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Atmospheric
Chemistry
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Discussions

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. Teyssèdre et al.

H. Teyssèdre et al.

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We thank you for your numerous remarks and suggestions. 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 your 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 (CNRS/Laboratoire d’Aérologie), and A. Volz-Thomas (FZ/Juelich).

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General comments

We have reworked all the figures and hope that they now better support the objectives of our article. We do not fully agree that this article is just “another realisation of chemistry-transport model driven by too fast (as authors pointed out) ECMWF circulation.”

Firstly, although a number of articles report on CTMs driven by the ECMWF ERA 40 reanalyses (Hadjinicolaou et al., GRL, 2005; Noije et al., JGR, 2004; Feng et al., ACP, 2007), very few papers, to our knowledge, describe experiments of CTMs driven by the ECMWF operational analyses of the last six years. Nevertheless, a recent paper justifies the use of the current operational analyses (Monge-Sanz et al., GRL, 2007) stating that with recent ECMWF analyses (of the year 2000) CTMs can produce stratospheric tracer transport over multiannual timescales more realistically than with other previous (re)analyses. This is in agreement with the conclusions of Chipperfield, QJRMS, 2006 that used (among other forcing fields) the operational ECMWF analyses of 2001. We decided to drive our CTM with these analysed fields, even though the age of air we computed was too young. Indeed, analysed meteorology has the advantage of making CTM results directly comparable with observations, since the model run is constrained by a realistic representation of the real atmosphere. Our use of a multi-year forcing (2000 to 2005) is, to our knowledge, without any other precedent.

Secondly, we think that the development of a new model is a scientific achievement and that is needs to be published in the peer-reviewed literature and not only in the grey literature. This seems to be fully accepted for new instruments. We also adhere to the principle that a model should be shown, again in the literature, to reproduce present observations before it can be used to predict the future. Our model simulates the chemistry composition of the atmosphere up to the mid-mesosphere without any artificial boundary conditions, neither in the troposphere nor in the stratosphere. Exchanges between the two layers are thus simulated freely, without any prescribed conditions, and this is in our opinion a scientific step forward.

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In the revised manuscript we have modified a number of descriptions of the chemical processes involved according to your remarks. In several cases we believe that these remarks arose because certain statements were unclear. We have tried to clarify them. Although these descriptions may not seem appropriate in a scientific paper, we believe that they help appreciate both the performance of the model and the main axes for future improvements. The option to move our paper to a technical note is not completely satisfactory as we do not really describe a new parameterization or a new version of data, but we rather use different ‘boxes’ that are gathered within our code. Since the present paper introduces the model in its entirety, size considerations do not allow for in-depth presentation of all aspects and we rather have to present a selection of our results.

Specific comments

1. *1. Title*

   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. MOCAGE-Climat is a version of the MOCAGE model, specifically adapted for multi-year simulations. We performed this validation here,

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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.’

2. *2. CTM/CCM*

   We have added in our introduction some sentences that clarify the differences between a CTM and a CCM. We have also emphasised the fact that the simulations we performed were those of a CTM and that our next objective was to couple the chemistry of MOCAGE-Climat with the ARPEGE-Climat GCM.

   We did not present any specific details on CCMs as it is not our purpose here. However, we investigated further the literature to get a better sense of what our position will be once our CCM is up and running. In particular, Eyring et al, JGR, 2006 presents, together with results from simulation of 13 CCMs, their limitations and deficiencies, for instance in terms of chemistry (in particular Cl_y or bromine chemistry), or in terms of transport. Though a number of current CCMs include a description of the tropospheric chemistry, they mostly do not consider this chemistry when simulations are made over several decades as it is currently too expensive in terms of computer time. One of the limitations in representing the tropospheric chemistry is linked to the treatment of the VOCs, both the representation of their emissions and of their oxidation. Quantitative variations arise also from the removal of soluble species (Shindell et al., JGR, 2006).

   In their article, (Shindell et al., JGR, 2006) indicated that most of the models were run in a CTM configuration. For the various models presented that considered a rather sophisticated tropospheric chemistry scheme, a couple only included a full description of the stratospheric chemistry, namely the ULAQ-GCMCM that has a very coarse horizontal resolution and the LLNL-IMPACT model; for the other models, chemical contents of the stratosphere were constrained by the use of climatologies.

3. *3. Objectives of our work*

We present here the ability of our CTM to simulate realistically the chemical composition of both the stratosphere and the troposphere. Only then can the results of a coupled mode be read with some confidence. This paper will be our reference for subsequent papers.

Since the present paper introduces the model in its entirety, it cannot do more than touch many features that are themselves a selection of our results. In our opinion, splitting the article into two would cut much of its scientific interest. However, the revised manuscript is now shorter as (1) we have shorten the part that illustrates the stability of the 6-year and 12-year simulations, and as (2) we have moved the part that describes the observations in an appendix, following your recommendations and the recommendations from the other reviewer. We still have a rather detailed description of the observations as we think it is crucial to know precisely which data sets have been used, even in the case of well-known instruments such as the TOMS. This is required as a lot of processing is put in the preparation of the data for later use by modellers or for comparisons with other instruments, and as new versions of the data are regularly prepared.

A run in a CTM mode not only points out the capabilities and limitations of the model but also helps in understanding processes in the atmosphere of the model.

We evaluate our model in various configurations in order to be able to choose the best one for long-term simulations. In particular, we compare results using two horizontal resolutions, simulations with and without a simplified treatment of the boundary layer, and simulations with and without deposition velocities calculated on-line. Given the discussions going on on the impact of the troposphere onto climate change, we thought it was interesting to show results from our sensitivity studies on boundary layer processes.

We do not agree that the experimental set-up does not fit our first goal. Firstly, we think that the recent ECMWF analyses are among the best in terms of reproducing the real state of the atmosphere, including both the troposphere and the stratosphere, even if it still has flaws. As mentioned, MOCAGE-Climat covers layers up to the middle mesosphere and ECMWF analyses on that vertical extension are available since 2000 only. Using this ECMWF forcing, we reduce the uncertainties related to the meteorology. Therefore, the main differences between our results and measurements should essentially be related to the representation of the chemistry (with all related physical processes). Secondly, we think that forcing our CTM with six years of recent ECMWF analyses (2000-2005) enables us to derive a realistic climatology of the chemical composition of the atmosphere as simulated by our CTM. This climatology can then be compared to observed climatologies. In addition, we could directly compare model outputs with some concomitant observations such as the MOZAIC, MOPITT or the ODIN observations. Forcing the CTM with GCM fields has already begun and will be pursued along with on-going tests to develop the model.

4. *4. Mass conservation in semi-Lagrangian*

   Please find the revised sentences in paragraph 2.1.

5. *5. Radiation*

   By using the meteorological forcing, wind, temperature and pressure fields, we indirectly take into account the full radiation, solar and terrestrial. But we also consider the solar radiation alone to compute off-line photolysis rates that affect chemical reactions.

   We have reworded the corresponding sentences (see paragraph 2.1).

6. *6. Species REPROBUS/RELACS*

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We have identified in Tables 1 and 2 the chemical species of the RELACS scheme that are also chemical species of the REPROBUS scheme.

7. *7. Description of observations*

   See our response to remark 3.

8. *8. Age of Air*

   The differences between our AOA and those presented in Bregman et al. 2006, calculated from their TM5 CTM, may be explained by two factors. The first factor is the transport scheme used: TM5 is an Eularian CTM while MOCAGE-Climat uses a Semi-Lagrangian Transport (SLT) scheme. This leads to differences in AOA as reported in Chipperfield, QJRMS, 2006 that used different configurations of their CTM to perform tests on stratospheric tracer transport. Chipperfield, QJRMS, 2006 investigated the impact of the advection scheme and concluded that changing from the Prather (1986) Eularian advection scheme to an SLT scheme resulted in lower ages of air for all variations of the model. The second factor could be the preprocessing, involving interpolations, that the ECMWF data undergo, in particular to recalculate the vertical motion, before they can be used to drive the CTM, as reported in Monge-Sanz et al., GRL, 2007.

   We investigated further in the literature to try to explain the difference we found between our T21 and T42 AOA. Two recent articles support our results: Scheele et al., ACP, 2007 indicated that a striking feature was that especially the 4D-Var data generated more cross-tropopause transport, corresponding to smaller, less realistic age, when the horizontal resolution was increased. They suggested that some transport across the tropopause by small-scale meteorological systems occured in the 4D-Var data set. Chipperfield, QJRMS, 2006 tested the stratospheric transport with different configurations of their CTM and concluded that, although increasing the horizontal resolution of an off-line model was expected to improve the overall simulation, this decreased the age of air in the tropical upper stratosphere from values which are already too low in some cases.

Our realised AOA should be taken as a feature of our simulation and is shown as such.

We have included the revised sentences in our article (see paragraph 3.2).

For an easier comparison with what appears in other articles, we replaced Fig 1 in our draft article that showed a monthly distribution for the month of December (last month of our simulation) by the annual mean of the last year of our 20-year simulation. We have now a figure that is more directly comparable to the figures generally presented on the AOA subject. We have made only small changes in the text however.

9. *9. Methane, water vapour and nitrous oxide*

   We agree that the paragraphs on CH_4 and N_2O need both some rewording and more discussion and explanations. We have done that, exploiting as you suggest the results obtained in the T42 ECMWF simulation and in the simulation driven by the ARPEGE-Climat GCM. We also have now included some H_2O comparisons. Paragraphs in the revised article are under 3.4.1.

   We have reworded the paragraph on N_2O (see paragraph 3.4.3).

10. *10. Tropopause and horizontal resolution*

    Improving the horizontal resolution allows to better describe the horizontal variations of the height of the tropopause, and as such better resolves the tropopause.

11. *11. Role of nitrogens in ozone chemistry*

    We have added the reference Crutzen, QJRMS, 1970. Indeed, the source of N_2O you mention in included in our chemical scheme. We have amended the text. We have also reworded the paragraph on N_2O (see above our response to remark 9).

12. *12.* NO_y

    We agree that the comparison between model NO_y and MOZAIC data only partly validates the model as the MOZAIC data are confined to the UTLS region. Based on a suggestion from the other reviewer of this article we have compared the model NO_y throughout the stratosphere to the UARS climatologies of $HNO_3 + NOsunset + NO_2sunset$.

    The revised paragraph on NO_y is under 3.4.4.

13. *13.* NO_x

    We agree that the role of HO_x and ClO_x is very important, and that the discussion on NO_3 can be shortened. N_2O_5 is not included in the NO_x family of the model as its lifetime is longer than the dynamical time step. It is thus transported by itself separately though it strongly interacts with the NO_x family.

    To follow your suggestion, we made a comparison between the monthly mean output of model NO_x and the average of the day-time values for that month (July). Relative differences were lower than ±10% throughout the entire stratosphere. Therefore, we have reworded the introduction of the paragraph on NO_x (see 3.5.1).

    Following your comment, we have checked the $ClONO_2$ field. It does not present any significant increase with respect to older simulations using REPROBUS coupled to ARPEGE-Climat (WMO, 1998), and clearly the first explanation that we gave was wrong. The problem seems to be related to the photo-dissociation rate of NO_2 that includes now the effect of the surface albedo. J(NO_2) is quite sensitive to this parameter that tends to reduce the photolysis rate (see figure provided as supplementary material). This results in a NO_x equilibrium at a higher value, and it should be investigated in future studies.

    Many thanks to the referee to point-out this particular problem and to help us understand our results.

In the revised version of the article we have now revised sentences (see 3.5.1).

Although the comparisons with the SCIAMACHY NO_2 columns reveal quite large differences that can be questioned, we have decided to keep them in the revised article. They outline what we think is one of the deficiencies of MOCAGE-Climat. A comparison with surface NO_x measurements in Europe would not be more appropriate to validate MOCAGE-Climat as they represent very local effects, that reveal surface characteristics and sparse anthropogenic sources. It is indeed not the purpose of MOCAGE-Climat, with its coarse horizontal resolution and the objective of being integrated over decades, to represent the features of such measurements. This is among the objectives of the air quality version of MOCAGE (see Dufour et al., AR, 2004 for instance).

14. *14. ClO and ozone hole*

The strengths of the paper from Farman et al. were (i) to highlight the Antarctic ozone depletion in the stratosphere but also (ii) to suggest the linkage with the chlorine amount in the atmosphere. Even if their explanation was very preliminary and imprecise, they gave the right ‘direction’ to follow to explain the ozone hole phenomenon, initiating the extensive associated literature that is regularly summarised within the WMO/UNEP publications. This paper is more than 20 years old now and its mention in our article is a modest tribute to the authors.

We agree that the role of HO_x is important, but this was implicit in our draft paper as HO_x participate in controlling the ozone budget over the whole atmospheric column, in association with other families that have different weights. For the troposphere, nitrogen oxides and ‘VOCs’ (that are more a class of chemical species rather than a family) are predominant. In the lower stratosphere, HO_x and nitrogen oxides mainly control the ozone budget, but the role of chlorine increases with altitude and is of primary importance within the vortex. In the upper stratosphere, the role of the NO_x chemistry decreases as the role of the chlorine chemistry increases, still in association with the HO_x chemistry. Therefore, we have mentioned the HO_x chemistry in the revised article to avoid any confusion.

We did not transform our data from latitudes to equivalent latitudes that are used in the Randel et al., JAS, 1998 climatology (please note that the Grooss and Russel, ACP, 2005 climatology we used is on latitudes). This certainly introduces differences, but principally at high latitudes during boreal winters. While this kind of transformation is mandatory when dealing with process oriented studies, that we will perform within the CCMVal framework, we thought that the present configuration was sufficient for our current purpose. Other groups have proceeded similarly (see Egovora et al., ACP, 2005).

15. *15. Spivakovsky data set*

    We have modified the wording in the text.

16. *16. Chlorine reservoir*

    Dr. Grooss has provided us with an extended HALOE climatology that covers the period 1991-2005. This new climatology is very close to the one described in Grooss and Russel, ACP, 2005, built from 1991-2002 data, even for the HCl field. This shows the solid representativeness of the Grooss and Russel, ACP, 2005 climatology. We will have simulations with the GCM meteorology that will cover the HALOE period when we will use our coupled system, in future studies.

    As for model Cl_y, following your suggestion, we have displayed them in the same way as presented in Eyring et al, JGR, 2006 Figure 12. We displayed a maximum of 3.8 ppbv, higher than we expected. We looked into the problem and discovered that our initialisation state, build from previous simulations WMO report 1998 (see para 3.1 in our article), imposed this mixing ratio for Cl_y obviously too high for 2000. We have corrected this “bug” in our code.

17. *17. Stratospheric ozone*

We do agree that the amount of CO_2 in the stratosphere is larger than that of ozone, but CO_2 is almost chemically inert, except at high altitudes where it can be photolysed. In fact, we mentioned ozone as the most abundant trace gas that is chemically reactive. We have updated the text accordingly in the revised article.

Regarding the O_2 photolysis, it is of course efficient everywhere in the atmosphere under sunlight conditions, but the maximum of ozone production is found in the tropical middle stratosphere, hence our sentence. We have reformulated it to be clearer.

About the sentence "It should be mentioned that though ozone mixing ratios in the stratosphere can be greater than 10 ppmv, it is in ‘chemical equilibrium’ with trace species whose mixing ratios can be from one thousand to one million times smaller.": the point is that the ozone budget is affected by other chemical species that have much lower concentrations, explaining why they should be considered within atmospheric CTMs to get reasonable ozone distributions. We recognize this contributes to the text-book style of our paper.

18. *18. Mesospheric ozone*

    As suggested, we have compared a comparison between the monthly mean output of O_3 from the model and the average of the day-values for that month (July). Indeed, zonal mixing ratios for altitudes above 0.3 hPa were about 0.4 ppmv lower. We have reworded this part of the paragraph (see 3.7.2).

19. *19. Tropospheric ozone*

    We have plotted the differences. However, this did not result in clearer figures as differences in the troposphere are two to three orders of magnitude smaller than differences in the stratosphere. We kept the presentation of the draft article as it allows to show results from two simulations both in the troposphere and in the stratosphere, together with the standard deviation of the observations. We agree that it is not optimal, but with the work on the labelling we made we think that these figures are informative.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 11295, 2007.