Regional modelling of the tropospheric multiphase system using COSMO-MUSCAT: Sensitivity on detail of cloud microphysics and chemical mechanism

R. Schrödner[1], A. Tilgner[1] and R. Wolke[1]

[1]Institute for Tropospheric Research, Leipzig, 04318, Germany
Keywords: Aerosol cloud interaction, Multiphase chemistry, Modelling (regional), Cloud microphysics.
Presenting author email: roland.schroedner@tropos.de

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., 2004a) and the forecast model of the German Weather Service (DWD) COSMO (Schättler et al., 2008). Both models are coupled online. MUSCAT was extended to consider 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 (Deguillaume et al., 2009).

With the advanced model system, sensitivity-studies have been conducted for urban and remote cases. The comparison of different chemical mechanisms have revealed agreements but also interesting differences for important chemical subsystems e.g. in the modeled multiphase HO_x budget and pH whereas more simple mechanism (without organics) lead to less acidic cloud droplets than CAPRAM. Investigation of reaction fluxes show that this is mostly due to organic acidification in CAPRAM. The difference in pH leads consequently to different regimes for e.g. the S(IV)-oxidation. A simple size-resolved microphysical scheme was introduced which shows the different chemical behavior of the droplet-size-bins.

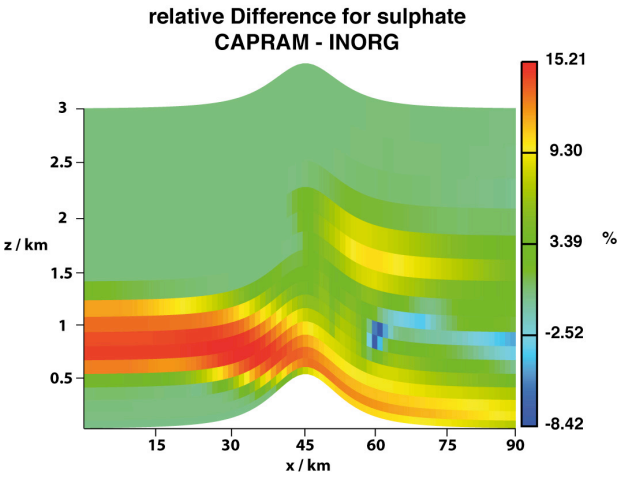

Figure 1. Height profile in x-direction of the simulated mass of S(VI) simulated with CAPRAM compared to the run with simple inorganic chemistry. The cloud is located left of the mountain (red and yellow area). Species stream in from the left-hand side.

This work was supported by the Deutsche Bundesstiftung Umwelt (DBU).

Deguillaume, L., et al. (2010), *Journal of Atmospheric Chemistry*, *64*(1), 1-35.

Ramanathan, V., Crutzen, P. J., Kiehl, J. T. and Rosenfeld, D. (2001) *Science*. 294, 2119 - 2124

Schättler, U., Doms, G. and Schraff, C. (2008): *A Description of the Nonhydrostatic Regional COSMO-Model Part VII: User's Guide.*

Tilgner, A. and Herrmann, H. (2010) *Atmospheric Environment*, *44*(40), 5415-5422

Wolke, R., Knoth, O., Hellmuth, O., Schröder, W. and Renner, E. (2004a) *Parallel computing: Software technology, algorithms, architectures and applications*, 363-370