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Regional modelling of the tropospheric multiphase system using COSMO-MUSCAT

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Clouds play a major role in the atmosphere due to their influence on the Earth's radiative budget, on the hydrologic cycle and on the tropospheric chemical composition. Cloud lifetime is driven by the dynamics of the atmosphere at the synoptic scale and, in close interaction, by microphysical processes (e.g. nucleation of cloud droplets and ice crystals, condensation and evaporation, collision/coalescence processes, freezing, sedimentation of hydrometeor) on the small scale.
These processes depend on the chemical composition of particles and cloud droplets. In addition, microphysical processes redistribute chemicals among the various reservoirs: gaseous, particulate, liquid and ice phases. Clouds favor the development of "multiphase chemistry": (1) clouds support very efficient photochemical processes inside droplets; (2) certain homogeneous chemical reactions within clouds can be faster than the equivalent in the gas phase, and reactions such as those involving ionic species, can be important; (3) finally, interactions between the aqueous and solid phase can contribute additionally to chemical processes in clouds (for example dissolution of soluble particulate species).
The coupled model system COSMO-MUSCAT (chemical transport model MUSCAT and the forecast model of the German Weather Service (DWD) COSMO) was extended to consider cloud-chemical processes on the regional scale replacing the former aqueous phase parameterization.
Based on the increasing kinetic and mechanistic knowledge on chemical aqueous phase reactions in the last two decades, advanced aqueous phase chemical mechanisms such as the Chemical Aqueous Phase Radical Mechanism (CAPRAM) are continuously developed. CAPRAM is an almost explicit mechanism which describes relevant chemical aqueous-phase conversions of both inorganic and organic compounds.
With the advanced model system, 2D-sensitivity-studies have been conducted for an urban and a remote case. The comparison of two different mechanisms (simple inorganic and detailed organic mechanism CAPRAM) have revealed agreements but also interesting differences for important chemical subsystems e.g. in the modeled multiphase HO_x budget and pH whereas the simple mechanism leads to always less acidic cloud droplets than CAPRAM. The difference in pH leads consequently to different regimes for the S(IV)-oxidation leading to about 5-10% diffrence in S(VI) between CAPRAM and the simple mechanism.
Additionally reaction flux studies were performed to investigate the cause of the occuring differences of chemical species.