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Atmospheric Chemistry and Physics Discussions

Interactive comment on "CO_2, $\vec{\delta}O_2/N_2$ and APO: observations from the Lutjewad, Mace Head and F3 platform flask sampling network" *by* I. T. van der Laan-Luijkx et al.

Anonymous Referee #2

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This paper presents an interesting and likely important dataset on trends and cycles in CO2 concentration and O2/N2 ratio from sites in Ireland, the Netherlands, and a platform in the North Sea. The data call attention to an interesting gradient in atmospheric concentration between Ireland and the Netherlands which appears to have grown with time. The interpretation is left open, but the result is thought provoking. The data are also used to estimate global sources and sinks. The paper includes a very useful summary of the relationship between trends and seasonal cycles observed at these sites in relation to other sites where O2/N2 data have been reported in Europe. Overall, the data are useful in the context of improving our understanding of both global and regional sources and sinks of carbon dioxide, and their presentation makes an important

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Interactive Discussion

Discussion Paper

contribution.

The paper has a few significant problems, however, that need attention via new analysis and extensive revision.

My greatest concern involves the approach used to interpret global sources and sinks of CO2. The authors advance a new approach which takes account of regional variations in the oxidative ratio of fossil fuels. The local ratios are somehow established using a regional atmospheric transport model (REMO) taking account of fossil-fuel emission, fuel types, and atmospheric transport. One problem is that the approach used to compute these ratios is not well described. But more troublingly, the authors then apply this local oxidative ratio in a global budget calculation to estimate global land and ocean CO2 sinks. The approach is not defended in terms of a rigorous mass balance.

Even in a partly contaminated site or a site impacted by regional emissions, the long-term trends in CO2 and O2/N2 must mostly follow the global background. The interpretation of trends from such a site therefore still requires information, such as global oxidative ratios, needed to interpret global trends, but may additionally require information about trends in regional or local emissions. It is troubling, therefore, that the authors appear to interpreting long-term trends (Section 4.5) taking only regional oxidative ratios, but not also global ratios into account.

If the authors feel that the trends at their stations are strongly influenced by local or regional emissions (or meteorlogy), but nevertheless wish to interpret the trends in terms of global sources and sinks, they will need first to come up with a means to estimate the regional/local contribution to the trend, allowing background trends to be computed by difference. The inferred background trends can then be fed into the global budget equations for computing global sources and sinks, which would use the global average oxidative ratio. Note that to estimate the regional/local contribution to the trend would not just require information on local oxidative ratios, but also require information

ACPD

10, C6499–C6504, 2010

about the absolute magnitude of the CO2 and O2/N2 signals contributions from fossil-fuel combustion. The latter will require careful analysis, as it will depend on the complex interplay between high frequency variability and sampling protocols. Any model used for this purpose, such as REMO would need to be very carefully tested against other tracers of local sources at the site.

Undertaking the above steps would require extensive new analysis and major revision of the manuscript. A simpler approach may be to use only the Mace Head data, which is presumably less impacted by regional emissions, for the global sink calculation. Taking this approach will also eliminate the need for the analysis presented with the REMO model, which would then need to be cut as it becomes irrelevant to the analysis and without purpose. I'm afraid there does not seem to be a fix that avoids major revision.

Turning to other general issues, the interpretation of the change in the O2/N2 and APO gradients between Mace Head and Lutjewad overlooks the possibility that these may be driven by interannual variations in air-sea O2 fluxes, e.g. in the North Atlantic. Significant interannual variations have recently been inferred from O2/N2 measurements as recently reported by Hamme and Keeling (2009) and Roedenbeck et al (2009) both in Tellus B. Hamme and Keeling particularly emphasize the potential for changes in ocean ventilation to drive variability. A change in ventilation would be expected to drive changes in CO2 and O2/N2 of opposite sign, but with much larger changes in O2/N2 on a molar basis, consistent with the Mace Head/ Lutjewad observations. This mechanism therefore could possibly also account for the changing CO2 gradient. This possibility needs to be discussed. The issue is also relevant for the use of Mace Head data in global sink calculations.

Although the flask data are clearly useful for resolving seasonal and long-term trends, the scatter (e.g. Figure 3) looks high compared to other sites where observations have been made. Some analysis of the sources of this scatter is needed. In particular, it is important to know whether the authors think this scatter can be interpreted as real variability.

ACPD

10, C6499–C6504, 2010

The organization is mostly good, but I notice that problems with assessing the seasonal amplitude at Lutjewad are discussed in two places (page 13067 lines 3-7 and page 13068-9, lines 22. These discussions would better be grouped together.

Some attention is needed to improve the clarity of the presentation, as discussed more specifically below.

More specific points:

Page 13069, line 11 onwards: The offset between Mace Head and Lutewad and particularly any trend in the offset is barely visible on this plot. The discussion of Figure 4 is therefore hard to follow, because the figure does not show well the features being discussed. The main points to be drawn from Figure 4 are not helped showing data from station F3, which adds clutter to the plot. Even better than removing F3, however, would be swapping out Figure 4 for another figure showing Lutejwad Mace Head differences versus time, perhaps based on monthly data, or some such, rather than a fit.

Page 13069, line 11 onwards: The sign convention in discussing gradient changes is also not very clear. The problem might be reduced if a new figure were drawn (as suggested above) so that the trends in the gradient was visible by eye, but the discussion in the text could also be clearer on this point. Best would be to rephrase the text so that the reader doesn't need to keep a sign convention in mind. For example, the discussion could focus on the Mace Head CO2 deficit, or the Mace Head O2/N2 excess.

Page 13069, line 20. The change of 0.5 ppm seems to lie outside the range found by Ramonet et al, which at face value is at odds with the statement that the result "fits well into the general picture presented by Ramonet et al.". There is doubtless a valid point to be made here, but it needs to be more carefully worded so that it doesn't appear to be contradicted by the numbers.

ACPD

10, C6499–C6504, 2010

Interactive Comment

Page 13069, line 24-26. It's not clear why changes in boundary layer height and emissions changes would cause the gradient to change. A more precise statement about the direct of the trends in boundary layer height and emissions would help. Where are emissions changing, and in what direction? Where is the boundary layer height changing, and in what direction?

Page 13069, line 26-27. Why would an increasing negative gradient in CO2 be expected to be accompanied by an increasing negative gradient in O2/N2? Combustion and land exchanges cause changes of opposite sign, not the same sign. What process is being assumed in making this estimate?

Page 13070, line 5, change "is observed in" to "is recorded in"

Page 13070, line 5. "oxidative ratio at Mace Head" This cannot refer to combustion at the site itself. How should this be interpreted?

Page 13070, line 6-8. An example of a sentence that cannot be understood without knowing the sign convention applied to the gradients.

Page 13072, first paragraph It's a challenge to understand what is being presented here and why. The concept of an oxidative ratio at a specific site is problematic, as an oxidation ratio typically refers to a source mechanism, e.g. combustion, rather than as a ratio of tracer concentrations. This concept needs a more precise definition, and probably the use of a different term instead of oxidative ratio, to avoid confusion. More detail is needed to understand how the ratio is computed. For example, does the calculation include concentration changes caused by European emissions that leave and then reenter the REMO domain? If not, why not? It is also not clear why these calculations are of interest. What question is being posed? Magnitudes would appear to be as interesting as ratios. How large is the CO2 contribution at each site, for example?

Page 13073, line 14, What is meant by minimum daily oxidative ratio? Is the ratio

ACPD

10, C6499–C6504, 2010

Interactive Comment

related to diurnal variations as simulated in REMO? Why the term “ minimum”?

Page 13075, line 2-3. “gradient between both locations” A gradient is always between two more locations, so why both? Sign convention is again here unclear. See above comment. Better to discuss in terms of the Mace Head excess or deficit compared to Lutjewad, or vice versa.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 13055, 2010.