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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemistry-transport modeling of the satellite observed distribution of tropical tropospheric ozone" *by* W. Peters et al.

W. Peters et al.

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First Referee comment (Rolf von Kuhlmann)

We thank the referee for his suggestions and comments, and with the general positive tone in which the comment is written. The comments placed are all very relevant, and will all be taken into account in the revised manuscript. We agree with the remark of our misplaced used of the term 'family concept', and will correct this as suggested. The label 'well captured' for the low NOx chemistry will be replaced by the suggested 'better represented', which is better justified here. The way we reported the outcome of our sensitivity experiments will be reviewed in the next version, to better reflect differences and to include more information on the simulations that did not qualify as improvements.

Our suggestions concerning the poor modeling of the diurnal PBL growth were based

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on Rn^{222} simulations published in Dentener et al [1999], we have included this reference and revised our text on this point. Concerning the questions and remarks with respect to our definition of the measure of error, we want to stress that by using the strongly aggregated values, our measure of error better reflects the larger (planetary) scale that we have defined as the topic of this study. If all individual points in the observations were included in the definition of ϵ_2, local over- or underestimates would strongly dominate the value of ϵ_2 obscuring the information that we want to obtain, and local improvements might be misinterpreted as being very important. As we state in the discussion, it is the fact that these improvements show up in a strongly aggregated measure of error that lends most credibility to our analysis.

In order to further assess the significance of the small changes in ϵ_1 and ϵ_2 that we report, we have performed a sensitivity test for ϵ_1 and ϵ_2. By creating 30 3-dimensional ozone distributions that are randomly perturbed by an average of $\pm$ 10 %, and calculating the respons in ϵ_1 and ϵ_2, we have determined that these significant perturbations only inflict a maximum $\pm$ 0.005 change in ϵ_1, and a $\pm$ 0.015 change in ϵ_2. We can thus say that the small differences we report in Section 6 and Table 2 represent a systematic difference in the ozone fields from different simulations. In this respect, we agree that ideally, the values of ϵ_2 would also improve when the correct meteorology is used. However, due to the lack of inter-annual variability in the emissions and their trend over the 15-years simulated, a measure of the absolute magnitude (ϵ_2) will not adequately reflect the changes one expects to see from year-to-year. Thus, the referee is right to conclude that other model deficiencies still have a large impact, as we did in Section 6.

Second Referee comment (Larry Horowitz)

We thank the referee for his elaborate comments, clearly he has read the paper thoroughly and has given our work a lot of thought. We will include many of the smaller comments, suggestions and requests for additional information in the revised manuscript. Also, we have included a list that answers the referee's short questions. First, we will address the more important remarks and questions put forward by the

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referee.

Labeling of ozone through the origin of its precursor NO_x is a technique that was incorporated into TM3 in the work of Lelieveld and Dentener [2000]. Besides NO_x, several NO_y species are labeled to account for recycling of NOx through their longer-lived reservoir species. The assumption that ozone production is NOx controlled holds over much of the troposphere, the exception is regions close to strong NO_x sources where NO_x abundances are extremely high and ozone production can be VOC limited. These regions occupy only a very small fraction of the tropospheric boundary layer, and it can be expected that both NO_x and VOC molecules, which together determine the production of ozone, are from the same source in this case. The overall effect on the ozone chemistry are small, since the sum of the labeled ozone tracers explains more than 90 % of the unlabeled ozone concentration [Lelieveld and Dentener 2000]. We want to stress however that the results from the labeled ozone simulations we present are interpreted in a qualitative way, and that the details in the labeling technique do not affect the conclusions in this work.

The Modified-Residual method that was applied to produce the 1979-1992 satellite record of TTOC is described extensively in Hudson and Thompson [1998], and is one of several methods to retrieve tropospheric ozone columns. The results of the different methods are not always consistent, although most large-scale features and also the inter-annual variability agree well. Of the several methods available, the MR method generally produces the highest values of TTOC, specifically over the Northern Hemispheric part of the Atlantic. This could be an artefact introduced by a number of uncertainties in the retrieval method (interference of sand and dust aerosols, limited applicability of regional ozone soundings, differences in amplitude of the zonal wave one pattern). In order to take such uncertainties into account in our quantitative approach, they would have to be reflected in a larger error bar on these locations, thus introducing a weighing in the calculation of ϵ_2 that would almost exclude such points from our comparison. However, such uncertainties can only be better quantified through studies

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such as the one presented here, which are very limited to this date. We have therefore taken the observations 'as is', to set a simple and quantitatively defined benchmark for our comparison. That is also one of the reasons why we do not tune our model to these observations, and limit ourselves to prioritizing directions for further work. Finally, we want to mention that the MR method is likely to be succeeded by a new method that overcomes many of the problems reported previously. We will state our reasons for not explicitly questioning the value of the MR method more clearly in the text of the revised manuscript.

The referee mentions the role of lightning NO_x emissions and the fact that the interannual variability in these emissions is not separated from interannual variability in the meteorology. Indeed, these emissions are calculated interactively, depending on the position and intensity of deep convection. However, in Peters et al [2001] we showed that the largest component of interannual variability is associated with shifting convection patterns during ENSO events, and that the resulting increases and decreases in TTOC were not accompanied by increases in NO_x concentrations. Rather, convective redistribution of ozone was responsible for the 2-3 DU differences in modeled TTOC. We agree that this should be mentioned more clearly in the paper, and we will include this in the discussion of Section 6.

We also agree that the strong influence of lightning NO_x emissions should have a more prominent place in the paper. The uncertainty in these emissions, and their poor representation in global models such as ours is an obvious, and ill-quantified source of errors. However, their influence on extra-tropical ozone concentrations are quite strong, and a doubling of lightning NO_x would lead to serious discrepancies with observations in these regions. Suggesting that other parameters, such as STE, might be wrong in these locations and therefore mask a possible improvement in the lightning NO_x emissions is of course valid, but we cannot test these suggestions in our model. Naturally, performing a global, completely observation constrained optimization of the TTOC field including the effects of STE is beyond the limits of our capabilities. Thus,

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we can only constrain the extra-tropical domain of our model based on previous comparisons. Therefore, our conclusions cannot go beyond stating that the lightning NO_x emission magnitude is a sensitive parameter, but cannot be altered freely in the model. Several times, the referee asks for more detailed explanations on the sensitivity simulations and comes with very relevant questions and suggestions on possible causes and solutions for observed discrepancies. We want to stress however, that the nature of possible improvements should be investigated in detailed studies, in a 3D perspective, and using more appropriate tools for model evaluation. For instance, whether deficiencies in the treatment of biomass burning are due to insufficient model resolution or due to shortcomings of our photochemistry can be answered only by comparison of CO, NO_x, O_3, and VOC vertical profiles, from several locations and times. A dataset such as from TRACE-A is highly useful in this respect. Answering this question through this work is not only impossible, but also risks coming to premature and possibly even false conclusions. The same argument holds for for instance the land/sea distribution of lightning NO_x emissions; our model indicated that the sensitivity of this parameter is not very large, and that the changes we introduced are not resulting in a better agreement with observations. From this, we cannot conclude that our current treatment of the land/sea distribution is correct, and also not that this parameter does not require further attention. However, it is not on our list of priorities, and it is not a parameter that will bring our model and these observations in better agreement. Since this last remark is also stated as a question in the referee report, we will put this more clearly in our conclusions, and also try to be more concise in formulating our goals in the introduction of the revised manuscript.

Answers to short questions and requests for additional info:

- The 1979-1993 ECMWF reanalysis was done on a spectral resolution of T106.
- EDGAR contains all biomass burning sources and not just anthropogenic.
- The scaling of the alternate fire calendar was done on grid box scale.

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- The tropopause definition for STE calculations in the model is based on the temperature gradient (WMO definition).
- The sensitivity of the calculations to tropopause height is minor ($<$2DU) and discussed in Peters et al [2001]. The definition of tropopause height for calculation of ozone columns is similar to Craig [1965], which is virtually the same as WMO.
- Emissions of NO_x from soils are distributed 50/50 among tropics/extratropics.
- The 2 DU increase reported for a doubling of biomass burning emissions is an average for July-August-September and not an annual average.
- Additional biomass burning emissions mentioned in Section 9 refer to the double biomass-burning scenario.
- The comparisons in Fig.7 show model output at the time and date of the actual sonde launches.
- Comparisons with seasonal cycles from multi-year ozone-sonde launches and the model were presented in Peters et al. [2001].
- The vertical resolution used in the model is inadequate to represent the fine-scale features in the sondes. The 'large-scale features that are dominated by transport' refer to model resolved scales, which we expect to reproduce. Indeed, the fine scales are also influenced by (vertical) transport.
- The term 'forcing' in the discussion is used to describe the effect of changing emissions on the photochemistry of ozone.

Interactive comment on Atmos. Chem. Phys. Discuss., 1, 337, 2001.