

Responses to the Editor

Comments to the author:

Dear Dr Ishidoya

many thanks for your revised submission to ACPD.

I believe that you have addressed most of the reviewers' comments satisfactorily (with a few exceptions as noted below).

However, I've found an error in one of the coefficients in Eq. 10 and have a few other concerns with respect to the presentation, calculations and use of units. To give you sufficient time to carry out the necessary corrections, I have indicated that major revisions are necessary even if the time required may well be shorter than the corresponding time allowance.

Best regards

Jan Kaiser

Editor ACP

Thank you very much for your significant and useful comments on the paper "Spatiotemporal variations of the $\delta(\text{O}_2/\text{N}_2)$, CO_2 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.

1) ACPD asks for the SI System of Units the Recommendations of the IUPAC Green Book to be followed. Amount fractions should therefore be reported using their SI unit "mol mol⁻¹" or derivatives thereof (e.g., " $\mu\text{mol mol}^{-1}$ "). This also avoids potential confusion between "ppm" and "per meg". There are various instances in the text and the axis labels of Figs. 4 to 7 and 9 where "ppm" needs to be replaced with " $\mu\text{mol mol}^{-1}$ ".

Throughout the paper: The word "ppm" has been changed to " $\mu\text{mol mol}^{-1}$ ", as suggested.

2) There are a couple of errors in equation 7:

a) The factor " $\times 10^6$ " after $y(\text{CO}_2)$ needs to be deleted.

b) The final term (2000) needs to be divided by 10^6 .

c) Also, amount fractions are not reported in "per meg", so "in per meg" should be removed from l. 144.

Lines 147-148: The errors in the equation have been corrected, as suggested.

3) The dry amount fractions should be written with the species enclosed in parentheses, i.e., $X(N_2)$ and $X(O_2)$, in line with the IUPAC Green Book and the practice you have adopted for other symbols in this paper.

Throughout the paper: The dry amount fractions, including $X(O_2)$ and $X(N_2)$, have been written with the species enclosed in parentheses, as suggested.

4) There is an inconsistency between $\delta(APO)$ in Eq. 7 and Eq. 8. Looking at Eq. 1, the first term in parentheses on the right-hand side of Eq. 8 needs to be divided by $y(N_2)/X(N_2)$ to match the corresponding term in Eq. 7. I acknowledge that there may be differences between measurement and modelling reference scales, but at least the underlying mathematical relationships of the quantity definitions should be the same.

5) Again, the first term on the right-hand side of Eq. 10 needs to be divided by $y(N_2)/X(N_2)$. Also, the coefficient in the final term (for TB) should be α_B , not α_F . Please check whether your calculations based on this equation are correct.

Lines 165-181: The sentences have been rewritten as follows, considering your comments.

“Model-based changes in $\delta(APO)$, CO_2 amount fraction and $\delta(O_2/N_2)$ ($\Delta\delta(APO)$, $\Delta y(CO_2)$ and $\Delta\delta(O_2/N_2)$) were calculated using the following equations (e.g. Nevison et al., 2008; Tohjima et al., 2012) in per meg, $\mu mol mol^{-1}$ and per meg, respectively:

$$\Delta\delta(APO) = \left(\frac{\Delta y^{SA}(O_2)}{X(O_2)} - \frac{\Delta y^{SA}(N_2)}{X(N_2)} \right) + \left(\frac{-\alpha_F \cdot \Delta y^{FF}(CO_2) + \alpha_B \cdot \Delta y^{FF}(CO_2)}{X(O_2)} \right) + \frac{\alpha_B \cdot \Delta y^{OC}(CO_2)}{X(O_2)}, \quad (8)$$

$$\Delta y(CO_2) = \Delta y^{FF}(CO_2) + \Delta y^{OC}(CO_2) + \Delta y^{TB}(CO_2), \quad (9)$$

$$\Delta\delta(O_2/N_2) \cong \left(\frac{\Delta y^{SA}(O_2)}{X(O_2)} - \frac{\Delta y^{SA}(N_2)}{X(N_2)} \right) + \frac{-\alpha_F \cdot \Delta y^{FF}(CO_2)}{X(O_2)} + \frac{-\alpha_B \cdot \Delta y^{TB}(CO_2)}{X(O_2)}, \quad (10)$$

where $\Delta y(O_2)$, $\Delta y(N_2)$ and $\Delta y(CO_2)$ are changes in dry amount fractions of the respective gases calculated using NICAM-TM. The superscripts “SA”, “FF”, “OC” and “TB” denote the seasonal anomaly of the air-seas O_2 and N_2 flux, CO_2 flux from fossil fuel combustion, ocean and terrestrial biosphere, respectively. X_{O_2} and α_B have the same meaning as in Equation (7), while $X(N_2)$ is the dry amount fraction of N_2 in the atmosphere and α_F is the global average OR for fossil fuel combustion. In this study, we adopted $X(N_2) = 0.7808$ and $\alpha_F = 1.37$. The α_F was calculated from the fossil fuel and cement production emissions by fuel type for the period 2012-2019, reported by GCP (Friedlingstein et al., 2020), and the oxidative ratios for the different fuel type were taken from Keeling (1988). It should be noted that we assume initial amount fractions of $y(O_2)$ and $y(N_2)$ are equal to $X(O_2)$ and $X(N_2)$, respectively, in eqs. (8) and (10). We can rewrite equation (8) as:

$$\Delta\delta(\text{APO}) = \Delta\delta^{\text{SA}}(\text{APO}) + \Delta\delta^{\text{FF}}(\text{APO}) + \Delta\delta^{\text{OC}}(\text{APO}). \quad (11)$$

Finally, $\delta(\text{O}_2/\text{N}_2)$, $\gamma(\text{CO}_2)$ and $\delta(\text{APO})$ obtained from NICAM-TM are reported as respective deviations of $\Delta\delta(\text{APO})$, $\Delta\gamma(\text{CO}_2)$ and $\Delta\delta(\text{O}_2/\text{N}_2)$ from arbitrary reference points, in other words, reported on different scales from those of observations.”

5) Again, the first term on the right-hand side of Eq. 10 needs to be divided by $y(\text{N}_2)/X(\text{N}_2)$. Also, the coefficient in the final term (for TB) should be α_B , not α_F . Please check whether your calculations based on this equation are correct.

Eq. 10: The coefficient in the final term has been changed to α_B . This is just a literal error, and our actual calculations of $\Delta\delta(\text{O}_2/\text{N}_2)$ are correct.

6) The "C" in "CO2" in Eqs. 8 and 10 should be written in upright font.

Eq. 8 and 10: We have revised them according to your instruction.

7) l. 22: Please split this sentence into a readable form, e.g., "annual net sea-to-air marine biological O₂ flux during El Niño and net air-to-sea flux during La Niña." Parentheses should be used for clarification, not saving space (cf. Alan Robock's EOS piece, "Parentheses Are (Are Not) for References and Clarification (Saving Space)", <https://eos.org/opinions/parentheses-are-are-not-for-references-and-clarification-saving-space>).

Line 22: The sentence has been modified, as suggested.

8) l. 32: The equal sign "=" and the unit "mol mol⁻¹" should be deleted. If you want to emphasise that you are referring to an amount ratio, this should be done by an appropriately name quantity, e.g., "oxidative amount ratio", but I don't think that is necessary.

Line 32: The equal sign "=" and the unit "mol mol⁻¹" have been deleted.

9) l. 43 and elsewhere: Height should be replaced with altitude. Altitude is the vertical distance between an object and sea level. For an aircraft, this would be 100s or 1000s of metres. In contrast, the height of an aircraft would typically be of the order of metres.

Throughout the paper: The words "height" have been changed to "altitude", as suggested.

10) l. 87: I think the position of the word "only" is not as intended. Do you mean to say, "we only use Teflon tube in the upstream ..." (i.e., not elsewhere)? Or do you mean to say that you only use it to avoid absorption? Part of the reason I am asking is because Teflon performs much worse in terms of O₂ absorption and permeability than glass or steel. Also, Langenfelds (PhD Thesis, 2002) reported PTFE (Teflon) o-rings to have the worst performance for O₂/N₂ stability in storage experiments.

Lines 87-89: The sentence has been modified to make the meaning clearer as “It is noted that we only use Teflon tube in the upstream of the diaphragm pump to avoid absorption and/or permeation of O₂ due to a pressurization of the Teflon tube”.

Was January 2016 the only flight when you used Teflon as tubing material downstream of the pump? What tubing material did you use downstream of the diaphragm pump during other flights? If you don't have that information, please explain why.

Lines 91-92: The sentence “During other flights, we use a flexible tube made of stainless steel in the downstream of the diaphragm pump” has been added.

11) l. 96: Please add the specific DOI (cf. section "Data availability").

Lines 98-99: We have added the specific DOI.

12) l. 118: The unit should be written "per meg (per meg)⁻¹".

Lines 120 and Fig. 2: The words "per meg per meg⁻¹" has been changed to "per meg (per meg)⁻¹".

13) l. 131: Please split this sentence into a readable form.

Lines 133-135: The sentence has been modified, as suggested.

14) l. 146: Which value of X(O₂) did you use for your calculations?

Lines 151: We have added the information.

15) l. 190: How do you explain the much lower variability in $\delta(\text{Ar}/\text{N}_2)$ data for periods of C-130R aircraft use? It seems odd that a mere a change of aircraft type should lead to such a reduction. Were there differences in the inlet or sampling system? If you don't have that information, please explain why.

Lines 77-79, 84-87 and 202-206: Details of the air sampling line from the inlet to flask sampler have not been informed to researchers, so that it is difficult to explain the cause(s) of the much lower variability in $\delta(\text{Ar}/\text{N}_2)$ data for periods of C-130R. The visible difference is the location of the air-conditioning blowing nozzles of C-130H and C-130R. The former and the latter are attached at the roof in the cockpit and at the front of the assistant driver's seat, respectively, and air sampling tubes are inserted into the nozzles.

16) l. 199: This description is a bit too sparse to reproduce what you have done. Please include more detail on what filter parameters you used, e.g., the cut-off periods, filter orders, etc. When you write, "fundamental and first harmonics", do you mean two frequencies? Or a linear change (secular trend)

and just one frequency (presumably annual)?

Lines 209-215: The sentences have been rewritten to add the description of the digital filtering technique.

17) l. 237 & elsewhere: Please avoid abbreviations such as "w/o". Also, it would be clearer to always write the expression "without-SH-flux run" with hyphens.

Throughout the paper: The word "w/o" has been changed to "without-SH-flux run", as suggested.

18) l. 279, Figs. 8b and 9b: The text refers to 6 km, but the values in Fig. 9b are relative to 1.3 km. More importantly, the queries from referee 3 about the different reference altitudes (panels 8b and 9b) and, I'd like to add, also the reference latitudes (panels 8a and 9a) have not been addressed.

Lines 264 and 294, Figs. 8a, 8b, 9a and 9b: The text has been corrected to refer to 25.5°N and 1.3 km. The figures and related captions have been modified to address the referee's and your queries.

19) l. 280, 395, 396, 399: Is NICAMT-TM different to NICAM-TM, or is this a typo? If not a typo, please explain the differences between NICAMT and NICAM.

Throughout the paper: The typo "NICAMT-TM" has been corrected to "NICAM-TM".

20) l. 311: Why 0.84? The solubility of O₂ is about 0.91 times the solubility of Ar.

Line 326 and Fig. 11: The word "0.84" has been changed to "about 0.9", and we have updated Fig. 11 using the coefficient of 0.9. As you know, the coefficient changes slightly depending on sea water temperature.

21) l. 332 & 345: "yr" should be replaced with "a".

Lines 347 and 360: The word "yr" has been changed to "a".

22) Caption Figs. 4 & 5: Please include a reference describing how you obtained the "best-fit" curves and secular trends.

Caption Figs. 4 & 5: The reference "Nakazawa et al. (1997)" has been added to the captions.

Finally, the font size of the labels in Figs. 2 to 10 is too small to be easily legible. Please increase it to match at least the size of the main text (when the figure is scaled to the size intended for reproduction).

Figs. 2 to 10: The font size of the labels have been increased. If it is still small, please point them out.