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Interactive comment on "Modelling study of the impact of deep convection on the UTLS air composition – Part II: Ozone budget in the TTL" *by* E. D. Rivière et al.

Anonymous Referee #2

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General Comments:

This paper, which is the second part of a series of two papers aiming to study the local impact of deep convection on the chemical composition of the Upper Troposphere and Lower Stratosphere (UTLS), focus on simulating the vertical ozone distribution and the Tropical Transition Layer (TTL) budget during a severe convective event over Barau, Brazil in February 2004. Examining and quantifying the ozone budget of the TTL region of the atmosphere is an important and scientifically interesting topic. The study is clear and well presented and the model tool utilized is appropriate. However, for the present version of the manuscript, I feel there is a strong need for a clarification of the original contribution of this study. How are the conclusions drawn in the present study new and/or different from what is found in the studies by e.g. Ridley et al. (2004), Tulet et al., (2002), Wang and Prinn (2000), Williams et al. (2002) and Kley et al. (1996)?

Other general comments:

1. The authors calculate the TTL budget for only one convective event. This makes it very hard to draw any general conclusions. In order to make a better estimate, I think it is essential to perform simulations for several cases, or at least to perform some type of sensitivity simulations, to estimate the robustness of the results.

2. There is a large focus on the production of ozone in this study, but it is never discussed how ozone is destroyed in the upper troposphere and how this influences the budget. In the study by Ridley et al. (2004) it is indicated that convection appears to have two different roles over the continent and over the ocean, where ozone and its precursors are added to the upper troposphere over continental regions during convection and depleted over maritime regions. It would be interesting if the authors could comment on this matter and also more generally discuss the destruction mechanisms for ozone in the UT.

3. The authors are aiming at studying the UT/LS region, but do not perform simulations that would be able to resolve any STE exchange. Also, the simulated convective event did not involve any overshooting into the stratosphere which would most likely be an important factor for this type of study.

Specific comments:

Title: "Modelling the study of the impact of deep convection on the UTLS air compositionĚ" I think the title is somewhat misleading, as the LS is never really evaluated in the study.

Abstract: It is stated that the different processes contribute to 75%+23.5%+11% of the ozone increase. The total number then becomes 109.4%. Please correct this.

Abstract: The authors give precise numbers on how much each process contributes to the ozone gain in the TTL during the convective event. I think it's important to clarify that these numbers are only for a single convective event and hence are only indications of the importance of different processes.

Introduction: The authors need to clarify what is original in this study. There is especially a need to emphasize differences compared to other model studies by e.g. Wang and Prinn (2000) and Tulet et al. (2002), e.g. the region over which study is performed, and how that may influence the results.

Page 9171, last paragraph: It is not clear what the authors mean when they say that the ozone behavior in the TTL "can be less regular and can lead to a local bluge or a sharp increase in the vertical profile", when there is an increase in the vertical profile in the TTL during the dry season as well. I guess the authors mean that the increase is sharper and that you can have a local maximum in this region?

Page 9174, last paragraph: It would be interesting to know the initial surface concentration of ozone. If the surface ozone concentration is high, ozone can actually be transported from the surface to the upper troposphere and significantly contribute to the ozone budget in this region.

Page 9174, 1st paragraph: The authors state that the variability between 8 and 13 km is relatively low (typically about 12%), however in Figure 2 the variability in this layer is actually higher for the later part of the simulation than the variability in the TTL.

Page 9177, 1st paragraph of the evaluation of the model results: No observations are available for comparison for February 2004. Why do the authors then choose to simulate this period? Would it not be better to simulate the period of February 2001 for which there are soundings available? Or to simulate both periods?

Page 9179, 2nd paragraph: The authors claim that the observed vertical variability structure is qualitatively reproduced by the model. However, in the model, the variability is actually not lower between 8 and 12 km, than between 12 and 17 km and there is a maximum at 12 km which is seen in the observations but not clearly in the model simulation. In general, I think it is also not really clear (at the moment) what the authors really want to say with the comparison of the variability between models and observations. At the moment I can not really see what it adds to the discussion of Figure 1 and 3.

Page 9180, last paragraph: The authors say that there is also a non-negligible contribution of the other ozone precursors (CO and NMVOCs), but do not mention the contribution from any NOx not produced by lightning.

Page 9182, ozone budget: The budget is calculated over 24 h so that a full ozone production/destruction cycle is encompassed. But since the authors aim to study the effects of convection on the ozone budget, wouldn't it be better to study the time period of convective activity?

Page 9184, chemical production inside the TTL: Which reactions are really important for governing the ozone budget? I think it would be helpful if the most important reactions were listed and/or discussed. For Figure 8, I think it is also important to clarify that it is not only the ozone production terms that are included in the budget, but also the destruction terms. At least I assume this is the case as the "production" term is negative during the last part of the simulation period?

Page 9185, last paragraph: It is not clear to me why the ozone flux for the reference run is only 3% higher than for the no LNOx run. How efficient is the photolysis of NO2 to produce ozone inside the convective cloud? How fast is the NO produced from lightning converted to NO2? Maybe this could be clarified by looking more carefully into the individual terms of the ozone production/destruction budget?

Page 9187, first paragraph: The contribution from the vertical flux at 17 km is relatively small, when integrated over the whole 24 h period. But why integrating over 24 h?

Would the contribution be larger if only the actual period of convection is considered?

Page 9188, budget summary: The budget summary shows that the behavior of ozone in the TTL is mainly driven by dynamics, the chemistry being responsible for ˜23.5%. How sensitive is this number? What would happen with different initial conditions? If the budget was extended over a larger period? If the convection started earlier/later?

Technical corrections:

Page 9170, line 14 of Abstract: "...of the ozone increase if NOx production by lightning..." change to "...of the ozone increase, if NOx production by lightning...

Page 9170, line 15 of Abstract: "It is shown that downward motions at the..." change to "It is shown that downward motion at the..."

Page 9175, line 3: Remove "the" from "...altitude range given by the several definitions of the TTL".

Page 9175, last sentence line before section 2.3: Change to "... with values typically below 10% and tends to decrease with altitude".

Page 9180, 3rd paragraph: "...leading to large concentrations in the 7-17 km layer..." change to "...leading to relatively large concentrations in the 7-17 km layer...".

Page 9189, 1st paragraph: "... represents 75% of the total ozone increase on the 24 h ..." change to "... represents 75% of the total ozone increase over the 24 h..."

Page 9190, line 22: "...the chemical production is mainly due to the presence..." change to "...the chemical production of ozone is mainly due to the presence..."

Figures 4, 5, 6, 8 and 9 would all benefit from including grid-lines.

References

D. Kley, P. J. Crutzen, H. G. J. Smit, H. Vömel, S. J. Oltmans, H. Grassl, V. Ramanathan, Observations of Near-Zero Ozone Concentrations Over the Convective Pacific: Effects on Air Chemistry, Science, 274, 230-233 , 1996.

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Interactive comment on Atmos. Chem. Phys. Discuss., 5, 9169, 2005.