

Modelling multiphase aerosol-cloud processing with the 3-D CTM COSMO-MUSCAT: Application for cloud events during HCCT-2010

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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 (e.g. Ramanathan et al., 2001). 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" since they are an ideal reaction medium for this: (1) clouds support very efficient photochemical processes inside droplets; (2) certain homogeneous chemical reactions within clouds can be usually faster than the equivalent reactions 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 evaluation of multiphase chemistry versus overall tropospheric chemistry and its role in the Earth's radiative budget is challenging since microphysical and chemical processes occurring at different time scales within clouds are still poorly known.

The model system COSMO-MUSCAT consists of MUSCAT (Wolke et al., 2012) and the forecast model of the German Weather Service (DWD) COSMO (Schättler et al., 2014). Both models are coupled online. MUSCAT was extended to consider size-resolved cloud-chemical processes (chemical aqueous phase reactions and phase transfer 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 (Tilgner and Herrmann, 2010). CAPRAM is an almost explicit mechanism which describes relevant chemical aqueous-phase conversions of both inorganic and organic compounds. A reduced version of the mechanism, applicable for 3D chemistry transport models was created (C3.0RED, Deguillaume et al., 2009).

The enhanced model system was applied for case studies connected to the field experiment HCCT-2010 (Hill Cap Cloud Thuringia, 2010, van Pinxteren et al., 2012) that took place in autumn 2010 at Mt. Schmücke, Germany (see Fig. 1). Interesting spatial effects of real clouds on e.g., tropospheric oxidants and inorganic mass have been studied for the first time with a chemistry transport model (CTM) considering an aqueous phase mechanism with the complexity of C3.0RED. The comparison of the model output with available measurements revealed many agreements and also interesting disagreements.

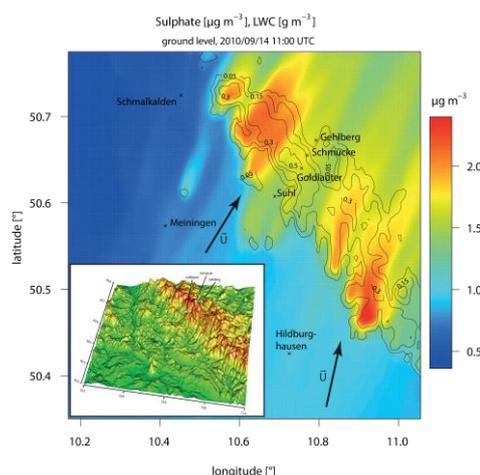


Figure 1. Modelled sulphate mass and topography of the domain (lower left corner).

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