Atmos. Chem. Phys. Discuss., 9, C6996–C6998, 2009
www.atmos-chem-phys-discuss.net/9/C6996/2009/

Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Secondary organic material formed by methylglyoxal in aqueous aerosol mimics – Part 1: Surface tension depression and light-absorbing products" *by* A. N. Schwier et al.

A. N. Schwier et al.

vfmcneill@columbia.edu

Received and published: 10 November 2009

We thank Professor Laaksonen for his constructive comments.

. . . I believe that the estimate of critical supersaturation lowering due to decreased surface tension has not been carried out in a way that would reflect current understanding, and the authors should redo the exercise for the final paper. In small droplets, surface active organics will be highly concentrated in the surface layer. At the same time, they are depleted from the bulk of the droplet, which causes the Raoult term to become

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

smaller, counteracting the surface tension influence on critical supersaturation. The depletion of the surfactant also causes the surface tension itself to increase. This "surface partitioning" effect, which follows from Gibbs' surface thermodynamics and finite size of the droplet, has been discussed in a number of recent papers. Lab experiments (Sorjamaa et al., Prisle et al.) have provided strong evidence that the effect is real, and usually leads to much smaller depression of critical supersaturation than what is estimated based on simple Köhler theory that doesn't account for surface partitioning. I recommend that for the final paper, the authors provide an estimate of the critical supersaturation lowering that accounts for surface partitioning. A rough estimate for 100 nm particles might be possible using Fig. 2 of Sorjamaa and Laaksonen (2006).

You raise excellent points for discussion. In a heterogeneous SOA formation scenario the bulk solute content of the aqueous aerosol particle is expected to be dominated by salt. For example, at 60% relative humidity an ammonium sulfate particle is predicted to contain 65 wt% $(NH_4)_2SO_4$ (Tang and Munkelwitz, 1994), whereas methylglyoxal was found to comprise roughly 0.005-0.05% of aerosol mass in urban environments (Kawamura and Yasui, 2005; Ho et al., 2007). Furthermore, gas-phase methylglyoxal will enter the particle through heterogeneous uptake at the aerosol surface and may never leave the surface region because of its surface-active nature, and that of its reaction products. Therefore we feel that for this system it is reasonable to assume that the loss of solute from the bulk due to surface partitioning is negligible and the effect of methylglyoxal and its reaction products on equilibrium CCN activity is purely surface tension based. We have modified the manuscript to include a discussion of these issues and a citation for Sorjamaa et al. (2004).

REFERENCES

Sorjamaa,R.; Svenningsson, B.; Raatikainen, T.; Henning, S.; Bilde, M.; Laaksonen, A. The role of surfactants in Kohler Theory reconsidered. Atmos. Chem. Phys. 2004, 4, 2107-2117.

Ho, K. F., Cao, J. J., Lee, S. C., Kawamura, K., Zhang, R. J., Chow, J. C., and Watson, J. G.: Dicarboxylic acids, ketocarboxylic acids, and dicarbonyls in the urban atmosphere of China, J.Geophys. Res.-Atmos., 112 (D22), 2007.

Kawamura, K. and Yasui, O.: Diurnal changes in the distribution of dicarboxylic acids, ketocarboxylic acids and dicarbonyls in the urban Tokyo atmosphere, Atmos. Environ., 39 (10), 1945-1960, 2005.

Tang, I. N. and Munkelwitz, H. R.: Water Activities, Densities, and Refractive-Indexes of Aqueous Sulfates and Sodium-Nitrate Droplets of Atmospheric Importance, Journal of Geophysical Research-Atmospheres, 99 (D9), 18801-18808, 1994.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 15541, 2009.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper