA and the black contours represent RCP2.6 with AOA for each experiment

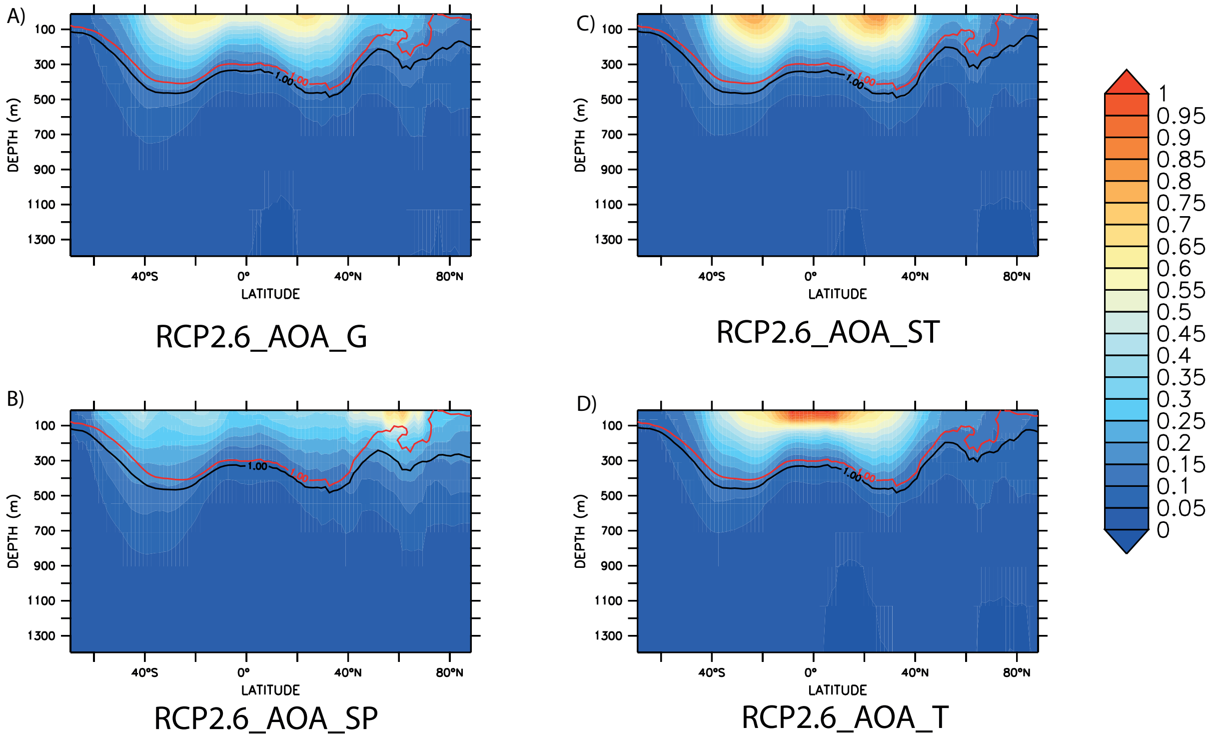


Figure 14 The ~~relative~~ zonal mean differences in aragonite saturation state in 2090 (mean; 2081-2100), associated with global and regional AOA under RCP2.6, relative to RCP2.6 with no AOA. Contoured on each map are the values of aragonite saturation state of 1~~,~~; please see the text for more explanation. The red contours represent RCP2.6 without AOA and the black contours represent RCP2.6 with AOA for each experiment~~.~~