**Atmospheric
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Discussions**

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 #3' 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

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the manuscript (in blue) when necessary. Also, during the ACPD on-line discussion 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.

ANSWERS TO THE COMMENTS

We agree that the weakness of the paper is that no observed vertical profiles of chemical species are available to evaluate the model. 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. We have used as much as possible the meteorological data to check the validity of the meteorological simulation. As for the chemistry, the comparison of our results with previous studies shows a good consistency. Nevertheless, we agree that the conclusions of our study need to be confirmed by future campaigns. This is stated in the conclusion of the revised paper.

Modification of the conclusion: To assess the quality of the simulation results, we have used the available chemical observations and data from the literature. Unfortunately, they are not sufficient to validate the model outputs. Therefore, the conclusions of the present paper need to be confirmed by field campaigns. This will be possible using the data from the coordinated HIBISCUS/TroCCiNOx/TroCCiBras field campaign that took place in Brazil in February and March 2004.

Following your suggestion, we added the Barth et al. (2001) and DeCaria et al. (2005) references in the introduction.

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 extended cluster composed of intense convective cells that possibly interact. This type of non-organized system is not easy to simulate. This is why the location of the cells is not exactly reproduced by the model. In particular, the main convective band that flooded Bauru is extended too far west in the model. Note that the maxima of precipitation below 23.4° latitude are out of the radar range which is displayed in Figure 4b as a solid line circle. 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. As for the radar rainrates, they are likely underestimated because of the relationship used to convert radar reflectivity to rainfall rate.

Modification of subsection 4.2: The model agrees fairly well with the observations although the location of the cells is not exactly reproduced by the model. In particular, the convective band that flooded Bauru extends too far west in the model. There is little rain below 23.4° latitude in both the radar observations and the model. 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 extended cluster composed of many intense convective cells. For this type of non-organized system, it is not possible to simulate exactly the development of each individual cell.

Following your interesting suggestion, we added in the revised version a discussion on the analysis of HOx (see corresponding manuscript modifications in the new section 6 that is given below). We compared the model results to those found by DeCaria et al. (2005) for HOx. On average over the grid 2, there is a decrease of HO2 and an increase of OH when lightning NOx are taken into account. OH mixing ratio being weaker that HO2, HOx mixing ratio is decreased by lightning NOx. This is consistent with DeCaria et al. (2005) except that they found a decrease of OH. The increase or decrease of OH depends on the relative quantity of NOx and VOCs. In our simulation, VOC mixing ratios are high in the UT because of the convective uplift and consecutive

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outflow of the surface emissions. For the reference run, NOx mixing ratio is very high in the UT where lightning is trigger. This leads to two types of mechanisms: 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), the ratio of VOC versus NOx is such that OH is produced. 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 COV emissions since the area considered are very different in the two studies: Colorado for DeCaria et al. (2005) and Brazil for us. The new section 6 is reproduced below after the comment on the LNox parameterization.

Following your suggestion, we added more details on the lightning NOx parameterization.

Additional text in section 3: Basically, the computation of the LNOx production is performed at each horizontal grid point. In this parameterisation, LNOx can be computed for two different cloud layers. The first layer made of mixed water and ice cloud lies up to the -15°C level. The second layer just above contains only ice phase cloud. For each horizontal grid point, the maximum vertical velocity in the corresponding air column is used to calculate the flash rate within the column. A proportion number of flash within each layer is then computed, depending on the thickness of the layer. Finally for each cloud layer, a specific parameterization is used to compute the NOx production at each level (Price et al., 1997), depending on the layer flash rates and the thickness of two consecutive model layers.

Following your interesting suggestion, we added in the revised version a new section (section 6) on the analysis of HOx and its precursors (H2O2, HCHO, H2O and organic hydroperoxides). In this section, a comparison is done with the results presented in DeCaria et al. (2005). Our results are consistent with those found by DeCaria et al. (2005). The HOx analysis is now discussed in a new section (section 6) that is given below. Note that the formaldehyde which is a HOx precursor is now discussed

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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 reproduced in the present document.

6. RESULTS FOR HOx AND ITS PRECURSORS The distribution of HOx (OH+HO2) in the atmosphere is of major importance in the ozone budget since the ozone precursors are oxidized through reactions with HOx to form ozone. Figure 12 represents the mean profiles for HOx at 1800 UT on February 8, 2001 for the reference and the "No LNOx" runs. The 2200 UT profile is not shown since this is the sun set time corresponding to a rapid decrease of HOx mixing ratios. The HOx mixing ratio for the "No LNOx" 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): 3. in

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very localized places where lightning NOx are produced, the ratio of VOC versus NOx is small enough to lead to OH depletion forming HNO3. 4. 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 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 NOx tends to deplete organic hydroperoxydes and H2O2. 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 NOx 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 HOx 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 HOx mixing ratio over Grid 2 domain as a function of altitude for the reference run (solid line) and for the " No LNOx " run (dashed line) on 08/02/2001 at 1800 UT.

Figure13 : Same as Figure 12 but for (a) H2O2, (b) organic hydroperoxides (ROOH) and (c) formaldehyde.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 9127, 2005.

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