Interactive comment on “Scavenging of ultrafine particles by rainfall at a boreal site: observations and model estimations” by C. Andronache et al.

Anonymous Referee #1

Received and published: 6 July 2006

In this manuscript, the authors try to model the scavenging coefficients of ultrafine aerosol particles calculated from field measurements by Laakso et al. (2003). Generally, the scavenging coefficients calculated taking into account only the below-cloud processes are lower than those estimated from measurements. Therefore, the authors consider into their scavenging model other aerosol sinks as in-cloud removal and activation to enhance the scavenging coefficients. Then, they carried out a sensitivity study of the model to dynamic (fraction of BL mixed into cloud) and microphysical parameters such as aerosol activation fraction, in-cloud efficiency of collection, cloud droplet concentration, intercept parameter of Marshall and Palmer rain distribution. The entrainment of aerosol into cloud and its subsequent activation into cloud condensation nuclei are not explicitly considered in the model, but they are roughly estimated based on pre-
vious experimental studies. The manuscript is interesting because it shows the impact of various parameters on the calculated scavenging coefficients, but it does not explain the values of the scavenging coefficients calculated by Laakso et al. (2003). The scavenging coefficients of Laakso et al. were computed using simultaneous measurements of aerosol and rain at ground, just before and during the rain event. Therefore, the aerosol depletion described by these coefficients is more probably due to downward advection of cleaner air caused by falling droplet than due to upward advection and entrainment in cloud. Can the authors describe the mechanism that move the aerosol from the surface to cloud base in the downdraft outflow region of the cloud where the measurements are performed? Vertical transport, aerosol activation and in-cloud scavenging are definitely important processes for large scale modeling of aerosol removal, but they are not taken into account appropriately in the present closure study. The zero dimensional scavenging model used in this manuscript is adequate for more detailed investigations of the impact of precipitation and aerosol microphysics. For example, the parameterization of the raindrop size distribution accounts for an important part of the discrepancy between the measured and calculated scavenging coefficients (Figs. 12 and 13 here). Therefore, given the lack of experimental data on the raindrop size distribution, a more extensive investigation of the sensitivity of scavenging coefficients to other parameterizations than Marshall Palmer is needed. I also suggest the addition of other two collecting mechanisms for aerosol particles as thermophoresis and diffusiophoresis since all the scavenging coefficients calculated in this manuscript for aerosol particles smaller than 100 nm are much lower than those observed. Chate et al. (2005, Atmos. Environ) have shown that the phoretic forces can increase substantially the scavenging coefficients for particles of this size. Another concern regards modeling of aerosol activation and in-cloud scavenging. To the understanding of this referee, first term in the eq. 3 says that the fraction of aerosol particles activated in cloud \(f_2 f_1 n(dp)\) are further removed due to in-cloud collection of cloud droplets by falling drops \(LIC_{coll}\). Which is the reason to consider this process that is not directly involved in the aerosol balance? The aerosol disappearance due to collection on cloud
droplets can be compensate by aerosol release due to evaporation of cloud droplets. Why is applied the in-cloud coagulation of UFP and cloud droplets to all aerosol particles entering the cloud and not only to the fraction \( f_1(1 - f_2) n(dp) \)? I also suggest to the authors to eliminate all the information already published in Laakso et al. (2003) (for example, at the beginning of Section 2, the description of the site and measurements) and to focus more the discussions on the purpose of this study: modeling the scavenging of ultrafine particles by rainfall. I think that the manuscript worth publication if the authors will change it to include the answers at the above comments.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 3801, 2006.