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Impact of in-cloud OVOC chemistry on tropospheric ozone

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In-cloud aqueous-phase chemistry is known to decrease tropospheric ozone (O_3) via O_3+O_2^- with the major source of O_2^- being hydroperoxyl radicals (HO_2). Therefore, tropospheric O_3 is sensitive to aqueous-phase HO_x (HO_x=HO_2+OH) chemistry. However, most global atmospheric models do not represent this sink reasonably well since they lack explicit representation of in-cloud aqueous-phase chemistry. In this study, a new detailed aqueous-phase mechanism for the oxidation of water soluble oxygenated volatile organic compounds (OVOCs) is developed, suitable for global scale modelling. This improves the representation of aqueous-phase HO_2 and thus the removal of tropospheric O_3. The mechanism focuses on OVOCs containing up to three-carbon atoms. A detailed box-model analysis under low and high NO_x conditions is performed. Afterwards, the developed mechanism is implemented into the global atmospheric model ECHAM/MESSy (EMAC), which is capable to represent the described processes explicitly and integrates the corresponding ODE system with a Rosenbrock solver. EMAC is then used to estimate the global impact of the proposed mechanism with a focus on monsoon systems and biomass burning events. The implemented changes are evaluated using airborne campaign data like OMO for the Asian monsoon. The OVOC oxidation leads to an increase in ozone scavenging and a substantial reduction in tropospheric gas-phase chemical production of ozone. These changes in the free troposphere significantly reduce the modelled tropospheric ozone column, which is known to be overestimated by EMAC and global atmospheric models in general.