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Review of PhD thesis 'Chemical aspects of cloud-aerosol interactions' by Anna Jaruga

This thesis reports the development, testing and application of a numerical scheme representing liquid water-phase chemical reactions in aqueous particles and a statistical formulation of collision-coalescence in a Lagrangian microphysics framework. Such a particle-based description of cloud-related processes is innovative, avoids numerical artifacts that plague conventional schemes (that discretize cloud particles on fixed size grids) and has a great potential for enhanced physically-based analyses of complex, multi-dimensional numerical simulations of low-level clouds. Particle tracking methods have become increasingly popular in the cloud modeling community and need to be more thoroughly studied. Therefore, the challenging topic of the thesis is very timely.

Chapter 1 presents the larger context and summarizes specific goals of the thesis. Chapter 2 provides a succinct introduction into the basic elements of warm (liquid-phase) cloud microphysics and chemistry as needed for this work and provides essential relationships to represent aqueous chemistry in cloud models. Chapter 3 provides details about a particle-based representation of droplet microphysics and heterogeneous chemical processes relevant for tropospheric conditions. Chapter 4 describes the numerical cloud model developed and employed for the first time (in 0-D and 2-D kinematic frameworks) in the thesis. Chapters 5 and 6 present model applications and first scientific results obtained by combining the novel tools with an idealized, low-level (stratocumulus) cloud model, respectively. Appendices A and B complete the thesis editorially, while appendix C provides information regarding code usage.

The scientific part of the thesis investigates how collisions between and chemical reactions within cloud droplets affect the size and mass distributions of aerosols on which the cloud droplets initially formed. Aqueous-phase chemistry is simulated using masses of neutral and ionized chemical species as attributes of simulation water particles ('super droplets'). Concentrations of protons–required to diagnose particle acidity–are determined using species equilibrium and electrical charge neutrality constraints in the dissociation step. Aqueous chemistry calculations in highly concentrated aerosol particles are not performed due to numerical issues; in a cloud, the mass budget affected by those reactions is likely small compared to changes due to reactions within cloud droplets. Adiabatic parcel simulations are used largely for validation and center around effects of in-cloud sulfate

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production; the 2D kinematic framework includes precipitation formation. A number of sensitivity studies around an idealized cloudy 2D-scenario partially constrained by observational data are performed and analyzed to explore systematically the impact of changed conditions regarding drop collision efficiencies, ambient and chemical conditions, and aerosol size distributions. In comparison to results from other numerical models, these studies show the quality of the novel numerical tools on the one hand and reinforce the importance of collisions and in-cloud liquid-phase chemistry in affecting aerosol particle size and mass spectra on the other. While the thesis demonstrates the significance of cloud processing with regard to the onset of precipitation, future studies of longer term impacts of cloud-processed aerosol particles on cloud development and lifetime require the use of more comprehensive driver models with appropriate meteorological boundary conditions and probably a wider set of aqueous oxidation reactions.

Anna Jaruga has mastered with great skill the task of developing, testing and applying a novel numerical scheme representing aqueous-phase chemical reactions in a Lagrangian microphysics framework that includes precipitation formation. She thereby created an efficient and accurate modeling tool that will prove useful in process-based studies of liquid-phase clouds within meteorological models.

The material in the thesis is presented with commendable clarity and conciseness, and its structure is logical and well thought-out. It is especially strong in numerical model development and validation, as this forms the central part. The scientific part is not quite as well developed as I might have hoped, but – given the goals of the thesis – this does not detract too much from my overall positive appreciation.

I anticipate that her model framework, applied in future studies, will help deepening our understanding of the role of clouds in atmospheric chemistry, in particular effects of heterogeneous chemical processing on cloud development. This work is showing her ability to act as a competent member of a large modeling team and share her results in the form of peer-reviewed publications.

I therefore judge this dissertation acceptable for public defense to obtain a Ph.D. degree in Physics at the University of Warsaw.