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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric measurements of the terrestrial $O_2 : CO_2$ exchange ratio of a mid-latitude forest" *by* Mark O. Battle et al.

Anonymous Referee #1

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In this paper, Battle and co-authors, conducting precise observation atmospheric CO2 and O2 concentrations at the Harvard Forest during 2006-2013, examine the correlative changes to evaluate the O2:CO2 exchange ration associated with terrestrial biospheric processes, αB. Although the value of αB is basic parameter to understand the atmospheric O2 variation and the value of 1.10 is commonly used for long time, there are still discussions regarding the absolute value. Finding the average correlation slope in the forest is significantly lower than 1.1, the authors conclude that the value of 1.1 for αB should be adjusted to slightly lower value as several other studies have already suggested. I believe this study considerably contributes to the atmospheric O2 study field and contains material that should be published in Atmospheric Chemistry and

Physics. However, I find a problem in processing the observed data and incomplete descriptions of the modeling studies to evaluate the contributions from the biotic and combustion emissions. An effort should be made by the authors to improve the data treatment and clarity of the manuscript before acceptance.

General comments: Although the authors carried out highly precise measurements of the atmospheric O2, I have a concern about the units of the O2 measurements used in this paper. The authors use units of mole fraction, μmol per mol, to express the O2 variations as well as CO2. However, the use of mole fraction to express variations in major atmospheric constituents like O2 are very confusing because of the influence of dilution effect (e. g. Keeling et al., 1998). For example, adding 1 μmol of CO2 to and removing 1.1 μmol of O2 from an air parcel containing 1 mol of dry air with 0.21 mol of O2 results in a 1 ppm increase in the CO2 mole fraction and 1.08 ppm decrease in the O2 mole fraction. Therefore, the correlation slope based on the atmospheric observation is -1.08, which is lower than the original O2:CO2 ratio of 1.1. In the similar way, if the authors truly use the mole fractions both for O2 and CO2, the correlation slope of 1.10 corresponds to the O2:CO2 exchange ratio of 1.126. To avoid such confusing situation, units of per meg was defined by Keeling and Shertz (1992). The authors, therefore, should use "per meg" or ppm equivalent calculated as a product of "per meg" and 0.2094.

To evaluate the value of αB from the field observation in the forested area, it is critically important to quantitatively assess influences of emissions from the fossil fuel combustions. The authors examine the contributions from the fossil fuel and biogenic fluxes in Section 4 by using a Lagrangian transport model. However, the results of the examinations are not clearly shown as are described in detail in specific comments as follows. In addition, except for such a modeling approach, I think that the observations like CO and 14C are useful to assess the influence of the fossil fuel fluxes. If such observations were carried out at the site, it would be better to use those data.

Specific comments: Page 1, line 7: It says here that the upper intake is placed about 6

m above the forest canopy. But, the height is about 5m in Method (page 3, line 4).

Page 1, line19 and 20: It would be better to number the equations. In addition, I understand that these budget equations were original forms given by Keeling and Shertz (1992), but recent studies generally include ocean O2 outgassing term in the O2 budget equation. So, I think it would be better to add the O2 outgassing term because it's considered that its uncertainty is the largest source of error for the carbon budget estimation based on the atmospheric O2 measurements.

Page 3, line 3: I think there is no need to show the position (latitude and longitude) of the site with a 0.1 m precision.

Page 3, line 21-23: It would be better to mention the size and the material of the calibration tanks. What are the uncertainties of the concentrations of O2 and CO2 of the standard cylinders listed in Table 1. Were the O2 concentrations in the calibration tanks stable during the entire observation? Could you show any experimental evidences of the O2 stability? In addition, "per meg" units are used for the O2 concentrations in Table 1, but there is no explanation of the units in the text.

Page 5, line 30: Does the "intake selector valve" correspond to the "Cross-over ball valve" in Figure 1? It should be clarified.

Page 6, line 7-10: I think that the long-term stability in the O2 and CO2 concentrations in the reference tanks is also critically important to accurately assess the O2:CO2 exchange ratio. Thus, it would be better to show some experimental results to confirm the stability of the reference tanks.

Page 6, line 27: During the 6-year duration, the O2 concentration of the background air decrease by more than 20 ppm. But it is difficult to see such decreasing trend in the O2 time series shown in Fig. 3.

Page 6 line 27: It would be better to change "due to fossil fuel combustion" to "mainly due to . . .".

Page 7, line 29: It is mentioned that in the 1-deminsional box model the air parcels travel forward in 12-minute steps. I think that the air parcels move along with the backward trajectories computed by a Lagrangian transport mode. Is it right? If so, the authors should clarify that.

Page 8, line 3-4: Here, the authors mention that the PBL height used in atmospheric transport models like STILT is used for h. However, in the following section, the authors mention that climatological hourly averages from the NAM12 dataset are used as the PBL values. Are these PBL heights same? Please clarify it.

Page 8, line 11-12: I don't really know what the sentence, "We performed sensitivity . . . and PBL height", means. How are the landscape composition, surface flux magnitude, and PBL height varied? Are they varied within the uncertainties? What is the ranges of the variations? Please clarify it.

Page 8, line 16-17: Here the authors mention that the results can be found in Conly (2018). But, Conly (2018) is a bachelor's thesis, not a peer-reviewed paper. So, I think the authors should show at least main results of the 1-D model experiments here.

Page 8, Section 4.2 (Region of influence): Why don't the authors show the average footprint of the atmospheric observation at the EMS tower? Would comparison between the average footprint and the maps showing biogenic and fossil fuel fluxes convince the

Page 8-9: Why don't the authors quantitatively evaluate the fossil fuel-derived CO2 contributions by using the above mentioned 1-D box model and the CO2 biogenic and fossil fuel-derived fluxes described in Section 4.3? I believe that such an approach is simple and straightforward.

Page 9, line 4-5: How do the authors obtain the number "10-20 times"?

Page 10, line 2-3: Can the large αB value for the soil respiration explain the high/low difference of the observed -O2/CO2 exchange ratio?

Page 10, section 5.3 (Temporal variability in slopes): Since the seasonal variation in the slope and αB is discussed in this section, the seasonal O2/CO2 slopes should be shown. The authors mention "The seasonal cycle in slopes may be due to seasonal changes in αB itself" (page 10, line 32). But immediately after that, it's mentioned that the fossil fuel contribution has significant seasonal variability: strong in winter and weak in summer. Do the authors consider that αB changes seasonally? If so, seasonal variation in αB estimated from the experimental result of this study should be clarified because it is very important to understand the atmospheric O2 and APO variations.

Page 17, Figure 1: Were the standard tanks placed vertically on the floor as shown in the figure? The standard tanks used for the O2 measurements are usually placed horizontally in the thermally insulated box to reduce fractionation effect on the O2 concentration due to the gradients of temperature and hydrostatic pressure within the tanks.

Page 22, Figure 6: What are the dashed-dotted lines in the figures? There is no explanation in the text and the figure captions.

Page 23, Figure 7: Why the backward trajectories after the observation period are adopted in the figure?

Page 24, Figure 8: These histograms are not discussed in the text.

Page 27, Table 2: Why is the order of the four data subsets for the 4-hour interval length (night-h, night-l, day-h, day-l) different from that for the 6-hour interval length (day-h, day-l, night-h, night-l)?

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-1041/acp-2018-1041-RC1-supplement.pdf

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