Reply to the short comments of Johannes Lelieveld

Compliments for the excellent article that represents a major step forward in the discussion of cloud chemical effects on tropospheric composition. The development of the JAMOC scheme, accounting for comprehensive VOC chemistry, and the successful implementation in the EMAC model is an important accomplishment. The use of JAMOC brings the model significantly closer to observations of VOCs and ozone. Impressive. The results on VOCs and OVOCs, notably of aqueous phase chemistry and considering that most clouds evaporate rather than precipitate, will also offer new angles of approach in studies of organic aerosols.

It should be mentioned that this work was possible as it could build on the EMAC modelling framework, being the effort of a team (of which I am happy to be a member). It has set the stage for comprehensive, global atmospheric chemistry modelling, including the explicit and comprehensive account of VOCs and multiphase processes (e.g. Tost et al., 2007; Taraborrelli et al., 2009; Sander et al., 2011, 2019; Jöckel et al., 2010). I hope the article will be accepted for publication in ACP, while having a few minor comments in view of the interpretation of my past work.

We are very grateful for this positive feedback and for seeing the potential of our contribution to the community. We are fully aware that our work builds on the works from contributors within and outside the EMAC community. Please find in black the original comments and in red our replies.

1.27/28: This was posed by Lelieveld and Crutzen (1990), as HO2 transfers to the aqueous phase, so that gas phase ozone formation through NO+HO2 ceases and dissolved HO2 (through superoxide) reacts with ozone, effectively turning O3 production into O3 loss. To a lesser degree this also applies to RO2.

Thank you for spotting this. We added the appropriate reference to the revised manuscript.

1.40-43, and 1.480: Lelieveld and Crutzen (1990) concluded that net O3 production at particular locations, being subject to cloud processing, can be reduced by 40 % (comparable to your CAABA results). Liang and Jacob (1997) referred to the troposphere in the tropics and midlatitudes. On 1.480 you are doing the same, although we did not predict a 40 % global ozone reduction. Comparing the black and red (ScJAMOC) curves in the lower right panel of Fig. 2, O3 production appears to be strongly reduced indeed. Even the results for ScSTa in Fig. 2 show a substantial reduction in O3 production. Further, Lelieveld and Crutzen (1990) introduced the effects of NOx decrease through nighttime heterogeneous loss of N2O5 on cloud droplets. A few years later it was shown that N2O5 is also significantly removed by aqueous aerosols, which moderates the impact of clouds on N2O5, NOx and oxidants predicted by us in 1990.

We agree that our last statement might be misleading. Therefore, we specified our statements in line 40-43 and removed parts of the statement in line 480. It now reads: “The predicted O3 loss by clouds is significantly higher than the global estimates by Liang and Jacob (1997) and regional changes might be in the same order of magnitude as predicted by Lelieveld and Crutzen (1990).”

References


