The impact of iron distribution among cloud droplets or aqueous aerosol particles on multiphase oxidant levels

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Reactive oxygen species (ROS), such as hydroxyl radical (OH•), hydroperoxy radicals (HO2•/O2-), and hydrogen peroxide (H2O2), are produced in cloud droplets and aqueous aerosol. Multiphase model studies suggest that the Fenton reaction, i.e. the oxidation of Fe(II) by H2O2 represents one of the main sources of the OH radical in the aqueous phase.

Current cloud and aerosol multiphase chemistry models are usually initialized with equal iron concentrations in all droplets or particles as derived from bulk samples of cloud water or aerosol composition. However, analysis of single aerosol particles has revealed that only a small number fraction of particles and, thus, of cloud droplets contain iron.

The aim of our study is to identify the impacts of the iron distribution in cloud droplets or aqueous aerosol particles on the total (gas + aqueous) budgets of OH, HO2, H2O2 and O3 in the multiphase system.

By means of model studies, we compare predicted oxidant budgets based on the assumptions of iron distributed among all droplets or particles versus the same iron mass concentrated in a few droplets (or particles) in the total population only. Our results suggest that the traditional approach based on bulk iron concentrations may significantly underestimate total OH budgets, whereas the predicted levels of H2O2, HO2/O2- and ozone are less affected. The reasons for the different findings between (i) the various oxidants and (ii) cloud droplets vs aerosol particles will be discussed. In summary, our model studies suggest that oxidant levels and oxidation potentials of particulate matter in the atmosphere can only be accurately assessed if particle- and size-resolved aerosol composition is accounted for.