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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The impact of anthropogenic chlorine, stratospheric ozone change and chemical feedbacks on stratospheric water" *by* T. Röckmann et al.

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We thank the referees for reviewing our paper. Here are our comments:

Referee 1:

The main point of criticism of the referee is that the concepts presented in this paper are not new and that there are no novel ideas or striking new results compared to papers by Nedoluha et al., (1998) and Considine et al., (2001). Although we were indeed not aware of the first paper, we disagree on this judgement and we show here that our paper presents important new ideas and concepts, as well as new results.

We thank the referee for bringing the paper by Nedoluha et al., (1998) to our attention, which will be referred to in the revised version . Those authors carried out a study simi-

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lar to ours, but - except for one chemical effect - suggested very different concepts and came to largely different conclusions. Regarding the three mechanisms put forward in our paper, Nedoluha et al., 1998

1) investigate the effect of increasing stratospheric chlorine on CH4 oxidation, but conclude in contrast to our work that the effect is almost negligible.

2) investigate a solar cycle effect, but again conclude that the effect is very small except for high altitudes. In particular, they only discuss the variation of UV photolysis due to variation in the Lyman a intensity. In contrast, in our paper we suggest that changes in the upper atmospheric ozone column can lead to higher O(1D) below and thus to intensified CH4 oxidation, and that this effect can be significant and may have strongly affected H2O and CH4 during the measurement period of HALOE.

3) do not mention the feedback of increased stratospheric water on OH levels and thus on intensified oxidation of CH4.

Thus, in reference to Nedoluha et al., [1998], our study presents new concepts and new ideas (2 and 3), as well as new results, namely that all three effects can have a significant effect on stratospheric water levels.

Considine et al., (2001) study primarily the influence of the Mount Pinatubo eruption on stratospheric H2O and CH4 trends. In contrast to what the referee cites, they conclude that "atmospheric observations lack the strong signature of a pulse propagating from the tropical lower stratosphere as seen in the model simulation" and that "the aerosol from the Mount Pinatubo eruption could have contributed to the observed changes in CH4 and H2O following the eruption but was probably not the sole driver of the observed changes." In their study, there are substantial differences between the model results and the observations. Thus additional processes, like the one proposed in our paper, are clearly called for rather than refuted by their conclusions.

Regarding the three mechanisms investigated in our paper, Considine et al., (2001)

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1) mention that "the smaller upper stratospheric trends compared to the lower stratosphere may be due to Cl....", but do not investigate this effect in detail.

2) mention a possible role of changes in lower stratospheric Ox due to increasing Cl levels. However, in their argumentation, this is briefly mentioned to explain a feature of decreasing water levels in the lower stratosphere, in sharp contrast to the mechanism that we propose, i.e. that ozone depletion in the upper stratosphere leads to higher actinic flux and thus more O(1D) formation at lower altitudes.

3) do not mention the feedback of increased stratospheric water on OH levels and thus on intensified oxidation of CH4.

Thus, in comparison to Considine et al., (2001), our paper clearly presents new ideas and concepts (2 and 3), investigates process (1) in detail and yields striking new results in showing quantitatively that all three effects can have significant impact on stratospheric water.

A further new result is that in our paper we investigate the importance of the chemical mechanisms for various time periods and show how they have varied significantly with time in the past.

Regarding the suggestion to add an additional figure, we note that we do not calculate time series and thus cannot provide such a figure. We have clearly stated in the paper that the chemical mechanisms put forward cannot explain the "large interannual and spatial variability" seen in the observations. It is hard to say it more clearly.

Referee 2:

The main point of criticism of referee 2 is the use of a box model to study the chemical effects. We have discussed pros and cons of models of different complexity in the paper and argue why we use our approach. Despite the deficiencies of a box model as stated clearly in the paper, we still think that the present approach is the most adequate one to separate the individual effects, actually because we can in this model (at

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least to first order) "decouple" the three effects. Although the box model does indeed not calculate the actinic flux interactively, we stress again that the ozone fields used for the actinic flux calculations (and actually all quantities in the paper for which there exist observations) are taken from actual atmospheric measurements. Furthermore, the results are supported by the results from the 2D model, which use interactive calculations of the actinic flux as noted by the referee.

Following the suggestions of the referee with slight modifications, we present in the revised version additional 2D model results which show the effect of intensified CH4 oxidation due to the increase in CFC emissions on stratospheric H2O and CH4 levels. The new Figure 2 shows 2D model results for the scenarios "CH4 increase only", "CFC increase only" and "CH4 + CFC increase" (the old figure 2). When only CH4 increase is considered, CH4 trends always remain positive. When only the CFC increase is considered, trends are always negative, which shows the pure effect of intensified oxidation without the simultaneous increase in the tropospheric CH4 mixing ratio. When both trends are considered, the intensified oxidation of CH4 is clearly seen by the atmospheric trends being positive in the lower stratosphere and becoming negative in the upper stratosphere. Since the 2D model simulations do calculate the actinic flux interactively, runs b) and c) already include part of the feedback from ozone changes, and also from the feedback of increasing water itself. Solar cycle effects are not considered in the 2D model runs, however. We note again that in the coupled 2D model, it is not possible to clearly separate the individual effects, and that is one reason why we decided to investigate the three effects using the box model in the original paper.

The technical comment has been included in the revised version.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 833, 2004.