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Comment

***Interactive comment on “A new chemistry-climate tropospheric and stratospheric model MOCAGE-Climat: evaluation of the present-day climatology and sensitivity to surface processes” by H. Teyssède et al.***

H. Teyssède et al.

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Thank you for your detailed review of our article. We have reworded the text of our draft article following your recommendations. Apart from these changes in the text, please find below our responses. **Please note that, in these responses, we often refer to the revised article where we have modified sentences or paragraphs according to your comments. Furthermore, due to the large number of remarks associated with such a long article, we did not repeat in our responses our comments. The numbering of the sections, tables and figures refers to the revised manuscript.** Note also that we have three new co-authors: P. Nédélec and V. Thouret

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## 1. *Title and CCM/CTM*

We fully agree that we present in this article results from a model run as an off-line chemistry and transport model. The confusion of using the term “chemistry-climate model” to qualify our model arose from the fact that, in our mind, models of the CCMVal activity (Eyring et al., JGR, 2006), that include radiative and dynamical feedback, are so-called coupled chemistry-climate models, with the term “coupled” being mandatory. We drove our CTM with several years of meteorological analyses and derived a climatology of chemistry fields; hence our denomination of simulations from a “chemistry-climate model”. We will change this denomination to CTM throughout the text to avoid confusion.

The aim of the present work was to validate the chemistry part of the CCM that we will use in the future. This CCM will be composed of the ARPEGE-Climat GCM and of MOCAGE-Climat for the chemistry part. The GCM has already been quite well documented while on the other hand MOCAGE-Climat requires a comprehensive validation. We performed this validation here, driving MOCAGE-Climat as a CTM with the most realistic meteorological forcing available, hence the meteorological analyses of the ECMWF NWP. We propose a new title for our article: ‘A new tropospheric and stratospheric Chemistry and Transport model MOCAGE-Climat for multi-year studies: evaluation of the present-day climatology and sensitivity to surface processes’.

We have revised a few sentences in the introduction (section 1).

## 2. *Figures*

As you say, we could not show the results from each and every simulation performed (a total of six simulations). We made a selection and chose to present results based on the general principle to show model results whose horizontal resolution was the closest to the resolution of the corresponding observation. In particular, observed climatologies of the stratospheric  $CH_4$ ,  $N_2O$ ,  $NO_x$ ,  $HCl$ ,  $O_3$ ,  $HNO_3$ , and  $ClO$  are zonal fields on 4, 5 or even 10 degrees latitudes. The  $OH$  climatology is on a 8 degree lat by 10 degree lon grid. We therefore present T21 results against these climatologies. On the other hand, observations from MOZAIC,  $CO$  and  $NO_2$  columns have a much higher horizontal resolution, so we show T42 model outputs. This principle does not apply to the  $O_3$  column field nor to the profiles in the troposphere ( $HNO_3$  or  $O_3$ ). For these fields we plotted model outputs that were the closest to the observations. We indicate in the text the deficiency of the T42 circulation that results in  $O_3$  columns further away from the observations. For the sake of completeness and to provide a general overview, we present a paragraph (entitled Summary statistics) that describes synthetically how the various simulations perform. As for the sensitivity to the deposition velocity, we performed an additional T21 simulation only due to limited computer time resources.

We have added the following sentence in the article (paragraph 3.1): “A summary of the six simulations appears in Table 5. Model outputs retained to appear in the various figures correspond to results whose resolution is the closest to the observations (e.g., T21 for stratospheric zonal comparisons, T42 for tropospheric  $CO$ ), unless explained otherwise in the text. Statistics compiled from all simulations appear in paragraph 3.8”.

The figure labelling has been enlarged in all figures and special attention has been paid to legibility.

### 3. Introduction : List of references for MOCAGE

We agree that we have created some confusion. We propose to rephrase the

sentence into: “This set-up enables the model to be used for a wide range of scientific applications from the study of global-scale distributions of species (Josse et al., 2004) to “chemical weather” forecasting, down to the regional scale (Dufour et al., 2004; Drobinski et al., 2007), and chemical data assimilation (Cathala et al., 2003; Geer et al., 2006; Pradier et al., 2006; Clark et al., 2007).

#### 4. Model Description : water vapour

In the revised version of the manuscript, we have included more details on the description of the water vapour field of our model :

“At the crossroads between dynamics, physics and chemistry, we use the mixing ratios of the ECMWF analyses up to the 380 K isentropic level for the representation of water vapour. Above this level,  $H_2O$  is calculated by the chemical scheme of MOCAGE-Climat (see below) and advected by its semi-lagrangian transport scheme. Prescribing the water vapour field between the surface and the 380 K level allows MOCAGE-Climat to benefit from the ECMWF analyses and from their modelling of the physical processes in the troposphere and the UTLS region. At middle and high latitudes the 380 K surface may be partly in the stratosphere, depending on the meteorological situation. The numerical diffusion of  $H_2O$  into the stratosphere is thus reduced and the stratospheric profile is still satisfactory since the ECMWF analyses include a simple parameterization of water vapour production by methane oxidation (Oikonomou and O’Neill, JGR, 2006).”

#### 5. Reference of emissions

The correct year for reference is 1985 and not 1895.

#### 6. Description of the observations

The description of the observation data sets we used has been moved in an appendix. We deleted Section 3 of the draft article “Observations used for the evaluation” and in the section “Model results and evaluation” of the revised article

the first subsections are now the following: “Description of the simulations performed”, “Age of air (AOA)”, and “Observations used for the evaluation”, with only a few lines and a reference to Table 6.

## 7. Reference and Age of air

- The reference Brewer, QJRMS, 1949 has been added in the text.
- *Details on the simulations of AOA*

We have deleted this paragraph.

## 8. Numerical stability of simulations

We have followed your recommendations, moving Fig. 2 into supplementary material and including a shorter description of the stability of the simulations in the section entitled “Description of the simulations performed (3.1)”.

## 9. Methane (CH<sub>4</sub>) and water vapour (H<sub>2</sub>O)

We indeed looked at the water vapour of the model and compared it with the climatology of Grooss and Russell, ACP, 2005. We arrived at the conclusions, presented in the paragraph “Methane and water vapour”, that we thought were self explanatory. Because both reviewers insist that we show these comparisons they are now included in the revised article. We also have reworded this paragraph to include more discussion, as requested by both reviewers (paragraph 3.4.1) :

““ ....

Comparisons between outputs of the MOCAGE-Climat T21 simulation and the zonal climatology of Grooss and Russell, ACP, 2005 are shown in Figure 2, between 100 and 0.1 *hPa*, for the months of March and September. Even though

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the T21 AOA is too young (see paragraph 3.2), the overall model  $CH_4$  distribution resembles the observed one:  $CH_4$  decreases with height and latitude. However, the diabatic descent in the southern polar vortex in September does not seem as marked in the simulations (although observations exist up to 65S only), and the distinctive ‘rabbit-ears’ shape in the observations in March is not clearly simulated. Nevertheless, the seasonal shift of the maximum towards the winter hemisphere appears similarly in the model and in the observations. For  $H_2O$ , the shape of the zonal distribution is qualitatively well simulated, with low equatorial mixing ratios above the tropical tropopause and generally a positive gradient towards higher altitudes and latitudes. Moreover, the dehydration is very similar in the simulations and in the observations. More quantitatively,  $CH_4$  mixing ratios from the model appear generally too low, 5 to 30% ( $\pm 0.05$  to  $-0.25$  ppmv), and in parallel  $H_2O$  mixing ratios are underestimated throughout the stratosphere, with relative differences between simulations and observations varying from -10 to -25% over large parts of the stratosphere ( $\pm 0.9$  to  $-1.3$  ppmv). These differences can have several causes including the chemical destruction of  $CH_4$  (consequently production of  $H_2O$ ), the underestimation of the mixing ratios at the entry level, or the deficiencies of the meridional transport. Further light on this is provided by the analysis of the T42 simulation and of an additional simulation with MOCAGE-Climat driven by the ARPEGE-Climat GCM.

As expected in agreement with increasing age of air (ECMWF T42 AOA < ECMWF T21 AOA < Arpege-Climat T42 AOA, see paragraph 3.2), there is much more  $CH_4$  in the entire stratosphere in the T42 simulation than in the T21 simulation, the T42 simulation overestimating observations. T21  $CH_4$  outputs are in turn larger than those of the ARPEGE-Climat simulations. For  $H_2O$ , mixing ratios are lower in T42 than in T21. They are similar in T21 and in ARPEGE-Climat, but the shape of the distribution from the ARPEGE-Climat simulation, being more centered around the equatorial latitudes, is more realistic (see the figures provided as supplementary material). The underestimation of the T21  $CH_4$  could

explain the negative deviation of  $H_2O$ , however this explanation does not hold for the T42 outputs. The entry levels of  $CH_4$  are overestimated in March and September. Therefore, there seems to be some deficiency in the oxidation chain of  $CH_4$  as one would expect to obtain  $CH_4$  mixing ratios that are too large in the T21 simulation since the circulation is too fast. As for  $H_2O$ , both that the circulation is too fast and that the entry-level mixing ratios are too low are plausible explanations of its deficient simulation. Indeed, if the circulation is too fast, there is insufficient time for moistening through methane oxidation. This problem is even greater in the ECMWF T42 simulation, hence the lower  $H_2O$  mixing ratios in this case. The rate of methane oxidation could also possibly be too slow. All this requires further investigation.”

#### 10. Carbon monoxide

The sentence in the text is now: “It thus affects two of the most important greenhouse gases.”

#### 11. N<sub>2</sub>O and BD circulation

- *Section order*

We had first included the section on  $N_2O$  immediately after that of  $CH_4$ . Then we changed our mind so that all paragraphs on nitrogen species follow each other. We think that both choices can be defended. We kept the presentation of our draft article.

- The mean age of air provides information on integrated transport within the stratosphere. We reworded our text so as to mention not only the too fast Brewer-Dobson circulation. Indeed it is not straightforward to link a difference in a tracer field to a particular transport process, and the situation is not as clear as implied in the first draft. We have further analysed our outputs of  $N_2O$  and the revised paragraph is under 3.4.3 as follows:.

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“... Nevertheless, Ricaud et al., ACPD, 2007 showed from ODIN  $N_2O$  observations that this compound can have spatial variations in the UTLS, especially in the tropics where troposphere to stratosphere exchange sometimes takes place in association with convective events.

Figure 6 presents the UARS climatology of Randel et al., JAS, 1998 and model outputs from the T21 simulation for March and September: the modeled  $N_2O$  field is consistent with the observations, maximizing in the lower stratosphere and decreasing as the altitude increases. In the lower stratosphere, MOCAGE-Climat simulates a smoother  $N_2O$  field as a function of latitude than UARS, with higher mixing ratios than the measurements. In the upper stratosphere (from 10 to 1 hPa), at equatorial and mid-latitudes, the model underestimates the observations, in March and September. This tends to indicate that the destruction of  $N_2O$  (photolysis + reaction with  $O(^1D)$ ) may be somehow too strong as we would expect the contrary on the basis of the too quick circulation alone. This hypothesis is confirmed by the outputs of both the ARPEGE-Climat driven simulation and of the T42 simulation (see the figures provided as supplementary material). Indeed, in the former case, with a relatively realistic AOA (see paragraph 3.2)  $N_2O$  mixing ratios are lower than observations by 20% or more throughout the stratosphere. In the T42 case, the model overestimates observations, again throughout the stratosphere by 20% or more which reveals that, though too strong, the destruction of  $N_2O$  is not too far off to counteract the fast T42 circulation. Further light is thrown on the deficiencies of the  $N_2O$  field by analysing the  $NO_y$  field (see paragraph 3.4.4). At high latitudes, differences between the model and the UARS observations have a seasonal cycle; this is also visible in the ODIN/SMR observations. For comparison with these observations, the simulated  $N_2O$  fields have been averaged in 10 degree latitude boxes. Figure 7 shows the evolutions between 2001 and 2005 of the zonal averages over three latitude bands with different dynamical

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characteristics, 80S-70S, 10S-EQ, and 50N-60N. In the tropical high stratosphere (10S-EQ), the underestimation of the model, already mentioned in the comparison with the UARS observations, appears in the time series for all years, though in a limited way for the year 2002. In the tropical high troposphere, that corresponds to the lowest altitudes ODIN can observe,  $N_2O$  is rather well mixed both in MOCAGE-Climat and in the satellite observations. However, as mentioned before, some variability appears around the 100 hPa level observed by ODIN with a minimum occurring during the spring of 2004; this minimum is not reproduced by the model. At high southern latitudes (80S-70S), the seasonal cycle of larger and smaller mixing ratios at a given altitude is not as marked in the model as in the observations. This is related, in winter to a too weak mesospheric subsidence in the ECMWF analyses, and in summer to the bias of the chemical destruction already mentioned. It results in alternating underestimations and overestimations of the observations. In the 50-60N latitude band, differences are generally not as important as for the other latitude bands, except for 2003, which reflects that the circulation is better simulated in the northern hemisphere.”

## 12. Total nitrogen oxides

- We have adopted now in the article: “The  $NO_y$  family consists of all nitrogen compounds excluding  $N_2O$ .”
- We plotted the fields following your recommendations. We now have a revised paragraph (see 3.4.4 Total nitrogen oxides).

“The  $NO_y$  family consists of all nitrogen compounds excluding  $N_2O$ .  $NO_y$  is produced from one branch of the reaction of  $N_2O$  with  $O(^1D)$ .  $O(^1D)$  itself comes from the photolysis of  $N_2O$  or  $O_3$ . We have first validated the simulated  $NO_y$  in the UTLS with the MOZAIC observations. The comparisons between MOCAGE-Climat T42 outputs and observations in DJF and

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JJA appear in Figure 4, while histograms of  $NO_y$  classes for several regions of the world are presented in Figure 5 (observations versus T21 and T42 simulations). The model always presents a positive bias, over all regions of the world. This overestimation is higher in the summer months when the chemistry that controls  $NO_y$  is the most effective. It appears clearly in the plotted distributions of Figure 5: peaks of the distributions of the model are 2 to 4 times higher than peaks of the observations, depending on the region. Furthermore, shapes of the distributions differ: observations have asymmetric distributions with large occurrences of very low mixing ratios ( $<0.4$  ppb), mainly observed in winter, while distributions of the model are quite symmetric and show no occurrence of these low mixing ratios. In the UTLS, various sources can contribute to augmenting the  $NO_y$  content, including lightning and aircraft emissions, transport from the troposphere and stratospheric intrusions. In our case, the first two sources are not relevant as we did not take them into account in the present simulations. On the contrary, transport from the troposphere can be incriminated for this positive bias in the UTLS: we will see later in the paragraph on nitrogen oxides (paragraph 3.5.1) that MOCAGE-Climat shows a general overestimation of the  $NO_2$  tropospheric content, in particular in winter. The impact on the UTLS is the positive deviation against the MOZAIC observations that we see here. As for the last hypothesis (intrusions from the stratosphere), we got a sense of the validity of the stratospheric  $NO_y$  of the model by comparing it to the sum of  $HNO_3$  and sunset  $NO + NO_2$  from the UARS observations, along the recommendations of Park et al., NASA, 1999. In March and September (not shown), months presented in our  $N_2O$  comparison, the model overestimates observations (by 10 to 20% between 50 and 2 hPa) over most latitudes and altitudes, for both months. This positive bias in the stratosphere could well play a role in the positive bias in the UTLS, and it is in agreement with the destruction of  $N_2O$  being too large (see paragraph 3.4.3)."

## 13. Spivakovsky data set

- We agree and have changed “Both model and observations reveal ...” into “Both model and the ‘reference’ data set (Spivakovski et al. 2000)...”

14.  $HNO_3$ 

The sentence has been replaced by “However, simulations are generally higher than observations in the layer between the surface and 3 km. ”

15.  $HCl$ 

You are right, hydrochloric acid is a recommended name (along with hydrogen chloride) by WMO for  $HCl$ . Therefore, we have now used this term throughout the text.

## 16. Total ozone column and ozone hole

- It is true that comparing CTM results, forced by real analyses, to CCM ones that recompute their own meteorology, is not the best way to proceed. Therefore, we have deleted the sentences : ‘It should be noted that a similar positive bias exists in most of the thirteen coupled chemistry-climate models (CCMs) evaluated in Eyring et al., JGR, 2006.’ and ‘Most of the CCMs assessed in Eyring et al., 2006 underestimate the area and the mass deficit of the ozone hole, and this is related to an overestimation of the total global ozone column’.
- We agree that this sentence is confusing. In Eyring et al., JGR, 2006, many of the CCMs simulate positively biased ozone columns, at all latitudes. Eyring et al., JGR, 2006 argues that, in this case, even if the chemical ozone loss is correct, the ozone hole will be underestimated. In MOCAGE-Climat, (1) we overestimate ozone columns, essentially in the northern hemisphere

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because of the too fast circulation, and (2) our Antarctic hole is not deep enough. We investigated further the second point, and concluded that it was mainly related to a too weak sedimentation of  $HNO_3$ , as detailed in the next response. We have deleted the reference to Eyring et al., JGR, 2006.

## 17. Stratospheric ozone

- The fact the ozone hole is not deep enough in MOCAGE-Climat is in apparent contradiction with the large  $ClO$  amounts that are found during the Antarctic winter (see our comparison of  $ClO$  to UARS observations in Figure 10). We provide as supplementary material a figure that helps understand why the ozone hole of MOCAGE-Climat is not deep enough. This figure shows the evolution of the mixing ratios of a number of species in the vortex at 50 hPa. The amount of  $ClO$  increases during winter to reach a maximum in September, when sunlight is back onto the vortex and initiates ozone depletion. However, starting in September, as  $NO_2$  produced by the photolysis of  $HNO_3$  increases,  $ClO$  mainly reacts with  $NO_2$  to produce its reservoir form,  $ClONO_2$ , rather than deplete more ozone. This is not conform to what should happen, as in the real atmosphere,  $HNO_3$  mixing ratios should drop to very small values throughout the winter because of the sedimentation of PSC ice particles produced by heterogeneous chemistry.  $HNO_3$  and therefore  $NO_2$  should be much lower in September. Sedimentation appears too weak in MOCAGE-Climat and should be checked.

The revised paragraph is under 3.7.1.

## 18. Summary statistics

- We guess you mention Figure 21 and more precisely, the Taylor diagram for model and MOZAIC comparisons. Unfortunately, the T21BL1 data interpolated in time and space for comparisons with MOZAIC observations got lost

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during technical operations. However, the effects of BL1 versus non-BL1 runs can be inferred from the simulations at T42.

## 19. Mixing ratios

- We have changed the text, using ‘maximum of relative differences’ throughout the paragraph. We also added the following sentence:  
“... and for each model level we looked the maximum of these relative differences. In the lines below, the maxima (respectively the mean) presented correspond to the maxima (respectively to the mean) of these maximum values”.

## 20. Conclusions

- *Page 50, last line. This sentence requires a reference.*  
We have added the Shindell et al., JGR, 2006 reference.
- *Page 51, paragraph 2. It would be helpful to add the Stevenson reference on the second line, ... recent model inter-comparison exercises (Stevenson et al. 2006), though ...*  
We have added this reference.

## 21. References

- Thank you for detailing the bibliography! We have updated it as some papers have been published between the first draft and the revised version of our paper.  
The simulation shown in figure 7 is T21BL1. It has been added to the figure caption.