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## Comment on acp-2021-787

Anonymous Referee #2

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Referee comment on "Spatiotemporal variations of the  $\delta(\text{O}_2/\text{N}_2)$ ,  $\text{CO}_2$  and  $\delta(\text{APO})$  in the troposphere over the western North Pacific" by Shigeyuki Ishidoya et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-787-RC2>, 2021

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General comments:

Ishidoya and colleagues present ~8 years of airborne observations of  $\text{O}_2/\text{N}_2$ ,  $\text{CO}_2$ , and  $\text{Ar}/\text{N}_2$  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  $\text{CO}_2$  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.

### ABSTRACT

L11 and throughout: "amount fraction" is not a term I'm familiar with, I suggest "abundances" for referring to both  $\text{O}_2/\text{N}_2$  and  $\text{CO}_2$ , and "mole fraction" for referring to  $\text{CO}_2$ .

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

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

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

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

## INTRODUCTION

L27: "ratio" should be "ratios"

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 CO<sub>2</sub>."

L31: suggest "terrestrial biosphere exchanges"

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

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"

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/N<sub>2</sub>.

## METHOD

This section could use subsections for easier reading.

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

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

L78: "The cruising altitude is about 6km"

L79: "titanium" (not capitalized)

L101: Could the authors specify the scales these species are measured on, at least for O<sub>2</sub>/N<sub>2</sub> and CO<sub>2</sub>?

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.

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  $(Ar/N_2)/(O_2/N_2)$  and results in the same tracer  $d(O_2/N_2)^*$

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.

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.

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

L119-125 and Fig3: Could the authors include a panel showing the vertical profiles of detrended  $(O_2/N_2)_{cor}$ , 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?

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.

L150: is the "seasonal anomaly" of  $O_2$  the Garcia and Keeling 2001 climatology?

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.

L156: I don't understand this sentence: "driven by an annual mean air-sea O<sub>2</sub> and N<sub>2</sub> 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 O<sub>2</sub> 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?

## RESULTS

I suggest to cut some of the L161-177 text, the decrease in O<sub>2</sub>/N<sub>2</sub>, the rise in CO<sub>2</sub>, and their seasonal cycles are well known. I would start the paragraph at "The average rates of change..." with figure citation.

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

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.

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

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".

L198/199: should read "the seasonal cycle in CO<sub>2</sub> mole fraction" or just "seasonal cycle in CO<sub>2</sub>".

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?

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.

L224: Suggest changing "highly" to "more"

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

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.

L265: This is a rough approximation of the thermal component of APO, which is a combination of air-sea fluxes of O<sub>2</sub>, N<sub>2</sub>, and CO<sub>2</sub> caused by solubility changes. The "netbio" APO will also have a contribution from fossil fuel burning/CO<sub>2</sub>.

L273-287: I am not fully convinced that this exercise accomplishes more than very roughly constraining the global APO and O<sub>2</sub> 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.

L301: I thought 1.35 was used?

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.

CONCLUSIONS

I would change this section heading to "Summary"

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

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