Atmos. Chem. Phys. Discuss., 5, S5053–S5058, 2005
www.atmos-chem-phys.org/acpd/5/S5053/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

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.

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Received and published: 13 January 2006

We would like to submit a revised version of the paper entitled "Modelling study of the impact of deep convection on the UTLS air composition: Part 2 ozone budget in the TTL" for publication in ACP. The present paper is the second part of a series of two articles dedicated to the study of the impact of a severe unorganised deep convective system on the UTLS (upper troposphere and lower stratosphere) chemical composition, with a particular attention paid on the Tropical Transitional Layer (TTL). Following the ACPD interactive comments period we were asked in a comment by Tom Karl about the first paper (Marécal et al.) to correct the isoprene emission routine. This was done and the simulations were rerun after this correction. This has a quantitative impact (but

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not qualitative) on the calculations made in the present paper for two reasons. Firstly more isoprene is now transported to the upper troposphere, and this modify locally the associated chemistry. Secondly our model optimisation compilation option was such that dynamical simulations with the model were not perfectly reproducible. The consequence is that the dynamical simulation we provide in the revised manuscript is slightly different from the one we had presented in the ACPD manuscript. Now we have changed this option. Both changes are not significant on the ozone average profiles shown in Figure 1, but this is more sensitive in the dynamical budget calculation. You will note that this does not change our conclusion on the ozone budget. A new Table 1is provided in this answer.

Now we answer point by point to the referee's comments.

Ozone budget : We agree with your remark. This is now stressed in the comments in section 5. It is recalled in the revised abstract that "The horizontal flux at a specific time is the main contribution in the budget, since it drives the sign and the magnitude of the total ozone flux. However, when averaged over the 24 hour period, the horizontal flux is smaller than the vertical fluxes, and leads to a net decrease of ozone molecule number of 23%." In section 5, we propose to reorganise the orders of subsections. Each dynamical flux (horizontal, vertical at 13 km and vertical at 17 km respectively) is discussed in subsection 5.1, 5.2, 5.3, respectively. Now we stress the correlation between the horizontal flux and the vertical fluxes during the period of convection, mainly due the conservation of mass. For mass conservation reasons below the tropopause where air masses cannot rise anymore, the vertical motion turn to horizontal motions, so that the horizontal motions increase in the TTL with an increasing convective activity.

Abstract : It is now appearing that the ozone budget is calculated over a 24 hour period. The important role of the horizontal flux is now stressed (as explained above). The correlation between the large negative horizontal flux and the convective activity, now discussed in the comment of Figure 8 (section 5) is also mentioned in the abstract. In section 5, the subsections have been reorganised, in order to stress the importance of the dynamics and the modification of the budget induced by convection. The comment on the ozone production within the TTL appears now is subsection 5.4.

Concerning §2 and the aqueous chemistry scheme, a more detailed description of the aqueous chemistry scheme appears in the revised version of manuscript 1 by V. Marecal et al.. We recall that the aqueous phase chemistry is available for 9 species in RAMS (O3, HNO3, SO2, H2O2, OH, HO2, HCHO, ROOH, and RO2). This routine act on the chemistry in two ways. Firstly it allows the dissolution of the most soluble species into cloud droplets and raindrops. When the aqueous phase chemistry is taken into account, this implies that the most soluble species are scavenged by rainfall. Secondly, there are chemical reactions in the liquid phase. The reactions are given in Grégoire et al. (1994). Prior to the present study, sensitivity tests have been performed with and without the liquid phase chemistry. The conclusion is that convection leads to a severe scavenging of HNO3, and to a lesser extend, H2O2 which is less soluble than HNO3. This implies very low values of HNO3 in the upper troposphere and a decrease of H2O2. This result is in agreement with the conclusion of Mari et al. (2000). The importance of the HNO3 scavenging on the upper troposphere composition has already been stressed in previous study. Thus, taking into account this process is necessary to perform a realistic simulation. In the new version of the manuscript, we now refers to Grégoire et al. (1994) and recall the principle of the aqueous chemistry routine

Concerning §2 the LNOx production switch off during the spin-up period: during the spin-up period the RAMS model simulates unrealistic very short time scale convective cells. In order not to produce LNOx associated with these unrealistic features, the LNOx production has been switched-off during the spin-up period of the model. Please note that in their study, DeCaria et al. (2005) have also switched off the LNOx production routine during the spin-up period for the same reason. In order to avoid any confusion, the sentence has been rephrased

Section 5.3 (now section 5.2): “the largest contribution in the ozone budget” has been changed into “the largest source of ozone”.

Role of lighning NOx: Referee#1 is wondering how can the LNOx be directly produced in the TTL if the bottom of the TTL is the defined as the level just above the maximum of the convective outflow. In our case, several convective cells are reaching the cold point tropopause (˜17 km) (see Marécal et al., part I) but the majority of the convective outflow is around 13 km, which shows that our definition of the bottom of the TTL is compatible with the one given in Highwood and Hoskins (1998). This is also consistent with Folkins et al. (1999) who defines the bottom of this layer as the altitude of the increase of the ozone profiles. In our case this increase starts at 13 km altitude. Another way to define the bottom of the TTL is using the lapse rate. Dessler and Sherwood (2000) define the bottom of the TTL as the zero net radiative level. From the model temperatures, it was found that the local minimum in lapse rate is around 13 km also. Therefore, we think that the choice we have made using 13 km as the bottom of the TTL is robust because it is compatible with several definitions of the TTL given in the literature. For all these reasons, we do not agree that the 14 km level is a good candidate for the bottom of the TTL. In our simulations, the number of very high convective cells is not as unfrequent as supposed by referee#1 especially during the very active convective period, so that the local production of ozone inside the TTL due to LNOx is not negligible. A detailed analysis shows that this local production mainly occurs in the altitude range [13 km;14 km]. Above 14 km, the LNOx deposition is close to zero so that no associated ozone can be produced. We think anyway that our definition of the bottom of the TTL is correct. Following referee #1’s recommendations, we have checked what the 24h ozone budget would be using 14 km and 12 km as the bottom of the TTL. Logically, the conclusions of each level are different than for the 13 km calculation. The 24 h ozone budget calculation using 12 km for the bottom of the TTL leads to a higher chemical production of ozone inside the TTL, a higher bottom flux contribution. When using the 14 km level as the bottom of the TTL, the 24 h ozone budget calculation shows a weak bottom flux and a weak local chemical ozone production as compared to the other contributions. This is logical since 14 km is above the region of maximum O3 production, as shown in Figure 5. The conclusion is that the budget calculation depends on the level of the bottom of the TTL used. Considering that the far best choice for the bottom of the TTL is 13 km (as explained above), we have retained and discussed the budget using 13 km as the TTL bottom altitude. In the revised manuscript, we now refers to . Dessler and Sherwood (2000) and the change in the lapse rate behaviour from 13 km. This justifies our choice for the bottom of the TTL.

Section 5 : In the first paragraph, we remind the altitude range (between 13 and 17 km altitude) we have used for the TTL and we refer to section 2.2 where the justification for this altitude range is given.

After a comment by referee #2 about the results shown in Figure 9 we have decided to remove Figure 9 from the manuscript. The new simulation shows differences that are generally not significant between the “reference” run bottom flux and the “No LNOx” run bottom flux except after 2200 UT when the “reference” run bottom flux is significantly higher than the “No LNOx” run bottom flux.

Figures : the label for 0400 UT in Figures 1 and 2 has been corrected. Because the new simulations slightly change the results, new figures are provided (Figure 2, 5, 6, 8, 9).

According to referee #2’s recommendation, other changes have been made. The most important change are listed below:

We were asked from referee #2 to perform the same budget calculation on the convection period timescale because it corresponds to a different dynamical regime. In the first version on the manuscript, we had chosen a 24 hour period in order to encompass the full ozone production/destruction cycle. In the new version, we have added a budget calculation for the period of maximum convection, that is between February 8, at 1600 UT and February 9 at 0000 UT. We have shown from the 8 hour budget that convection modifies the dynamical contributions to the ozone budget compared to

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pre-convective conditions. This is now added in a new version of Table 1 (see below)

..................../total / Top(17 km) / Bottom(13 km) / Horiz. / Chemistry

24 h period /97.5 / 23.3 / 68.7 / -23.1 / 28.7

(1E30 O3 molec)

% of the total /100 / 23.9 / 70.4 / -23.7 / 29.4

O3increase

Convective Period /11.0 / 34.2 / 52.2 / -91.8 / 16.4

(8 h)

Table 1. Integrated number of molecules of ozone entering the domain drawn in Figure 7 during a 24 hour period starting from 2001/02/08 0000 UT, and during the 8 hour convective period starting from 2001/02/08 1800 UT (in 1030 molec) for the “reference” run. The horizontal, the top, bottom and the chemical contributions are reported. A positive value means a gain for the domain. Also shown are the percentage contributions to the ozone molecule increase for the 24 hour period.

References: taking into account comments by referees #1 and #2, the following reference are now added:

Barth, et al., J. Geophys. Res., 108(D7), 4214, doi:10.1029/2002JD002673, 2003. DeCaria, et al., J. Geophys. Res.,110, D14303,doi:10.1029/2004JD005556, 2005. Grégoire, et al., J. Atmos. Chem., Vol. 18 , No. 3 , p. 247-266, 1994. Ridley., et al., Atmospheric Environment, 38, 1259-1274, 2004. Sherwood and Dessler, Geophys. Res. Lett., 27, 2513-2516, 2000. Tulet, Atmospheric Environment, 36, 4491-4501, 2002.

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