

## ***Interactive comment on “Modelling study of the impact of deep convection on the UTLS air composition – Part I: Analysis of ozone precursors” by V. Marécal et al.***

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### ANSWER TO REFEREE #2' COMMENTS

INTRODUCTION 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 I analysis of ozone precursors” for publication in ACP. In this version, we have modified the text according to the remark made by JL Attié during the ACPD on-line discussion process, to the remarks posted by the other referee and to your remarks. The detailed answers to your comments are given in the following pages with the corresponding changes in the manuscript (in blue) when necessary. Also, during the ACPD on-line discussion

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process, there was a comment on the low values of isoprene from our simulations. After investigation, we found a mistake in the isoprene emission module leading, after correction, to an increase of the mean isoprene content. Since isoprene is a reactive compound, this increase slightly impacts on some of the other ozone precursor contents and the results are only slightly changed (except for isoprene), leading to no modification of the paper conclusions.

The modification of the isoprene emission routine has mainly modified the isoprene content. See below the new figures 10 and 11.

## ANSWERS TO THE COMMENTS

### General comments

1. One of your major remarks is that the paper did not contain enough new material to be published separately. Thanks to your comments and those from referee #3, additional material and scientific discussion were added to the paper for submission to ACP. In particular, we added comparisons with the recent study by DeCaria et al. (JGR 2005) and a section (section 6) on the analysis on the effect of lightning NO<sub>x</sub> on the HO<sub>x</sub> distribution and the HO<sub>x</sub> precursors (H<sub>2</sub>O, H<sub>2</sub>O<sub>2</sub>, ROOH = organic hydroperoxides and formaldehyde). From our point of view, this new material improves the paper significantly. The HO<sub>x</sub> analysis is now discussed in a new section (section 6) that is given below. Note that the formaldehyde which is a HO<sub>x</sub> precursor is now discussed in section 6 instead of in section 5 in the ACPD manuscript and the ACPD Figure 12 is now Figure 13c. Unfortunately, the figures cannot be included in the present answer.

6. RESULTS FOR HO<sub>x</sub> AND ITS PRECURSORS The distribution of HO<sub>x</sub> (OH+HO<sub>2</sub>) in the atmosphere is of major importance in the ozone budget since the ozone precursors are oxidized through reactions with HO<sub>x</sub> to form ozone. Figure 12 represents the mean profiles for HO<sub>x</sub> at 1800 UT on February 8, 2001 for the reference and the “No LNO<sub>x</sub>” runs. The 2200 UT profile is not shown since this is the sun set time corresponding to a rapid decrease of HO<sub>x</sub> mixing ratios. The HO<sub>x</sub> mixing ratio for the “No LNO<sub>x</sub>”

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run is nearly constant between 10 and 13.5 km altitude. This is related to the vertical transport by convection of the HOx precursors as illustrated in Figure 13 showing a bulge mainly for organic hydroperoxides (noted ROOH) and formaldehyde (HCHO) in the UT. This result is consistent with the model results obtained by DeCaria et al. (2005) within the anvil of a mid-latitude convective system. Another important HOx precursor (not shown here) is the water vapour. Its mean values are increased by 12 % at maximum in the UT during the convective period favouring the HOx production. As illustrated in Figures 12 and 13, there is a significant impact of the lightning NOx on HOx and its precursors. The HOx mean profiles for the two runs are similar except in the 10-16 km altitude range where there is an important decrease of the reference run compared to the “No LNOx” run. This result is consistent with the mean HOx profile calculated by DeCaria et al. (2005). In their case, this decrease was associated to a decrease of both HO2 and OH while in the present study the model simulates on average a decrease of HO2 but an increase of OH. The mechanism responsible for the HO2 decrease is similar in both studies: HO2 reactions with NO and NO2. For OH, its production/loss depends on the relative quantity of NOx and VOCs (Volatile Organic Compound). In both simulations, VOC mixing ratios are high in the UT because of the convective uplift of the surface emissions and consecutive outflow. For the reference run, NOx mixing ratio is very high in the UT mainly where lightning is triggered. This leads to two types of mechanisms (Chapter 16 in Finlayson-Pitts and Pitts, 2000): 1. in very localized places where lightning NOx are produced, the ratio of VOC versus NOx is small enough to lead to OH depletion forming HNO3. 2. in other places in the vicinity of convective updrafts, a detailed analysis shows that the ratio of VOC versus NOx is large enough to lead to OH production. On average, this is mechanism 2 that dominates in our simulation leading to a mean increase of OH while in DeCaria et al. (2005) this is mechanism 1. This difference can be explained by different VOC emissions since the geographical regions considered are very different in the two studies. As in the present study, Wang and Prinn (2000) found an increase of OH during the daytime when NOx are produced by lightning from 2D simulations of a cloud resolving model

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including chemistry. Using a global modelling approach, Labrador et al. (2004) and Jourdain (2003) obtained similar results on average. As shown in Figure 13a and 13b, lightning NO<sub>x</sub> tends to deplete organic hydroperoxydes and H<sub>2</sub>O<sub>2</sub>. This result is in agreement with DeCaria et al. (2005). The mean formaldehyde mixing ratio is enhanced in the 9-15 km layer by the increase of NO<sub>x</sub> by lightning. This is related to the fact that formaldehyde is formed and depleted at the same time by a complex chain of reactions. In fine, the loss term is of lesser importance, particularly at night time. In the LS, there is no impact of convection on HO<sub>x</sub> and its precursors since the simulated convection cells do not cross the isentropic barrier at the tropopause. This result is similar to that found for the ozone precursors.

Figure 12: Mean HO<sub>x</sub> mixing ratio over Grid 2 domain as a function of altitude for the reference run (solid line) and for the “ No LNO<sub>x</sub> ” run (dashed line) on 08/02/2001 at 1800 UT.

Figure13: Same as Figure 12 but for (a) H<sub>2</sub>O<sub>2</sub>, (b) organic hydroperoxides (ROOH) and (c) formaldehyde.

2. We agree that the weakness of the paper is that no observed vertical profiles are available to evaluate the simulated ozone precursors. There were no observations gathered because the selected convective system was extremely intense (very strong wind gusts and very intense precipitation) making a balloon launch impossible in the vicinity of the system. Despite this lack of data, we feel that this is interesting to study this convective event because of its extreme characteristics. In particular, this cluster of convective cells was composed of many individual cells, several of them reaching very high altitude (around the cold point tropopause). Some of the convective cells are close in space and time leading to a possible dynamical interaction between them. The system we have chosen is original in this respect since in the literature, the cases studied are either individual convective cells or organized convective events (convective line + anvil). Since there are no chemical data available for our case study we have used as much as possible the meteorological data to check the validity of the meteorological

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simulation. The comparison with the meteorological data is fairly good considering the complexity of the convective cluster (convective cells that initially develop individually and then merge into a cluster). In the revised manuscript we emphasized the originality of the chosen case study compared to previous studies.

3. We agree that the transport of CO, NO<sub>x</sub> and NMVOCs from the surface to the UTLS has been shown in previous mesoscale modelling studies. In the revised version, following your suggestion, we have stressed more on the results concerning the impact of convection on the lower stratosphere.

4. In the simulation, the cloud top of several of the convective cells reach the model level located at 16.9km altitude. This altitude is close to the cold point tropopause which is located around 17km altitude. This means that, even if the model does not simulate “overshooting”, it provides convective cells nearly reaching the tropopause and therefore could have an impact on the lower stratosphere. The model results show that, even if the tropopause is nearly reached by the convective cells, there are no changes either on average or locally on the ozone precursor contents above the tropopause during the simulation. The isentropic barrier is not penetrated by convection even in the case of the deepest modelled cells. This means that, during the convective event, there is no significant transport from the upper troposphere to the lower stratosphere by the very deep convective cells. This is explained in the revised version.

The modifications for the last two comments (3 and 4) are listed below:

Abstract: During the simulation time, the impact of convection on the air composition of the lower stratosphere is negligible for all ozone precursors although several of the simulated convective cells nearly reach the tropopause. There is no significant transport from the upper troposphere to the lower stratosphere, the isentropic barrier not being crossed by convection.

Modification in subsection 5.1: Note also that the space variability of CO is much smaller in the stratosphere than in the troposphere as illustrated by the grey area in

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Figure 5a. This indicates that the CO in the LS does not originate from the troposphere. The influence of the dynamics of the deep convective cells on the stratospheric CO is negligible since even the highest cells are not able to cross the isentropic barrier at the tropopause.

Modification of subsection 5.2: As for CO, this indicates that there is no dynamical impact of the convective cluster on the stratospheric NO<sub>x</sub> even if the convective cells sometimes nearly reach 17km altitude.

Modification of subsection 5.3: In the LS (above 17 km), there are no significant changes of all the NMVOC contents even if the cloud top of several of the simulated convective cells nearly reach 17km altitude. As for CO and NO<sub>x</sub>, there is no penetration of any of the simulated convective cells through the isentropic barrier that could lead to troposphere-to-stratosphere transport.

Addition in section 6: In the LS, there is no impact of convection on HO<sub>x</sub> and its precursors since the simulated convection cells do not cross the isentropic barrier at the tropopause. This result is similar to that found for the ozone precursors.

Modification of the conclusion: Another important result is that there is no modification of the mean ozone precursor contents in the LS for this extreme convective event. Even at the location of the highest simulated convective cells of the cluster, there is no upward transport through the tropopause isentropic barrier.

#### Specific comments

Page 9128: We agree that in the ACPD paper there were very few comments on the LS composition. The reason is that there are no significant changes in the LS composition during the simulation. This is an original result since several of the simulated convective cells nearly reach 17km altitude (the cold point tropopause) and could have had an impact on the LS composition by mixing through wave breaking for instance. Nevertheless, the increase of ozone precursors in the UT by transport and lightning

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NO<sub>x</sub> production does not modify the LS composition on the considered time scale but are possibly of importance at longer time scales. This is stated in the revised version (in the abstract, in the result sections and in the conclusion). The modifications are listed above for point 3 and for (general comments)

Page 9129: The sentence “The tropical UTLS has not<sup>Ē</sup>” has been modified to be clearer.

Modification of the introduction: Although tropical regions are of major importance for stratospheric ozone, there have been only few field campaigns in the tropics documenting the tropical Upper Troposphere and Lower Stratosphere (UTLS). There is still a need for measurements of both tropospheric and stratospheric species in the whole altitude range of the tropical UTLS in the vicinity of convective events to improve our understanding of the impact of deep convection on the UTLS air composition.

As you suggested we have completed the references concerning the modelling studies. As for the references on the observational studies, we have chosen to only reference those including observations over Brazil because our case study took place in Brazil and because tropical convection characteristics depend on the considered region (continental or maritime, organized or non-organized).

Page 9131: In the revised version we have shown more explicitly the originality of the type of system studied (see remarks in the general comments above point 2).

Page 9135: A sensitivity simulation to the pH value was performed with a slightly less acid pH. It showed that there is a no significant impact on the ozone precursors presented in the paper except for the reactive compounds (ethene and isoprene) that are slightly increased by a few tens of pptv during daytime above 13 km. This is related to changes in HNO<sub>3</sub>. We added a sentence on this sensitivity test in section 3.

Page 9135: The global simulation used for the initialization was started 15 days before the beginning of our simulation. We change the text to make this point more clear.

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Page 9136: Following your interesting suggestion, we have made a statistical evaluation of the model water vapour field. Unfortunately, no specific humidity or relative humidity observations were available from ECMWF. Only relative humidity observations were available from INMET. We have used these observations to make a statistical evaluation (Table 3). The comparison with the model is generally good showing the consistency of the modelled water vapour field as illustrated by the table below. This table and corresponding comments have been added in the revised version.

Number of observations Mean from observations Mean from model STD from observations STD from model Index of agreement

08/02/2001 at 0000 UT 19 78.5 76.8 10.5 8.3 0.66

08/02/2001 at 1200 UT 16 76.1 80.8 5.2 2.6 0.56

08/02/2001 at 1800 UT 16 70.8 69.6 15.4 12.3 0.66

09/02/2001 at 0000 UT 15 87.8 83.3 8.0 7.9 0.75

Table 3: Statistical results for the 2-metre relative humidity. STD stands for the standard deviation. Mean and STD values are in %.

Additional comments in subsection 4.1: For relative humidity, only the INMET data were available. The model means and standard deviations at different times show a generally good agreement with observations. The model tends to slightly overestimate the temperature on average at night (up to 1.7K difference) and slightly underestimates it during daytime (up to 1.3K difference). The model provides less variability for the relative humidity than in the observations. For relative humidity, the index of agreement is slightly weaker than for temperature and wind. Considering that the humidity varies very rapidly when and where convection occurs, the model performs fairly well in simulating near-surface relative humidity for the studied convective cluster.

Page 9137: When evaluating the model rainrates versus the radar rainrates, one has to take into account the high complexity of the convective event studied. This is an

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extended cluster composed of intense convective cells that possibly interact. This type of system is not easy to simulate. This is why the location of the cells is not exactly reproduced by the model. We agree that the main convective band that flooded Bauru is extended too far west in the model. We changed the comment in the revised version saying now that the model agree fairly well (and not just well) with the observations and discussing the fact that the individual cells are not exactly located in the model as in the observations. Concerning the radar rainrates, they are likely underestimated because of the relationship used to convert radar reflectivity to rainfall rate. This is why we have chosen not to make a quantitative comparison but a qualitative comparison of the radar versus model rainrates.

Page 9138: Although there is no “overshooting” in the model results, several modelled convective cells nearly reach the tropopause. These cells could possibly induce changes in the lower stratosphere because of their high vertical extent. The model simulations show that there are no significant exchanges at the tropopause level because the isentropic barrier is not crossed by the convective cells or associated waves. This is explained in the revised version (see modification for point 3 and 4 general comments).

Page 9138: The issue of the ozone production/destruction budget is addressed in part 2 of this series and in the answer to referee#2 of part 2 for page 9184. The model results show that the main ozone production source is due to the photolysis of NO<sub>2</sub>. LNO<sub>x</sub> is an important term as illustrated by the large decrease of ozone production when the LNO<sub>x</sub> are not taken into account in the model (see part 2). Nevertheless, it is difficult to discriminate the role of CO and NMVOCs versus NO<sub>x</sub> in the ozone production since CO and NMVOCs are involved in cycles that transform NO into NO<sub>2</sub>.

#### Technical corrections

P 9129 Proposed change: “TTL” to “tropical TTL”. We have not made the change since the word tropical is already in the TTL acronym that means tropical transitional layer or tropical tropopause layer. Page 9132: we have done the modification.

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