Atmos. Chem. Phys. Discuss., 12, C7637–C7641, 2012
www.atmos-chem-phys-discuss.net/12/C7637/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The sensitivity of stratospheric ozone changes through the 21st century to N_2O and CH_4" *by* L. E. Revell et al.

Anonymous Referee #2

Received and published: 1 October 2012

General

This paper analyses several scenarios of greenhouse gas development in the 21 century, in particular increases in nitrous oxide and methane, and its impact on ozone. This is an interesting and important topic which is well appropriate for ACP. The paper is well presented and Figures are of good quality.

I have some points of critique as well. I suggest more discussion of changes in the Brewer Dobson circulation, of chemistry driven by stratospheric temperature change (e.g., N_2O + O(1D)), and of the stratospheric chlorine loading and its impact on the predicted behaviour of ozone. The decreasing stratospheric chlorine loading is certainly the dominant effect on stratospheric ozone over the time period considered.

Furthermore, I suggest more discussion about the observed latitudinal structure of the response of stratospheric ozone on N_2O and methane changes. And I am not convinced that CO oxidation is the reason for enhanced ozone production and an enhanced N_2O scenario (Fig. 4).

In summary, I suggest several revisions to the paper (see also details below) but expect to find a revised version acceptable for publication in ACP.

Specific Comments

The NOx levels in the stratosphere are determined by the reaction N_2O with O(1D), which is temperature dependent. Thus, an increase in N_2O in the future does not necessarily lead to an increase in NOx. Further, changes in greenhouse gases lead to changes in the Brewer-Dobson circulation which likely have an impact the relation between N_2O (and methane) increases in the troposphere and stratospheric ozone loss cycles. I suggest more discussion of these issues in the paper. In part, the issues mentioned above have been discussed in a recent publication by the authors (in *Geophys. Rev. Lett.*) so that it might be helpful to refer to this paper in the discussion. This could also help clarifying in what respect the results in the present paper reach beyond those in the earlier paper (I do not dispute that they do reach beyond).

Furthermore, increase in stratospheric water vapour has an impact also on polar ozone (e.g., Kirk-Davidoff et al., 1999; Feck et al., 2008; Vogel et al., 2011) and possibly even on mid-latitude ozone (e.g., Ravishankara, 2012) via a coupling with chlorine chemistry. This aspect is not treated in the paper. Possibly, such effects are less an issue for the present study as it aims at a situation when chlorine levels will have significantly decreased compared to the maximum values. Nonetheless, this issue should be discussed in the paper. In particular as Figs. 4 and 5 demonstrate that (apart from the tropical lower stratosphere) the greatest effect of N_2O and methane

increases occur in the tropics in the SOCOL simulations discussed here. What is the reason for this model behaviour? Can any impact of chlorine chemistry on this model behaviour be excluded?

Moreover, chemical ozone production via R2-R5 (and driven by NOx increase) is put forward as a reason for the positive ozone signal in the tropics visible e.g. in Fig. 4. Note that the positive ozone signal reaches up to altitudes of about 30 hPa. Are there references which could support this idea? Could not tropical ozone might also change through a change in tropical up-welling (e.g., Randel et al., 2007; Ploeger et al., 2011), which might well occur between the scenarios compared in Fig. 4? The paper could be clearer here to preset the ozone production via R2-R5 either a *possible* explanation or to back up the conclusion with information (e.g., ozone production rates) from the model simulations.

Finally, global averages of ozone loss rates are shown in Fig. 3. However, ozone loss rates have a different meaning in a photochemically controlled regime, where loss rates and chemical production are in close balance (upper stratosphere and tropics) and in a dynamically controlled regime (e.g., polar spring and lower stratosphere). So I am not sure how to interpret a global mean of this quantity. My suggestion is to discuss and show only regional averages of this quantity not global averages. Investigating the ozone loss rates in regions might also help explaining why ozone reacts differently in different latitudes on N_2O and methane increases in the simulations (Figs. 4 and 5).

Comments in Detail

- page 17584, l. 9: quantify 'decrease'
- l. 11: state why the overall effect is an increase in total ozone
- l. 24: The radiative impact of methane and N_2O should be discussed. Could they

  contribute to stratospheric cooling in the model results?
- p. 17585, l. 17: add 'and adjustments'
- p. 17586, l. 1,2: this statement is not true for polar ozone depletion. Although R1 is important for Antarctica, the concentration of methane is not relevant.
- p. 17589, l. 17: What is the evidence from the model results for this conclusion? As it stands, this statement is too speculative.
- l. 28,29: again, what is the evidence from the model results for this attribution?
- p. 17590, l. 4: why is the strongest effect in the Arctic?
- l 14: 'significantly different' from zero ...
- l. 19./20: why is the sensitivity enhanced in the polar regions?
- What is the evidence from the model results that enhanced N_2O leads to ozone production?
- p. 17591, l. 10.,11.: This increase in ozone is driven mainly by the the decrease of the stratospheric chlorine levels. This should be clarified.
- l. 21: state the result of the 'vertically-resolved relationship' not just the fact that it was 'shown'.
- p. 17592: Ref. Brasseur and Solomon: publisher is missing.
- Fig. 5: it is obvious from this figure that the strongest impact of methane increase is at about 20–50 hPa at the poles. This result seems important to me and I suggest that it should be discussed in more detail.

- Figs. 6, 7, 8, and 9: These fits and the estimated uncertainty are based on only *four* points. I think this needs to be taken into account in the interpretation of these results.

References

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Kirk-Davidoff, D. B., Hintsa, E. J., Anderson, J. G., and Keith, D. W.: The effect of climate change on ozone depletion through changes in stratospheric water vapour, Nature, 402, 399–401, doi:10.1038/46521, 1999.

Ploeger, F., Fueglistaler, S., Grooß, J.-U., Günther, G., Konopka, P., Liu, Y. S., Müller, R., Ravegnani, F., Schiller, C., Ulanovski, A., and Riese, M.: Insight from ozone and water vapour on transport in the tropical tropopause layer (TTL), Atmos. Chem. Phys., pp. 407–419, doi:10.5194/acp-11-407-2011, 2011.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 17583, 2012.