nature portfolio

Peer Review File

Integrated Analysis of Carbon Dioxide and Oxygen Concentrations as a Quality Control of Ocean Float Data

Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality.

29th Oct 21

Dear Professor Tyrrell,

Please allow us to apologise for the delay in sending a decision on your manuscript titled "Comparing Carbon Dioxide and Oxygen Disequilibria reveals Ocean Processes and Sensor Performance". It has now been seen by 3 reviewers, and I include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file. Specifically, your response should 1) provide more information regarding the SOCCOM quality control process, 2) address the rationale for using dissolved carbon dioxide rather than other dominant carbonate species at ocean pH and 3) discuss real processes that could also cause the observed y-intercept discrepancies.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter) and the completed checklist:
[link redacted]
** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Annie Bourbonnais, PhD
Editorial Board Member
Communications Earth & Environment
orcid.org/0000-0001-7247-5230

Joe Aslin
Associate Editor
Communications Earth & Environment

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Editorial Policy: Policy requirements

Furthermore, please align your manuscript with our format requirements, which are summarized on the following checklist:
Communications Earth & Environment formatting checklist

and also in our style and formatting guide Communications Earth & Environment formatting guide .

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All Communications Earth & Environment manuscripts must include a section titled "Data Availability" at the end of the Methods section or main text (if no Methods). More information on this policy, is available at http://www.nature.com/authors/policies/data/data-availability-statements-data-citations.pdf.

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specific, community-recognized repositories, where possible and a list of recommended repositories is provided at http://www.nature.com/sdata/policies/repositories.

If a community resource is unavailable, data can be submitted to generalist repositories such as figshare or Dryad Digital Repository. Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

Please refer to our data policies at http://www.nature.com/authors/policies/availability.html.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Dear authors,

this is a nice work, explores the relationship between O2 and CO2 saturation in the surface ocean from a high quality data base as GLODAPv2.2020 and purposes this approach to detect biases in sensor derived CO2 data, in fact, directly measured sensor pH.

This a non trivial issue, as the BGC-argo commnuty is much worried about the pH QC procedure as clear unconsistencies were found between spectrophotometric pH measurements by different labs and also between measured and calculated pH from DIC and TA. Consequently a clear reference to compare sensor pH with is lacking and consequently derived CO2 data might be uncontrolled biased.

Consequences for our estimate of the CO2 balance or acidification estimate in the ocean are clear ... no agreement between platforms. Higher release of CO2 in the Southern Oc. might be the result of a bias in the sensor based (Corrected to.. whatever) pH.

COMMENTS
1- ABSTRACT:
1.A) whenever using the term "deviation" please add "air/atmospheric saturation deviation", in this way it clearer to the reader what You refer to
1.B) it might be clearer to state that " linear fits of DCO2 vs. DO2 in this high quality data set the y-intercept informs about .. bla bla ...
in this way the reader clearly knows which is the y and x

variables
1.C) At the end of the abstract instead of CORS it might be
more catching to expand the acronym and write a longer
but conclusive and decisive statement.
1.D)SOCCOM and GLODAP might be defined before use

2- whenever you use GLODAPv2 please refer to the corresponding year update .. in this case GLODAPv2.2020

3- INTRODUCTION
line 34 -- pCO2 it is not defined previously, but in line 35 (!)

- it might be worthy citing this paper in the second paragraph of the introduction
Peter G. Brewer and Edward T. Peltzer Limits to marine life. Science • 17 Apr 2009 • Vol 324, Issue 5925 • pp. 347-348 • DOI: 10.1126/science.1170756
- lines 53-54-- here you define CORS with ...Saturation but in the title you use the term "Disequilibrium".. it might be good unifying both.
- lines 57-60, it is nice to see a reference to GEOSECS .. but I guess for the sake of simplicity, just a reference to the suplement figure and a short comment of this FigS1 in the supplement might be better.
- I suggest a clear statement of the two main objectives of this work: 1) global evaluation of CORS, 2) CORS-like evaluation of SOCCOM O2 and pCO2 derived data.
- lines 78-82 .. a bit repetitive

4. RESULTS
4.1. OVERALL PATTERNS
I suggest exchanging Fig S2 with figure 1 in the main text, although it is interesting to see the spatial and temporal coverage of the sample data, FigS2 is more informative about processes. It is just a suggestion.

4.2. PROCESSES CAUSING ...
line 121.. please refer the reader to the corresponding section in the methods to explain the inset, here in the text or in the figure caption.
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I suppose the float data plotted here contain all "corrected" data both good and bad. Corrected means that pH has been corrected for bias as described in Williams et al
Figure 4 and 6 ...instead of providing the slope and intercept, as the slopes are not commented, it might be relevant to provide the intercept and the corresponding uncertainty.
Line 201 onwards--- I wonder if within the SOCCOM documentation for each float the pH offset applied is clearly stated, it would be very interesting to check whether those floats with identified by CORS as anomalous have or not any pH adjustment.

In Table 1. it is not clear the meaning of ** .. and in the methods neither. ** means the y intercpet is very different?

DISCUSSION
Please make clear the meaning of y_diff in TAble 1 and discuss which of the floats deviate more along with Fig6. Maybe this point should be placed in the results section. I miss a description of Fig 6 along with Table 1, for example, F9275 seems biased and the y_diff is 2.47 with no ** .. it means it is OK?

It might be worthy to comment on how the pH bias detection is done for the SOCCOM floats, as far as I know, they use crossover analysis for deep waters but also predictions from neural networks calculating pH from PRES, TEMP, SAL and O2.

I suggest performing a sort of simulacro for one or two very biased float where CORS works and check if the other SOCCOM QC methods also show the same bias.

Best regards

Reviewer #2 (Remarks to the Author):

Wu and coauthors compare patterns of surface concentrations of dissolved oxygen and carbon dioxide across the global ocean. They do this by computing concentrations of these gases relative to their saturation values, an approach they call CORS ("carbon and oxygen relative to saturation"). The authors present CORS plots (ΔCO₂ vs. ΔO₂) created from the Global Ocean Data Analysis Project dataset (GLODAPv2) and discuss the broad patterns observed. They also present CORS plots created from biogeochemical Argo floats deployed in the Southern Ocean as part of the Southern Ocean Carbon and Climate Observations and Modeling (SOCCOM) program. The authors make the case that CORS plots are useful for identifying erroneous data from autonomous platforms and that the y-intercepts of CORS plots derived from SOCCOM floats are on average higher than those derived from GLODAPv2.

The manuscript is well written and presents a useful strategy for applying the CORS method to oceanic waters, as it has been applied before to freshwaters (Vachon et al., 2020). The insights from the analysis of GLODAPv2 data are valuable in establishing how CORS plots should be evaluated and in elucidating a number of ocean processes. And the discussion of the utility of CORS plots for data QC is timely as autonomous platforms are relied upon more and more in studies of ocean biogeochemistry. I support the publication of this manuscript after addressing some minor concerns, which are organized below into general comments and line-by-line comments.

<B>General comments</B>

One point that I'm not completely clear on is why [CO₂] is more appropriate to use in this context than DIC. The authors argue that analyzing O₂ and CO₂ using the same units is useful for taking advantaging of stoichiometric relationships. However, due to the behavior of dissolved carbon dioxide, only a small percentage of the inorganic carbon utilized or produced by biogeochemical processes exists as dissolved CO₂ in seawater, rather it is

mostly HCO₃⁻ or CO₃²⁻. So, wouldn't DIC (which encompasses all dissolved inorganic carbon) be more useful in representing stoichiometric relationships? Of course, calculating [CO₂] relative to saturation is straightforward, while calculating DIC relative to its saturation value would require an estimate of total alkalinity. In any event, since stoichiometric relationships are referenced, I think a sentence or two discussing this issue may be helpful.

I'm not entirely convinced that a y-intercept from a SOCCOM float-based CORS plot that differs from the Southern Ocean average y-intercept is indicative of a problem with the float sensor performance. While an average of a large amount of data may have a y-intercept that crosses near the origin (or at -1.10 umol kg⁻¹ due to ice melt in the Southern Ocean), can it be said that measurements from one float in particular (in a limited region for a limited amount of time) should have the same intercept? Perhaps comparatively less ice melt has occurred in that float's spatiotemporal observation window, biasing the intercept high due to a real process rather than sensor bias. I'm not sure the subsampling procedure addresses this concern either, since it involves taking random subsets of GLODAPv2 data across the entire Southern Ocean, rather than limiting the subsets to a region and time period corresponding to the float observations. In short, I don't think the analysis presented here points definitively to biases in SOCCOM float pH sensors (though that certainly is a possibility), so some additional discussion of potential alternative sources of the y-intercept discrepancies would be valuable.

<B>Line-by-line comments</B>

Line 48: Vachon et al. (2020) state that they build upon the approach of Torgensen and Branco (2007), so perhaps also cite that paper here.

Lines 72–74: I'd suggest mentioning here the current push for a global biogeochemical Argo array (https://www.go-bgc.org), which will provide float coverage similar to that provided by the SOCCOM project across the globe. I see that this is included in the Discussion and Conclusions section, but I'd advocate for mentioning it here as well to emphasize upfront the importance of float carbonate data QC.

Lines 99–100: Eliminate extra phrase "in the Atlantic"

Lines 128–129: NO₃ may be so low in this quadrant due to photosynthetic uptake that correlation is difficult to assess.

Line 157: Capitalize "Figure"

Line 176: Move reference to Fig. 4 to the end of this sentence.

Lines 177–178: "CORS plots" rather than "CORS plot"

Line 190–192: How many of the float intercepts were significantly higher than the GLODAP-derived intercepts? I'm not sure I understand why the selected null hypothesis here was that the float y-intercept was greater than or equal to the GLODAP reference. From Table 1, it looks like the intercepts are relatively evenly distributed between greater than and less than -1.10 umol kg⁻¹.

Line 270: Minus sign in parentheses should be equals.

Line 286: ΔCO₂ should be [CO₂]

Figure 1: Could you add to this figure caption what ΔO₂ and ΔCO₂ represent (i.e., [CO_{2,obs}] – [CO_{2,sat}])

Line 578: I believe the period in this figure caption should be a semicolon.

<B>References</B>

Torgersen, T. and Branco, B., 2007. Carbon and oxygen dynamics of shallow aquatic systems: Process vectors and bacterial productivity. Journal of Geophysical Research: Biogeosciences, 112, 1–16.

Vachon, D. et al., 2020. Paired O₂–CO₂ measurements provide emergent insights into aquatic ecosystem function. Limnology and Oceanography Letters, 5, 287–294.

Jonathan D. Sharp
September 29, 2021

Reviewer #3 (Remarks to the Author):

The manuscript by Tyrell et al. is looking to address an important issue of quality control of the float pH data collected within the SOCCOM project. This work relies on using GLODAP, CO2SYS and LIAR datasets and data products, and a novel carbon dioxide and oxygen relative to saturation (CORS) method. The authors are using a method of comparing variations in [O2] and [CO2] and their deviations from the origin to the 'expected' relations to identify possible outliers or bad data. Although the method is well described from the theoretical perspective and is a useful tool to study the superposition of processes driving carbon and oxygen fluxes, it lacks the precision and accuracy required for quality control of a vast amount of the SOCCOM floats' pH data. While clear outliers and questionable data could be identified with the current QC procedures (e.g. Fig. 5), an additional level of QC as proposed is based on the (subjective) assumptions of seasonality, geographical division and processes governing [O2] and [CO2] variability. Applying such a procedure, therefore, seems like an additional tool at the level where a subset of SOCCOM data is used for a specific study; the CORS method is too crude to be used on a QC level. Below are a couple of lines of reasoning.
Beyond obvious outliers due to the sensor malfunctioning, pressure hysteresis, drift, etc., most of the questionable data resides in the upper 200m of the water column. This is the region where multiple processes can influence [O2] and [CO2] variability even with a presence of a dominant force. Therefore, constructing a robust relationship model is a priori challenging.
With a model that forces 'good' data to obey certain rules (y-intercept, slope), there is a risk of misidentifying potentially good data as 'questionable' or 'bad'. This is particularly important for the Southern Ocean, where a combination of vigorous mixing, air-sea exchange, deep water mixing may cause 'abnormal' signals in delta[O2] and delta[CO2] relationship.
The core problem of using [CO2] is that, as a part of the carbonate system, its variability cannot be linked directly to [O2], neither in the biotic (DIC will work here) nor in abiotic processes. While the

definition of departure from equilibrium has a concrete meaning of O2 @ 100% (S,T,P) – O2@ (S,T,P), the similar definition for CO2 cannot be constructed simply because it will have a different meaning at each state of the carbonate system (DIC, TA concentration for example). Coming back to air-sea exchange, delta[O2] and delta[CO2] will vary dependent on the strength of the flux, which is non-linear and different for both gases, and on the depth of the mixed layer, which will add CO2 and O2 in odd proportions – this is just the Southern Ocean case. Lastly, equilibration times of CO2 is roughly 10x slower than those of O2, therefore comparing instantaneous changes in [O2] and [CO2] (like the ones from the float) bears a risk of identifying both false-positives and false-negatives.

Response Letter

Please note that all the line numbers mentioned below refer to the track-and-change version, and response is in blue.

Editor's Comments:

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file. Specifically, your response should 1) provide more information regarding the SOCCOM quality control process, 2) address the rationale for using dissolved carbon dioxide rather than other dominant carbonate species at ocean pH and 3) discuss real processes that could also cause the observed y-intercept discrepancies.

Response: We thank the editor for evaluating our manuscript and providing us the opportunity to revise. As requested, we have resubmitted a revised manuscript (amendments shown with track-and-change) as well as a point-by-point response below. Specifically,

1) we provide more information regarding the SOCCOM QC in L301-308, as well as in our response to Reviewer #1;

2) we explain our rationale for using [CO_2] rather than other variables in L391-394, as well as in our response to Reviewers #2 and #3; in principle, using [CO_2] has advantages of straightforward relation to pCO_2 and air-sea CO_2 exchange and avoiding artificial errors during conversions (using CO2SYS) from other variables (e.g., DIC) to pCO_2; and

3) we acknowledge and discuss other real processes that could possibly cause deviations in the observed y-intercepts in L219-225, as well as in our response to Reviewer #2.

We also update the MS to include recently-published advances relevant to our paper: a few other studies have commented on the accuracy of the SOCCOM CO_2 flux estimates (L227-230, L260-261); they used different approaches from ours but came to a similar conclusion of float-overestimated CO_2 outgassing.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Dear authors,

this is a nice work, explores the relationship between O2 and CO2 saturation in the surface ocean from a high quality data base as GLODAPv2.2020 and purposes this approach to detect biases in sensor derived CO2 data, in fact, directly measured sensor pH.

This a non trivial issue, as the BGC-argo commnuty is much worried about the pH QC procedure as clear unconsistencies were found between spectrophotometric pH measurements by different labs and also between measured and calculated pH from DIC and TA. Consequently a clear reference to compare sensor pH with is lacking and consequently derived CO2 data might be uncontrolled biased.

Consequences for our estimate of the CO2 balance or acidification estimate in the ocean are clear ... no agreement between platforms. Higher release of CO2 in the Southern Oc. might be the result of a bias in the sensor based (Corrected to.. whatever) pH.

Response: we want to thank the reviewer for the positive assessments and insightful understanding of the Argo-related issues.

COMMENTS
1- ABSTRACT:
1.A) whenever using the term "deviation" please add "air/atmospheric saturation deviation", in this way it clearer to the reader what You refer to 1.B) it might be clearer to state that " linear fits of DCO2 vs. DO2 in this high quality data set the y-intercept informs about .. bla bla ... in this way the reader clearly knows which is the y and x variables 1.C) At the end of the abstract instead of CORS it might be more catching to expand the acronym and write a longer but conclusive and decisive statement. 1.D) SOCCOM and GLODAP might be defined before use

Response: A) changes made and applied throughout the manuscript;

B) we change to "*linear fits of CO_2 and O_2 deviations from atmospheric saturation (ΔCO_2 against ΔO_2) yield y-intercepts close to zero, suggesting a requirement for data validity*";

C) we change the last sentence to "*CORS analysis implies overestimations of float-based CO_2 release in the Southern Ocean; the technique can be applied to data from other autonomous platforms for quality assessment.*";

D) due to the word count limitation, we decide to define SOCCOM and GLODAP in the main text. In the abstract, we use 'ship-collected data' and 'float-collected data' to refer to these datasets.

2- whenever you use GLODAPv2 please refer to the corresponding year update .. in this case GLODAPv2.2020

Response: changes made in the way suggested. We note at the first time GLODAPv2.2020 appears that in the following text it is also referred to GLODAPv2. See L68: "GLODAPv2.2020[25-27]; used throughout this study, for simplicity, it is referred to as GLODAPv2 hereafter." Also in L276.

3- INTRODUCTION
line 34 -- pCO2 it is not defined previously, but in line 35 (!)
- it might be worthy citing this paper in the second paragraph of the introduction Peter G. Brewer and Edward T. Peltzer Limits to marine life. Science • 17 Apr 2009 • Vol 324, Issue 5925 • pp. 347-348 • DOI: 10.1126/science.1170756

Response: changes made and recommended paper cited.

- lines 53-54-- here you define CORS with ...Saturation but in the title you use the term "Disequilibrium".. it might be good unifying both.

Response: The title has been changed. It now reads:

Integrated Analysis of Carbon Dioxide and Oxygen reveals Ocean Processes and Sensor Performance.

The two terms are now unified in the introduction (L54-55).

- lines 57-60, it is nice to see a reference to GEOSECS .. but I guess for the sake of simplicity, just a reference to the supplement figure and a short comment of this FigS1 in the supplement might be better.

Response: we now move the description of GEOSECS O_2-temperature relationship to Supplementary Figure 1's caption.

- I suggest a clear statement of the two main objectives of this work: 1) global evaluation of CORS, 2) CORS-like evaluation of SOCCOM O2 and pCO2 derived data.

Response: we thank the reviewer for this suggestion. We have reformulated the final paragraph of the introduction to make a clear statement of the two main objectives, see L89-95.

- lines 78-82 .. a bit repetitive

Response: these sentences have been rewritten.

4. RESULTS

4.1. OVERALL PATTERNS

I suggest exchanging Fig S2 with figure 1 in the main text, although it is interesting to see the spatial and temporal coverage of the sample data, FigS2 is more informative about processes. It is just a suggestion.

Response: we agree that FigS2 is more informative about processes, and we have exchanged FigS2 with Fig1.

4.2. PROCESSES CAUSING ...

line 121.. please refer the reader to the corresponding section in the methods to explain the inset, here in the text or in the figure caption.

Response: information added to guide the readers.

line 143.. there is no Fig 3e, c?

Response: we are sorry for the typos, it is changed to Figure 3c.

4.3 NEAR ZERO y-INTERCEPT

line 161.. might be worthy to state here that the y-intercept corresponds to the value of DCO2 when DO2 = 0.

Response: we thank the reviewer for this suggestion, we now add definition of y-intercept which reads as: "value of ΔCO_2 when ΔO_2 is zero." See L176.

4.4 CORS PLOTS FROM ALL FLOAT DATA

I suppose the float data plotted here contain all "corrected" data both good and bad.

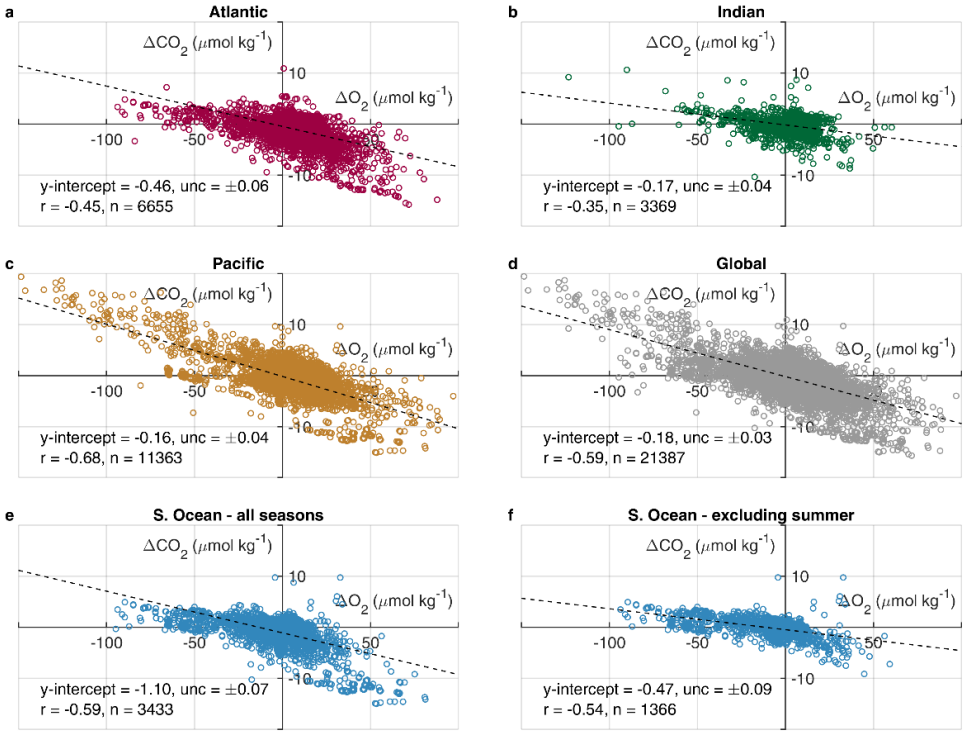
Response: yes, the float data used and plotted in this study were corrected for bias as described in Williams et al. (2017), and given quality flags 'good' or 'bad' depending on SOCCOM's quality assessment. As described previously in the Methods (sub-section titled 'CORS plots from all float data, regardless of QC flag'), for Figure 5 we used all available data including both good and bad, using different symbols for the two.

Corrected means that pH has been corrected for bias as described in Williams et al Figure 4 and 6 ...instead of providing the slope and intercept, as the slopes are not commented, it might be relevant to provide the intercept and the corresponding uncertainty.

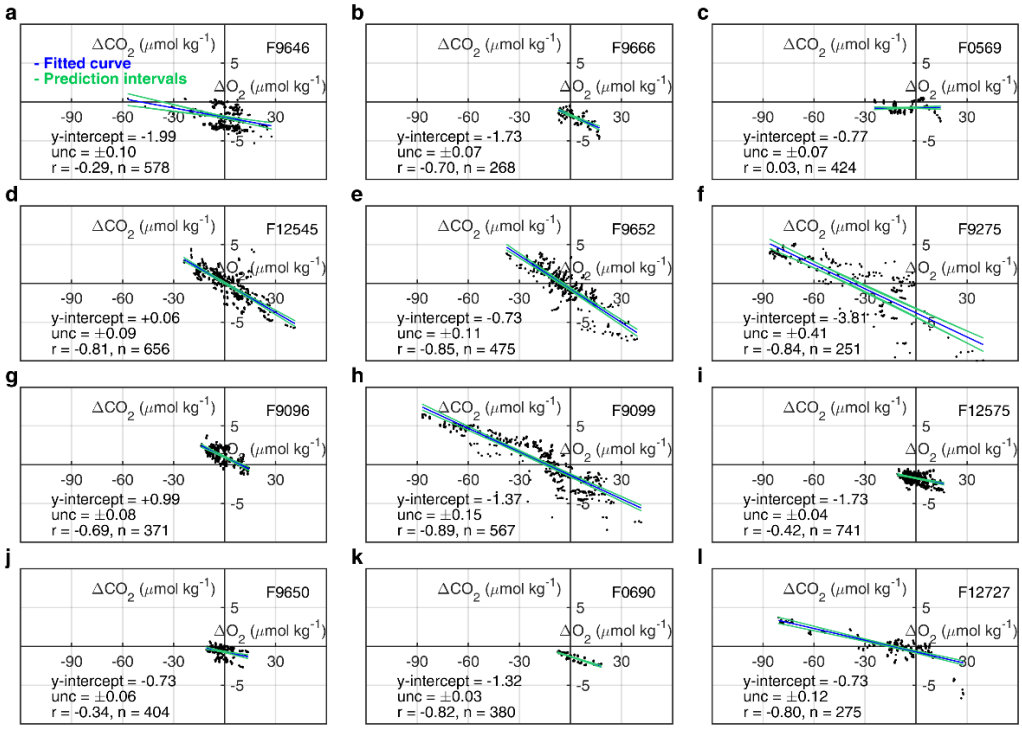
Response: we thank the reviewer for this constructive suggestion. We agree with the reviewer that providing intercept and the associated uncertainty makes our point more straightforwardly and that information on slopes can be omitted. We have now revised Figs. 4 and 6 in the way suggested (see below).

In addition, we have replaced Fig. 6 with the all-season version. The original Fig. 6 incorrectly excluded summer data. This issue only affected this figure. Analyses and text were correctly based on data from all seasons.

The revised Figure 4:



The revised Figure 6:



Line 201 onwards--- I wonder if within the SOCCOM documentation for each float the pH offset applied is clearly stated, it would be very interesting to check whether those floats with identified by CORS as anomalous have or not any pH adjustment.

Response: We agree with the reviewer that it would be interesting and more straightforward to check how SOCCOM identified and calibrated suspicious data. Unfortunately, in the SOCCOM data centre, there is no such documentation explaining how they did the adjustments. It is only noted that "Calibration for BGC sensors follows methods described in Johnson et al. (2017)". Each float with its entire profile in the SOCCOM dataset has been quality-controlled and adjusted (Page 3 in Williams et al., 2017), but the data are only given a quality flag. It is not like the GLODAP dataset, for instance, which includes an adjustment table providing all the calibration details. From our experience of using SOCCOM data, we found that some conspicuous errors (e.g., questionable data in Figure 5) are correctly flagged as 'bad'; however, other potentially erroneous data (as identified in this study) are not.

In Table 1. it is not clear the meaning of ** .. and in the methods neither. ** means the y intercpet is very different?

Response: To avoid confusion, we revised the text in the Methods (L408-416), deleted ** in Table 1, and revised * to: **GLODAP-derived y-intercepts minus float-derived y-intercepts; negative (positive) values mean that the float y-intercept is greater (lower) than the GLODAP y-intercept.*

The same revisions were applied to Supplementary Table 2.

DISCUSSION

Please make clear the meaning of y_diff in TAble 1 and discuss which of the floats deviate more along with Fig6. Maybe this point should be placed in the results section. I miss a description of Fig 6 along with Table 1, for example, F9275 seems biased and the y_diff is 2.47 with no ** .. it means it is OK?

Response: the note explaining y_diff in Table 1 is revised. We have now added information and description as to which floats deviate more strongly (L205-210). It now reads:

"We adopted a subsampling strategy (see Methods) to treat the GLODAP and float datasets identically, with the result showing that half of the selected floats have y-intercepts greater than the GLODAP-derived value of -1.10 µmol kg^{-1} (Fig. 6, Table 1). Among these floats, F9096 and F12545 deviate more strongly from the GLODAP pattern along the positive y-axis direction, whereas some other floats, e.g., F9275 and F9646 deviate along the opposite direction (Fig. 6). The average difference in y-intercepts (calculated as GLODAPv2 minus float values) is -0.12 µmol kg^{-1}, implying that, overall, float y-intercepts are more positive (or less negative) than GLODAP ones (Table 1)."

It might be worthy to comment on how the pH bias detection is done for the SOCCOM floats, as far as I know, they use crossover analysis for deep waters but also predictions from neural networks calculating pH from PRES, TEMP, SAL and O2.

Response: As suggested, we have now added more description of how pH bias detection and quality control is done for the SOCCOM floats (L301-308).

It should be noted that, even after correction following their procedures, SOCCOM pCO_2 estimates are still biased high by 4 µatm compared to shipboard measurements (Bushinsky et al., 2019; Gray et al., 2018). Although Bushinsky et al (2019) declared that such a bias is too small to influence their estimates of CO_2 flux, Long et al. (2021) (Figure 4 therein) showed that a systematic 4 µatm offset doubles the air-sea CO_2 flux estimate. And even if a -4 µatm correction is applied to all data, the SOCCOM-estimated flux still deviates far from shipboard measurements and aircraft observations (Long et al., 2021). The biased CO_2 flux issue has also been investigated by a few other studies (Mackay and Watson, 2021; Sutton et al., 2021); they used different approaches, independent of ours, with both indicating overestimation of SOCCOM CO_2 fluxes. We added a few sentences in L227-230 and L260-261 to discuss this:

"Our finding is in line with some recent studies[47-49] based on different approaches (airborne observations of atmospheric CO_2 gradients, uncrewed surface vehicle observations of circumnavigation of Antarctica, and reconstructed estimates of winter observations and CO_2 fluxes) that indicate possible overestimation of CO_2 outgassing from SOCCOM float data."

and:

"Our approach provides a more straightforward way to assess and potentially improve CO_2 data quality by comparison to other float measurements."

I suggest performing a sort of simulacro for one or two very biased float where CORS works and check if the other SOCCOM QC methods also show the same bias.

Response: We acknowledge this constructive comment. This is a big issue and requires plenty of analyses which is beyond the scope of this study. Our study provides more of a diagnostic way to evaluate SOCCOM dataset, rather than a robust correction way (more works are still needed to achieve this goal). One of our co-authors, Dorothee Bakker, has been looking into the algorithm approach (e.g., MLR and LIAR) that SOCCOM used to calibrate pH, and she has some misgivings of such method when applying in the data sparse subantarctic eastern Pacific waters. This work is currently in preparation and she prefers to keep it confidential.

Since we do not have access to all of the original data SOCCOM used (data from SOCCOM deployment cruises and their Southern Ocean data atlas), it is difficult to reproduce their exact QC process. The QC methods have been reported to apply to

each float, and a constant offset of +0.0054 pH units (on average) is added to the float data regardless of the vertical variability of this offset (Álvarez et al., 2020; Williams et al., 2017). This is a problem particularly when we are analyzing the surface ocean data and it probably contributes to the y-intercept discrepancies.

References:

Álvarez, M., Fajar, N. M., Carter, B. R., Guallart, E. F., Pérez, F. F., Woosley, R. J., & Murata, A. (2020). Global ocean spectrophotometric pH assessment: consistent inconsistencies. *Environmental Science & Technology, 54*(18), 10977-10988. doi:10.1021/acs.est.9b06932

Bushinsky, S. M., Landschutzer, P., Rodenbeck, C., Gray, A. R., Baker, D., Mazloff, M. R., . . . Sarmiento, J. L. (2019). Reassessing Southern Ocean air-sea CO2 flux estimates with the addition of biogeochemical float observations. *Global Biogeochem Cycles, 33*(11), 1370-1388. doi:10.1029/2019GB006176

Gray, A. R., Johnson, K. S., Bushinsky, S. M., Riser, S. C., Russell, J. L., Talley, L. D., . . . Sarmiento, J. L. (2018). Autonomous biogeochemical floats detect significant carbon dioxide outgassing in the high-latitude Southern Ocean. *Geophysical Research Letters, 45*(17), 9049-9057. doi:10.1029/2018GL078013

Long, M. C., Stephens, B. B., McKain, K., Sweeney, C., Keeling, R. F., Kort, E. A., . . . Wofsy, S. C. (2021). Strong Southern Ocean carbon uptake evident in airborne observations. *Science, 374*(6572), 1275-1280. doi:10.1126/science.abi4355

Mackay, N., & Watson, A. (2021). Winter air-sea CO2 fluxes constructed from summer observations of the Polar Southern Ocean suggest weak outgassing. *Journal of Geophysical Research: Oceans*, e2020JC016600. doi:10.1029/2020JC016600

Sutton, A. J., Williams, N. L., & Tilbrook, B. (2021). Constraining Southern Ocean CO2 Flux Uncertainty Using Uncrewed Surface Vehicle Observations. *Geophysical Research Letters, 48*(3), e2020GL091748. doi:10.1029/2020GL091748

Reviewer #2 (Remarks to the Author):

Wu and coauthors compare patterns of surface concentrations of dissolved oxygen and carbon dioxide across the global ocean. They do this by computing concentrations of these gases relative to their saturation values, an approach they call CORS ("carbon and oxygen relative to saturation"). The authors present CORS plots (ΔCO_2 vs. ΔO_2) created from the Global Ocean Data Analysis Project dataset (GLODAPv2) and discuss the broad patterns observed. They also present CORS plots created from biogeochemical Argo floats deployed in the Southern Ocean as part of the Southern Ocean Carbon and Climate Observations and Modeling (SOCCOM) program. The authors make the case that CORS plots are useful for identifying erroneous data from autonomous platforms and that the y-intercepts of CORS plots derived from SOCCOM floats are on average higher than those derived from GLODAPv2.

The manuscript is well written and presents a useful strategy for applying the CORS method to oceanic waters, as it has been applied before to freshwaters (Vachon et al., 2020). The insights from the analysis of GLODAPv2 data are valuable in establishing how CORS plots should be evaluated and in elucidating a number of ocean processes. And the discussion of the utility of CORS plots for data QC is timely as autonomous platforms are relied upon more and more in studies of ocean biogeochemistry. I support the publication of this manuscript after addressing some minor concerns, which are organized below into general comments and line-by-line comments.

Response: We are grateful to the reviewer for the positive evaluation of our manuscript.

General comments

One point that I'm not completely clear on is why [CO_2] is more appropriate to use in this context than DIC. The authors argue that analyzing O_2 and CO_2 using the same units is useful for taking advantaging of stoichiometric relationships. However, due to the behavior of dissolved carbon dioxide, only a small percentage of the inorganic carbon utilized or produced by biogeochemical processes exists as dissolved CO_2 in seawater, rather it is mostly HCO_3^- or CO_3^{2-}. So, wouldn't DIC (which encompasses all dissolved inorganic carbon) be more useful in representing stoichiometric relationships? Of course, calculating [CO_2] relative to saturation is straightforward, while calculating DIC relative to its saturation value would require an estimate of total alkalinity. In any event, since stoichiometric relationships are referenced, I think a sentence or two discussing this issue may be helpful.

Response: We thank the reviewer for this comment. A lot of our former efforts have been focused on deciding which one ([CO_2] or DIC) to use. As the reviewer

acknowledges, there are arguments either way. Our reasons for favouring CO_2 include: (1) the principle behind our method, comparing deviations from atmospheric equilibrium of two dissolved gases, is more easily appreciated when both gases are plotted, rather than one gas and one associated parameter; and (2) as the reviewer notes, going from CO_2 to DIC requires alkalinity, so is less straightforward. If we use DIC and a DIC-based y-intercept, we have to convert the y-intercept deviation back to pCO_2 and $[CO_2]$ for a quantitative purpose, during which the uncertainty (from dissociation constants and CO2SYS; Orr et al., 2018) is introduced. That is to say, by directly using $[CO_2]$, we can avoid some artificial errors.

We have added a couple of sentences in L391-394 to discuss this issue:

"*Although the Redfield ratio refers to changes in DIC and $[O_2]$, our choice of plotting $[CO_2]$ rather than DIC versus $[O_2]$ has advantages: 1) the y-axis value on the CORS plot relates directly to the tendency for air-sea CO_2 exchange to occur; 2) assessing $[CO_2]$ relative to saturation is quite straightforward whereas assessing [DIC] relative to its saturation value requires an additional step involving alkalinity.*"

I'm not entirely convinced that a y-intercept from a SOCCOM float-based CORS plot that differs from the Southern Ocean average y-intercept is indicative of a problem with the float sensor performance. While an average of a large amount of data may have a y-intercept that crosses near the origin (or at -1.10 umol kg^{-1} due to ice melt in the Southern Ocean), can it be said that measurements from one float in particular (in a limited region for a limited amount of time) should have the same intercept? Perhaps comparatively less ice melt has occurred in that float's spatiotemporal observation window, biasing the intercept high due to a real process rather than sensor bias. I'm not sure the subsampling procedure addresses this concern either, since it involves taking random subsets of GLODAPv2 data across the entire Southern Ocean, rather than limiting the subsets to a region and time period corresponding to the float observations. In short, I don't think the analysis presented here points definitively to biases in SOCCOM float pH sensors (though that certainly is a possibility), so some additional discussion of potential alternative sources of the y-intercept discrepancies would be valuable.

Response: We thank the reviewer for this very constructive and insightful comment, and we agree that additional discussion of this issue is needed. We accept the reviewer's point. In fact, ongoing work in our group (by PhD student Amavi Silva in Southampton) is investigating persistent CORS y-intercept offsets in the Labrador Sea and finding them to be caused by mixing with deep waters (winter mixing extends to great depths at this location). This work will be the subject of a separate paper and is too lengthy to be included in this paper. We have however modified the text of this current paper in light of the results of the Labrador Sea study and the reviewer's suggestion, L219-225:
"*While unusual y-intercept values are suggestive of data quality issues, they are not necessarily definitive proof. It is also possible, we believe, that, in some locations,*

local processes produce real CORS patterns that differ from those normally seen. For instance, surface water near to where rivers enter the sea, or in regions of strong mixing with deeper waters, could potentially exhibit persistent unusual CORS patterns. Data displaying unusual behaviour on CORS plots should therefore not be immediately discounted but should instead be flagged as requiring further investigation before it can be accepted as valid."

Line-by-line comments

Line 48: Vachon et al. (2020) state that they build upon the approach of Torgensen and Branco (2007), so perhaps also cite that paper here.

Response: citation added.

Lines 72–74: I'd suggest mentioning here the current push for a global biogeochemical Argo array (https://www.go-bgc.org) which will provide float coverage similar to that provided by the SOCCOM project across the globe. I see that this is included in the Discussion and Conclusions section, but I'd advocate for mentioning it here as well to emphasize upfront the importance of float carbonate data QC.

Response: We edited to L80-84:

"Funding has recently been announced (https://www.us-ocb.org/implementation-of-the-global-ocean-biogeochemistry-go-bgc-array-request-for-community-engagement/) for the construction and deployment of 500 floats (as a contribution towards an anticipated eventual fleet of 1000 floats) to provide float coverage similar to that provided by the SOCCOM project but across the global ocean."

Lines 99–100: Eliminate extra phrase "in the Atlantic"

Response: revised.

Lines 128–129: NO_3 may be so low in this quadrant due to photosynthetic uptake that correlation is difficult to assess.

Response: But if nutrients are always low (as in oligotrophic oceans) then we would expect an absence of blooms and hence CORS plots in which all points lie close to the origin (as in our Supplementary Figure 8).

Line 157: Capitalize "Figure"

Response: revised.

Line 176: Move reference to Fig. 4 to the end of this sentence.

Response: revised.

Lines 177–178: "CORS plots" rather than "CORS plot"

Response: revised.

Line 190–192: How many of the float intercepts were significantly higher than the GLODAP-derived intercepts? I'm not sure I understand why the selected null hypothesis here was that the float y-intercept was greater than or equal to the GLODAP reference. From Table 1, it looks like the intercepts are relatively evenly distributed between greater than and less than -1.10 umol kg^{-1}.
Response: Among the selected 12 floats with better overlaps with shipboard measurements, half of them have higher/greater y-intercepts, but on average y-intercept is greater than -1.10 umol kg^{-1}. And from all the QCed floats in the high-latitude (south of 55°S) Southern Ocean, 28 out of 48 have greater y-intercepts. On average, it is implied a 0.36 umol kg^{-1} overestimation by float measurements. We did not present the way we ran the subsampling well in the previous version, we have now revised and the null hypothesis is "*float-derived y-intercept is less than or equal to the GLODAP-derived y-intercept*".

Line 270: Minus sign in parentheses should be equals.
Response: revised.

Line 286: ΔCO_2 should be $[CO_2]$
Response: revised.

Figure 1: Could you add to this figure caption what ΔO_2 and ΔCO_2 represent (i.e., $[CO_{2,obs}] - [CO_{2,sat}]$)
Response: We replaced Fig. 1 with supplementary Fig. 2 as suggested by one of the reviewers, because supplementary Fig. 2 is more informative about processes. The suggested change has been made to the new supplementary Fig. 2.

Line 578: I believe the period in this figure caption should be a semicolon.
Response: revised.

References:

Long, M. C., Stephens, B. B., McKain, K., Sweeney, C., Keeling, R. F., Kort, E. A., . . . Wofsy, S. C. (2021). Strong Southern Ocean carbon uptake evident in airborne observations. *Science, 374*(6572), 1275-1280. doi:10.1126/science.abi4355

Mackay, N., & Watson, A. (2021). Winter air-sea CO2 fluxes constructed from summer observations of the Polar Southern Ocean suggest weak outgassing. *Journal of Geophysical Research: Oceans*, e2020JC016600. doi:10.1029/2020JC016600

Sutton, A. J., Williams, N. L., & Tilbrook, B. (2021). Constraining Southern Ocean CO2 Flux Uncertainty Using Uncrewed Surface Vehicle Observations. *Geophysical Research Letters, 48*(3), e2020GL091748. doi:10.1029/2020GL091748

Reviewer #3 (Remarks to the Author):

The manuscript by Tyrrell et al. is looking to address an important issue of quality control of the float pH data collected within the SOCCOM project. This work relies on using GLODAP, CO2SYS and LIAR datasets and data products, and a novel carbon dioxide and oxygen relative to saturation (CORS) method. The authors are using a method of comparing variations in [O2] and [CO2] and their deviations from the origin to the 'expected' relations to identify possible outliers or bad data. Although the method is well described from the theoretical perspective and is a useful tool to study the superposition of processes driving carbon and oxygen fluxes, it lacks the precision and accuracy required for quality control of a vast amount of the SOCCOM floats' pH data. While clear outliers and questionable data could be identified with the current QC procedures (e.g. Fig. 5), an additional level of QC as proposed is based on the (subjective) assumptions of seasonality, geographical division and processes governing [O2] and [CO2] variability. Applying such a procedure, therefore, seems like an additional tool at the level where a subset of SOCCOM data is used for a specific study; the CORS method is too crude to be used on a QC level. Below are a couple of lines of reasoning.

Response: We thank the reviewer for assessing CORS as a useful tool for studying biogeochemical processes, and for the positive evaluation of CORS on detecting clear outliers and questionable data. We acknowledge, as commented by the reviewer, that the CORS method in its current form/stage is not precise enough for correction of the SOCCOM dataset. As described in the response to reviewer 2, we have revised the MS to mention possible limitations to the CORS approach in its current form. The CORS approach described and applied in this study has been shown capable of identifying low quality float data; it is useful as a diagnostic tool for quality checks. This is the reason that we do not look to correct SOCCOM CO_2 data or flux estimates in this manuscript. Future research, such as the work described above in the Labrador Sea, will lead to improvements in CORS analysis as a robust QC tool.

We note that some recent papers, published after we carried out our study and arrived at our conclusions, independently came to similar conclusions with respect to concerns about SOCCOM CO_2 data. These recent studies (Long et al., 2021; Mackay and Watson, 2021; Sutton et al., 2021) used different approaches (e.g., airborne observations of atmospheric CO_2 gradients, reconstructed estimates of winter observations and CO_2 fluxes, and uncrewed surface vehicle observations during circumnavigation of Antarctica) (L227-230, L260-261).

Beyond obvious outliers due to the sensor malfunctioning, pressure hysteresis, drift, etc., most of the questionable data resides in the upper 200m of the water column. This is the region where multiple processes can influence [O2] and [CO2] variability even with a presence of a dominant force. Therefore, constructing a robust relationship model is a priori challenging. With a model that forces 'good' data to obey certain rules (y-intercept, slope), there is a risk of misidentifying potentially

good data as ‘questionable’ or ‘bad’. This is particularly important for the Southern Ocean, where a combination of vigorous mixing, air-sea exchange, deep water mixing may cause ‘abnormal’ signals in delta[O2] and delta[CO2] relationship.

Response: We take a more positive view. CORS is applied in the surface ocean because that is the only part of the ocean directly exchanging gases with the atmosphere. We did not try to develop or claim to have developed a robust relationship model, although we do note that the data support strong influences of photosynthesis and upwelling that lead to characteristic patterns (second-to-fourth quadrant trends intercepting close to the origin). It is perfectly valid to use comparison to patterns in a high-quality dataset as a means of identifying possible issues in a lower quality dataset. It is not necessary to have a full and complete understanding of all processes (a robust relationship model) in order to do this, as long as the statements made about data quality issues are suitably caveated so as not to overstate levels of certainty.

The core problem of using [CO2] is that, as a part of the carbonate system, its variability cannot be linked directly to [O2], neither in the biotic (DIC will work here) nor in abiotic processes. While the definition of departure from equilibrium has a concrete meaning of O2 @ 100% (S,T,P) – O2@ (S,T,P), the similar definition for CO2 cannot be constructed simply because it will have a different meaning at each state of the carbonate system (DIC, TA concentration for example). Coming back to air-sea exchange, delta[O2] and delta[CO2] will vary dependent on the strength of the flux, which is non-linear and different for both gases, and on the depth of the mixed layer, which will add CO2 and O2 in odd proportions – this is just the Southern Ocean case. Lastly, equilibration times of CO2 is roughly 10x slower than those of O2, therefore comparing instantaneous changes in [O2] and [CO2] (like the ones from the float) bears a risk of identifying both false-positives and false-negatives.

Response: We connect [CO_2] to [O_2] via DIC and CO2SYS calculations (Methods section). The reason for not using DIC as the y-axis is explained in L391-394 as well as in our response to Reviewer #2. More studies (Torgersen and Branco, Vachon et al., 2020) have shown the utility of [CO_2]-[O_2] comparisons to identify biogeochemical processes and to be used as quantitative metrics. The reviewer is mistaken in thinking that we attempt a precise mechanistic model. We do not, nor do we claim to. While the points the reviewer makes would indeed be concerns for a mechanistic model, they are not for the approach used here.

References

Long, M. C., Stephens, B. B., McKain, K., Sweeney, C., Keeling, R. F., Kort, E. A., . . . Wofsy, S. C. (2021). Strong Southern Ocean carbon uptake evident in airborne observations. *Science, 374*(6572), 1275-1280. doi:10.1126/science.abi4355

Mackay, N., & Watson, A. (2021). Winter air-sea CO_2 fluxes constructed from summer observations of the Polar Southern Ocean suggest weak outgassing. *Journal of Geophysical Research: Oceans*, e2020JC016600. doi:10.1029/2020JC016600

Sutton, A. J., Williams, N. L., & Tilbrook, B. (2021). Constraining Southern Ocean CO_2 Flux Uncertainty Using Uncrewed Surface Vehicle Observations. *Geophysical Research Letters, 48*(3), e2020GL091748. doi:10.1029/2020GL091748

Torgersen, T., & Branco, B. (2007). Carbon and oxygen dynamics of shallow aquatic systems: Process vectors and bacterial productivity. *Journal of geophysical research, 112*(G3). doi:10.1029/2007JG000401

Vachon, D., Sadro, S., Bogard, M. J., Lapierre, J.-F., Baulch, H. M., Rusak, J. A., . . . del Giorgio, P. A. (2020). Paired O_2–CO_2 measurements provide emergent insights into aquatic ecosystem function. *Limnology and Oceanography Letters, 5*(4), 287-294. doi:10.1002/lol2.10135

22nd Feb 22

Dear Professor Tyrrell,

Please allow me to apologise for the delay in sending a decision on your manuscript titled "Integrated Analysis of Carbon Dioxide and Oxygen reveals Ocean Processes and Sensor Performance". It has now been seen again by our reviewers, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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Joe Aslin
Senior Editor,
Communications Earth & Environment
https://www.nature.com/commsenv/
Twitter: @CommsEarth

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Dear authors,

the manuscript has improved according to the suggestions from all three reviewers. In addition, all my suggestions and questions were very well adressed.

This is a valuable work that would be useful for the community and on top is very timing.

congratulations and keep using GLODAP and argo floats

best regards

Reviewer #2 (Remarks to the Author):

The authors have responded extensively and effectively to the comments submitted during the first round of review. I have no further concerns with this manuscript.

While I don't think the CORS method is ready to be adopted as a robust QC procedure, the authors

demonstrate the utility of the method for identifying obviously flawed data and for flagging data for further examination. Notably, other independent analyses suggest that the data flagged by the CORS method in the Southern Ocean may be problematic as well. I believe this is a timely contribution that has the potential to influence analyses of data quality from autonomous platforms.

Jonathan D. Sharp
January 17, 2022

Reviewer #3 (Remarks to the Author):

The authors put a significant effort in revising the manuscript and addressing the comments raised by all reviewers. The authors have also made emphasis on the fact that the CORS method in its current form should be used with caution and only as a diagnostic tool for detecting outliers e.g. in ARGO data. I agree with this assessment. Further work is required to investigate the true meaning of the slope and intercept in CORS plots in the different geographical regions (such as the Labrador Sea work mentioned in the study).

Professor Toby Tyrrell
Southampton Ocean and Earth Science

University of Southampton, Waterfront Campus,
European Way, Southampton SO14 3ZH, UK
t +44 (0)23 8059 6110 **f** +44 (0)23 8059 3059
e tt@noc.soton.ac.uk www.noc.soton.ac.uk

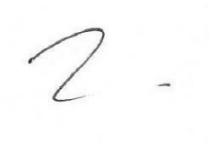
01 March 2022.

Communications Earth & Environment

Dear Editor,

The reviewers were all satisfied that their concerns have been addressed, so no response is required. We made a few necessary revisions regarding the format requirements.

Yours sincerely



Prof. Toby Tyrrell on behalf of all authors