Atmospheric Chemistry and Physics
An Interactive Open Access Journal of the European Geosciences Union

Atmos. Chem. Phys., 4, 2107-2117, 2004
www.atmos-chem-phys.net/4/2107/2004/

The role of surfactants in Köhler theory reconsidered

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Abstract. Atmospheric aerosol particles typically consist of inorganic salts and organic material. The inorganic compounds as well as their hygroscopic properties are well defined, but the effect of organic compounds on cloud droplet activation is still poorly characterized. The focus of the present study is the organic compounds that are surface active i.e. tend to concentrate on droplet surface and decrease the surface tension. Gibbsian surface thermodynamics was used to find out how partitioning between droplet surface and the bulk of the droplet affects the surface tension and the surfactant bulk concentration in droplets large enough to act as cloud condensation nuclei. Sodium dodecyl sulfate (SDS) was used together with sodium chloride to investigate the effect of surfactant partitioning on the Raoult effect (solute effect). While accounting for the surface to bulk partitioning is known to lead to lowered bulk surfactant concentration and thereby to increased surface tension compared to a case in which the partitioning is neglected, the present results show that the partitioning also alters the Raoult effect, and that the change is large enough to further increase the critical supersaturation and hence decrease cloud droplet activation. The fraction of surfactant partitioned to droplet surface increases with decreasing droplet size, which suggests that surfactants might enhance the activation of larger particles relatively more thus leading to less dense clouds. Cis-pinonic acid-ammonium sulfate aqueous solutions were studied in order to study the partitioning with compounds found in the atmosphere and to find out the combined effects of dissolution and partitioning behavior. The results show that the partitioning consideration presented in this paper alters the shape of the Köhler curve when compared to calculations in which the partitioning is neglected either completely or in the Raoult effect. In addition, critical supersaturation was measured for SDS particles with dry radii of 25-60nm using a static parallel plate Cloud Condensation Nucleus Counter. The experimentally determined critical supersaturations agree very well with theoretical calculations taking the surface to bulk partitioning fully into account and are much higher than those calculated neglecting the partitioning.

Final Revised Paper (PDF, 307 KB) Discussion Paper (ACPD)

Citation: Sorjamaa, R., Svenningsson, B., Raatikainen, T., Henning, S., Bilde, M., and Laaksonen, A.: The role of surfactants in Köhler theory reconsidered, Atmos. Chem. Phys., 4, 2107-2117, 2004. Bibtex EndNote Reference Manager