

Chemical aspects of cloud - aerosol interactions

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In the atmosphere cloud droplets form and grow on aerosol particles - tiny solid or liquid particles suspended in the air. When trapped inside water drops aerosol particles are affected by the processes happening in clouds such as collisions between water drops or aqueous phase chemical reactions inside water drops. It can be said that clouds and aerosol particles coexist, continuously changing each others properties. This dissertation studies the interactions between aerosol particles and water drops in warm (i.e. ice free) boundary layer clouds. The main focus is on the collisions between water drops and the aqueous phase oxidation of sulfur to sulfate occurring inside cloud droplets and the impact of those processes on the size distribution of aerosol particles.

The study is carried out using numerical simulations employing a Lagrangian representation of cloud microphysics and aqueous phase chemical reactions. The Lagrangian methods, also called particle tracking or particle based, allow to represent cloud microphysical and chemical processes as if from a point of view of an aerosol particle or a water drop. They are computationally expensive but allow to resolve numerically the changes to both aerosol particle and water drop size distribution.

This study uses the Lagrangian microphysics scheme developed at the Faculty of Physics at the University of Warsaw. For the purpose of this study the Lagrangian scheme was extended to cover also the aqueous phase processes in cloud droplets. The list of chemical processes includes the dissolving of trace gases into water drops their dissociation and the oxidation reaction of sulfur to sulfate by ozone and hydrogen peroxide. Six trace gases are included in the scheme: sulfur dioxide, ozone, hydrogen peroxide, carbon dioxide, ammonia and nitric acid. The scientific software created during this study is available for further use as a part of an open source library of schemes. Parts of this dissertation may serve as the first documentation of the design, performance and the user interface of the created Lagrangian microphysics and aqueous phase chemistry scheme.

The dissertation contains a short theoretical introduction to the microphysical and chemical processes occurring in warm clouds. It presents the details of the Lagrangian representation of microphysics in a numerical scheme. The new module of the Lagrangian scheme responsible for aqueous phase chemical reactions is described and tested in an adiabatic parcel model framework. Next, the created Lagrangian microphysics and aqueous phase chemistry scheme is used in a 2-dimensional kinematic setup representing a vertical slice through a boundary layer topped with a stratocumulus deck. The discussion focuses on the impact of collisions between water drops and the in-cloud oxidation of sulfur to sulfate on the size distribution on aerosol particles. The sensitivity of the obtained results to different microphysical and chemical conditions is also investigated.