

Responses to Referee 2

General comments:

Ishidoya and colleagues present ~8 years of airborne observations of O₂/N₂, CO₂, and Ar/N₂ from cargo aircraft flights over the North Pacific. The data are significantly impacted by sampling artifacts. The authors correct this data convincingly, and, from the corrected data, calculate the apparent global ocean and terrestrial CO₂ sinks. They also show that the seasonal cycle in APO is influenced by interhemispheric transport. An analysis of the interannual variability in the observations shows a signal which can be attributed to ENSO.

I think it could be made clearer in the text that the transported fluxes are derived from climatologies, which have known deficiencies. The Garcia and Keeling fluxes, for instance used the Wanninkhof 92 gas transfer velocity which is now known to be biased. The authors might consider using potential temperature or pressure as a vertical coordinate for binning instead of altitude.

I think the study is well conducted and worthy of inclusion in ACP. I have raised some minor points to be addressed below.

Thank you very much for your significant and useful comments on the paper “Spatiotemporal variations of the $\delta(\text{O}_2/\text{N}_2)$, CO₂ and $\delta(\text{APO})$ in the troposphere over the Western North Pacific” by Ishidoya et al. We have revised the manuscript, considering your comments and suggestions. Details of our revision are as follows. The line numbers denote those of the revised manuscript.

ABSTRACT

L11 and throughout: "amount fraction" is not a term I'm familiar with, I suggest "abundances" for referring to both O₂/N₂ and CO₂, and "mole fraction" for referring to CO₂.

We recognize the phrases you suggested are more familiar with our research field, however, I have used the phrase following the Editor's comment.

L11: "Observations were corrected for significant..."

Line 11: We have modified the sentence, as suggested.

L18 and throughout: usually "northern hemisphere" and "southern hemisphere" are capitalized, but I would refer to the specific style guidelines of the journal.

We leave the words as they are. If the Editor requests the revision before publication, then I will revise the words to be capitalized.

L22: "indicated a clear evidence of influence" -- suggest change to "indicated a clear influence"

Lines 21-22: The words have been changed to "indicated a clear influence", as suggested.

L24: What is a "C equivalent"? Do you mean simply petagrams of carbon? If so, Pg C a-1 is a widely used unit.

We recognize the unit you suggested are more familiar with our research field, however, I have used the unit following the Editor's comment.

INTRODUCTION

L27: "ratio" should be "ratios"

Line 27: The words "ratio has" have been changed to "ratios have", as suggested.

L27: "marine biospheric activities" is a little unclear to me. I might suggest simply "...early 1990s, for the primary application of constraining the marine and terrestrial exchange of CO₂."

Lines 27-28: The sentence has been modified, as suggested.

L31: suggest "terrestrial biosphere exchanges"

Lines 31-32: The words have been changed to "terrestrial biosphere exchanges", as suggested.

L36: suggest "carbon dissociation effect" read something like "the carbonate buffer system"

Line 36: The words have been changed to "the carbonate buffer system", as suggested.

L39: I think there is a missing sentence here explaining that airborne observations are useful because they quickly map a large spatial area. Suggest cutting "from this point of view" and moving the sentence to the first sentence of the paragraph beginning with "Aircraft observations"

Lines 39-40: The sentence has been moved to the first sentence of the paragraph, as suggested.

L40-45: Steinbach (2010) might be worth citing here, since it pre-dates the Ishidoya and van der Laan references. Bent (2014) might be worth citing here as well, since he also reported Ar/N₂.

Lines 41: Steinbach (2010) and Bent (2014) have been cited, as suggested.

METHOD

This section could use subsections for easier reading.

L76: Suggest changing "Method" to "Methods" in the section heading.

Line 36: The word "Method" has been changed to "Methods", as suggested.

L78: Suggest rewording to "Minamitorishima, Japan, a small coral atoll (MNM; 24.28N, 153.98E)".

Line 78: We have modified the sentence, as suggested.

L78: "The cruising altitude is about 6km"

Line 78: The words "flight altitude" have been changed to "cruising altitude".

L79: "titanium" (not capitalized)

Line 79: "Titanium" has been changed to "titanium".

L101: Could the authors specify the scales these species are measured on, at least for O₂/N₂ and CO₂?

Lines 93-95 and Lines 112-113: The sentences have been added to specify the scales, following your suggestion.

L102-103: The authors cite Ishidoya et al 2014, which cites Niwa et al 2014, which cites Tsuboi et al 2013. I will admit I only scanned the papers but it seems Tsuboi is the only reference that describes the intake and flask sampling apparatus. So I would point directly to this paper on L81 to save the reader time. This paper does not seem to have a diagram of the flask sampler, so unless I missed it in one of these papers I think it would be nice to include either in the text or in a supplement. I think this is important because the fractionation of the samples is quite considerable. I am still not clear on what kind of inlet is actually outside the airfract. From Stephens et al 2021 it was clear that the design and orientation of the inlet is critical for avoiding fractionation. Where is the air conditioning inlet located on the plane, and what does it look like? The thermal fractionation the authors identify is so massive it is probably obscuring other sources of artifacts, like at the inlet or somewhere in the air conditioning system. Since sample air passes through this, could the authors include it in the plumbing diagram? I am also surprised the authors don't have serious problems with surface effects, given that the teflon tubing is used, flasks are only partially dried, they are pressurized fairly high to 0.4 MPa, and then analyzed (I think) without a push gas. I am sure the authors have worked all this out, and probably have already published details on it, but without details here or specific citations it's hard to understand the sampling conditions.

Lines 80-91: We have changed the sentence as "Details of the air sampling method has been described elsewhere (Tsuboi et al., 2013)" to point directly to the paper (lines 90-91). Unfortunately, details of the air sampling line from the inlet to flask sampler have not been informed to researchers from Japan Ministry of Defense. Therefore, that was the only thing we can do was to add some information from Tsuboi et al. (2013) and an issue associated with the using o Teflon tubing (Lines 80-90). As to drying, water vapor is removed by using not only Mg(ClO₄)₂ during air collection but also a cold trap at -50°C during analyses. The analyses have been carried out without a push gas, as you expected. Consequently,

the relationships between the measured $\delta(\text{Ar}/\text{N}_2)$, $\delta^{18}\text{O}$ and $\delta^{40}\text{Ar}$ with $\delta^{15}\text{N}$ show total fractionation including the processes you pointed out.

L113: Perhaps cite here that the ratio of the scaling factors 4.57/16.2 is close to the Keeling et al 2004 diffusion factor for $(\text{Ar}/\text{N}_2)/(\text{O}_2/\text{N}_2)$ and results in the same tracer $d(\text{O}_2/\text{N}_2)^*$

Lines 127-128: The sentences have been added considering your suggestion.

L115: Since all of the samples are from the same region, why not use the monthly mean at Tsukuba? I expect there will be some lag to consider, but otherwise it seems like this is introducing an unnecessary approximation.

Lines 130-132: Your suggestion is reasonable, but we leave the correction processes as they are. In this study, we simply evaluate the uncertainty associated with the seasonal $\delta(\text{Ar}/\text{N}_2)$ cycle since the spatial variations in the surface $\delta(\text{Ar}/\text{N}_2)$ have not been clarified yet sufficiently, and as you pointed out, there is some lag due to atmospheric transport to be considered.

L116: How was the uncertainty evaluated, and what terms are contributing to the total uncertainty? Are you accounting for natural variations in Ar/N_2 ? Is this what is meant in L117? How much does the annual mean of Ar/N_2 vary? Have the authors considered forcing to a constant value of Ar/N_2 ? This might be preferable since the paper deals with interannual trends.

Lines 130-138: The sentences have been added to discuss the matters you pointed out.

L119: This seems like the beginning of the "Results" section to me, since data is presented.

Lines 185-196: We have moved the sentences to the beginning of the "Results and discussion", as suggested.

L119-125 and Fig3: Could the authors include a panel showing the vertical profiles of detrended $(\text{O}_2/\text{N}_2)_{\text{cor}}$, perhaps separated by season? It is hard to evaluate the quality of the data when only shown as a time series. Also, I don't see what the bottom plot of panel b in Fig 3 is adding, since the data are also shown in the bottom plot of panel a. The authors could replace this with profiles or I change the y axis of Ar/N_2 , since there is not much that can be seen at that scale. Is it correct that the red line is the annual mean Ar/N_2 for the Tsukuba time series?

Lines 192-196, and Fig. 3(c): We have added Fig. 3(c) showing the vertical profiles of detrended $\delta_{\text{cor.}}(\text{O}_2/\text{N}_2)$ separated by season, and modified Fig. 3(b), as suggested.

L124: It would be good to indicate which samples were taken with which type of aircraft with a vertical line or some other indicator. Is it correct that only two aircraft were used? Could this be given in the

Methods? It appears that the change caused the thermal fractionation effect to be reduced by more than 400 per meg.

Line 190, and Fig. 3(a): Fig. 3(a) have been modified to show the periods when the C-130R were used by pale red shade. Some aircrafts are used as C-130H and C-130R, respectively (i.e. not limited two just two aircraft).

L150: is the "seasonal anomaly" of O₂ the Garcia and Keeling 2001 climatology?

Line 167: The words "the seasonal anomaly" have been changed to "the seasonal anomaly of the air-seas O₂ and N₂ flux", to make the meaning clearer. The flux is from Garcia and Keeling (2001), as you expected.

L153: Where does the 1.35 value for the global OR for fossil fuel combustion come from? It is lower than the values given in Keeling and Manning 2014 and I think lower than what the CDIAC data would suggest.

Lines 170-172: We have corrected the global OR for fossil fuel combustion to 1.37 calculated based on fossil emission by category summarized in GCP (Friedlingstein et al., 2020).

L156: I don't understand this sentence: "driven by an annual mean air-sea O₂ and N₂ fluxes...that was considered by Tohjima...was ignored". Also, in the Tohjima et al 2012 reference there is an unnecessary hyphen in "annual". In the Tohjima paper this seems refer to the Gruber et al ocean inversion O₂ fluxes? Was there a separate run of the Gruber fluxes? Or just two products: 1) Garcia and Keeling + Takahashi + CDIAC and then 2) simulated - observed APO?

Lines 176-178: The sentence has been changed to "In this calculation, $\delta(APO)$ driven by an annual mean air-sea O₂ and N₂ fluxes (hereafter referred to as the " $\delta^{AM}(APO)$ "), which were estimated by Gruber et al. (2001), was ignored", considering your comment. The typo in the reference has been corrected. Thank you for pointing out.

RESULTS

I suggest to cut some of the L161-177 text, the decrease in O₂/N₂, the rise in CO₂, and their seasonal cycles are well known. I would start the paragraph at "The average rates of change..." with figure citation.

We understand your suggestion, but we leave the sentences as they are, for the convenience of the readers who are not familiar with the O₂/N₂ study.

L171/Fig4: there are multiple fit lines to the data but this is not explained in the caption. Maybe a legend? I think it

I think we have explained the fit curves as “Best-fit curves to the data (solid lines) and secular trends (dashed lines) are also shown” in the caption of Fig. 4(a).

L186/Fig 6: I think it would be better to plot each latitude bin as a separate plot with observations and model together, it is a little difficult to compare as is. It would also be nice to see the seasonal cycle in the detrended observations along with the fits.

Lines 223-225, Lines 229-230, and Figs. 6 and 7: We have added not only Fig. 6(c) and (d) but also Fig. 7(c) and (d) to plot each latitude bin as a separate plot with observations and model together, as suggested. The seasonal cycles in the detrended observations along with the fits have also been added in the figures. Related sentences have been also added in the text.

L194: Suggest changing "anti-phase nature in the seasonal APO cycles" to the "opposing phase of the seasonal APO cycles"

Line 235: The words “anti-phase nature” have been changed to “opposing phase”, as suggested.

L197: I don't understand "superimposing the anti-phase seasonal cycles through the inter-hemispheric mixing of air". From this I would think you are running only the SH flux, but the "w/o SH flux run" would imply it was northern hemisphere fluxes only. Earlier in L192 it says "northern hemisphere flux only".

The "w/o SH flux run" means northern hemisphere flux only in the seasonal air-sea O₂ and N₂ fluxes, which yields significant seasonal APO cycle in the northern hemisphere only. On the other hand, in the “control run”, the anti-phase seasonal cycles in the northern and southern hemisphere are superimposed through the inter-hemispheric mixing of air in the lower latitude. We examined an effect of the inter-hemispheric mixing of air from the comparison between the “control run” and "w/o SH flux run".

L198/199: should read "the seasonal cycle in CO₂ mole fraction" or just "seasonal cycle in CO₂".

Line 240: The words “seasonal CO₂ amount fraction cycle” have been changed to “the seasonal cycle in CO₂ amount fraction”. We leave the phrase “amount fraction” as it is following the Editor’s comment.

L205: From figure 6 it looks to me that most of the seasonal cycle is due to NH fluxes, as one would expect...it does not look that much smaller to me. Can you give the amplitudes of the two runs?

From Fig. 6(b), the peak-to-peak amplitudes of the seasonal APO cycle are 8.7, 10.0, 11.6, 13.8 and 15.8 per meg at 25.5, 27.5, 29.5, 31.5 and 33.5 °N, respectively, for the “control run”, while those are 12.1, 11.9, 12.5, 14.5 and 15.4 per meg for the “w/o SH flux run”.

L213-215: I don't fully follow this--it's the gradients in the fluxes, with contributions from atmospheric transport, that causes a gradient in the amplitude in the atmosphere. I think it would be clearer to say simply that the SH makes a significant contribution to the amplitude and phase of the lower latitude observations. I would also caution against over interpreting the model results--the transport model could be over or underestimating the interhemispheric transport.

Lines 254-255: The sentence has been rewritten as “These results indicate that the SH makes a significant contribution to the amplitude and phase of the seasonal $\delta(\text{APO})$ cycle at lower latitude”, considering your comment.

L224: Suggest changing "highly" to "more"

Line 269: The word “highly” has been changed to “more”.

Fig8/9: Typically one plots altitude on the y axis, but I leave it up to the authors.

Thank you for suggestion, but we leave the figures as they are in the present study.

Fig10: This looks like a fit to the data, can you include the points as well? The data look a little odd, I would expect observations of the APO growth rate to look noisier.

Lines 289-290 and Fig. 10 (a): The figure has been added to show the deseasonalised values of $\delta(\text{APO})$ at each latitude with the secular trends. Uncertainties of the secular trends can also be found from the error bands in the figure, which will also be useful to imagine the uncertainties in the change rates.

L265: This is a rough approximation of the thermal component of APO, which is a combination of air-sea fluxes of O₂, N₂, and CO₂ caused by solubility changes. The "netbio" APO will also have a contribution from fossil fuel burning/CO₂.

Lines 312-313: The sentence has been added to note the thermal component of APO estimated in this study is a rough approximation, considering your comment.

L273-287: I am not fully convinced that this exercise accomplishes more than very roughly constraining the global APO and O₂ flux. The number of simplifying assumptions is extensive, and using aircraft observations from a comparatively small region, sparse data coverage, and enormous sampling artifacts is not an ideal approach. To me what this shows is that the corrected data have a coherent signal the authors can explain, and helps to prove that the data are of good quality. But I would caution against over interpreting and overselling the data.

Lines 334-337: The sentences have been added to include your suggestion and caution in the text.

L301: I thought 1.35 was used?

As mentioned above, we have corrected the global OR for fossil fuel combustion to 1.37 calculated based on fossil emission by category summarized in GCP (Friedlingstein et al., 2020).

L312: Where does the 4-5 year period come from? Just visual inspection? Missing here is a citation for the Nevison et al 2008 study, which (also) showed that the errors from assuming a constant zeff over short time scales (e.g. 5 years) are quite large.

We have added Nevison et al. (2008) to the reference (line 362). Tohjima et al. (2019) also examined the period based on their observational data (Fig. 7 in their study).

CONCLUSIONS

I would change this section heading to "Summary"

Line 375: "Conclusions" has been changed to "Summary".

L327: Better something like "Regular air samples were taken on cargo aircraft flights from..."

Line 376: The words "Cargo aircraft C-130 flies once per month from Atsugi Base to MNM" have been changed to "Regular air samples were taken on cargo aircraft C-130 flights from Atsugi Base to MNM".

L341 and throughout: "Superposition" is a slightly strange choice of words, I might suggest something simple like "combination".

"superposition" has been changed to "combination", as suggested.

Other changes

Lines 146-147: The sentence has been modified and Tohjima et al. (2005) has been added to reference since we have noticed that we used X_{O_2} of 0.2094 in their study to calculate the observed $\delta(APO)$.

Lines 201-203, Figs. 4 and 5: The sentence has been added to note the data selection in the digital filtering technique, and the observational data deviated from the best-fitted curves more than $\pm 3\sigma$ have been excluded from Figs. 4 and 5.

Figure 8 caption: The sentence to show the method to calculate the amplitude of seasonal APO and CO_2 cycles have been added.