

Response to review comments on "Global carbon budgets estimated from atmospheric O₂/N₂ and CO₂ observations in the western Pacific region over a 15-year period"

Anonymous Referee #2:

We would like to thank the anonymous referee for his/her helpful comments and suggestions on our paper. We have revised the manuscript as is described in the following. The referee's comments are in *blue italics*, and the modifications are shown in **red**.

Reply to main comments:

1) It's unclear how uncertainty in estimating Z_{eff} comes into play in the carbon budget estimates, especially given its relevance for the shorter timescales considered. The authors do show trends without Z_{eff} (figure 8) but it's not evident if this is incorporated in the carbon sinks and trends calculation (i.e. unclear if incorporated in grey shading in figure 8 or error estimates in carbon sinks column in Table 2). Perhaps, the authors could evaluate uncertainty in Z_{eff} from using the upper/lower bounds with and without Z_{eff}? Further, it's unclear how correcting for Z_{eff} in the carbon budgets plays out in the 5 year timescales described (as also discussed in Nevison et al. 2008 and elsewhere). It seems, as referred to by the authors, that the ventilation events of 1999-2001 could impact the pentad trends, and thus similar variability during other years probably could have similar effects on other pentad periods (e.g. 2004-2005 dip in pentad ocean sink seems to co-occur with inter-annual variation that don't seem to be fully suppressed?). Finally, an additional and not insignificant component of Z_{eff} not included in this study is the atmospheric deposition effect, as detailed in Keeling and Manning 2014, which adds about 0.1 (+/-0.1) Pg C/yr, and which should raise Z_{eff} from 0.1-0.9 Pg C/yr, to 0.2-1.0 Pg C/yr. Overall, I feel the treatment of Z_{eff} uncertainty within the shorter timescales considered here merits further clarification.

As both Referee #1 and #2 indicated, the description of the uncertainty in the ocean outgassing effect was unclear in the original manuscript. We assumed a $\pm 100\%$ uncertainty for Z_{eff} for the corresponding budget calculation period. To make it clear, we have added a sentence in the second paragraph in the revised manuscript (see reply (2) for more details): "**Since the ocean outgassing effect is rather speculative, we assumed that the values of Z_{eff} for the individual periods had $\pm 100\%$ uncertainties in accordance with previous studies (e.g. Manning and Keeling, 2006; Tohjima et al., 2008).**"

As Referee#2 suspected, it seems that the five-year average cannot fully suppress the APO variations associated with the anomalous air-sea gas exchanges. As we discussed in Section 3.1 and as is shown in Fig. 7, the pentad APO trends have a temporal variability of about +1.2 per meg yr⁻¹ or +0.5 PgC yr⁻¹, which is comparable to the 2004-2005 dip in the pentad ocean sink shown in Fig. 8. To emphasize the limitations of the pentad averaging, we added the following sentences after the first sentence of the second paragraph in Section 3.3: "**Nevison et al. (2008) suggested that a decadal or longer period is needed to suppress the influence of the interannual variation in the ocean O₂ flux on the carbon sink estimation within ± 0.1 PgC yr⁻¹ based on an ocean ecosystem model and an atmospheric transport model. In addition, the pentad APO changing rate still contains an uncertainty corresponding to ± 0.5 PgC yr⁻¹ as is discussed in Section 3.1. Therefore, the anomalous dip in the ocean sink for 2004-2005 might be an error caused by the anomalous ocean O₂ flux variations.**"

In response to the suggestion of Referee #2, we have included the anthropogenic N deposition effect in the Z_{eff} estimation. To explain the anthropogenic nitrogen deposition effect (Z_{anthN}), we have separated Z_{eff} into Z_{anthN} and the global ocean warming component (Z_{gow}), and added the following paragraph after the fourth paragraph in Section 2.5:

“In addition to the ocean warming effect, Keeling and Manning (2014) introduced recently another ocean outgassing effect caused by atmospheric deposition of excess anthropogenic nitrogen to the open ocean. The excess nitrogen is considered to enhance the ocean biotic production of organic matter, which is associated with the O₂ production. Keeling and Manning (2014) evaluated the anthropogenic nitrogen-induced outgassing as being about 0.1×10^{14} mol O₂ yr⁻¹. Since the outgassing effect caused by the anthropogenic nitrogen deposition is small but rather significant, we adopted the effect as Z_{anthN} , with a magnitude of 0.12 PgC yr^{-1} ($=0.1 \times 10^{14}$ mol O₂ yr⁻¹ $\times 12.01 \text{ gC mol}^{-1}$). Eventually, the total outgassing effect, Z_{eff} , is expressed as the summation of Z_{gow} and Z_{anthN} :

$$Z_{eff} = Z_{gow} + Z_{anthN}. \quad (9)''$$

2) I see the need perhaps for a section dedicated to clarifying and detailing the sources, contribution, and methods for calculating carbon budget uncertainty, as it can help clarify the confidence in the pentad trends and conclusions presented here. Table 2, for instance, could incorporate uncertainty due to Z_{eff} in the carbon sinks column, and in Figure 8 (gray shading). Uncertainty due to undersampling the global signal has also been shown by Nevison et al. (2008) to contribute to uncertainty in estimating budgets on the shorter timescales evaluated here. What is impact of sampling over the western Pacific (40S-40N) vs. full global sampling on the carbon sink trends? How does uncertainty in α_B in using values of 1.1 vs 1.05 affect the uncertainty in carbon sink budgets? Finally, it's unclear how the contribution of measurement uncertainty, due to span calibration of the gas chromatographic technique and potential longterm drift shared across all cylinders, is incorporated in the uncertainty analysis.

In the original manuscript, we didn't incorporate the anthropogenic N deposition effect on Z_{eff} , the uncertainty associated with the global average of APO from limited samples, uncertainty due to span sensitivity of the gas chromatographic technique, and the potential long-term drift among the O₂/N₂ reference cylinders. In addition, the uncertainty of ± 0.05 for α_B in the original manuscript should be increased to ± 0.10 . Considering these uncertainties, we have carefully reevaluated the uncertainties. To make it clear, we have modified the original manuscript as follows:

To clarify the long-term stability of the O₂/N₂ scale, we have added the following sentences at the end of Section 2.3: “However, this stability test cannot exclude the possibility that the O₂/N₂ ratios of the reference gases drift across all the cylinders rather uniformly. There are several mechanisms that affect the O₂/N₂ ratios of the gases within the high-pressure cylinders, including corrosion of the inner surface, leakage, thermal diffusion and gravitational fractionation. Keeling et al. (2007) assessed carefully and comprehensively the influences of those potential mechanisms on the long-term stability of the O₂/N₂ ratio of the reference gases and obtained an estimated uncertainty of ± 0.4 per meg yr⁻¹. We also treated the reference cylinders which were kept horizontally in a thermally insulated box, with the greatest care (Tohjima et al., 2008). Therefore, we adopted the value of ± 0.4 per meg yr⁻¹ as the long-term drift of the reference gases caused by the above degradation effects. Consequently, we assumed that the total uncertainty of the long-term stability of the O₂/N₂ reference scale was ± 0.45 per meg yr⁻¹ ($= (0.2^2 + 0.4^2)^{1/2}$) in this study.”

As for the uncertainty of α_B , we have added the following sentences at the end of Section 2.4: “Considering the recent reports about the global net -O₂/CO₂ exchange ratio, Keeling and Manning (2014) revised the uncertainty of α_B upward from ± 0.05 (Severinghaus, 1995) to ± 0.10 . Thus, we also adopted ± 0.10 for the uncertainty of α_B in this study.”

Finally, to clarify how we computed the total uncertainties associated with the global sink estimations, we have added the following paragraph after the first paragraph in Section 3.2:

“The uncertainties in the parameters used for the carbon budget calculation (Eqs. (6) and (7)), which are also listed in Table 2, are briefly discussed here. Note that in this study the estimated uncertainties are $\pm 1\sigma$. Since the ocean outgassing effect is rather speculative, we assumed that the values of Z_{eff} for the individual periods had $\pm 100\%$ uncertainties in accordance with previous studies (e.g. Manning and Keeling, 2006; Tohjima et al., 2008). We adopted uncertainties of $\pm 5\%$ for the fossil fuel-derived CO_2 emission rate and $\pm 0.2 \text{ PgC yr}^{-1}$ for the atmospheric CO_2 increasing rate from Le Quéré, et al. (2018). As for the uncertainties of the observed APO changing rates, we adopted the standard deviations among the sites shown in Fig. 7 ($\pm 0.37 \text{ per meg yr}^{-1}$ for longer than 10 years and $\pm 0.54 \text{ per meg yr}^{-1}$ for 5 years). The estimated uncertainty of the O_2/N_2 scale stability ($\pm 0.45 \text{ per meg yr}^{-1}$) discussed in Section 2. 3, the uncertainty of the O_2/N_2 span sensitivity ($\pm 3\%$), and the uncertainty in the global averaged APO associated with the limited atmospheric sampling ($\pm 0.2 \text{ PgC yr}^{-1}$) discussed in Nevison et al. (2008) were also included in the calculation of the uncertainties in ΔAPO . The uncertainties of α_B and α_F were ± 0.10 (Keeling and Manning, 2014) and ± 0.04 (Tohjima et al., 2008), respectively. Finally, these uncertainties were propagated to the ocean and land sink uncertainties in accordance with Eqs. (6) and (7).”

In accordance with the uncertainty revision, we have also revised the uncertainties of the O_2/N_2 and APO changing rates listed in Table 1. The uncertainties of ± 0.2 or $\pm 0.3 \text{ per meg yr}^{-1}$ have increased to $\pm 0.8 \text{ per meg yr}^{-1}$. In addition, we have also redrawn Fig. 8 as follows:

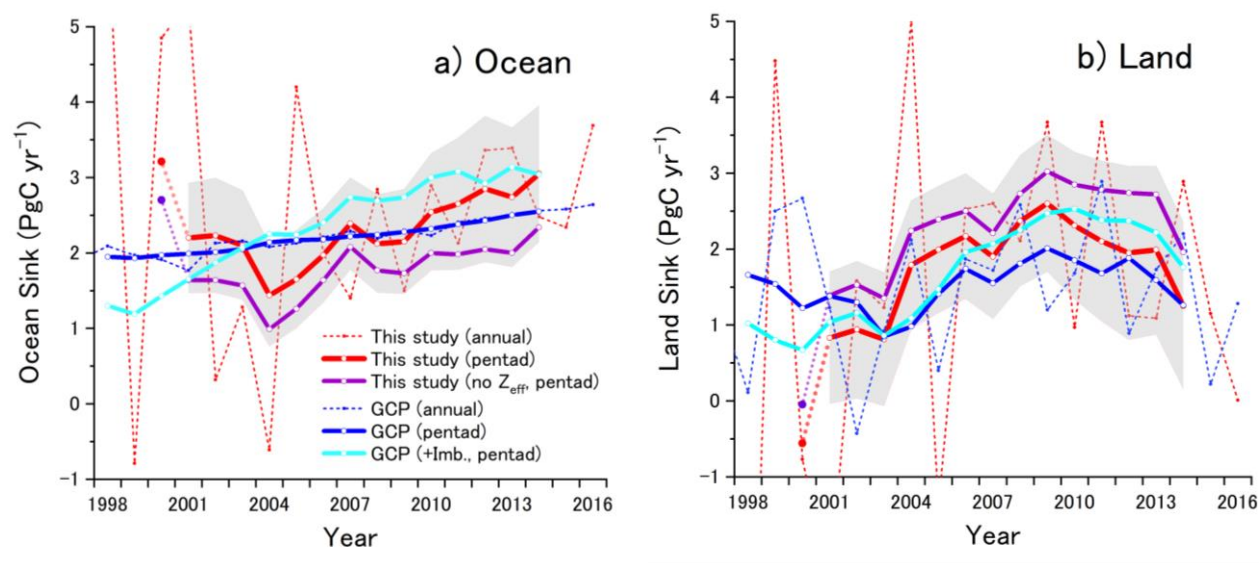


Fig. 8. Temporal variations in (a) ocean and (b) land biospheric sinks estimated from APO variations of this study (red) and process-based models of GCP (blue). The thin broken lines represent the annual sinks and the thick lines represent the pentad sinks. The purple lines represent the pentad sinks based on APO without ocean outgassing correction (Z_{eff}) and the light blue lines represent the sinks of GCP with the imbalance sinks added. The uncertainty associated with the pentad sinks with Z_{eff} corrections are shown as shaded area.

3) The comparison against the GCP could be elaborated on a bit more, as it raises important issues in the field. The authors could elaborate further (through existing or new figure/table) how different estimates reported by GCP compare to the APO method, including hindcast ocean models and ocean observation based

products, all of which are readily available in the GCP product as globally integrated fluxes: <https://www.icoscp.eu/GCP/2018>. It is interesting that the comparison to the GCP mean showcases similarities in magnitude and in temporal evolution of pentads. The point that the uptake of carbon by the ocean is larger than expected from atmospheric increase alone is very interesting. How do the decadal trends (2000-2016) in this study compare to the pCO₂ based air-sea flux timeseries by Landschutzer et al (2016) and Rodenbeck et al (2013), as both of these estimates seem to show larger decadal variability than the ocean models? These items may be beyond the current scope of this study, but could substantially improve the impact of this paper with (hopefully?) relatively minor figure/text additions.

We have carefully compared our pentad ocean sinks based on APO with those of GCP and the pCO₂-based estimations and found that the increasing trend of the ocean sinks based on APO was close to those based on the pCO₂ observations. To explain this clearly, we have added the following sentences after the second to the last sentence of the second paragraph in Section 3.3: “For a detailed comparison, the global ocean sinks based on pCO₂ observations and interpolation techniques (Landschutzer et al., 2016; Rödenbeck et al., 2014) for the period of 1990-2017 are plotted in Fig. 9 together with the ocean sinks of this study and GCP. Note that the extended pCO₂-derived ocean sinks were given as supplementary data of Le Quéré, et al. (2018) and those sinks were uniformly inflated by 0.78 PgC yr⁻¹ to compensate for the pre-industrial steady state source of CO₂ derived from riverine input of carbon to the ocean (Resplandy et al., 2018). As you can see, both the GCP and pCO₂-derived ocean sinks show changes in the trends between before and after 2001 while the magnitude of the changes in the pCO₂-derived sinks are larger. The increasing rates determined by a linear regression during 2001-2014 are 0.08 ± 0.01 PgC yr⁻² in Landschutzer et al. (2016) and 0.07 ± 0.02 PgC yr⁻² in Rödenbeck et al. (2014), which are more consistent with the rate found in this study. Therefore, our result seems to support a previous conclusion that the recent increase in the ocean sinks exceeds the increasing trend of ocean sink expected only from the atmospheric CO₂ increase (Landschutzer et al., 2015; DeVries et al., 2017).” In accordance of this change, we have modified the third sentence of Conclusion 3) as “The pentad ocean sinks showed an overall increasing trend for the entire period (2001-2014) with a linear increasing rate of 0.08 ± 0.02 PgC yr⁻². This increasing rate was about two times larger than that for the GCP ocean sinks (0.04 ± 0.01 PgC yr⁻²) but was consistent with those for the global ocean sinks based on pCO₂ observations and interpolation techniques (Landschutzer et al., 2016; Rödenbeck et al., 2014).” We have also added Resplandy et al. (2018) to Reference and Fig. 9 showing ocean sinks based on the APO data, process-based models (GCP), and pCO₂ observations as follows:

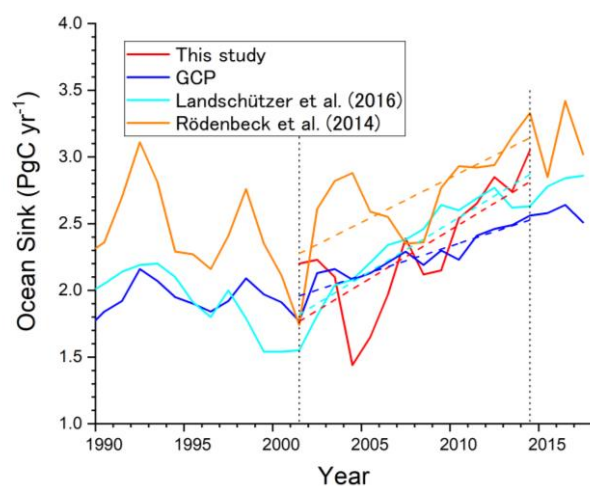


Fig. 9. Comparison of the temporal variations of the ocean sinks based on the APO data of this study (red), global ocean biogeochemistry models (GOBMs) of GCP (blue), and pCO₂ data of Landschützer et al. (2016) (light blue) and Rödenbeck et al. (2014) (orange). The broken lines represent the regression lines for the corresponding data during 2001-2014. Note that the pCO₂-based ocean sinks are adjusted for the pre-industrial ocean CO₂ emissions ($\pm 0.78 \text{PgC yr}^{-1}$) caused by riverine CO₂ input to the ocean (Resplandy et al., 2018).

In addition to the above modifications, we have changed the GCP-reported data (fossil fuel emissions, atmospheric accumulation, and global sinks) from Global Carbon Budget 2017 to the data from Global Carbon Budget 2018 (Le Quéré, et al., 2018). We have used the updated GCP data for recalculating the global carbon budgets in the revised manuscript. Because the fossil fuel-derived CO₂ emission rates have been slightly downwardly revised, the ocean and land sinks based on the APO data have been slightly decreased. But the changes are at most 0.1 PgC yr⁻¹. Consequently, this change has affected very little the conclusion of the original manuscript.

Reply to minor issues:

Pg2 L27: “The estimated value for α_F is about 1.10 ± 0.05 (Severinghaus, 1995) and that for α_B is about 1.4 (Keeling, 1988).” Should be the other way around: α_B is 1.10 and α_F is 1.4.

“The estimated value for α_F is about 1.10 ± 0.05 (Severinghaus, 1995) and that for α_B is about 1.4 (Keeling, 1988)” has been changed to “The estimated value for α_B is about 1.1 (Severinghaus, 1995) and that for α_F is about 1.4 (Keeling, 1988).”

Perhaps add citations for Equations (1), (2), and (3)?

Citation for Eq. (1), (2), and (3): We have added the citation, Manning and Keeling (2006), for these equations.

P3 L1, this paragraph could use a brief explanation of APO concept as a tracer for those not familiar with APO, i.e. cancellation of terrestrial influence, etc

In accordance with the suggestion, we have added the following sentence after the first sentence of the paragraph: “Since the APO is defined to be invariant with respect to the land biotic exchange, the secular trend in the APO is determined by fossil fuel combustions which cause a gradually decreasing trend in APO, and the air-sea gas exchange.”

Pg 7 L29, shouldn't Z_{eff} be in PgC/yr?

The unit “TgC yr⁻¹” has been altered to “PgC yr⁻¹”.

Pg 9 Line 20, the ENSO topic deserves a bit more clarification here. It would be good to preface the ENSO sentence with the findings of Rodenbeck et al 2008, who suggest anomalous outgassing of APO during El Niño, while Tohjima et al 2015 show a suppressed peak instead, and clarify that Eddebbbar et al (2017) reconcile this apparent discrepancy through a model-simulated zonal dipole-like ENSO response in the equatorial Pacific, and that enhanced observational zonal coverage in this region is needed to constrain the full basin ENSO response.

In response to the Referee’s suggestion, we have modified the relevant part as “Conducting atmospheric inversion analyses based on the APO data from the Scripps observation network,

Rödenbeck et al. (2008) suggested anomalous outgassing of APO from the equatorial region during El Niño periods, while Tohjima et al. (2015) found a suppressed equatorial peak during El Niño periods based on the western Pacific observations. Eddebbar et al. (2017) reconciled these conflicting results by predicting the existence of a zonal dipole-like ENSO response in the equatorial Pacific based on several ocean process-based models and an atmospheric transport model. These results suggest that an enhanced zonal coverage of the atmospheric observations in the equatorial Pacific is needed to constrain the full basin-scale ENSO response. We can see a considerable suppression of the equatorial peak during the strong 2015/2016 El Niño event in Fig. 6c, which was not reported in Tohjima et al. (2015). Any detailed discussion about the temporal variation of the equatorial peak during the 2015/2016 El Niño event is, however, beyond the scope of this study and will be given elsewhere.”

Suggested editing notes:

Pg 2 Line 5: remove “still”, and add year by which emissions rose to 10 Pg C/yr?

“...the global fossil fuel-derived CO₂ emissions in recent years still increased gradually and rose toward 10 PgC yr⁻¹ (Boden et al. 2017)” has been changed to “...the global fossil fuel-derived CO₂ emissions in recent years still increased gradually and rose to 9.9 PgC yr⁻¹ by 2014 (Boden et al. 2017)”.

Pg 2 Line 6: “Paris Agreement . . . aimed to balance the anthropogenic greenhouse gas emissions and natural removals in the second half of this century. . .”, I suggest editing to: “. . . aimed to reduce anthropogenic greenhouse gas emissions to maintain the increase in global mean surface temperature well below 2_C by 2100, . . .”?

The relevant part has been modified to “..., the Paris Agreement adopted at COP21 in 2015 aimed to **reduce** the anthropogenic greenhouse gas emissions to maintain the increase in global **mean surface** temperatures well below 2°C **by 2100**, ...”.

Pg3 L8, suggest deleting “In these days”.

“In these days” has been deleted.

Pg3 L21, “which reduces the ventilation of the seawater.”, suggest instead: “which reduces the ventilation of interior water masses.”

We have changed the relevant sentence from “the ventilation of the seawater” to “the ventilation of interior water masses”.

Pg3 L26: replace “huge” with “large”

“huge” has been replaced with “large”.

Pg 14 L 20: Not sure I understand this sentence: “This means that the changing trends of carbon budgets may be evaluated by the at least decadal APO data.” Suggest rephrasing and/or elaborating further?

The ambiguous fourth conclusion has been deleted. We have also modified the first two sentences of the last paragraph in Section 3.3, “From the above discussions, we feel ...in the temporal resolution.”, to “From the above discussions, we feel that a five-year duration effectively suppresses **to some extent** the anomalous variations in the carbon budget estimations

based on APO, which are considered to be caused by the imbalance of the seasonal air-sea O₂ exchange. **Probably, the five-year average suppresses the variability of Z_{eff} to a level of ±0.5 PgC yr⁻¹ as is discussed in Section 3.1.**”

Pg 12 L 32. Replace “stagnant” with “stagnancy”

“stagnant” has been replaced with “stagnancy”.

Pg13 L 1: replace “in spite of” with “despite”

“in spite of” has been replaced with “despite”.