Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-652-RC1, 2019

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Consumption of atmospheric O_2 in an Urban Area of Tokyo, Japan derived from continuous observations of O_2 and CO_2 concentrations and CO_2 flux" *by* Shigeyuki Ishidoya et al.

Anonymous Referee #2

Received and published: 27 November 2019

In this paper, the authors use continuous and simultaneous measurements of O_2 and CO_2 to determine the net oxidative ratio (OR) of atmospheric influences in a densely settled urban location. They then use an independent set of CO_2 flux measurements to calculate an eddy diffusion coefficient (K). With K in hand, they use vertical gradients in O_2 and CO_2 (in particular, differences in measured O_2 and CO_2 at two different heights on their sampling tower) to infer local fluxes of O_2 and CO_2, and from these, the net OR for fossil fuels. Assuming only two fossil fuel types (gaseous fuels and liquid gasoline), each with its own distinct OR, the combination of total emissions and net OR constrains

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Discussion paper

the relative fractions of the two fuel types.

Overall, the paper presents high-quality measurements that are definitely worthy of publication. The use of the high-low differences, in conjunction with the eddy-flux measurements of CO_2, is clever and yields valuable information about surface O_2 fluxes. Unfortunately, there is a significant problem with the interpretation of the measurements that needs to be addressed. In addition, there are many spots throughout the manuscript that require clarification.

Primary scientific concern:

One of the major challenges of working with tower data is determining the region of influence (the "footprint") for the tower. Calculations of the OR from $O_2 - CO_2$ covariation are particularly challenging, since the lower-frequency data from a paramagnetic analyzer lend themselves to aggregating data over extended periods. The authors acknowledge this in lines 133-135. However, the problem in this analysis is more profound than simply scaling footprints (inversely) by data-rate. This is because the OR slopes shown in Fig. 4 (lower panel) include data from the entire 18 month set of observations. Consequently, this is effectively a global average number with local influences superimposed.

To understand this, first consider a point in the plot with very low O_2 (and high CO_2). Maybe this parcel started with relatively high O_2 and was influenced by a great deal of local combustion. Or maybe it's part of an air mass that arrived from some distant location (highly influenced by combustion) and was relatively unaffected by local fluxes. Compare this to a point with relatively high O_2 (and low CO_2). If this point was measured hours before the low-O_2 one, and the wind pattern was roughly constant, chances are good that O_2 fell due to local combustion. In contrast, if this high-O_2 point was measured days (or months) before, it might have come from a totally different region and the difference from the first point reflects local influences to a much smaller degree. One solution is to choose much shorter aggregation periods when determining

OR_{atm}.

In short, all of the analysis of OR_{atm}, and the comparisons of OR_{atm} with OR_f need to be reconsidered.

For this reason, I will not comment further on the parts of the manuscript that involve the interpretation of OR_{atm}.

Other scientific concerns:

Line 30: In addition to Mitchell et al., please cite Sargent et al., PNAS 2018.

Lines 59-60: Does the vegetated area actually change seasonally? Or is it that the vegetation is mostly dormant in the winter?

Lines 71-72: As I understand it, the samples are measured with a paramagnetic analyzer relative to secondary standards. It's the secondary standards that are measured against the primary standard with a mass spectrometer. This is not what this sentence says.

Lines 73-75: Air is being drawn down at 10l/m and a very small subset of that airstream is being analyzed. There is no mention here of the possibility of fractionation at this sampling, of tests to detect fractionation, nor of measures to prevent it. This is something that Stephens et al. (DOI: 10.1175/JTECH1959.1) discusses extensively. Perhaps this is discussed in the original methods paper, but it should at least be mentioned here.

Lines 75-76: If air is measured first at one height, then the other, and air is measured for 10minutes at each height, isn't each measurement cycle 20minutes long (and thus, 9 cycles is 180 minutes)?

Line 79: How is a correction made for Ar? The paramagnetic analyzer doesn't measure this species. Again, this might be presented in 2014 Tellus paper, but a few words of explanation here would be welcome.

Line 83: Why are uncertainties being quoted for 30minute averages when atmospheric measurements are only made for 10-minute intervals on each intake, and standards are measured for 5-minute intervals.

Line 91: What does “span-difference” mean? Please clarify.

Lines 114-115: Downward excursions in O_2 may be due to consumption within the canopy, or non-local influences being transported to the tower. If they coincide with positive excursions in deltaO2, then I would be convinced that the cause is consumption within the local canopy, but until you show that the two excursions are coincident, you can't claim local consumption is the cause.

Line 154: If errors in both species are non-negligible, a standard least-squares linear regression will give the wrong slope. Instead a Deming regression is required (which reduces to an orthogonal fit in the case of equal areas).

Lines 183ff: A very basic back-of-the envelope calculation would be appropriate here to indicate whether human respiration really was utterly negligible or not. For example, the population density given for this area is 0.016 people m^{-2}. If each requires 2000 kcal/day, this could be supplied by metabolizing 3.34 moles of glucose, with a resulting consumption of $3.7 \mu mol m^{-2} s^{-1}$ of atmospheric O_2. This seems to be about 20% of the smallest values quoted on line 232: A modest, but non-negligible correction to the results presented here.

Minor editorial comments:

Line 44: Change to “In this paper, we first present the. . .”

Line 74: should read “and 37m was introduced”

Line 75: should read “100mL min^{-1} with the pressure stabilized to 0.1 Pa and measured”

Line 85: should read “We used the gravimetrically prepared air-based”

Line 86: should read “1991) to determine”

Lines 87 and 90, “gravimetrically standard” should be replaced with “gravimetrically prepared standard”

Line 107: should read “activities. In contrast, the atmosphericO_2”

Line 111: should read “Therefore, we attribute the opposite phase” and “in this study mainly to fossil”

Line 124: Remove “by”

Line 131: End the sentence with “troposphere” and simply remove “whereas. . .”

Line 134: should read “1994). We note that”

Line 204: should read “standard error ($\sigma/\sqrt{n}$)” (i.e. use symbols instead of writing it out).

Line 205: should read “negative values respectively, indicating”

Line 206: end the sentence with “the year.” and remove “respectively”.

Figure 6: There is no legend explaining the filled and unfilled symbols in the upper panel.

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