Atmos. Chem. Phys. Discuss., 6, S2269–S2273, 2006
www.atmos-chem-phys-discuss.net/6/S2269/2006/

Atmospheric
Chemistry
and Physics
Discussions

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

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Received and published: 3 August 2006

We thank the reviewer for many useful comments and suggestions that improved the revised paper. Based on reviewer's comments we made the changes given below to the revised manuscript.

Abbreviations used: A - Authors R - Reviewer

R: Can the authors describe the mechanism that moves the aerosol from the surface to cloud base in the downdraft outflow region of the cloud where the measurements are performed?

A: The revised paper presents a better description of the role of mixing and microphysical processes that can contribute to observed rates of aerosol scavenging. Concerning the mechanism by which aerosol particles (and other tracers) are lifted through the boundary layer to reach cloud base, we have these clarifications:

(a) While the measurements are made at a fixed point near the ground during 1-2 hours, the cloud system above the boundary layer is advected horizontally with a speed of about 10-20 m/s (for this discussion we use some typical values). Thus the cloud system can travel over 100 km during the sampling of one rain event, which implies that above the sampling points we have many individual cloud elements passing, and rainfall intensity varies as well. If someone measures or simulates the instantaneous rainfall rate R, this will vary significantly during the passage of various clouds, a reflection of the fact that some clouds are more convective, R is intense in updraft regions of individual clouds and R is very small or negligible in spaces where downdrafts are present. These features are evident in radar data as well as in mesoscale models that describe frontal systems with precipitation. Whereas for an observer at the ground the rain appears as a continuous process from a single cloud entity, empirical evidence suggest that a rain observed at a fixed point is the result of passage of many clouds that form a mesoscale structure with significant fine scale variations (updrafts and downdrafts);

(b) The existence of clouds over a mesoscale area is due to large-scale low-level convergence of the wind field. This convergence forces air, moisture and CN to be lifted, enabling the cloud to form, grow, and eventually reach the stage of precipitation. (There are some exceptions from this mechanism, such as for orographic and breeze clouds). For frontal systems with precipitation, large-scale (a scale of over 100 km) convergence creates an average ascent that maintains the cloud system. Within such system, there are some intense updrafts and downdrafts, but the vertical velocity averaged over the large-scale area is upward. This is most evident in simulations with 3D cloud models or in mesoscale models of frontal systems. In the revised paper, we review some representative work in this area and present a justification of our assumption that f1 = 0.1 (the net fraction of BL aerosol that reaches cloud base). Some relevant work by Niewiadomski (1986), Tremblay (1987) and Agusti-Panareda et al. (2005) is described. We note also that Laakso et al. (2003a) (denoted as ‘L2003a’ in this discussion) presented the criteria for data selection that can eliminate factors such as significant vertical mixing and dilution. Wet scavenging and vertical mixing were considered as remaining mechanisms in the analyzed data. Clearly, more work is needed in future studies to determine the contributions, from various mechanisms involving vertical transport, to measurements made near the ground.

R: 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.

A: We illustrate the model sensitivity to the choice of raindrop size distribution (RSD), by performing extra runs with a Gamma-function for the RSD. Parameters reported in literature (Zhang et al. 2001) are applied in the Gamma-function. This extra run is compared with results from the run using the Marshall and Palmer (MP) RSD. We note that the MP RSD overestimates the number of very small and very large raindrops. Zhang et al. (2001) determined three parameters of the Gamma-function from radar measurements: reflectivity, differential reflectivity and a constrained relation between the shape and slope parameters derived from disdrometer observations. Values of the shape parameter of the RSD in these extra runs are: 2, 3 and 4. For these values of shape parameter, predicted values of Leff are compared with Lo in a new figure. Comparisons with calculations based on the MP RSD demonstrate that the run with the Gamma-function produces higher values of the scavenging coefficient. Overall, variations in the RSD function and its parameters are a source of significant uncertainty in evaluation of the scavenging coefficients.

R: 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.

A: Thermophoretic and diffusiophoretic effects were included in the scavenging coefficient, and the sensitivity to the temperature difference between air and raindrop surface (Ta - Ts) is shown. Generally, a difference of 1-3 degC is typical, while in deep convective precipitation the difference can be higher and phoretic effects are enhanced, as shown by the sensitivity analysis. The magnitude of Ta - Ts is largely uncertain and can vary significantly from case to case.

R: 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 (f1 f2 n(dp)) are further removed due to in-cloud collection of cloud droplets by falling drops (LICColl). 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 compensated 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 f1(1- f2) n(dp) ?

A: In this model, LICColl is the rate of in-cloud removal of droplets due to collection by raindrops. It allows an estimation of how much of f1 f2 n(dp) is removed after such particles are in cloud droplets. We agree with the reviewer that some cloud droplets can evaporate, releasing aerosol back into the atmosphere, most of them will be evaporated above the BL. Some will be mixed back in the BL. We do not have an estimation of the fraction of droplets that evaporate and which are mixed back into the BL during a rain event. The overall effect of cloud droplet evaporation is to diminish the fraction of cloud droplets that are collected by raindrops. The sensitivity of Leff to EIC is used to illustrate what the overall effect of this process can be. For the case of very low values of EIC, we find that Leff is significantly reduced, especially for dp larger than 100 nm. We corrected the above problem related to in-cloud coagulation of UFP, which is now applied only to f1(1-f2)n(dp).

R: 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.

A: All information published already by L2003a (such as the material at the beginning of Section 2, with the description of the site and measurements) has been eliminated. That section now contains new information that was not presented by L2003a. We have also added more elements to the discussion of the dependence of scavenging coefficients on several other factors as mentioned above (phoretic effects, dependence on RSD function, and changes of UFP size distribution due to wet scavenging). The paper is now focused better on processes that can impact the scavenging of aerosol by rainfall.

References

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