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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The influence of the vertical distribution of emissions on tropospheric chemistry" *by* A. Pozzer et al.

A. Pozzer et al.

pozzer@cyi.ac.cy

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We thank the referee #1 for her/his positive and constructive review, which will clearly improve the quality of the paper. We will expand the introduction with more information about the physical needs to inject the emissions at altitude.

Comments to the main concerns:

- We indeed agree that a comparison with ozone sonde observations would give additional information on the two simulations. It must be stressed, however, that we compare the two simulations with large published datasets which are (or could be) considered as climatologies (see below). In this sense, we believe that the usage of single ozone sondes will not give very robust results. In addition, we are

reluctant in downloading data and generating our own climatology, because the climatology creation is error prone if the data are not carefully analysed. Neverthless we performed a comparison for two specific sites, whose location was carefully selected: Uccle station (Belgium) and Wallops island station (USA). Both stations are at locations with large differences between simulation *S1* and *F1*, and, in addition, data for the period 1995-2005 are available, The climatology obtained from these two sondes have been compared with the model results, confirming our main conclusions. Focusing the analysis at the surface ($\sim$ 1000 hPa), we compared the seasonal cycle of model results and the calculated climatology. Although the correlation between the model simulations and the climatology shows no significant differences (only $\sim$ 3%), the results from simulation *S1* agree better with the observations than results from simulation *F1*. In fact the average bias at Uccle island station between simulation *S1* and the observations is $\sim$ 50% lower (-3.6 nmol/mol) than the average bias between simulation *F1* and the observations (-7.14 nmol/mol). Coherently, the average bias at Wallop station between simulation *S1* and the observations is $\sim$ 20 % lower (-17.8 nmol/mol) than the average bias between simulation *F1* and the observations (-21.2 nmol/mol).

On the other hand, we think that a well establish climatology obtained from ozone sondes could give us additional and by far more robust results. The dataset presented by Logan (1999) is very well suited for such an analysis. This climatology, however, contains only a few levels close to the ground. A first analysis based on the lowest two levels (for all sondes at 1000 and 900 hPa) confirm the result presented in the paper. The correlation between simulation *S1* (with vertically distributed emissions) and the ozone sonde climatology is about 8-10% larger than that of simulation *F1* (with emissions concentrated at the surface). A further dataset based on ozone sondes which should be mentioned, is the one based on the SHADOZ project (Thompson et al., 2003), which covers tropical regions. However, this dataset is not totally useful in our case, due to the peculiar locations of the ozone sondes. The observations are mainly located in the Pacific

Ocean where the differences between simulation *S1* and *F1* are only a few percent. Also the few observations over the Atlantic ocean are either located in coastal or in open ocean regions. Since there the differences between simulation *S1* and *F1* are in the order of $\sim$ 0-5% only, they canot be regarder as being significant. In this dataset only onle location (namely Irene, South Africa), shows a somewhat strong difference at the surface between the two simulations. Based on this single station, we obtain a $\sim$ 10% better correlation between simulation *S1* and the observations ($R^2 = 0.22$) than between simulation *F1* and the observations ($R^2 = 0.20$), based on data below 900 hPa (lowest measurement level). The low correlation, however, does not allow us to draw any conclusion. Nevertheless, the average difference at the lowest level of the measurements between simulation *S1* and Irene ozone sonde (-1.19 nmol/mol) is smaller than the one calculated with the same observational data but model results from simulation *F1* (-1.71 nmol/mol). We will add this analysis to the revised version of the paper.

- As mentioned by Emmons et al. (2000), the field campaigns used are "data composites" of different species and are a basis to create observation-based climatologies. The measurements have been averaged over the entire field campaign region and organized in vertically gridded data. The data are hence representative for a certain region and for a certain period (generally 1 or 2 months) of one specific year. We indeed agree that the specific field campaign data strictly speaking, is only representative for the spacific year, but, the averaging of the measurements partly solves the problem. Nevertheless, the interannual variability is indeed present and depends on the location in space/time and the tracer considered. As shown by Pozzer et al. (2007), as example, for a relatively long lived tracer as CO a clear interannual variability due to the different meteorology is present at the surface. Hence the simulated year 2000 has been compared with climatologies or datasets averaged over large time/space regions. To confirm that our results are independent on the analysed year, we calculated the

correlation between the model results (CO of simulation *S1*) of different years (1999-2004) with the data-composites from the field campaigns. As expected, the difference in the correlation is in the range of a few percent ($\pm$ 5%). This is also a reason why, when compared to the observations, differences lower than 5% in the correlation or bias have been considered insufficient to draw any firm conclusion. To resume, the interannual variability cannot be neglected per se. However, thanks to the usage of data composites obtained from long aircraft field campaigns, the results obtained are almost independent on the selected year.

1 Specific comments

1. A few examples are shown in the electronic supplement. For instance, it is generally well established that in the process of power generation (from solid, liquid and gaseous fuels) the emission temperature causes an updraft of the plume. This has also been confirmed by the measurements of Pregger and Friedrich (2009). We will add this example (and others) in Section 2.

   As mentioned in the paper, debates are ongoing about biomass burning emission elevations. In general, the altitude depends on fire activities (flaming/smouldering) and location (boreal/tropical or others). It is, however, accepted that the main emissions are injected within the Planetary Boundary Layer (PBL) and emissions outside play a smaller role. Based on this assumption we used a constant altitude of 140 meters for the emissions. This implies that $\sim$ 67 % of the biomass burning emissions are injiected within the PBL. With a lower emission height, almost all biomass burning emissions will be injected into the PBL, while any higher emission height would imply a too strong injection outside the PBL.

2. As $HNO3$ is mainly formed by the reaction of NO_2 with OH, it follows mainly the behavior of these two tracers. Over regions with strong NO_x emissions we see

an increase (up to 60%) of HNO_3 at the surface in simulation *F1* compared to simulation *S1*, while a decrease outside the lowest layer is observed. In remote regions, instead, a small decrease in simulation *F1* compared to simulation *S1* is present, which is also in accordance with the NO_x and OH changes between the two simulations.

3. We agree to change the sequence in the subsection and move the discussion of CO before the HO_x section. We further agree with the referee and will modify the text accordingly. We will clarify the sentences, stating that in remote regions, the lower level of OH in simulation *F1* produced an increase of the CO concentrations compared to simulation *S1*.

4. We will include a surface map of O_3 differences (as for CO) in the revised manuscript. At the surface over polluted regions, the ozone mixing ratios are lower in simulation *F1* than in simulation *S1*, despite the increase in NO_x concentrations. This is mainly due to the increase of NO_2 deposition, which does not convert to $O3$ in simulation *F1* as in simulation *S1*, where the injection at higher altitude gives enough time for the interconversion. Although this gives generally smaller differences (in the range of 5-10 %), in some very polluted areas (mainly China), this gives a difference of up to 30%. In remote regions the differences in $O3$ are smallest, where simulation *F1* predicts lower mixing ratios than simulation *S1* ($\simeq 2\%$), due to the lower abundance of NO_x. We will also add this explanation to the revised manuscript.

5. As mentioned before, the aircraft observations are composite data, obtained by averaging different flights from aircraft campaigns, to be representative for the region at a specific time of the year. The model results have therefore been averaged in the same region and time of the year of the field campaign. In addition, we transformed the model results to the same vertical grid as the observational dataset.

6. We indeed selected two different field campaigns to show also the different effect of vertically distributed emissions for cases with strong biomass burning or anthropogenic emission influence. The main point is to show that, despite the emissions altitude, in both cases the free troposphere is almost not influenced, while at the surface we find significant differences. This finding is also derived from the comparison to the quasi-climatological composite data-sets, i.e., if averaged over larger areas and time intervals.

7. We suppose that the referee refers to the special treatment of the CO dataset. We agree that the conclusions are the same as for the other station observations. However, we want to point out that the correlation between station observations and simulation *S1* and the correlation between station observations and simulation *F1* differ by only $\simeq$ 10%, which is less than what is observed for other tracers (where a difference of $\simeq$ 20 - 25 % is present). An explanation is that the masking of (observed) polluted air masses is decreasing artificially the possible effect of the vertically distributed emissions, since the background air is generally less affected by the injection altitude of the emissions.

We will modify the text according to the corrections suggested by referee #1 and we will increase the size of the titles in Figures 5 to 9. We will also change the colorbars in Figure 1.

We are, however, reluctant to change the colorbars in Figures 2 to 4. In fact the large and different ranges of the data (from -30% to 30%) render it difficult to have one colorbar which is meaningful for all figures. Nevertheless, we will improve the quality with a clearer colorbar and (where possible) by applying the same scale.

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