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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.

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Author's reply to anonymous referee #2

The authors first of all would like to thank the referee for the thorough review of the manuscript. The concerns expressed in the referee report have been studied by the authors and have been used to revise the manuscript substantially. A reply to each individual comment is given below and are grouped as 4 general points and 11 specific points.

GENERAL POINTS

Interpretation of the global sources and sinks: The authors are thankful that the referee has pointed out this issue. After studying the referee report, the authors agree with the referee that the APO trend at Lutjewad does not represent the global situation and is therefore not suited in a study of the global marine CO2 uptake. The trend at Lutjewad is supposed to follow the global average, but is deviating from this during the period of our observations. This situation cannot continue to exist and eventually the trend at Lutjewad should recover to the global situation by atmospheric transport and mixing. We have therefore chosen to use only Mace Head in the global budget calculations.

Section 4 has been reorganized in order to group the discussions of the long-term trends, the seasonal amplitudes and the comparisons to other European stations. The comparison to the GLOBALVIEW-CO2 background reference has been shifted to section 4.1 with the discussion of the seasonal patterns in order to leave out the repeated parts as mentioned by the referee. The old section 4.3 (APO) has been moved to become section 4.2. The new section 4.3 is now dedicated to the gradients between Mace Head and Lutjewad. The old section 4.4 is included in the discussion of the gradients. Following the recommendations of the referee, we have not used the Lutjewad record for the global ocean sink determination anymore. Nevertheless, we retain the part about the REMO model, because the concept of regionally different OR and their effect on the sites is illustrative and interesting.

The section on the gradients has been extended with the information on the concept of the perceived OR. For this purpose we use the perceived OR as obtained using REMO. The concept of perceived OR is described in a more clear way in the new version, to explain better how it was obtained and that it does in fact represent a combination of global and local fossil ORs. The perceived OR is a correction to the global 1.4. See also below with the specific comment addressing this issue.

The REMO simulations for Lutjewad have been tested against other tracers of local influences at the site. The comparison between modelled and observed 222Radon and fossil fuel CO2 has been presented by van der Laan et al. (2010) in Tellus B. A

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comparison study for CO has also been performed, but is not published. Both tracers agree well between model and observations.

Changing gradients: The section on the gradients has been changed considerably using the suggestions of the referee. The replies to the specific comments relating this issue are included below. The suggestion to include Hamme and Keeling (2008) and Rödenbeck et al. (2008) has been taken up and this has been included in the discussion of the Lutjewad-Mace Head gradients. (A calibration problem to explain the difference in the trends between Lutjewad and Mace Head is excluded based on the fact that flask samples from both locations are measured on the same instrument in the same lab).

Scatter in observations: The following paragraph has been included in section 4.1 in order to address this issue: "The observed signals for O2 and CO2 from Lutjewad and also F3 show a higher degree of variability compared to Mace Head. These stations are more influenced by local and regional sources and sinks of CO2 and O2 (e.g. terrestrial biosphere and fossil fuel emissions) and the sampling and filtering procedures do not adequately exclude these disturbances. The regional influence at Lutjewad has been presented by van der Laan et al. (2010) and is used to estimate national fossil fuel CO2 emissions."

Repeated discussion seasonal amplitude Lutjewad: This has been changed (see above).

SPECIFIC POINTS

P13069 L11 onwards 1st comment: It is true that the change in the gradients is not well visible from figure 4. The discussion of the comparison of the fits between the three stations and also GLOBALVIEW-CO2 has been shifted to section 4.1 (the results of the observations of CO2 and O2). Figure 4 is helpful in that section and the authors have therefore chosen to keep it in the new version. The section on the Lutjewad-Mace Head gradients is in the new version not anymore linked to the figure. The authors

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have tried to come up with a clearer figure to show the changing gradients. However, the monthly averaged data does not give a much clearer picture. The authors have chosen to not include an additional figure based on only the long-term linear trends, as this is not more clear than the numbers only.

P13069 L11 onwards 2nd comment: The text has been modified to avoid confusion on the gradient subject. The authors now use the terms CO2 excess and O2 deficit to address the difference between Lutjewad and Mace Head.

P13069 L20: The change of 1-2 ppm as mentioned from Ramonet et al. (2010) refers to the change observed at several Eastern European sites. As Lutjewad is not comparable to those stations, it was not a good choice to compare our gradient with these numbers. The text has therefore been modified to show the general picture for all sites studied by Ramonet et al. which is up to 2 ppm.

P13069 L24-26: The effect of the changes in the boundary layer height over the continent and the regional changes in emissions have been thoroughly studied by Ramonet et al. and are therefore not repeated in our manuscript. The changes in boundary layer height over the continent affect the mixing ratios at Lutjewad and not at Mace Head. Samples at Mace Head are taken under restricted baseline conditions and therefore represent background conditions, whereas Lutjewad is more affected by changes on the continent and therefore also by changes in the boundary layer height. The same is true for the regional changes in emissions, especially to changes in the fossil fuel mix which is shifting towards a higher share of natural gas.

P13069 L26-27: The authors meant to say that the CO2 and O2 gradients were changing with the opposite sign as the referee suggests. However, due to the chosen terminology this was not clear in the manuscript. The new terminology as explained before using the CO2 excess and O2 deficit should clarify this issue.

P13070 L5 (1): Changed accordingly.

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P13070 L5 (2): The text has been modified to clarify this issue. The oxidative ratio differs between the Netherlands and other countries in Europe (or even world wide) and this creates a gradient between Lutjewad and Mace Head.

P13070 L6-8: Also this sentence has become more clear using the new terminology as suggested by the referee.

P13072 paragraph 1: This section has been rewritten to clarify this issue and is in the new version included in section 4.3 covering the gradients between Lutjewad and Mace Head. This paragraph is dedicated to the results of our REMO simulations with the CO2 emissions and locally distributed oxidative ratios as described in section 3.4. The input of the model (i.e. CO2 and O2 concentrations split in three components: ocean, biosphere and fossil fuel) undergo atmospheric transport and mixing and the model simulations yield the CO2 and O2 concentrations for each grid cell. The fossil fuel component of these concentrations is used to calculate the perceived fossil fuel related OR (ORff) per grid cell: ORff = -O2 ff/CO2 ff. Since the semi-hemispheric model domain covers the whole area north of 30°N, the influence of the European emissions is transported with the westerly winds around the globe and reaches the European region again, of course after substantial mixing. The (comparatively small) part of the signal of European emissions that is transported southward across 30°N can re-enter the model domain as long as it is also represented in the global transport model.

P13073 L14: The text has been adjusted to clarify this sentence. Here the perceived OR is meant, which is explained with the previous comment.

P13075 L2-3: The new terminology as suggested by the referee has also improved the clarity of this sentence.

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