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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The MIPAS HOCl climatology" *by* T. von Clarmann et al.

T. von Clarmann et al.

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Authors' Reply

The authors like to thank the reviewers for their helpful comments. The original comments are inserted in italic font, the authors' replies are printed in normal roman font.

Anonymous Referee #1

General comments:
The paper presents a stratospheric vertically resolved HOCl climatology base on MI-

PAS satellite measurements. The climatology provides nearly two full annual cycles of the first global HOCl measurements and is therefore a valuable tool for the evaluation of coupled Chemistry-Climate Models (CCMs) and Chemical Transport Models (CTMs). In general the paper is well written and the important features of the HOCl climatology are presented clearly. However, some more detailed scientific background information on the presented features is missing. Also, the evaluation of the model data needs a more thorough discussion to understand the conclusions made regarding the rate coefficients. At the moment it seems that the model results presented here do not add anything new to the discussion of the rate coefficients. Publication of the manuscript is recommended after addressing these issues which are explained more in detailed in the specific comments below.

Calculations with various sets of rate constants for ClO + HO_2 (Stimpfle, JPL 2006, JPL 2009) have meanwhile been performed and will be discussed in the revised version.

Specific comments:
Page 20795, line 16: Only model calculations based on one set of rate coefficients are presented in the manuscript. However, I agree that a comparison of model calculations based on the two different rate coefficients would be interesting and add scientific value to the presentation of the model results.

We agree; model calculations based on different rate coefficients have meanwhile been performed and will be included in the revised version of the paper. See above for details.

Page 20796, line 5 and following: So the MIPAS HOCl has only been retrieved for 2002-2004? Please clarify.

We will add the following sentence: "Thus MIPAS HOCl retrievals are available only from June 2002 to March 2004."

Page 20797, line 6-7: What kind of earlier measurements? Please provide more information. Has the good agreement with the earlier measurements been published somewhere? What exactly is meant by 'good agreement'?

These are balloon-borne far-infrared measurements. The agreement is of the kind that the trend-corrected far-infrared measurements fall within the MIPAS zonal means of the related latitude band plus minus one standard deviation. This information along with a reference of the comparison will be provided in the revised version.

Page 20799, line 10-15: Additionally, there are in general higher mixing ratios in the summer hemisphere than in the winter hemisphere with a maximum in midlatitudes. Please provide and explain this information including relevant references.

This information will be provided but there is no easy explanation. The concentration of HOCl depends directly on illumination, via the photolysis rates, and indirectly, via the abundance of OH and HO_2. Further there is a temperature dependence of the HOCl formation and dependence on the ClO abundance. All these involved atmospheric variables have their own altitude and latitude dependent diurnal and annual cycles. The actual abundance of HOCl thus is the net effect of multiple superimposed processes and thus is in most cases resistant to an easy explanation. We will discuss the processes in more detail but we cannot assign the observed phenomena to a simple mechanism. Instead, we use a model to verify that the quantitative understanding of all involved processes is sufficient to explain the observed behaviour.

Page 20799, line 13-18: Please provide (if possible) a short explanation (including references) why the altitude of the maximum mixing ratio is lower in the summer hemisphere and during daytime. Also, the latitudinal structure (with higher mixing ratios in

the summer hemisphere) apparent from am panels in Figure 2 and 5 changes for pm measurements as the comparison between Figure 5, panel 2 and figure 6, panel 1 reveals.

Here applies the same as said above.

Page 20799, line 23-26: Is the semiannual oscillation of the peak mixing ratios connected to the SAO in zonal wind and temperature? Please provide some scientific background to explain the observed features.

The HOCl concentrations are driven by photochemistry and the availability of short-lived species of illumination-dependent abundance. Thus, it seems quite natural to us that the HOCl abundance is correlated to the noon solar zenith angle, which also is described by a semi-annual oscillation in the tropics. For a gas whose abundance is driven by reactions as fast that a pronounced diurnal cycle can be observed, winds and transport play a minor role. In the paper, we write about 'a' (indefinite article) semiannual oscillation, which should make clear that we do not talk about 'the' semiannual oscillation which would refer to the technical meteorological term SAO, but about a generic oscillation with the frequency 2yr^{-1} is observed.

Page 20800, line 1-4: Why are particularly low values found in early winter midlatitudes? To explain this one would need to understand the general latitudinal structure of the HOCl mixing ratios (see also comment for page 20799, line 10-15).

Comparison of am and pm measurements yields that this minimum occurs during daytime only. This hints at photolysis which destroys HOCl in midlatitudes but not in the polar night.

Page 20800, line 18-22: Why are there no elevated HOCl abundances in the very inner *part of the SH 2002 polar vortex (see Figure 6 panel 1). Please also provide some information if this feature is independent of am/pm separation of the measurements (which seems to strongly influence the latitudinal structure of the HOCl abundances).*

In the polar night there is no diurnal effect. Systematic differences between the am and pm measurements are restricted to parts of the atmosphere which are sunlit at am but dark at pm. Exactly over the pole there is no light to trigger HOx chemistry, thus no HO_2 to form HOCl. This explanation will be included in the text.

Page 20800, line 20: What means regularly? Is this phrase based on the two winters of MIPAS observations or on independent observations as well? Please rephrase and/or add references. The same argument holds for the phrase 'occasional' for the NH winter. Observations for two winters do not really imply the use of terms like regular or occasional. Also Figure 7 shows this event for the 2003 and 2004 NH winter!

We agree and will reword this.

Page 20801, line 12: Would the overestimation of HOCl during polar night be contradicting the assumption that Stimpfle rate coefficients are more appropriate?

We thank the reviewer for raising this interesting question. The answer is: No, we have found another cause for overestimation of HOCl during polar night by the model: The handling of HOCl with a family approach for transport modelling, which seems to be default in EMAC caused this artifact, which has disappeared when HOCl was explicitly modeled.

Page 2804, line 10-11: How do we know that observations agree better with kinetic data reported by Stimpfle? From this manuscript? Earlier publications?

In the revised version calculations with various kinetic data will be included and

discussed.

Figures 2, 3, 5, and 6: The panels are too small. A better way of presenting the figure would be a 2x2 panel plot which extends over the width of the whole page.

We agree that the figures are too small. The small reproduction of the figures is due to the particular ACPD landscape format. In the ACP format the figures will be reproduced larger.

Anonymous Referee #2

The authors present a valuable data set derived from MIPAS ENVISAT HOCl limb measurements in the mid-infrared spectral region covering the time domain from June 2002 to March 2004. Results from an up-to-date chemical climate model are also presented. The paper is based on earlier HOCl papers by the same group focussing on global distributions of HOCl for short temporal episodes (von Clarmann, 2006) and on the the Antarctic winter vortex HOCl chemistry (von Clarmann et al., 2009a). While the data set well deserves publication the present state of the paper is not suitable for publication before major revisions have been made. The first and main criticism is a missing or much to short discussion of the model-measurement intercomparison that shows several interesting features. Here the authors do not even attempt to give possible reasons for the major model overestimation of HOCl in the 30-40 km altitude regime for polar night conditions. This is a very intersting feature that has not been explained and is just mentioned in a few sentences.

The authors thank the reviewer for this point. The cause of this discrepancy has been identified: The handling of HOCl with a family approach for transport modelling, which seems to be default in EMAC caused this artifact; it has disappeared when HOCl was explicitly modeled. This will be discussed in the revised manuscript.

A total of eight figures showing measured and modelled HOCl distributions and evolutions is dealt with in a short paragraph of intercomparison which for my taste is not at all sufficient for publication in a peer-reviewed journal.

We have decided to restructure the paper, and will, after a short introduction of the model runs, discuss the model results along with the MIPAS results, following the organization used for the latter. This will give room for extended discussion.

Secondly, the data and model features are not really presented in an effective manner so the reader can easily grap the details. Several figures do not focus on the features they are supposed to present but standard global plots are used instead.

Plots with limited latitude coverage will be provided where appropriate.

In the introduction the JPL recommendation of 2006 is given as the current recommendation although the reaction in question has been updated in the 2009 issue.

The authors thank the reviewer for spotting this mistake. The revised version will include calculations both with Sander (2006) and Sander (2009) rate constants for ClO + HO_2.

Several other points are raised in the detailed comments below. In general the data and model results do have the potential for a solid paper but substantial effort is needed to discuss the features in an adequate way and to present the argumentation and results to the reader.

The discussion will be extended; additional model runs will be included.

At some places the paper suffers from Germanisms.

We will do our best to avoid these.

Major Detailed Comments p.20794, l.6:. The abstract should be more quantitative in places: Formulations such as "at lower altitudes" and "in the lower stratosphere" are both used leaving the reader unsure about the altitude regime.

Numbers will be provided where appropriate.

p.20795, l.12: Sander 2006 is NOT the current recommendation. For ClO+HO2 it has been superseeded by the 2009 update Sander et al. 2009 (JPL Publication 09-31).

This is correct; thanks again for spotting this mistake. We have meanwhile performed model calculations using the Sander et al. 2009 rate constants. These will be discussed in the paper.

p.20796, l.13: For the intercomparison of absolute HOCl mixing ratios with model results precision is not really the relevant error estimate. Accuracy of the measurements must be stated for this and used throughout the intercomparison process.

This is correct but this section does not cover the measurement model intercomparison but is a generic description of the MIPAS data. Thus we feel that it is adequate to report also the precision. In the revised version we will additionally report the estimated systematic error, which is 10%. The only known source of systematic errors is the uncertainty of spectroscopic data.

p.20797, l.6: The quality of agreement must be quantified in terms of the measurement error...

Since we do not compare collocated profiles but measurements from different decades, we compare the profiles by Chance et al with related zonal means of

MIPAS. For the latter the standard deviation of the zonal mean, not the measurement error, is the quantity to be used to assess how well the measurements agree. The trend-corrected profiles by Chance et al. fall within the range covered by the MIPAS zonal mean profiles plus/minus one standard deviation.

... it should be stated which trends have been used in the intercomparison.

The trend correction (actually we have assumed an increase of HOCl along with a total stratospheric chlorine increase by the same amount by 35% from 1988/89 to 2002) has been reported in von Clarmann et al., 2006. This paper will be referenced for the details of trend correction.

An intercomparison with the earlier publication from the same group (v.Clarmann et al. 2006) based on an earlier data version should be presented or it should be mentioned that the data compare very well.

We will mention that the data compare very well.

p.20797, l.8: The data set should be made publicly available. A download location should be given.

The download location will be given.

p.20797, l.8: A whole bunch of improvements is listed here but none is explained in detail. This should be done or a proper reference must be given.

These improvements are on a technical level and are related to details which are, in most other papers on remote sensing, not reported at all. While these improvements make the retrieval more robust, they do not substantially change the results. While we feel that we should inform the data user about the differences between data versions,

we do not consider the discussion of these technical details useful.

p.20799, l.16: Since the altitude of the peak mixing ratios is lower during daytime this should be a downward shift (NOT upward) during nighttime, right?

We will avoid the term ‘shift’ because it may be understood to characterize the process instead of the status.

p.20800, l.13: Which ensemble of data in terms of space and time does this standard deviation refer to? This must be clearly stated.

Since the calculation of related standard deviations is reported explicitly in the section “Method” and since the whole paper is about climatologies of monthly zonal mixing ratios in 5 degree latitude bins, we thought it was obvious that these are the ensembles the standard deviations refer to, but we will specify this again.

p.20800, l.15: The statement concerning high standard deviations in the polar night regime must be weakened since other potential causes have not been ruled out.

We will add rapid composition changes within the averaging period as a third reason. Since we have no indication for massively enhanced retrieval errors, what else than inhomogeneous composition of an ensemble (in space or time) can make standard deviations large?

p.20801, l.5: While the model data are prominently presented along with the MIPAS HOCl data the paragraph does only present an extremely short discussion on the model-measurement comparison. The improvement of the modeled peak HOCl mixing ratios by using the Stimpfle (1979) rate constant is just mentioned but not shown in any way. A figure showing the effect on the global distribution would have been very *worthwhile...*

Such calculations have meanwhile been performed and will be included in the revised version.

...Else just a few facts are mentioned and no single attempt is made to explain the strong model overestimation of HOCl for polar night conditions, which is the most obvious feature in several of the plots presented.

The cause for this overestimation has meanwhile been identified. The family approach used by default to model transport of ClOx species including HOCl causes this artifact. Model calculations with explicit HOCl modeling have been performed and will be used for the revised version of the paper.

p.20801, l.22: The behaviour discussed in the paragraph is not in any way obvious from Fig.8 that gives zonal mean HOCl over all latitudes (see comments below concerning the figure). In the way presented the arguments of the authors (although sensible) are not comprehensible to the reader.

The first lines of Section 5.1.1. are not meant to explain Figure 8 but to place Figure 8 in an appropriate scientific context. Figure 8 is used to give evidence that the September 2002 southern hemispheric polar values are exceptional and should not be considered as representative.

Figs.2,3,5,6: The plots are much to small in the size given in the discussion paper. They should be enlarged and reorganized in a 2x2 scheme which provides better intercomparison between measurement and model. Also, since the color bars are identical one per figure will be sufficient. A geometric altitude axis to the right might also help some readers.

We agree that these figures are too small. This is due to the ACPD landscape format. In the ACP format the figures will be reproduced much larger. We do not think that an approximate geometrical altitude scale is appropriate, because pressure varies with latitude by more than a factor of two for a given altitude.

Fig.2: The vast overestimation of HOCl inside the polar vortices by the model is obvious but not discussed in the text at all.

This problem has been solved. See above.

Fig.5: The major part of the figure does not add any significant new information as compared to Fig.2. The plot should be reduced to the regimes that are discussed in the text and enlarged.

We prefer to leave the figure as it is because we think that a global minimum can only be identified on a global plot.

Figs.5,6: What is the reason for the unnaturally steep vortex boundary gradients in the winter antarctic model data? These look like artifacts?

Also this is caused by the family approach used. This problem has been solved.

Fig.6: Regarding that Fig.7 shows the features discussed in the text much better Fig. 6 could be skipped completely.

We prefer to keep Figure 6 because it includes information about the altitude dependence.

Fig.8: The figure should be changed in a way to focus on the antarctic region discussed

in the appropriate paragraph in the text. At the first glance the figure seems incompatible with Fig.7 which as well shows the Sept. 2002 and 2003 averages over the anarctic region (although different color scales are used). This must be checked.

Figure 7 is pm, figure 8 is am. We agree with respect to the focus of this figure and will replace it by one which is limited to polar regions.

Appendix A: Not reviewed, due to other obligations. Minor Comments p.20794, l.2:. Use "period" instead of "episode"

ok

p.20794, l.22: Is Solomon et al. 1986 really a good reference for mid-lat. O3 loss? Why not use a WMO-report?

We will include two further journal references and the WMO report of 2011 here instead.

p.20794, l.25: Occasional measurements exist ...

p.20795, l.5 and in general:. Be more specific: ... MIPAS onboard ENVISAT ... (MIPAS-E was used in earlier papers)

p.20796, l.9: ... differences conStraint

p.20796, l.18: The Figure deserves better attention than being introduced in parentheses at the end of the sentence.

all agreed.

p.20799, l.3: It should be checked wether there is no better reference than a Ph.Thesis

of obviously German language which is of limited use for the community.

For this particular matter there is no better reference but the most important information is also included in the Kirner et al. 2011 reference.

p.20799, l.11: ... maximum ...

p.20799, l.12: (appr. 35-43 km), respectively.

all agreed.

p.20799, l.14 and following: Figures should not be introduced in parentheses.

We disagree. To dedicate a sentence to each figure interrupts the logical flow of the text and would add unnecessary length to the paper.

p.20799, l.25: Delete one "of"

agreed.

p.20800, l.8: ... to warm ...

We have changed this to "warmer than usual."

p.20801, l.5: ...maximum ...

p.20802, l.17: ...confirmed...

all agreed.

Fig.1: The title on top of the figure is superfluous

ok, will be removed..

Interactive comment on "The MIPAS HOCl climatology" by T. von Clarmann et al., M. Suzuki

Dear authors,

This paper is referring ISS/JEM/SMILES. Since SMILES has been launched and operated during Sep. 2009 and Apr. 2010, and some early results has been published. We would like to comment that the reference to the SMILES should be the following paper (1) which describes instrument, retrieval, real spectrum and sample of retrieved profiles.

(1) Kikuchi, Ken-Ichi, Toshiyuki Nishibori, Satoshi Ochiai, Hiroyuki Ozeki, Yoshihisa Irimajiri, Yasuko Kasai, Makoto Koike, et al. 2010. "Overview and early results of the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES)." J. GeoPhys. Res. 115 (D23) (December 7): D23306. doi:10.1029/2010JD014379.

Currently you are referring Y. Kasai et al 2009, it is reporting sensitivity study from one of the retrieval groups in the SMILES science team, and it is not relating operational L2 products. Algorithms and sensitivity analysis of SMILES Operational L2 products is already published in the following papers (2, 3). If you need references to the paper on the sensitivity studies of SMILES, you may refer also Ref (2) in addition to Ref (1). But we feel the reference for the SMILES is simply Ref (1).

(2) Takahashi, Chikako, Satoshi Ochiai, and Makoto Suzuki. 2010. "Operational retrieval algorithms for JEM/SMILES level 2 data processing system." Journal of Quantitative Spectroscopy and Radiative Transfer 111: 160–173.

doi:10.1016/j.jqsrt.2009.06.005.

(3) Takahashi, Chikako, Makoto Suzuki, Chihiro Mitsuda, Satoshi Ochiai, Naohiro Manago, Hiroo Hayashi, Yoshitaka Iwata, et al. 2011. "Capability for ozone high-precision retrieval on JEM/SMILES observation." ADVANCES IN SPACE RESEARCH (May 26): 1–10. doi:10.1016/j.asr.2011.04.038.

The original reference has been chosen because it explicitly mentions HOCl retrievals. However, we will include the suggested reference.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 20793, 2011.