Author Comment to Referee #1

ACP Discussions doi: 10.5194/acp-2018-1193 (Editor - Martin Dameris) 'Mechanism of ozone loss under enhanced water vapour conditions in the mid-latitude lower stratosphere in summer'

We thank Referee #1 for guidance on how to revise our paper. Following the reviewers advice we shortend the description of chemical processes and refer to a potential change of stratospheric dynamics due to an enhancement of the stratospheric sulphur abundance. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics. Passages from the revised version of the manuscript are shown in blue.

This paper presents a detailed chemical study for a potentially significant ozone depletion in the lowermost stratosphere, using a chemical box-model, with 7-day and 19-day back-trajectory analysis. The study is conducted under conditions of low temperatures (<205 K) and an elevated water vapour mixing ratio, up to 20 ppmv (resulting from convective overshooting events, rather frequent for summertime mid-latitude conditions). These convective events can transport ice crystals into the lowermost stratosphere, where the ice evaporates leading to a local water vapour increase. The sensitivity to high Cly mixing ratios is also addressed. The authors analyze with plenty of details the catalytic chemical cycles involving ClOx, NOx, HOx and leading to a perturbed balance of ozone production and destruction taking place in the lowermost stratosphere. The study takes inspiration from previous published works (Anderson et al., 2012; Anderson and Clapp, 2018); the authors conclude that the combined effects of temperature, water vapour and chlorine on the ozone loss process are consistent in their study with respect to these previous ones. I think that this study may help clarify important points regarding the ozone sensitivity to elevated water vapour conditions in the lowermost stratosphere and for this reason I recommend publication on ACP. In my opinion, a few improvements could be made in the manuscript, mainly for completeness and for improving readability.

Specific Comments

(1) Chemical cycles are essentially those leading to polar ozone depletion and widely described in previous literature. For this reason, I suggest moving large part of chemical details from the main text to a specific Appendix or in supplementary material. In particular, section 3.2.2 is in my opinion way too detailed and should be simplified focusing of the evidences of Fig. 5 (which is clear and exhaustive).

As proposed, we decided to shorten following aspects to focus on the analysis of lowermost stratospheric ozone chemistry occurring under enhanced water vapour conditions:

- We removed the chemical formulation of the ClO-Dimer-Cycle and the ClO-BrO-cycle in the description of ozone loss cycles leading to polar ozone loss and moved the formulation of cycle C3 from the introduction to section 3.2.2.
- The description of ozone formation at low water vapour mixing ratios is shortened by removing the formulation of ozone chemistry and NO reactions in Section 3.1.
- The detailed explanations of the water vapour dependence of R12 ($OH+O_3$) and R19 (OH+CO) in Section 3.2.2 (P.17, l.18-26, ACPD version of the manuscript) is removed.
- The description of the maintenance of activated chlorine is simplified by moving the chemical formulation of cycle C7 and C8 to the appendix. They are substituted by a new scheme (see Fig. 1 of this reply), which illustrates the important relations for the maintenance of elevated chlorine.

(2) The authors clearly state that their box-model ignores mixing with outside air poorer of water vapour, so that their calculated ozone losses should be interpreted as an extreme case. They also suggest a possible use of their findings in global modelling of the atmosphere for future experiment of sulphate geoengineering under changing climate conditions, or in case of major volcanic eruptions. It should be mentioned that in these cases the large scale

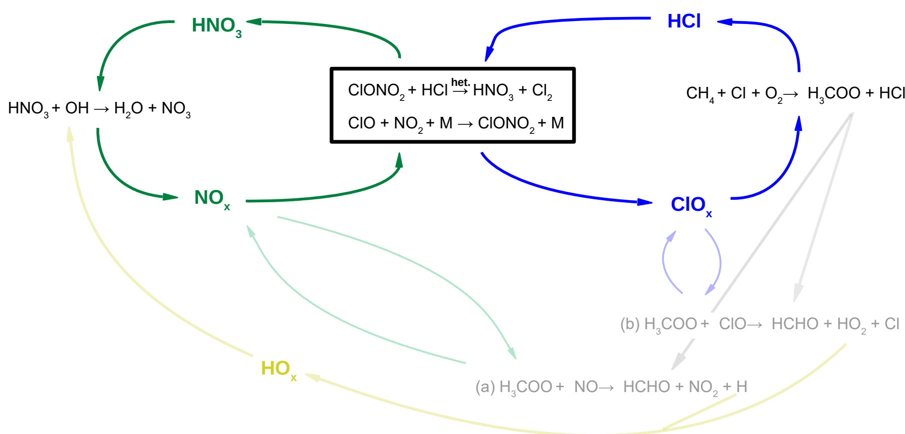

Figure 1: Scheme to illustrate the balance between chlorine activation and chlorine deactivation (blue, right) and NO_x activation and deactivation (green, left). The heterogeneous reaction $ClONO_2$+HCl (R1) links both cycles. Additional reaction pathways balancing radicals are shown in light colour.

latitudinal mixing of atmospheric tracers in the lower branch of the Brewer-Dobson circulation could be significantly affected in sulphate-perturbed conditions due to geoengineering or major tropical eruptions, both leading to a different level of isolation of the tropical pipe with the mid-latitudes. Eddy heat fluxes could also be perturbed, thus affecting mid-latitude temperatures in the lower stratosphere. Recent works have addressed these specific points (e.g., Visioni et al.,2017).

Visioni, D., et al. : Sulphate Geoengineering Impact on Methane Transport and Lifetime: Results from the Geoengineering Model Intercomparison Project (GeoMIP), Atmos. Chem. Phys., 17, 11209-11226, doi:10.5194/acp-17-11209-2017, 2017.

We mention the effect of sulphate geoengineering on lowermost stratospheric dynamics in Section 6 (P.27, l.27–31, revised version of the manuscript). We added the following paragraph.

Applying solar geoengineering would also affect the temperature in the lowermost stratosphere by perturbing the Eddy-heat fluxes and would change the lower stratospheric dynamic (Visioni et al., 2017). It would affect large scale latitudinal mixing of atmospheric tracers in the lower branch of the Brewer-Dobson-Circulation leading to a different level of isolation of the tropical pipe with mid-latitudes and would result in a different chemical composition of the lower mid-latitude stratosphere.

References

Visioni, D., Pitari, G., Aquila, V., Tilmes, S., Cionni, I., Di Genova, G., and Mancini, E.: Sulfate geoengineering impact on methane transport and lifetime: results from the Geoengineering Model Intercomparison Project (GeoMIP), Atmos. Chem. Phys., 17, 11 209–11 226, doi: 10.5194/acp-17-11209-2017, 2017.