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Review of the Ph.D. thesis of

Ms Anna Barbara Jaruga

« Chemical aspects of cloud-aerosol interactions »

In her Ph.D. thesis Ms Anna Barbara JARUGA has tackled the difficult task to quantify the growth of aerosol particles due to accumulated mass resulting from aqueous phase oxidation reactions inside cloud drops. Ms JARUGA is focussing here on the sulphate production through uptake of SO_2 into the liquid phase and subsequent oxidation by H_2O_2 and O_3 in warm clouds.

The manuscript is structured into 7 sections. After a short introduction, where Ms JARUGA reminds us of the importance of the size of aerosol particles for cloud processes, she gives in the second section an overview over cloud microphysical processes and the associated aqueous sulphate chemistry. She lists here also the six different combinations of collision kernels and efficiencies that are used for sensitivity studies later in the manuscript. Here, some graphs visualizing the zones of differences of the kernels would have been helpful in order to appreciate the resulting differences in the model results in the subsequent sections.

In section 3 Ms JARUGA explains the numerical implementation of the physics that was presented in the previous section. The scheme selected is quite original, as it presents an alternative to the mostly used bin resolved schemes. Into this already existing "super-droplet" microphysics scheme, Ms JARUGA implements the sulphate chemistry reactions. From own experience the reviewer is aware of the numerical problems that result from the extremely different reactions times involved, creating a stiff numerical system. From this section, we can barely appreciate the enormous amount of coding that must have been involved.

Section 4 introduces the two different dynamics driver that will be used subsequently, a parcel model and a 2D - kinematic setup. The explanation of the library-setup is quite technical and might have been better placed in an annex. However, a presentation of the 2D boundary conditions is missing.

Section 5 presents the results of the parcel model tests. It is a faithful reproduction of the results of Kreidenweis et al (2003). One can regret here, that these tests did not go beyond just reproducing the studies of the Kreidenweis paper. E.g. the CO_2 concentration has increased from the used values of the last century to current values beyond 400 ppm. What would be the impact on the chemistry results?

In section 6 Ms JARUGA coupled the microphysics and chemistry module to an imposed 2-D kinematic driver, and several sensitivity studies were performed. Here, the reader is left somewhat unsatisfied, as in the sensitivity studies differences are stated, however, not really explained. Thus, some questions remain, such as the origin of the differences in the kernel sensitivity studies (see comments above) and why these tests have been integrated into a work on aqueous phase chemistry. This is in particular regretful, as the kernel sensitivity potentially outweighs the differences due to chemical sensitivities. Ms JARUGA then presents the chemical sensitivity studies, identifying the most sensible cases for chemical processing of aerosol particles. She modified e.g. the concentration of NH_3, which is known as a powerful buffering agent in the atmosphere. But did this result, e.g., in a degasing of ammonia due to the dissociation of the ammoniumbisulphate aerosol particles?

The final remarks of section 7, summarizing the work done and the major results, terminate the thesis. The outlook presented, however, is without motivation and lacks enthusiasm.
This is particularly astonishing, as the method presented here provides important potential for new scientific insights. Even within a probably heavy coding work the study could have provided insights beyond what is known already since 20 years. Here, the reviewer regrets that these opportunities were not seized and exploited.
Nevertheless, the reviewer considers that the work done in the framework of this thesis is significant. It required a profound knowledge about cloud physics and chemistry as well as modelling (numerics and techniques of geophysical fluid mechanics). The super-droplet method is quite recent and original and the results reflect a careful procedure as well as numerical protocol, in order to merit the Ph.D. degree. The reviewer encourages Ms JARUGA to defend her work in front of a jury as soon as possible.

Aubière, the 23/03/17

Andrea Flossmann, PR

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