Re: “Diagnosing CO_2 fluxes in the upwelling system off the Oregon coast” by Zhimian Cao et al. (bg-2014-130)

Dear Editor,

Enclosed is a copy of the revised manuscript “Diagnosing CO_2 fluxes in the upwelling system off the Oregon-California coast” by Zhimian Cao et al.

In this revised MS, we have fully considered all comments from the reviewers. Revision details are described in the enclosure. We sincerely thank you and the reviewers for the constructive comments and valuable suggestions, which certainly improved the quality of the paper. We hope that this revised MS will now meet the highest standard of Biogeosciences.

Sincerely,

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Enclosure: Response to reviews

Response to the comments from Dr. Rik Wanninkhof

Cao and co-authors use a conceptual model to estimate CO_2 dynamics/fluxes in upwelling systems. The model is referred to as OceMar (Ocean dominated Margin) and is derived from the work by Dai et al. (2013) in the Caribbean region. In short, it is based on the idea that the carbon to nutrient ratio in upwelling water determines the surface water CO_2 levels. Biological consumption determined from decreasing nutrients concentrations will decrease the pCO$_2$. The final pCO$_2$ value when nutrients are drawn down to zero can either be greater (CO_2 source the atmosphere) or less than atmospheric (CO_2 sink) depending on the C:N ratio of the source water.
The concept is elegant and simple, and "works" in some environments and less so in others. For the Pacific upwelling system the authors essentially show that it does not work very well. As described in the paper for the method to work, the system has to fulfill the following:
1. It should be in steady state
2. Alkalinity needs to be modeled as a function of salinity
3. Biological uptake needs to follow Redfield stoichiometry, in particular the C:N ratio is assumed to be 6.6:1
4. Endmembers need to be well described.
5. The residence time of the water needs to be on the same order as biological response
The authors use two case studies in the USA West Coast upwelling system, one off the coast of Newport OR, and one at the Oregon-California border. The method "works" at the former and fails at the latter which is attributed to non-steady state conditions.
The fundamental issue is, that it is difficult to independently determine if the criteria for successful application will be met. Thus, if the calculated pCO$_2$ values end up being reasonable compared to observation it can be assumed that they are, or that compensating errors yield a reasonable value. When the values do not meet expectation it can be assumed that some of the criteria are violated. We know that upwelling systems are highly dynamic; that Alkalinity to salinity ratios are regionally constant for surface waters but not for subsurface (or many upwelling systems); that Redfield stoichiometry is an average that often does not hold in surface water analyses; and that endmembers are difficult to determine. Therefor the applicability and use of the approach seems limited.

[Response]: We appreciate that Dr. Wanninkhof is generally positive with the conceptual model of OceMar and our diagnostic approach which involves essentially two couplings: 1) physics-biogeochemistry; 2) carbon and nutrients. Dr. Wanninkhof well laid out a suite of criteria in the diagnostic approach, which we would further clarify as follows:

1) Steady state associated with the water transport (i.e., water input equals water output) in a given system is often assumed in any modeling studies, which is also the basis of our OceMar model. Decomposition of the biological component in a coupled physical-biogeochemical system assumes similar time scales between water mass mixing and biological reactions, and requires conservative tracers to address water mass mixing processes. TAlk is often used for the latter purpose because it is relatively widely

available and in most cases is conservative in the upper waters. Such decomposition shall also need to resolve the end-members of the mixing scheme.

2) When water mass mixing is decoupled from biogeochemical reactions, the diagnostic approach should be simplified by taking off the biological reaction, which is exactly the case of the nearshore area off the Oregon-California coast shown in the paper.
3) Our approach also assumes Redfield ratio in coupling carbon-nutrients as most of the numerical modeling scheme adopts. This is an important assumption that poses challenge to the community. While increasing cases and datasets have inferred non-Redfield C/N uptake ratios, defined departure from the Redfield ratio is very hard to be determined. We are further elaborating this issue below in addressing specific criteria.

The motivation of the exercise of this study is to further test the applicability of the OceMar model and the diagnostic approach to a well-known upwelling system exerting strong CO_2 sink except (and also known) at the nearshore with intensified upwelling. Specifically, we elucidate as follows the critical issues raised by Dr. Wanninkhof on the conservativity of TAlk, Redfield ratio and end-member determination.

- Total alkalinity-salinity (TAlk-Sal) relationship

The field observed TAlk values were generally well correlated with salinity, telling us that the system was dominated by two end-member mixing schemes in the upper waters off the Oregon-California coast. Although the non-conservativity of TAlk existed, it was not that significant as seen by the deviations of a few data points from each linear regression (Fig. 2 of the original MS and Fig. 3 of the revised MS). As a matter of fact, Fassbender et al. (2011) have estimated that the contribution of $CaCO_3$ dissolution to the TAlk addition at Transect 5 was <10 μmol kg^{-1} (<0.5% of their absolute contents in seawater), close to the analytical precision of TAlk. Such small non-conservative portions would not compromise the application of TAlk as a conservative tracer. On the other hand, the elevated DIC corresponding to the TAlk addition were even smaller (~half of the TAlk deviation), which were <5 μmol kg^{-1} (Fassbender et al., 2011) and slightly higher than the measurement uncertainties.

In another upwelling system on the northern South China Sea shelf, organic carbon production/remineralization rather than biocalcification/$CaCO_3$ dissolution exclusively induced the DIC variations during intensified upwelling events nearshore, as supported by the nearly constant salinity normalized values of both TAlk and dissolved Ca^{2+} (Cao et al., 2011).

As the organic carbon metabolism often dominates the biological activities, we contend that TAlk can be well served as a quasiconservative chemical tracer in many coastal upwelling systems, in which the influence of $CaCO_3$ production/dissolution on TAlk/DIC would be negligible. We have made this point clearer in our revisions (see Line 202-208 of the revised MS).

- Redfield ratio

We agree with Dr. Wanninkhof as well as the other two reviewers that, the real C/N uptake ratio in a given oceanic setting can be different from the Redfield one of ~6.6 (Redfield et al., 1963). However, since the precise estimation of the C/N uptake ratio via e.g. in situ incubation experiments is still problematic, such data are currently scarce over the world's oceans and the empirical stoichiometry is routinely applied into field studies investigating the dynamics and coupling of carbon and nutrients (e.g., Chen et al., 2008; Fassbender et al., 2011). In the work of Fassbender et al. (2011), another empirical C/N uptake ratio (~117/16=7.3; Anderson and Sarmiento, 1994) was applied to the same data set as this study. We thus have done a simple sensitivity analysis using this alternative value of 7.3 (see Line 451-472 and Table 3 of the revised MS). Since $\Delta DIC-7.3\Delta NO_3$ values were obviously smaller than $\Delta DIC-6.6\Delta NO_3$ ones, the new sea-air ΔpCO_2 values were halved. Correspondingly, the newly estimated sea surface pCO_2 were ~35-45 μatm lower than the estimation using the Redfield ratio, which were however consistent with the field measurements. Given that the Redfield ratio also works in our OceMar case studies of the South China Sea and the Caribbean Sea (Dai et al., 2013), we contend that this classic ratio could be preferentially employed if the field observed elemental stoichiometry is not available. Moreover, as Martz et al. (2014) point out, "treating the Redfield ratios as global or regional constants may be acceptable in the context of interpreting snapshots of the water column captured in shipboard bottle data".

The above notion was also supported by examining the slope of the linear regression between DIC and NO_3 normalized to a constant salinity, which provides an alternative to the C/N uptake ratio (Sambrotto et al., 1993; Wong et al., 2002; Ianson et al., 2003). Our new analysis with all data from the CO_2 sink zones off the Oregon-California coast revealed a slope of 6.70 ± 0.37. This value was within error comparable to that of 6.6, suggesting that using the Redfield ratio in our diagnostic approach was in order (see Line 473-491 and Fig. 6 of the revised MS).

- Identification of end-members

We note that identifications of initial end-members associated with individual water masses are highly complex in any given oceanic regime. To bypass and simplify this issue, the relationship between conservative chemical tracers and salinity was used to reveal the water mass mixing scheme and to identify the end-member values, which might have experienced physical or biological alterations from their original values. Such a method, as one of the core components of the diagnostic approach we are introducing, well worked in two contrasting environments, the coastal upwelling system (this study) and the deep basins of large marginal seas (Dai et al., 2013). Per the suggestion from Dr. Ianson as well, we have performed a sensitivity analysis of end-members including the combined freshwater end-member and the deep water end-member, showing that the influence of variations in either end-member on our diagnostic approach was indeed minor (see Line 420-450 and Tables 1 and 2 of the revised MS).

Taken together, the OceMar conceptual model is applicable to other marginal systems but the diagnosis will have to be adjusted to individual settings with different water mass dynamics and biological responses.

The paper is nicely written and well-researched. A minor issue is that the Revelle Factor appears to be misinterpreted and incorrectly used by assuming that a fractional change in pCO_2 is the same as the air-water concentration difference (plus that temperature, alkalinity, and salinity do not change).

[Response]: We thank the comment and have made it right in our revisions (see Line 299-303 of the revised MS).

References

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Response to the comments from Referee #1

Cao et al. apply a simple framework (OceMar) based on carbon/nitrate mass balance to semi-analytically predict CO_2 air-sea fluxes in the upwelling system off the Oregon coast. They find that they are able to represent observed fluxes in regions of the shelf that act as sinks of CO_2, but are unable to represent the source regions following the OceMar approach. Then, they add one extra assumption to the applicability of the method, namely the requirement of steady state conditions. I think the manuscript is interesting and worth of publication after some main issues are addressed.

[Response]: We are pleased that the reviewer is generally positive with our study.

I agree with Dr. Wanninkhof's comment about the overall applicability of the method and think that the manuscript would benefit from an expansion of this discussion.

[Response]: Please see our response to the comments from Dr. Wanninkhof.

Moreover, the source data the authors described (Feely et al 2008, Feely and Sabine 2011) show many more transects along the California Current System that may provide more insight into the un/applicability of OceMar in wind-driven upwelling margins (in particular, regions to the south, further away from the Columbia River). I am wondering whether the authors could incorporate some of these transects or otherwise explain their chosen focus on the Oregon region. In the latter case, they should discuss whether they would expect OceMar to work in the regions to the south and north of Oregon within the California Current System.

[Response]: We agree with the reviewer that diagnosis of additional transects would be helpful, and thus we have added in our revisions the diagnosis of Transect 6, which is off the northern California coast (see Fig. 1 of the revised MS) and was also significantly influenced by intensified upwelling. This additional diagnosis generated very consistent estimations of the CO_2 flux with those of Transect 5 (see Figs. 2, 3 and 4 and the corresponding texts of the revised MS), indicating that the upper waters on Transects 5 and 6 experienced nearly the same physical mixing and biogeochemical reactions. In this context, we would expect that our OceMar approach to work in other regions within the California Current system, in particular given that the physical transport and biological alterations would be more straightforward in areas without either costal upwelling or river plume.

Another suggestion would be to test the robustness of the results with respect to the choice of the carbon to nitrogen ratio. The authors could perform a sensitivity analysis where they repeat their calculations replacing the Redfield ratio by some deviations of the 6.6 value (maybe observed ranges of C:N in the region?).

[Response]: We have tested the OceMar approach with another C/N uptake ratio. Please also see our response to the comment from Dr. Wanninkhof.

Specific comments:

The abstract would benefit from a slightly more detailed explanation of the OceMar framework.

[Response]: Revised as suggested. See Line 25-30 of the revised MS.

Also in the abstract: I found lines 15-17 a bit misleading ("we showed significant CO_2 outgassing in the nearshore regions associated with intensified upwelling and minor biological consumption: : :"). In my opinion, it reads as if the method was able to capture the outgassing, while actually the pCO_2 observations showed the outgassing and the method failed to reproduce it. Then, the authors argued for a modification to the method to address this issue.

[Response]: This sentence has been rewritten for clarity. See Line 38-42 of the revised MS.

Page 7393, line 12-14: the region of interest is part of the California Current System. The southern part of this system has permanent upwelling-favourable winds.

[Response]: Among the entire research domain during the first NACP West Coast cruise, the most intensified upwelling was observed on Transects 5 and 6, where the subsurface water reached to the nearshore surface (see Feely et al., 2008, Figure 1). We have added the diagnosis of Transect 6 in our revisions. Please also see our response to the general comment from Referee #1.

Page 7397, lines 15-18: I'd suggest being explicit about how X^{eff} is calculated, so the reader doesn't have to dig into Dai et al. (2013) to understand this step in the calculation. The explanation could be added as an appendix.

[Response]: The following two paragraphs in the original MS (now Line 248-271 of the revised MS) had already explained how X^{eff} was calculated.

Page 7400, line ~26: why are these values not shown in figure 3?

[Response]: The estimated values of $\Delta DIC-6.6\Delta NO_3$ and sea-air ΔpCO_2 at station 25 (~82 $\mu mol\ kg^{-1}$ and ~157 μatm, respectively) were largely different from those at other stations on Transect 4, which could not be well illustrated in a single figure. Instead, we have directly added the two values in the text of the revised MS (Line 332).

We have also added a note that no values of $\Delta DIC-6.6\Delta NO_3$ and sea-air ΔpCO_2 were obtained at station 26 because the nutrient data at this station were not available (see Line 310-311 of the revised MS).

Technical comments:

Every now and then I had problems with specific sentences that I think could be improved, e.g.:

Page 7390: lines 10-11: English here could be improved - besides, this text is part of a really long sentence, line 10 to 15!

[Response]: This sentence has been rewritten for clarity. See Line 34-38 of the revised MS.

Page 7393: line 7: should say "broad" instead of "board"

[Response]: Modified as suggested. See Line 107 of the revised MS.

Page 7394: line 18: "to quantify the conservative portion of carbon and nitrate."? (or nitrogen nutrients, but I think the authors only use NO_3 in their calculations).

[Response]: Modified as suggested. See Line 142-143, Line 146 and Line 149 of the revised MS.

Page 7395: line 16: "parameters" should be replaced by "variables"

[Response]: Modified as suggested. See Line 187 of the revised MS.

Reference

Feely, R. A., Sabine, C. L., Hernandez-Ayon, J. M., Ianson, D., and Hales, B.: Evidence for upwelling of corrosive "acidified" water onto the continental shelf, Science, 320, 1490-1492, 2008.

Response to the comments from Dr. Debby Ianson

Cao et al. use a relatively simple end-member mixing scheme to estimate the air-sea CO_2 flux from two transects in the coastal upwelling zone along the west coast of North America during the spring of 2007. This method has been used elsewhere but not yet applied to coastal upwelling.

General comments

This work uses an end-member scheme that relies on conservative behavior of total alkalinity and salinity (and a suite of other assumptions nicely laid out in the Wanninkhof review). The simplicity of the method is appealing and a useful tool to investigate CO_2 fluxes, at least to first order, as long as assumptions are not violated (which is tricky in coastal upwelling zones as the authors point out). I have a few recommendations/comments (below) mainly involving the addition of sensitivity analyses, which would add significant value to the interpretation.

[Response]: We thank Dr. Ianson for the valuable comments and suggestions, which have been fully considered in our revisions.

1. Since upwelling regions push the assumptions involved I suggest a sensitivity/error analysis/discussion be added. The limitations of the method could then be discussed in a quantitative fashion and perhaps boundaries on its utility imposed. For e.g. the X^{eff} terms are nicely discussed in the context of CR data and TA intercepts (with S) but then a single number is used for each of DIC and NO_3 (eff).

[Response]: We agree that a single number of X^{eff} is to some extent not convincing. While we are certain that NO_3^{eff} from the Columbia River plume is zero, we have performed a sensitivity analysis showing the minor influence of the DIC^{eff} variations on our diagnostic approach (see Line 420-437 and Table 1 of the revised MS).

2. Including data below 200 m (or below a salinity of 34) in the TA-S plots in this dataset make the determination of the 'end-member' a little murky in my opinion. At about S=34 (a little shallower) the TA-S 'curves' become more steep in these data, presumably getting into the California Undercurrent and/or aragonite dissolution (relatively shallow in this part of the world). I don't dispute the author's choice of regression and end-member necessarily - but again recommend a sensitivity analysis (choose a range of end-members) and suggest showing the data below 200 m in Fig. 2 even if they are coloured differently (which could convince the reader that the end-member really was tight and unambiguous).

[Response]: Per the reviewer's suggestion, we have plotted the TAlk-Sal relationship through the entire water column on Transects 4, 5 and 6 (see Line 152-176 and Fig. 2 of the revised MS). It's now easy to see that the deep water end-member for the upper waters was a little shallower than ~200 m, corresponding to an average salinity of 33.9. As a result, we have changed "in the upper 200 m waters" to "in the upper 175 m waters" throughout the revised

MS and removed data points of ~200 m from the TAlk-Sal plot for the upper 175 m waters (Fig. 2 of the original MS and Fig. 3 of the revised MS).

In the original MS, we stated selecting ~200 m waters as the deep water end-member, whereas the real data used were those collected at ~175 m. We apologize for this misleading statement and have made this point clearer in our revisions (see Line 231-240 of the revised MS). Moreover, we have also performed a sensitivity analysis of this deep water end-member demonstrating that our choice of ~175 m was in order (see Line 438-450 and Table 2 of the revised MS).

3. Related to the previous point, the authors do not discuss the California Undercurrent (CUC) and its unique properties. At least along T4, the core of the CUC is above 200 m (Thomson and Krassovski 2010 JGR) and must be present at some stations on the transect.

[Response]: We have added a couple of sentences discussing the CUC based on the TAlk-Sal relationship through the entire water column (see Line 158-165 of the revised MS). But again, we don't intend to distinguish all of the water masses and identify their initial end-member values one by one. We used field observed values as end-members which might have experienced physical or biological alterations from their original water masses.

4. The authors discuss the method in the context of determining net source and sink (for CO_2) regions in a general sense - but with two transects (single visit) they are only able to look at a couple of snapshots in time in a system with large spatial and temporal variability. They would need seasonal data, in particular from the winter (downwelling) season where PP is light-limited on T4, to make a firm assessment of the source/sink capability of a region. (The steady state assumption is clearly violated.) This caveat should be more clearly stated.

[Response]: Our main objective is to test the OceMar conceptual model and our diagnostic approach in coastal upwelling systems and the Oregon-California shelf is selected as a representative case. It is not the scope of this paper to systematically investigate the distribution and seasonality of CO_2 source/sink nature in this area, which however has been well reported in relevant studies such as Evans et al. (2011).

5. There is evidence for 'excess' DIC uptake (uncoupled from NO_3) when phytoplankton become nutrient limited (Ianson and Allen 2002 GBC and Druon et al.
2010 ECSS model this uptake - but the concept is much older - e.g. Sambrotto et al. 1993 Nature) which would affect the estimation of air-sea flux if present. For these transects (esp. T5) this feature may not be an issue (although on T4 depletion of silicic acid suggests that it might), but it should at least be discussed in the paper and again sensitivity analysis would be valuable.

[Response]: We think the issue of excess DIC uptake relative to NO_3 is similar to that of non-Redfield C/N uptake ratio so that they were discussed together in our revisions (see Line 453-456 of the revised MS). We have tested the OceMar approach with another empirical

C/N uptake ratio of 7.3 (Anderson and Sarmiento, 1994), which to some extent suggests the excess DIC uptake. Please also see our response to the comments from Dr. Wanninkhof.

Specific comments

1. p.7391 l.2-5 for general source/sink discussion need seasonal context (state)

[Response]: Modified as suggested. See Line 51-52 of the revised MS.

2. p.7391 l.13 - why mention Ca ion if not measured?

[Response]: Ca^{2+} is used as a conservative chemical tracer in Dai et al. (2013).

3. p.7392 l.7 - eNP - add 'Subtropical Gyre' to distinguish from Alaskan Gyre - eNP.

[Response]: We have deleted this sentence in the revised MS.

4. p.7392 l.15-20 - a good place to mention the possibility of excess DIC uptake

[Response]: We don't think here the excess DIC uptake works because the undersaturated pCO_2 off Oregon were well predicted with the Redfield C/N uptake ratio in both Hales et al. (2005) and this study.

5. p.7393 l.3 300 m seems too deep for CC ?

[Response]: We have changed to 0-200 m. See Line 107 of the revised MS.

6. p.7393 l.9 - CUC is shallower than that (Thomson and Krassovski 2010 JGR)

[Response]: We have changed to 150-300 m according to Thomson and Krassovski (2010). See Line 109 of the revised MS.

7. p.7993 l.13 - I work further north, but these upwelling depths look too deep. Are you sure?

[Response]: Yes. The vertical sections of some physical/chemical parameters including temperature, pH and DIC show that the subsurface water is upwelled from the depths of 150 m to 200 m onto the Oregon-California shelf (see Feely et al., 2008, Figure 2), while other studies also prove these upwelling depths (e.g., Hales et al., 2005).

8. p.7396 l.1 - remove 'very good' or be quantitative

[Response]: We have added the r value to quantitatively show the good TAlk-Sal relationship. See Line 201 of the revised MS.

9. p.7396 l.9 and l.15 ‘should’ - why ‘should’ they? do they? suggest more explanation or better word choice.

[Response]: Modified as suggested. See Line 214 and Line 220 of the revised MS.

10. p.7397 l.2-3 - given that it’s an upwelling zone wouldn’t it be smarter to use a set isopycnal instead of depth? and I would choose a shallower depth in this zone as my end-member to stay in a linear zone of TA-S etc (see general comments above).

[Response]: While the isopycnal mixing dominated the nearshore upwelling zone off Oregon and northern California, upper waters in offshore areas beyond the upwelling circulation were largely fed by on-site deep waters via diapycnal mixing. In this context, we kept using depth which is more straightforward than density.

We have clarified that the values at ~175 m were selected as the deep water end-member (see Line 231-240 of the revised MS). Please also see our response to general comment #2 from Dr. Ianson.

11. p.7398 l.5 (and further) what about non-Redfieldian C:N?

[Response]: We have tested the OceMar approach with another C/N uptake ratio. Please also see our response to the comments from Dr. Wanninkhof.

12. p.7399 sect.3.3 - again - consider ‘excess DIC’ uptake

[Response]: Please see our response to general comment #5 from Dr. Ianson.

References

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