

## ***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. Rivièrè et al.***

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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 1 is provided in this answer.

We now answer point by point to the referee.

General comments:

One of the main comments from referee #2 concerns the original contribution of this study. Several modelling studies have already been carried out on the topic of the impact of deep convection on the upper troposphere composition. Some of them are listed by referee #2 at the end of his/her comment. However, the topic is large and complex, and investigating it needs studying several subtopics. Some of them are the importance of LNO<sub>x</sub> in the upper troposphere chemistry, the role of scavenging of soluble species within clouds or redistribution of species by ice clouds. A second point is that at the global scale, deep convection can take several different form and the impact of each kind of convective system on the UTLS composition might differ. Would an organized squall line have the same impact as a multicell unorganised system ? Would a continental convective system have the same chemical impact as an oceanic system ? Would a single cloud analysis reach the same conclusion as a multicloud convective system ? Has the mid-latitude deep convection the same consequence on the UTLS composition as the tropical deep convection ? All those issues need to be addressed to fully understand and to quantify the impact of deep convection on the UTLS composition. Here, our aim is to document and to study the impact of an

extreme continental unorganised convective system on the UTLS (and particularly on the tropical transitional layer). This now appears more clearly in the revised manuscript (see changes below). The impact of such a type of system on the UTLS air composition has never been studied in the past and the comparison with the conclusion of other studies is not always relevant. Some studies focus on the cloud scale and this is not easy to extrapolate the conclusions of the papers to the regional scale of our study. Some of them document convective systems over the ocean with specific chemical source at the sea level (especially halogen species which might destroy ozone) while our study is over a continental area (at least for the fine grid of simulation) with NO<sub>x</sub>, NMVOCs, and CO, well known to be ozone precursors, that are emitted at the ground. Some of the references are now added in the text and comparisons are made when possible with the conclusions of our study. Finally, to our knowledge, an ozone budget in the TTL has never been done. Thus we think that this study provide original material with respect to previous studies. This is now clarified in the text. We now stress that the case is severe (several cells almost reach the tropopause) and unorganised (is composed of several convective cells that interact with each others) and take place in a continental area. We also recall that the impact on the upper troposphere chemistry of such a system has never been studied before. now recall that several cells almost reach the tropopause. Concerning the comparison with other studies, we refer to Ridley et al., (2004), since in our case we find that the NO<sub>2</sub> photolysis is the main chemical source of ozone. Ridley et al. (2004) reach the same conclusion from measurements of continental air masses. Concerning Tulet et al. (2002) we have added the two following comments: “A comparable mesoscale model with online chemistry was successfully used by (Tulet et al., 2002) to simulate the redistribution of ozone by a convective observed at mid-latitude.” (section 2) and “Finally, it was shown in the simulations of Tulet et al. (2002), using a model similar to ours and with a coarser vertical resolution (700 m), that mid-latitude convective systems can induce the intrusion of stratospheric ozone into the upper troposphere.” (section 5). At the beginning of the conclusion, we now stress again the originality of this work with respect to other studies.

Other general comments:

1. We do agree that it will be needed in the future to compare our results with other comparable convective cases to test the robustness of the results. The simulation of one particular convective case requires a large amount of work (setup and initialisation of the simulation, validation of the meteorological and/or chemical results) and computing time (a simulation with chemistry is five times more costfull than a meteorological simulation). This is why only one case study is presented here. But the 2004 HIBIS-CUS/Troccinox/Troccibras field campaign that also took place in Bauru will be the basis for documenting other cases.

2. In the new version of the manuscript, we now refer to the paper of Ridley et al. (2004). Comparisons with this study are not often straightforward since most of the observations given in this paper are of marine origin. The only possible comparison is related to the air masses of continental origin sampled by the high altitude aircraft. The conclusion from these observations is that the main chemical source of O<sub>3</sub> is from NO<sub>2</sub> photolysis. In our study, we reach the same conclusion. However this production source include the potential role of NMVOC or CO in transforming NO into NO<sub>2</sub> which produces ozone after photolysis. A comment about this now appears in Section 4. of the manuscript. "A detailed analysis shows that the main contribution in the ozone chemical production is due to the photolysis of NO<sub>2</sub>, the other chemical sources such as the photolysis of NO<sub>3</sub>, or reactions involving HO<sub>2</sub> and RCOO<sub>2</sub> (peroxy acyl radicals) being of lesser importance. This analysis is compatible with the conclusions reached by Ridley et al. (2004) from aircraft measurements in the upper troposphere influenced by continental deep convection over central America."

3. As it is now explained in Marécal et al., and their answers to their referees, this convective case is severe with many cells reaching the cold point tropopause. This could have had an impact on the lower stratosphere composition. After analysis, it was shown that none of the cells are overshooting into the stratosphere, limiting the impact of the deep convective event on the LS composition on the simulation timescale.

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However, it is now well admitted that once in the TTL, chemical species can penetrate into the stratosphere by quasi-isentropic slow ascent. Even if the present study cannot assess directly the short time scale impact of deep convection on the LS composition, it can document the TTL composition. This can be the starting point of a forthcoming study about the slow quasi-isentropic transport to the stratosphere, using the TTL composition simulated here. In the revised manuscript we propose to recall that a significant number of convective cells are almost reaching the cold point tropopause: “The reasons for choosing this case study are the following. Firstly, the severity of the convection, almost reaching the tropopause, makes this case particularly interesting for studying the impact of deep convection on the TTL chemical composition.”

Specific comments: Title : we have chosen to keep the title since the severe convective event presented here could have had an impact on the LS composition. This issue is now documented in the companion paper by Marécal et al., part I. Their analysis shows that the impact of the convective system on the LS composition is negligible at the time scale of the convective event.

Abstract: the numbers given in the abstract did not mention the horizontal contribution (which was negative). The sum of all the contributions was 100%. Now in the new simulation, the numbers have changed, and all contributions appear so that the sum is 100 %. We now specify in the abstract that they are related to the 24h simulation including the convective system studied here. “The calculation of the ozone budget in the TTL during a 24 hour period in the area of the convective system shows that the ozone behaviour in this layer is mainly driven by dynamics. 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%. The upward motions at the bottom of the TTL, related to the convection activity is the main contributor to the budget over the 24 hour period since it can explain 70 % of the total ozone increase in the TTL, while the chemical

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ozone production inside the TTL is estimated to be 29 % of the ozone increase, if NO<sub>x</sub> production by lightning (LNO<sub>x</sub>) is taken into account. It is shown that downward motion at the tropopause induced by gravity waves generated by deep convection is non negligible in the TTL ozone budget, since it represents 24% of the ozone increase.”

Introduction: we are now more precise about the originality of the case study, that it is continental, and unorganised. The reference of Wang and Prinn (2000), Tulet et al. (2002), Ridley et al. (2004) are now added in the manuscript (as discussed before, see our answer to the general comments).

Page 9171: Yes we mean that the increase is sharper and can lead to a local maximum in the region. We also mean that the vertical profile structure is perturbed with respect to the dry season. This has been clarified in the text.

Page 9174: Now we give the mean ozone concentration at the ground level in Grid 2 (about 40 ppbv).

Page 9174: About the variability results in the 8-13 km layer, it is written later in section 2.3 that the variability increases with time in this layer. The value of 12 % given here is a typical value for this layer.

Page 9177: As previously explained, this case was chosen because: 1. it is an extreme case of deep convection and 2. the impact of this kind of unorganised deep convective system composed of many different cells on the upper troposphere composition was not studied before. In order to clarify this, the introduction and the conclusion have been changed as follows: Introduction. “The reasons for choosing this case study are the following. Firstly, the severity of the convection, almost reaching the tropopause, makes this case particularly interesting for studying the impact of deep convection on the TTL chemical composition. Secondly, this convective case is composed of a cluster convective cells that possibly interact with each others. Several modelling studies aiming at quantifying the impact of deep convection on the upper troposphere composition have already been published (Yin et al. 2001; Wang and Prinn, 2000; Barth et al.

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2001; Tulet et al. 2002, DeCaria et al., 2005). Most of them studied either an individual convective cell or a well organised system study (tropical or not). The present paper focuses on a more complex type of system: a non-organised and extreme tropical convective system.” Conclusion. “Most of the modelling studies on this aspect investigated the cloud scale approach (e.g Yin et al., 2001; DeCaria et al., 2005) or the large scale approach (Labrador et al., 2004). Here we focus on the study of the ozone behaviour in the upper troposphere, and particularly in the tropopause transitional layer (TTL), for an unpublished type of convective system: an extreme continental non-organised convective cluster.”

Page 9179: We think that qualitatively there are common features between the variability in our simulations and the variability derived from the DMI sondes, keeping in mind that the bottom of the TTL in the DMI sondes (starting altitude for the ozone increase) is lower on average than in our case study. We do agree that the values are not exactly the same but the following tendency can be underlined in both the simulation and the observations : a low variability below the TTL, a higher in the TTL, and a lower variability in the stratosphere. We propose the following changes in the manuscript: “However, one can note that the maximum of variability of the DMI O3 measurements occurs at a lower altitude than the maximum of variability in our simulation. This can be explained considering that for a few profiles (e.g. DMI 24/02/2004), the ozone increase with altitude starts significantly below 13 km while in our case study the bottom of the TTL is at 13 km altitude. Therefore, this comparison shows that the simulation results are realistic in the UTLS and capture rather well the observed ozone distribution in the Bauru region during the convective season.”

Page 9180: We had forgotten to add the contribution by NO<sub>x</sub> not produced by lightning. This is now added.

Page 9182: The choice to compute the budget over a 24 hour period was driven by the ozone production/destruction cycle. Since we wanted all the contribution to be comparable, we had chosen this period for the whole budget. However, the suggestion

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to compute the same budget over the most active period of convection is interesting. We have performed this calculation and corresponding results are provided in Table 1 (at the end of this comment) and discussed in subsection 5.1, 5.2, 5.3 and 5.4. Section 5.1: “Integrated on the 8 hours severe of the convective period, there is a net exiting horizontal flux of  $91.8 \cdot 10^{30}$  O<sub>3</sub> molecules while it is 4 times less then for the 24 h calculation. Despite the large negative horizontal flux on the 8 hour period, the total flux remains positive during this period but 9 times smaller ( $11 \cdot 10^{30}$  O<sub>3</sub> molecules) than for the 24 hour period ( $97.5 \cdot 10^{30}$  O<sub>3</sub> molecules). The positive value is reached because, during the 8 hour period, the vertical fluxes are large and positive and the chemistry contribution is significant, even if twice smaller than for the 24 hour period. It important to note that O<sub>3</sub> molecules entering or exiting horizontally the flux calculation domain shown in Figure 7 do not represent a loss or a gain for the TTL since most of the O<sub>3</sub> molecules exiting the domain horizontally will remain in the TTL. The real gain or loss of O<sub>3</sub> molecules for the TTL will be generated by the vertical fluxes or the O<sub>3</sub> chemical production/destruction.” Section 5.2. “For the 8 hours of the intense convective period, the vertical flux at the bottom of the TTL corresponds to  $52.2 \cdot 10^{30}$  O<sub>3</sub> molecules entering the TTL. This is 76 % of the same flux calculated for the 24 h period. This means that the main process responsible for the large vertical flux value at 13 km in the 24 h calculation is deep convection.” Section 5.3. “Integrated over the 8 hours of intense convection, the top flux contribution is  $34.2 \cdot 10^{30}$  O<sub>3</sub> molecules while it is  $23.3 \cdot 10^{30}$  O<sub>3</sub> molecules for the 24 h period. As for the vertical flux at the bottom of the TTL, this latter result shows that the 24 h tendency for the top vertical flux is mostly explained by processes related to the convective activity. In particular,É” Section 5.4 “. Integrated over the 8 h period of severe convection, the number of ozone molecules produced in the domain of calculation is roughly half (57 %) the number of molecules produced during the 24 hour period. This is due to the fact that the 8 h period encompasses the ozone destruction at sunset but also the period between 1800 UT and 2000 UT when the ozone production is maximum. This number of  $16.4 \cdot 10^{30}$  O<sub>3</sub> molecules produced in the domain during the 8 hour period is relatively small compared



to the dynamical fluxes during the same period. However, keeping in mind that the sum of the dynamical fluxes is slightly negative, this ozone production ensures the ozone budget to be positive during the severe convective period.” Section 5.5 “Calculating the same budget for the 8 most active hours of convection shows the importance of the vertical fluxes during this period since the total number of ozone molecule entering the TTL vertically during the 8 hour period is comparable to the same number calculated for the 24 hour period. The results have shown that there are two different chemical and dynamical regimes that have an impact the ozone budget: before and during the convective period.”

Page 9184. According to Referee#2’s suggestion, we have investigated what was the main ozone production source. It is mainly due to the NO<sub>2</sub> photolysis, the source from RCOO<sub>2</sub> compounds and the NO<sub>3</sub> photolysis being smaller. However, this source term from NO<sub>2</sub> includes the production by NO<sub>2</sub>, but also by CO and NMVOC since the last two species are involved in cycles that transform NO into NO<sub>2</sub> to later produce O<sub>3</sub>. It is however difficult to discriminate the relative importance of CO, NMVOCs and NO<sub>x</sub> in this source term. The difference between the reference run and the no LNO<sub>x</sub> run shows that the production term from NO<sub>2</sub> is the major one. A comment has been added on this point in section 4 (ozone production). We also now specify that the production term into the TTL is the net contribution of the raw production term and the raw destruction term (section 4 and 5). We also give the order of magnitude of the row destruction term with respect to the row production term. In the first kilometre of the TTL, during the sunlight convective period, the raw ozone chemical destruction term depending on the place with respect to the convective cells, is 40% to 90 % the raw ozone production term. This is added in the text in section 5.4.

Page 9185: concerning Figure 9. After a comment by referee #2 about the results shown in Figure 9 we have decided to remove this figure from the manuscript. The new simulation shows small differences between the “reference” run bottom flux and the “No LNO<sub>x</sub>” run bottom flux as in the previous simulations. The differences are only

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significant after 2200 UT when the “reference” run bottom flux is largely higher than the “No LNOx” run bottom flux. It is now shown that after 2200 UT, the bottom flux for the reference run is 11% higher than the bottom flux for the “no LNOx” run. The comment on this appears in section 5.2.

Page 9187: The contribution of the 17 km flux over the convective period is now given in Table 1(see below) and discussed in section 5.3.

Page 9188: About the contribution of the chemical ozone production in the budget: this is difficult to evaluate the sensitivity of this number since a large number of full simulations (which are computationally expensive) would be needed to estimate this. However the difference between this simulation and the previous one (submitted manuscript) can give an indication on this evaluation. It shows that the difference in the isoprene emissions has changed the results of a few percentage point. This means that the main contribution is the dynamics but that the contribution of the ozone chemical production is significant even if smaller, as stated in the budget summary.

All technical corrections have been included.

Table 1

...../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 1600 UT (in 1E30 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.

The following references have been added and discussed in the manuscript. Barth, et al. (2003); DeCaria, et al. (2005); Grégoire, et al., (1994) ; Ridley. et al. (2004); Sherwood, and Dessler, (2000).

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