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Importance of surfactant representations for predicted cloud droplet numbers using the ECHAM5.5-HAM2 aerosol-climate model

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The indirect radiative effects of atmospheric aerosols via their influence on cloud drop formation and cloud properties constitute the single largest uncertainty in predictions of global climate and future climate changes ipcc:2007. Cloud condensation nucleus (CCN) activity is determined by both aerosol particle size and chemical composition and in particular the CCN activity of organic aerosols remains to be firmly constrained hallquist:acp:2009. This in part owes to the surface activity (the tendency to preferentially accumulate in the surface region) in aqueous solutions of some of the organics comprising atmospheric aerosol particles. Surface active molecules (surfactants) have been demonstrated in atmospheric aerosol and cloud and fog water samples from marine, and rural and urban/polluted continental environments mochida:jgr:2003b and their aqueous extracts asa-awuku:acp:2008, and can collectively comprise a significant fraction of the organic aerosol mass.

Thermodynamic model calculations, as well as laboratory experiments, show that surface activity can significantly affect organic aerosol CCN potential prisle:acp:2010. The combined effect of partitioning of organic molecules to the droplet surface and reduced droplet surface tension for calculated cloud droplet activation can be determined from numerical solutions to thermodynamic relations: Gibbs adsorption equation gibbs:book:1928 and the Köhler equation for droplet growth at equilibrium conditions kohler:trans:faraday:soc:1936. Unfortunately, such calculations with several nested iterations are too computationally demanding for implementation into atmospheric models kokkola:grl:2006. As an alternative, topping:gmd:2010, raatikainen:gmdd:2010, and prisle:acpd:2010 have presented different parameterizations to account for the combined effects of surfactant properties in Köhler calculations for organic aerosols.

We compare predictions of cloud droplet number concentrations and radiative forcing, using the aerosol-climate model ECHAM5.5-HAM2 stier:acp:2005 with different representations of the aerosol organic carbon fraction in terms of the influence of surfactant properties on cloud microphysics. Specifically, we use the parameterizations of topping:gmd:2010, raatikainen:gmdd:2010, and prisle:acpd:2010, which account for the combined organic surfactant effects, in addition to the previously applied approaches using either reduced droplet surface tension without consideration of organic surface partitioning effects, or disregarding surfactant properties altogether prisle:acp:2010. The chemical compounds included in aerosol model HAM2 are: sulfate, black carbon, organic carbon, sea salt, and mineral dust. The aerosol microphysical processes are calculated within the modal aerosol microphysics module M7 vignati:jgr:2004, which is coupled to HAM2. The number of activated cloud droplets are calculated using the cloud activation parameterization by abdul:jgr:2000, which has here been extended to account for effects of organic surface activity by implementing surfactant surface partitioning and droplet surface tension according to each of the different approaches mentioned.

Our preliminary results show that including the surfactant effects on cloud microphysics can have a significant effect on predicted cloud droplet numbers, and thus radiative forcing, compared to assuming simple Köhler theory that disregards such effects altogether. This is especially the case in areas of high organic aerosol mass concentrations. These results are the first to include effects of detailed surfactant properties in global scale simulations, and somewhat contradicts the anticipation that the non-linear responses predicted from thermodynamics to a combination of several surfactant effects on droplet activation would largely cancel out in global scale predictions. Of the three parameterizations applied, which take the combined surfactant effects into account, that of prisle:acpd:2010 is readily implemented to the ECHAM5.5-HAM2 framework, whereas those of topping:gmd:2010 and raatikainen:gmdd:2010 still require one additional iterative scheme to obtain the droplet critical supersaturations. The former is therefore significantly less computationally demanding than the latter two.

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