

Dear Authors,
I am glad to accept your manuscript for publication after revisions. All reviewers, and myself are convinced of the validity of this study. I would like to point out, however, that my comments (mainly concerning typos and English) and some of the reviewers concerns (although addressed in your answers) were not taken into consideration in the submitted revised version of the manuscript. Further, all reviewers are still not entirely convinced with the assumptions used in the historical reconstruction (delta pCO₂ unchanged after correction for changes in atmospheric pCO₂) and demand a more critical discussion of the results. I would urge you to attend to the reviewer's previous and current comment in your answer as well as make sure the manuscript is modified accordingly (including proof reading) as this would speed up the review process.
Sincerely,
Christine Klaas

Dear Dr Klaas,

Please find our revised manuscript, and response to the reviewers in response to the request for minor corrections. We have worked hard to address the concerns in the current and previous review(s). We have removed the discussion of interannual variability and added details justifying our assumption of fixed air-sea disequilibria. I must humbly apologize for the numerous errors and typos in the version that I returned to you in the second round of reviews. I have been through the paper at length and I trust that you find it a much-improved manuscript. We have also worked at improving the figures in line with the reviewers' comments. Finally I must apologize for the delay in responding, with CSIRO in the process of dismantling its climate science work it been a very distracting (and troubling) time.

With Thanks

Andrew

Reviewer 1:

Review of Lenton et al: Historical reconstruction of ocean acidification in the Australian region

Lenton et al. have improved the manuscript considerably. They have addressed most comments of both reviewers. Yet, I'd like to ask the authors to work on their draft and uploaded files more thoroughly, there are still a number of typos, the abstract in the manuscript has not been updated as indicated in the answer to the reviewers and Figure 10 is still missing. See specific comments. The authors have to address these issues before publication of the manuscript.

Specific comments:

1) a) The abstract has been updated in the author comment response to reviewers. Yet, the abstract has not been updated in the main manuscript.

Apologies - this has now been updated

b) Please reconsider the use of the wording: "increase in ocean acidity levels"

(see: <http://www.imber.info/News/News/A-plea-to-ocean-acidification-scientists>:

It is unfortunate that the terminology used in some papers, presentations and media interviews is misleading. The definition of "acidic" in the Oxford English dictionary is "having the properties of an acid; having a pH of less than 7". Despite the process of ocean acidification (the acidity of seawater, or hydrogen ion concentration, has increased by 28% since preindustrial time), the oceans are alkaline (pH higher than 7) and will not become acidic in the foreseeable future in most regions of the oceans. Hence, while it is accurate to refer to an "increase in acidity" or to "ocean acidification", the terms "acid" or "acidic" should not be used when referring to seawater. Note that there are few exceptions, seawater can be acidic in some near-shore environments such as estuaries, in the immediate vicinity of CO₂ vents, or in purposeful perturbation experiments.)

Stating that there is an increase in acidity levels is very different than stating the waters are becoming acid and refers to

increasing hydrogen ion activity, which is happening. Stating the acidity level is increasing is a more correct statement than saying the ocean acidity is increasing, which is suggested as OK. Further, the Oxford dictionary may state acidic applies to waters with a pH less than 7, but this statement applies to infinitely dilute solutions and seawater is not infinitely dilute. The plea to change the wording on acidity is a good step, but the suggestions have issues and it is unfortunate that more marine chemists were not asked for input before issuing the plea. Editors should be aware it is a suggestion rather than a decree and could be improved.

c) In the last sentence of the abstract it says: "... an important to link biological observations...". This is grammatically incorrect and Reviewer 1 has mentioned this already in the first round of reviews. At least three of the five authors are native speakers and should be able to proofread their manuscript before submission.

We have remove 'to'

2) Response to reviewers, C5199 ("What about SOCAT data"/ method of Sasse et al): The authors state that SOCAT data is not used and refer to the next comment. In the answer to the next comment, they explain the method of Sasse et al., but not what the input data is. Are these pCO₂ data from SOCAT or from another source? Please indicate, what data is used in the main manuscript.

We now list the number of cruises (263) and time series sites (BATS and HOTS in text. As this comes from Table (A1) in Sasse et al (2013) we have cited this table, as this is published. Sasse et al (2013) in Table A1 further breaks the cruises down into the published datasets e.g. CARINA, PACIFICA, GLODAP etc and includes only data collected from 1980 onwards

Oceanic values of pCO₂ were taken from an updated version of Sasse et al. (2013) that used a self-organizing multiple linear output (SOMLO) approach to predict pCO₂ values around Australia on a 1° x 1° degree grid each month for the nominal year of 2000. The SOMLO approach utilizes a global network of bottle-derived pCO₂ and corresponding standard hydrographic parameters (SHP; temperature, salinity, dissolved oxygen and phosphate; N=17753), collected from more than 263 cruises and the HOTS and BATS time series sites, for more information on the individual cruise data please see Table A1 in Sasse et al (2013).....

3) p. 12, lines 349-353: These two sentences are opposing each other. Which stations are you referring to for "larger changes in the north" and "largest changes occur in the Tasman Sea and along the southern coast".

This has now been clarified and the text now reads:

There is also a strong latitudinal gradient in magnitude of the decrease, with larger changes occurring in Northern Australian waters (Ningaloo and North Stradbroke Island) and smaller changes to the south. However the largest decreases in aragonite saturation state (>0.6) have occurred in the Tasman Sea and south of Australia (e.g. Maria Island).

4) p. 12, line 354: Figure 10 is still missing.

This has been removed as per the previous comments, but due to an oversight the references to this figure and associated text was not adjusted.

5) p. 14, line 427ff: both reviewers have asked for a clear statement of the uncertainty for the assumption that Delta pCO₂ does not change. The authors have written more text and have clearly stated that Delta pCO₂ is assumed to be constant in time. They do, however, not discuss, how large the error is and what the implications are. Reviewer 2 wrote: "When combining this assumption with the atmospheric CO₂ record, this will give the upper limit of DIC increase and thereby the upper limits of aragonite saturation state decrease and pH decrease associated with the atmospheric CO₂ rise." This is the type of explicit statement that I still miss in the text.

The authors focus on pointing out that effects of primary production on pCO₂ will be small, which is probably true, however, biological effects are only one driver of Delta pCO₂. The other drivers should be addressed too, either explicitly or with a summarizing statement as the sentence from reviewer 2.

We agree that the time invariant Delta pCO₂ remains a limitation of this approach, which we openly acknowledge (in the methods and discussion) and as discussed in the previous response to reviewers we have removed the plots and discussion on interannual variability per se. However adding statement that the reviewer suggests in the text is clearly not correct, because it assumes that the disequilibria that we assume here is the maximum when in fact we simply do not know, due to the limitation of datasets that span the study period.

We have added the following text to the paper:

In this study we assumed that the seasonal air-sea disequilibrium ($\Delta p\text{CO}_2$) is seasonally time invariant i.e. no interannual variability. While some studies have argued this variability maybe important at shorter-term timescales e.g. Sitch et al (2015), it is less clear how important these are on longer time series e.g. McKinley et al (2011). This is further complicated as the existing products of oceanic $p\text{CO}_2$ fields e.g. Landschützer et al. (2014) only extend back in time several decades reflecting the limit of historical observations. Nevertheless to assess how important this term could be, we assumed an upper bound from the published study of McNeil and Matear (2008) who, in the more dynamically and biologically active Southern Ocean, calculated the error introduced by assuming a fixed (air-sea) disequilibrium term. From this study we can see that for an equivalent increase in (future) atmospheric CO_2 levels, that the error introduced into our calculation of pH and aragonite saturation state falls well within the reported uncertainty of our reconstruction.

6) Figures 5,6,7: The figures have been improved (x and y-axis labels and tick marks in Figures 5,6,7). The tick marks on the color scale are still hard to read, particularly in a BG type-set version of the figures which are generally very small. You can take out every other tick mark, but increase their size, that will enhance readability.

The same holds true for new Figure 8. It looks great on a full page, but it won't be published in that size.

Removing every second tick mark makes the results less useful, instead we have increased the font on the numbers, and this has been done for every figure requested

Please also increase x-and y-tick label marks in Figure 9. Main and minor ticks are not distinguishable in Figure 9.

Changed

Please also increase font size of ticks and labels in Figure 2. Figures 5+6 don't use the same color scale (5a: 1.8-4.2, but 6a: 1.5-4.2, 5b: 8.02-8.15, but 6b: 8.01-8.15)

This has been fixed.

Further technical comments:

p.2, line 53; than the preindustrial period → than in the preindustrial period or relative to the preindustrial period.

Fixed

p.3, line 85: here and at a number of places: e.g. (Shaw et al., 2012) → (e.g., Shaw et al., 2012)

p.4, line 105: dissolved inorganic carbon dioxide (DIC) → dissolved inorganic carbon (DIC)

p.4, line 114: mixed layer this region → mixed layer in this region throughout the manuscript: be consistent in using pCO₂ or pCO₂

p. 7, line 220: HadiSST represent → HadiSST represents

Fixed

line 222: local process → local processes

Fixed

p. 9, line 273: parenthesis around citation

Fixed

p. 10, line 310: “we can only compare our results T14 away from these regions” → compare our results TO T14...

Fixed

p. 11, line 327: T14 the product → the T14 product

Fixed

p. 14, please check your style of citations on the whole page.

p. 14, 416/7: freshwater water changes → freshwater changes

Fixed

p. 14, line 429: ... products the existing products... → the existing products

Fixed

p. 15, line 449: an new estimate --> a new estimate

Fixed

p. 16, line 487: impacts are likely vary → impacts likely vary

Fixed

Reviewer 2

In this manuscript by Lenton and colleagues, the authors reconstructed the evolution of pH and aragonite saturation state changes for the period between 1870-2013, using the present day ocean carbonate data in conjunction with atmospheric pCO₂ records for the same period.

The paper clearly and thoroughly describes the methods and the data used and the results obtained from analysis. I trust their honest efforts put into this work and thus I sincerely hope their efforts will finally come to fruition.

The manuscript consists of two parts. The first part is the production of present-day maps of pCO₂ and total alkalinity (TA), which were then used to calculate the similar maps of total carbon (DIC) and pH (or aragonite saturation state). I do not have any problem with this part. Certainly, the resulting products of pCO₂ and TA distribution are more accurate than the same maps produced by Takahashi et al., because the authors used a lot more data than did Takahashi et al.

However, I have great disquiet with the second part of this manuscript. In this part of the manuscript, the authors transformed the present-day yearly cycle of pCO₂ to yearly cycles for other years (years including all the way back to 1870) with the help of atmospheric pCO₂ records for 1870-2013. The underlying assumption in this analysis was that the present-day pattern of pCO₂ cycle in waters near Australia has remained unchanged for the analysis period (1870-2013). My wild guess is that their assumption may not be terribly wrong, but, at the same time, they may not be able to present convincing lines of evidence supporting that their assumption is reasonable. Therefore, I feel that this type of analysis is too much of a stretch of their present-day carbon observations to the past. I do find the second part of this manuscript (about the evolution of ocean acidification between 1870-2013), neither useful nor interesting.

To address the reviewers concern we now cite the paper by McNeil and Matear (2008) showing that error introduced by assumed a fixed air-sea disequilibrium, even under very large changes in to the future (IS92A) that the error introduced by this assumption is less than the uncertainty presented in our reconstruction.

The text now states:

In this study we assumed that the seasonal air-sea disequilibrium ($\Delta p\text{CO}_2$) is seasonally time invariant i.e. no interannual variability. While some studies have argued this variability maybe important at shorter-term timescales e.g. Sitch et al (2015), it is less clear how important these are on longer time series e.g. McKinley et al (2011). This is further complicated as the existing

products of oceanic pCO₂ fields e.g. Landschützer et al. (2014) only extend back in time several decades reflecting the limit of historical observations. Nevertheless to assess how important this term could be, we assumed an upper bound from the published study of McNeil and Matear (2008) who, in the more dynamically and biologically active Southern Ocean, calculated the error introduced by assuming a fixed (air-sea) disequilibrium term. From this study we can see that for an equivalent increase in (future) atmospheric CO₂ levels, that the error introduced into our calculation of pH and aragonite saturation state falls well within the reported uncertainty of our reconstruction.

I feel that the first part of this paper can stand by itself and thus still merit for publication. If the authors decide to do so, I have several technical comments. I also feel that the authors should greatly down play the second part of this paper.

With respect we strongly disagree.