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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.

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We thank referee #2 for her/his comments.

We appreciate the precision and the quality of the comments, and we believe that most of the main concerns are due to a partly unclear description of the model set-up, for which we apologize.

We did not describe any aircraft emissions in our model description section. However, we have certainly included this kind of emissions (see Jöckel et al., 2006) in the model simulation, and we did not change any emission heights for this emission type between simulation *S1* and simulation *F1*. The aircraft emissions are based on Schmitt and Brunner (1997). Hence, no aircraft emissions were used from the EDGAR database.

This explains why we used a constant factor of zero for the EDGAR F57 emission class (air transport), as mentioned in the electronic supplement. The EDGAR emissions have been masked this way in order to avoid double counting. This information will be added to the manuscript.

Regarding the other sources which the referee correctly mentioned in the review, we again apologize for the incomplete information caused by an automatic conversion of the table. In fact, emission classes L50 and L70 are denoted as "Agriculture" in the EMEP suggestion and we used the suggested elevation for these sources. Similarly, emission classes F58 (International shipping) have been treated as "Non-industrial combustion plants", F60 (Chem. Feedstock) and B30 (Charcoal production) as "Solvents and other product use", i.e., also following the EMEP suggestion. Because there is no suggestion from EMEP about biomass burning height injection, emission classes L42 to L45 have been chosen to be concentrated at 140 m (see below). We will therefore change the "personal judgment" with " specially treated", for source F57 and L42-L45 and provide the additional information in the revised manuscript.

We indeed agree that a highly detailed emissions plume model would be necessary for a realistic biomass burning (BB) injection (Freitas et al., 2006). However, due to the needs to specify a single value for different kinds of BB emissions, we used a single level (140 m) which allows us to have 67 % of the total BB emission injected within the Planetary Boundary Layer (PBL). As replied to referee #1, this altitude was based on the assumption that most of the injection should be in the PBL. This is in close agreement with the literature (Langmann et al., 2009). In addition, we performed further test simulations with injections at different altitudes and the best results were obtained with emissions concentrated at the chosen altitude.

In our study we showed that the global budget of the tracers and their mixing ratios in remote regions are hardly affected by the choice of the injection heights. Nevertheless, it is clear that injection heights are important on the small scale, for both, biomass burning (Luderer et al., 2006) or anthropogenic (Pregger and Friedrich, 2009) emis-

sions. We are hence very hesitating to perform additional simulations with separate emission heights for biomass burning and anthropogenic emissions,respectively, because we believe that this would not give additional information. Separate simulations with different injection heights for biomass and anthropogenic emissions will show a reduced signal in remote regions, a similar signal in those regions which are dominated by either biomass or anthropogenic emissions, and a reduced signal in regions influenced by both source categories. In this way, our study delivers an upper limit of the combined signal.

Regarding the nudging, we again apologize for the incorrect value presented in Table 1. These values were referring to the 8 years of simulation *S1* and therefore show the interannual variability (Pozzer et al., 2007, see also electronic supplement). The calculation for the year 2000 for soil NO_x emission gives 6.772 Tg(N)/yr and 6.769 Tg(N)/yr in simulation *S1* and simulation *F1*, respectively, i.e. a difference of 0.04 % In addition, the spatio-temporal distribution coincides, with a maximum difference of 0.3 %. Further, the lightning NO_x emissions for the year 2000 integrate to a total of 2.149 Tg(N)/yr and 2.145 Tg(N)/yr in simulation *S1* and simulation *F1*, respectively, which is a relative difference of 0.2 %.

We certainly agree that the meteorology between the two simulations cannot be binary identical. Nevertheless, because of the applied nudging, the meteorological parameters are sufficiently similar that they are not influencing the results. To support this statement, we performed additional tests by directly comparing the temperature and the specific humidity (two prognostic variables) between the two simulations. First, the temperature (a nudged quantity) has been compared by applying the same averaging procedure used for the comparison to the aircraft campaigns. The differences in temperature between simulation *S1* and simulation *F1* are less than 0.01%. In conclusion, the meteorological vertical profile is generally reproduced in both simulations. Further analyses using only monthly averages at the surface show slightly larger differences (a maximum of 0.4 $^\circ$ C, or 0.2% all over the globe). Second, we compared the simulated

specific humidity (a quantity which was not nudged). Whereas for a snapshot in time, the relative difference during one simulation year can reach up to 10-15%, the monthly averages differ by 5% at maximum, but with general differences of ≤ 1%. Finally, for the specific humidity averaged over the time and area of the aircraft campaigns, the differences in the tropospheric column are even smaller, namely $\simeq 0.5\%$.

We will correct the wrong footnote in Table 1, and add also the discussion on the nudging and the meteorological similarity between the two simulations.

We totally agree with referee #2 regarding the necessity to use climatological data, as also replied to referee #1. Not all used measurements are from the year 2000, but as explained in the reply to referee #1, the dataset is an aggregate of flight measurements. The results are almost independent of the year chosen for the comparison. As example, the correlation for CO and O_3 from different years from simulation *S1* with the aircraft observations exhibits differences within few percent ($\simeq$ 5% and $\simeq$ 4%, respectively) only. We agree that the interannual variability cannot be neglected per se, and that climatologies are necessary for any fair comparison. For this reason also the station observations are based on long time series of measurements.

It is generally very difficult to find in the literature datasets that describe vertical profiles of different tracers as climatology. This is the main reason why we used extensively the database compiled by Emmons et al. (2000). As correctly mentioned by the referee, however, a further analysis can be performed with the MOZAIC data, which are observed from aircraft (hence with vertical profiles), and are also multiannual and hence can be used to calculate real climatologies. However, to our knowledge, the only published climatology from this dataset present in the literature is the one from Zbinden et al. (2006). This *seasonal* climatology is certainly well suited for our comparison, although the vertical profiles are limited to a few places (Japan, New York, Paris and Frankfurt). In addition this climatology has been calculated only for ozone, CO and NO_x data are still in the calculation process (Zbinden, pers. com.). A correlation calculation with respect to the time would not give robust results, because the database

represents a seasonal climatology. However, we calculated the average bias between the observations and the model simulations for the altitude between surface and 3000 meters. These results do not conclusively confirm our findings: results from simulation *F1* have lower differences to the observations than simulation *S1* in Paris and Frankfurt, while simulation *S1* shows lower differences to the observations than simulation *F1* in Japan region and New York. The data will be further analysed and the results included in the revised version of the paper.

1 Specific comments

1. The EMAC model is a hydrostatic model with a hybrid pressure coordinate system. This implies that the levels are not constant with respect to the geometric altitude and that the number of levels within the first 800 m depends on the location and time. On average, the applied vertical resolution has 5 to 6 levels between ground and 800 m altitude.

2. The figures refer to the emission of CO above the PBL and the total one, as simulated in simulation *S1* (note, for example, the strong emissions from China, India and Europe, which are not present in the biomass burning emission source, only). We will correct the text to clarify the figure. The PBL calculated in the model is based on the work of Holtslag et al. (1990). The calculation is made interactively following the approach of Troen and Mahrt (1986), using the Richardson number, the horizontal velocity components, the buoyancy parameters and the virtual temperature. See Holtslag and Boville (1993, chapter 3) for a detailed and complete description of this method. We will add this information to the revised manuscript.

3. We agree with the referee that "Oxidation capacity" does not suit well as a title for the subsection. With "Oxidation capacity" we meant the OH mixing ratio, which

is clearly not correct. We will change the title accordingly and we will follow the suggestion of the referee to move the order of the paragraphs, so that the text will be better understandable.

4. As written in the reply to referee #1, only a vertical interpolation has been applied to the simulated data when compared to aircraft observations. For the comparison, the average over time and space of the corresponding aircraft campaign was used. The vertical interpolation was required to transform the hybrid pressure model levels to altitude levels as the observational dataset. For the comparison to the CO station observations, all simulated monthly data at the station location were used. Although this decreases the correlation between observations and simulated concentrations (see Pozzer et al., 2007) due to the inclusion of the effect of polluted air, both simulations show the same effect, since the meteorology is very similar.

5. We agree with the referee that the sentence is not clear enough. As noticed, only the anthropogenic emissions were distributed on different levels, while biogenic, online soil and biomass burning emissions were constantly injected at a single altitude (surface or 140 m). We will clarify the sentence.

References

L. K. Emmons, D. A. Hauglustaine, J.-F. Müller, M. A. Carroll, G. P. Brasseur, D. Brunner, J. Staehelin, V. Thouret, and A. Marenco. Data composites of airborne observations of tropospheric ozone and its precursors. *J. Geophys. Res.*, 105:20497–20538, 2000.

S. R. Freitas, K.M. Longo, and M.O. Andreae. Impact of including the plume rise of vegetation fires in numerical simulations of associated atmospheric pollutants. *Geophys. Res. Lett.*, 33: L17808,doi:10.1029/2006GL026608., 2006.

A. A. M. Holtslag and B. A. Boville. Local versus nonlocal boundary-layer diffusion in a global climate model. *J. Climate*, 6:1825–1841, 1993.

A. A. M. Holtslag, E. I. F. de Bruijn, and H.-L. Pan. A high resolution air mass transformation model for short-range weather forecasting. *Mon. Wea. Rev*, 118:1561Â£1575, 1990.

P. Jöckel, H. Tost, A. Pozzer, C. Brühl, J. Bucholz, Ganzeveld L., P. Hoor, A. Kerkweg, M.G. Lawrence, R. Sander, B. Steil, G. Stiller, M. Tanarhte, D. Taraborrelli, J. van Aardenne, and J. Lelieveld. Evaluation of the atmospheric chemistry gcm echam5/messy: Consistent simulation of ozone in the stratosphere and troposphere. *Atmos. Chem. Phys.*, 6:5067–5104, 2006.

B. Langmann, B. Duncan, C. Textor, J. Trentmann, and G. van der Werf. Vegetation fire emissions and their impact on air pollution and climate. *Atmos. Environ.*, 43:107–116, 2009.

G. Luderer, J. Trentmann, T. Winterrath, C. Textor, M. Herzog, H.-F. Graf, and M.O. Andreae. Modeling of biomass smoke injection into lower stratosphere by a large forest fire: Sensitivity studies. *Atmos. Chem. Phys.*, 6:5261–5277, 2006.

A. Pozzer, P. Jöckel, H. Tost, R. Sander, L. Ganzeveld, A. Kerkweg, and J. Lelieveld. Simulating organic species with the global atmospheric chemistry general circulation model echam5/messy1: a comparison of model results with observations. *Atmos. Chem. Phys.*, 7:2527–2550, 2007.

T. Pregger and R. Friedrich. Effective pollutant emission heights for atmospheric transport modelling based on real-world information. *Environmental Pollution*, 157, 2:552–560, doi:10.1016/j.envpol.2008.09.027, 2009.

A. Schmitt and B. Brunner. Emissions from aviation and their development over time. In *Pollutants from air traffic - results of atmospheric research 1992-1997. Final Report on the BMBF Verbundprogramm "Schadstoffe in der Luftfahrt", Tech. report, DLR - Mitteilung 97-04*, pages 1–301, 1997.

I. Troen and L. Mahrt. A simple model of the atmospheric boundary layer; sensitivity to surface evaporation. *Boundary-Layer Meteorology*, 37:129–148, 1986.

R. M. Zbinden, J.-P. Cammas, V. Thouret, P. Nédélec, F. Karcher, and P. Simon. Mid-latitude tropospheric ozone columns from the mozaic program: climatology and interannual variability. *Atmospheric Chemistry and Physics*, 6(4):1053–1073, 2006. ISSN 1680-7316. URL http://www.atmos-chem-phys.net/6/1053/2006/.

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