



Multiphase chemical processing by clouds: Modelling the HCCT-2010 hill cap cloud experiment with SPACCIM/CAPRAM4.0 α

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Tropospheric clouds are a complex multiphase and multi-component environment with simultaneously occurring gas and aqueous phase chemical transformations. Such multiphase cloud processes can proceed very efficient on short timescales and can potentially alter the physical and chemical composition and the deduced physical properties on a global scale. Further, 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 by means of field measurements and associated multiphase modelling. In Sept./Oct. 2010, the Lagrangian-type cloud experiment HCCT-2010 (Hill Cap Cloud Thuringia 2010) was conducted at Mt. Schmücke in Thuringia, Germany to investigate aerosol cloud interactions.

The main aim of the present model simulations was to investigate the multiphase chemical processing of aerosol constituents during real orographic clouds and to compare of the modelled and measured data. In detail, the model study focuses on the multiphase chemistry of important oxidants and organic compounds by means of the multiphase chemistry model SPACCIM (SPectral Aerosol Cloud Chemistry Interaction Model). The air parcel model SPACCIM combines a complex cloud microphysical and a detailed multiphase chemistry model with 11381 gas phase and 7118 aqueous phase reactions. The chemical multiphase mechanism (MCMv3.1 (Master Chemical Mechanism)/ CAPRAM4.0 α (Chemical Aqueous Phase RADical Mechanism)) incorporates a detailed near-explicit description of the inorganic and organic multiphase chemistry. 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 selected cloud events (FCEs), 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. For the first time, modelled interstitial gas phase in-cloud HO_x radical concentrations were compared with in-cloud measurements of a FAGE instrument. Model results show substantially reduced gas phase HO₂ radical concentrations under in-cloud conditions (<-90%) and increased H₂O₂ gas phase concentrations after the modelled cloud passage. Modelled in-cloud HO₂ gas phase concentrations at the summit are approx. 2 times higher than the average concentration measured by FAGE instrument at summit site. Moreover, the simulation results show both increased and decreased organic concentrations, e.g. a glyoxalic acid production of about 12 ng/m³ (FCE1.1). According to the organic in-cloud chemistry and the affected phase partitioning, the model simulations show increases in total organic mass of about 0.5 μ g/m³ during FCE1.1 and FCE1.3, respectively. Comparisons between modelled and measured concentrations have revealed good agreements for various measured cloud water constituents. However organic compounds with low water solubilities measurements have shown considerably higher concentrations than the model results.

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