Modelling the multiphase chemical processing of aerosol constituents in orographic hill cap clouds during HCCT-2010

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Tropospheric clouds and deliquescent aerosol particles are a complex multiphase and multi-component environment with simultaneously occurring gas and aqueous phase chemical transformations. Such aqueous phase chemical processes in cloud droplets are expected to proceed very efficient on short timescales and hence they are able to alter the chemical aerosol composition and the deduced physical properties on a global scale. Besides the physico-chemical aerosol processing, chemical aerosol-cloud interactions have significant effects on the whole multiphase oxidation budget.

In order to improve the still limited understanding of the aerosol-cloud interactions, Lagrangian-type field experiments, where an orographic cloud is used as a natural flow-through reactor, are used for studying such processes in more detail. In Sept./Oct. 2010, the Lagrangian-type cloud passage experiment HCCT-2010 (Hill Cap Cloud Thuringia 2010) was conducted at Mt. Schmücke in Thuringia, Germany to study aerosol cloud interactions (see Figure 1). As known from former cloud passage experiments, particularly associated model investigations (see e.g. Tilgner et al., 2005) including comparisons of model results with observations have considerably contributed to the interpretation of the measured field data.

Figure 1. Schematic representation of the campaign area of the cloud passage experiment HCCT-2010.

In the present study, the parcel model SPACCIM (SPectral Aerosol Cloud Chemistry Interaction Model, Wolke et al., 2005) was applied to investigate the effects of multiphase chemical processing of tropospheric aerosol particles and trace gases resulting from a passage through an orographic cloud at Mt. Schmücke (Germany) during HCCT-2010. The applied model combines a complex microphysical and a detailed multiphase chemistry model with 11381 gas phase and over 3700 aqueous phase reactions. The chemical multiphase mechanism (MCMv3.1 (Master Chemical Mechanism; Saunders et al., 2003)/ CAPRAM3.7 (Chemical Aqueous Phase RAdical Mechanism, modified following Herrmann et al., 2005)) incorporates a detailed near-explicit description of the inorganic and organic multiphase chemistry based on time-dependent size-resolved aerosol/cloud spectra. The measured physical and chemical data at the upwind site provided the basis for the model initialisation under real environmental conditions.

SPACCIM simulations have been carried out for few cloud events, which provided appropriate meteorological and overflow conditions fulfilling the cloud passage experiment requirements. Model results of the cloud passage simulation have been compared with experimental cloud water composition data at Mt. Schmücke (summit site) as well as gas and aerosol measurements at the downwind site in order to interpret the experimental data and to evaluate the model results. To this end, detailed analyses of the chemical multiphase system have been performed including chemical source and sink studies with special emphasis on radical and non-radical oxidants as well as important organic and inorganic chemical subsystems. For the first time, modelled interstitial gas phase in-cloud HOx radical concentrations were compared with in-cloud measurements. Moreover, a central objective of the study was to assess in-cloud oxidations of important C2 and C3 organic compounds. Total organic mass increases after the cloud passage of about 200 ng m⁻³ were modelled. Comparisons between modelled and measured concentrations have revealed good agreements for various measured cloud water constituents. However for organic compounds with low water solubilities measurements have shown considerably higher concentrations than the model results indicating an enrichment in the cloud droplets.

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References