# Surfactant effects in global simulations of cloud droplet activation

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[1] Atmospheric aerosols often contain surface active organics. We study the influence of these surfactants on predictions of particle cloud activation potential and aerosol indirect climate effects, by implementing different parametrizations of surfactant effects in the global circulation model ECHAM5.5–HAM2. A parametrization based only on droplet surface tension reduction produces significantly larger effects on predicted cloud droplet numbers than novel parametrizations based on detailed considerations of organic surface activity. It seems better to disregard surfactant effects altogether than employing parametrizations accounting only for effects on surface tension. We strongly recommend not using only the surface tension reduction to represent the surfactant effects in climate models. **Citation:** Prisle, N. L., et al. (2012), Surfactant effects in global simulations of cloud droplet activation, *Geophys. Res. Lett.*, *39*, L05802, doi:10.1029/2011GL050467.

## 1. Introduction

[2] Indirect radiative effects of atmospheric aerosols via their influence on cloud drop formation and cloud properties still account for major uncertainties in predictions of global climate and future climate change [*Intergovernmental Panel on Climate Change*, 2007]. In particular, cloud condensation nucleus (CCN) activity of the organic aerosol remains to be firmly constrained [*Hallquist et al.*, 2009]. Surface active organic molecules (surfactants) have been observed in atmospheric aerosol and cloud and fog water samples [*Cheng et al.*, 2004] and their aqueous extracts [*Dinar et al.*, 2006] from many types of environments and can collectively comprise a significant fraction of the organic aerosol mass.

[3] Surfactants concentrate in the surfaces and can reduce the surface tension of aqueous solutions, such as activating cloud droplets. Reduced surface tension at the point of droplet activation would enhance CCN activity by suppression of the droplet activation threshold [*Shulman et al.*, 1996], which could ultimately lead to a cooling effect [*Facchini et al.*, 1999]. Sub-micrometer solution droplets can also experience significant depletion of dissolved surfactants from the bulk phase due to surface partitioning,

leading to increased equilibrium water vapour pressure over the droplet surfaces [*Sorjamaa et al.*, 2004]. The combined effect on droplet activation of surfactant surface partitioning and surface tension reduction in aqueous solution, is highly non-linear as a function of aerosol size and composition and thus non-trivial to predict. The surface partitioning equilibrium and surface tension of a single activating cloud droplet can be determined from numerical solutions to thermodynamically consistent relations [*Prisle et al.*, 2008, 2010]. Such calculations are however computationally too demanding for implementation to larger scale models [*Kokkola et al.*, 2006]. In global climate models, surfactant effects on cloud droplet activation have therefore traditionally not been considered at all.

[4] *Abdul-Razzak and Ghan* [2000] presented a parametrization to treat multiple aerosol types in a cloud activation scheme and *Abdul-Razzak and Ghan* [2004] also included effects of surfactants on the surface tension of activating droplets, neglecting any direct influence of bulk-phase depletion on droplet water activity. Only recently have different parametrizations been developed to comprehensibly account for the combined effects of both surface partitioning and surface tension on cloud droplet activation of organic aerosols [*Topping*, 2010; *Prisle et al.*, 2011; *Raatikainen and Laaksonen*, 2011]. These parametrizations are based on different assumptions and approximations at the detailed process level description of surfactant surface partitioning [*Gibbs et al.*, 1928] and droplet activation from Köhler theory [*Köhler*, 1936], and consequently have somewhat different scope and applications.

[5] We investigate the significance of including surfactant properties of organic aerosol in global scale simulations of cloud droplet numbers and cloud radiative forcing (aerosol indirect climate effects), and the level of microphysical detail required in the representation to yield significant differences in such predictions. To our knowledge, this is the first time that comprehensive surfactant effects have been implemented in a climate model, and thus the first comparison of differences in modelled global cloud droplet concentrations using the novel parametrizations of detailed surfactant CCN activity and evaluating their climate relevance.

## 2. Surfactant Properties Parametrizations

[6] For the purposes of this work, we have extended the cloud activation parametrization of *Abdul-Razzak and Ghan* [2000] to account for effects of organic surface activity on critical supersaturations of different particle sizes, according to four different representations (see Figure 1): Traditional equilibrium Köhler theory, disregarding surfactant surface partitioning and assuming the constant surface tension of

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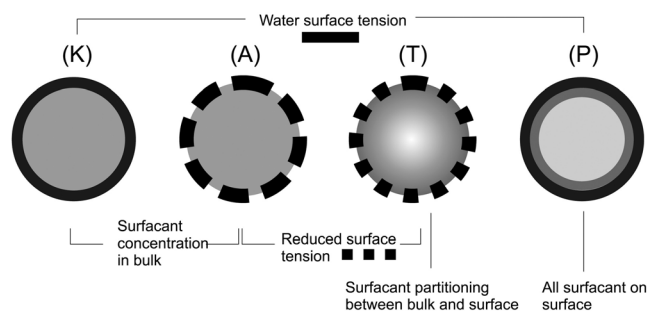
**Figure 1.** Overview of the different surfactant CCN activity parametrizations used in this work.

pure water **(K)**; Reduced surface tension in activating droplets according to total surfactant concentration, following the Szyskowski equation [*Szyskowski*, 1908] **(A)**; Parametrization by *Topping* [2010]: The extent of surfactant surface partitioning is calculated according to dry particle composition. Droplet surface tension is reduced according to surfactant concentration remaining in the bulk, following the Szyskowski equation **(T)**; Parameterization of *Prisle et al.* [2011]: The surfactant is assumed to be completely partitioned to the droplet surface, such that surfactant mass does not affect either concentration or surface tension of activating droplets, essentially treating the organic particle fraction as an insoluble, fully wettable, core **(P)**.

[7] Parametrization **(A)** does not explicitly account for the potentially significant bulk-phase depletion of dissolved surfactant in sub-micron activating droplets [*Prisle et al.*, 2010]. The **(T)** parametrization considers both bulk-phase depletion of surfactant and surface tension reduction in activating droplets, whereas **(P)** and **(A)** include only either bulk-phase depletion or surface tension effects, respectively. Evaluating the droplet surface tension according to total surfactant concentration in the droplet phase, as for a macroscopic solution of corresponding overall composition, can lead to severe overestimations of CCN activity. Furthermore, when surfactant surface partitioning is taken into account, the effect on bulk-phase depletion appears to dominate that of any surface tension reduction [*Sorjamaa et al.*, 2004; *Prisle et al.*, 2008, 2010]. We thus consider parametrizations **(T)** and **(P)** to yield the most thermodynamically consistent representations of surface active organic aerosol in CCN activation. The cloud activation scheme of *Abdul-Razzak and Ghan* [2004, hereinafter ARG2004] accounts for the influence of bulk-phase depletion on resulting droplet surface tension, but not for the direct implications for droplet water activity. Test calculations made using a cloud parcel model, for selected aerosol compositions and size distributions and updraft velocities, show a relative order of activated aerosol fractions as **(A)** > ARG2004 > **(T)** > **(K)** > **(P)** (see auxiliary material).[1] This trend is expected, but may however not be universal for all surfactant types and aerosol systems [*Prisle et al.*, 2011]. The largest differences between different surfactant representations are furthermore expected for conditions

of high organic aerosol mass fraction and high critical supersaturations.

## 3. Experiments

### 3.1. ECHAM5.5 Climate Model

[8] Global simulations were made using an experimental version of aerosol-climate model ECHAM5.5–HAM2 (P. Stier et al., Mechanistic aerosol–cloud coupling and indirect aerosol effects in the aerosol-climate model ECHAM–HAM, manuscript in preparation, 2012) consisting of fifth generation general circulation model ECHAM5.5 [*Roeckner et al.*, 2003] with aerosol model HAM2 (K. Zhang et al., manuscript in preparation, starting in 2011), developed from earlier HAM [*Stier et al.*, 2005]. Aerosol microphysical processes are calculated within the modal aerosol microphysics module M7 [*Vignati et al.*, 2004], coupled to HAM2, and aerosol–cloud interactions are included by a double-moment cloud microphysics scheme [*Lohmann et al.*, 2007].

[9] The activated cloud droplet number concentration (CDNC) was calculated using the cloud activation parametrization of *Abdul-Razzak and Ghan* [2000]. The parametrization estimates the maximum supersaturation according to the critical supersaturations of individual aerosol modes, while accounting for kinetic effects of condensation, using aerosol size distribution, aerosol composition, and updraft velocity. With representation **(K)**, where surfactant surface partitioning and droplet surface tension effects are not considered, droplet critical radius and supersaturation can be calculated directly. Representation **(P)** is implemented analogously, by subtracting the surfactant mass from the droplet solute, and no further iterations are required. When droplet surface tension is reduced, using **(A)** and **(T)**, critical supersaturations cannot be solved analytically and are iterated by the Newton–Raphson method, using the critical radii from **(K)** as starting values.

[10] We did separate 5-year experiments with each of the four representations of surfactant CCN properties. All simulations were conducted with prescribed meteorology and emission fields for the years 1998–2002, with a spin-up period of 3 months. Model meteorology was nudged towards ECMWF ERA Interim re-analysis data [*Dee et al.*, 2011] to keep conditions as similar as possible between individual runs. Model resolution in the simulations was T63L31.

[11] The entire aerosol organic carbon fraction is assumed to be surface active, with properties corresponding to Suwannee River Fulvic Acid (SRFA). SRFA is a reference compound that is frequently invoked as a simple proxy for surface active humic-like atmospheric organics; Szyskowski-type parametrizations of aqueous surface tensions are already available [*Topping et al.*, 2007, and references therein], as needed input for cloud activation parametrizations according to representations **(A)** and **(T)**.

### 3.2. Data Analysis

[12] In the comparison of different surfactant CCN activity scenarios, the traditional Köhler representation **(K)** was selected as base case, since it is the approach most commonly used in global modelling. We calculated normalized median differences (*D*) of monthly column mean simulated properties (*C*) between each experiment and the base case (subscript K) as $D(i,j) = \left( \widehat{C(i,j)} - \widehat{C_K(i,j)} \right) / \widehat{C_K(i,j)}$,
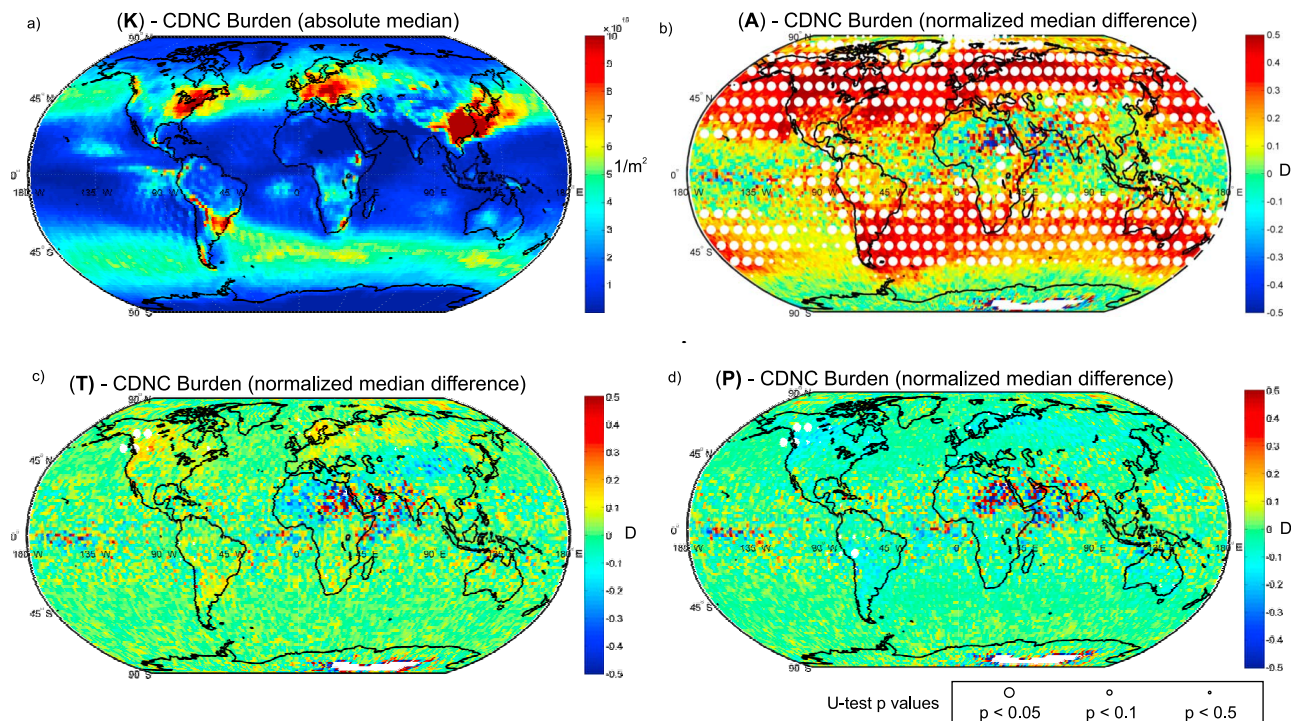
**Figure 2.** CDNC column burden differences between simulations with different surfactant CCN activity parametrizations: (a) 5-year medians in (K) simulation in absolute units, total column CDNC [m$^{-2}$], (b) normalized median differences of (A) and (K) simulations, (c) normalized median differences of (T) and (K) simulations, and (d) normalized median differences of (P) and (K) simulations. Sizes of the white dots indicate p-values of the U-test comparison to the (K) simulation in each case; p-values are only shown in areas with p < .5.

where $\widehat{C(i,j)}$ is the 5-year median of the quantity in question for the simulation horizontal coordinates $(i, j)$. For properties where monthly mean column value could be zero, the differences were calculated from non-zero values only. We also estimated the significance of differences between model experiments using the non-parametric Mann–Whitney U-test, comparing the property distributions of experiments in each column cell with the respective (K) representation distribution.

## 4. Results and Discussion

[13] This work focuses on predicted cloud droplet numbers and cloud radiative forcing. Selected results of the 5-year simulations are presented here, and in the auxiliary material.

[14] Figure 2 shows the modelled median CDNC burden values of the simulations. Figure 2a shows the five year median CDNC burden (in m$^{-2}$) of base case (K). Most of the CDNC burden is clearly concentrated in the regions of major anthropogenic emissions in North America, Europe and South-East Asia. CDNC burden is also pronounced over the oceans in the mid-latitudes. The overall mean value of CDNC burden in the (K) experiment was $2.9 \cdot 10^{10}$ m$^{-2}$. Figure 2b shows the normalized median differences between experiments (A) and (K). The (A) experiment generates very high CDNC burdens, especially over the mid-latitude oceans and northern continental areas. The increase over the continental source areas is over 50%, with a global mean increase of 24%. U-test results show that the increases almost everywhere exceed the normal variability of the CDNC burdens. In contrast, Figures 2c and 2d show similar median

differences for parametrizations (T) and (P), respectively. Even though (T) shows somewhat increased, and (P) decreased, global CDNC burdens (mean global differences of 1% and −3%), compared to (K), the differences are numerically much lower than in the case of (A) and the U-test does not show any statistically significant changes outside of a few scattered areas in the Northern boreal zone and South America. Over the oceans, the CDNC burden changes from the base case are minimal.

[15] CDNC cloud top concentrations display overall similar patterns as CDNC burdens, as seen in the median difference maps (auxiliary material), where only (A) has statistically significant increases over (K), especially over oceans.

[16] Surface partitioning parametrizations use aerosol organic fraction as one of the main parameters. Differences in organic fractions between different experiments were negligible. Mean column organic mass fractions over North America, Europe and South East Asia are low (0.1 to 0.4) in the accumulation mode and medium (0.4 to 0.8) in the Aitken mode, in contrast with high mass fractions over tropical continental areas in both modes (see auxiliary material). Notably low are the mass fractions of organics over the oceans, which stems from a lack of organic emissions from the sea surface within the current model framework.

### 4.1. Radiative Forcing and Climate Impact

[17] *Facchini et al.* [1999] estimated the potential magnitude for the effect of organic aerosol surface activity on cloud droplet numbers and resulting cloud radiative forcing, based on surface tensions measured in ambient precipitation samples conditioned to concentrations relevant for CCN

activation. In their considerations, a 30% decrease in droplet surface tension at the point of activation leads to a 20% increase in droplet numbers and, as an upper bound, a change in cloud radiative forcing of $-1$ W/m$^2$. To our knowledge, the present results are the first actual climate model predictions to include comprehensive surfactant effects of organic aerosol on cloud activation. Comparisons of short- and long-wave radiative fluxes showed much smaller variations between different experiments than CDNC concentrations. This is partly due to many other factors influencing the radiative balance besides cloud activation. In our experiments, the 5-year mean shortwave cloud radiative forcing (SWCRF) changed from the **(K)**-case value of $-46.7$ W/m$^2$ by $-1.27$ W/m$^2$ (3% of total SWCRF) in **(A)**, $-0.08$ W/m$^2$ (0.2%) in **(T)**, and $+0.17$ W/m$^2$ ($-0.3$%) in **(P)** parametrizations. Experiment **(A)** shows good agreement with the earlier estimate based on an analogous representation of surfactant CCN activity [*Facchini et al.*, 1999]. The results from representations **(T)** and **(P)** both suggest a significantly weaker overall effect of organic aerosol surface activity. It should be noted that only the first aerosol indirect effect was in practice included in our results, as the simulations were nudged to follow the same dynamics. The nudging is not done for cloud water and thus its values for different runs are not identical, but very close. The total effect could be larger if the cloud cover was allowed to change freely.

### 4.2. Implications for Global Modelling

[18] Results of this work demonstrate that surface activity of organic aerosol and its representation in cloud activation can have significant influence on global scale predictions, even though the effect may be limited to specific regions. Surfactant representation **(A)**, which only includes effects of reduced surface tension according to the total droplet concentration, gives strong and statistically significant differences from the conventional approach of disregarding surface activity altogether **(K)**, even in areas with relatively low organic aerosol content. On the other hand, parametrizations based on detailed considerations of both surface partitioning and surface tension effects on droplet activation (**T** and **P**) both give overall very similar predictions of CDNC to the base case. This clearly underscores how extending the climate model framework to account for the influence of surface tension on organic aerosol CCN potential, according to total surfactant concentration, may lead to significant overestimations of global cloud droplet numbers and cloud radiative forcing.

[19] *Reutter et al.* [2009] found that activated cloud droplet numbers in a cloud parcel model were not very sensitive to variations in aerosol chemical composition, due to buffering from meteorological feedbacks. We do also see evidence of buffering mechanisms dampening any influence of surface activity occurring at the microphysical process level, as the relative sensitivity of predicted radiative forcings to surfactant representations is diminished, compared to cloud droplet numbers. We do, however, observe strongly significant differences between experiments using different surfactant representations, especially upon application of representation **(A)**, showing that potential effects of organic surface activity are not entirely buffered by the many processes coupled within the climate system. The relatively close similarities of experiments **(T)** and **(P)** with the base case **(K)**, are *not* merely caused by feedback mechanisms buffering any perturbations introduced by using the different surfactant parametrizations, but also by cancellation effects in the microphysical process description [*Prisle et al.*, 2010].

[20] Cancellation of different surfactant effects on cloud activation may be a consequence of the choice of proxy for the organic aerosol fraction. We here assumed that this entire fraction is surface active with properties corresponding to SRFA. Although a rather extreme case in terms of organic surfactant properties, this does not necessarily translate into a correspondingly extreme case in terms of aerosol CCN activity, let alone in globally predicted cloud droplet numbers. Other combinations of organic aerosol compositions and surfactant molecular properties should therefore be explored in future studies.

[21] Within the ECHAM5.5–HAM2 framework, organic emissions only occur from land areas and high organic mass fractions are therefore not seen over oceans. Since the radiative effects of changed cloud albedo would be greatest over the oceans, and marine aerosol have been seen to comprise significant fractions of potentially surface active organics [*O'Dowd et al.*, 2004], our results might underestimate the global radiative effects of surfactants.

### 5. Conclusions

[22] For the first time, surfactant effects on cloud activation and cloud radiative properties were tested in a global climate model. Two novel parametrizations based on detailed considerations of surfactant cloud microphysics were implemented in the ECHAM5.5–HAM2 framework, in addition to approaches considering only surface tension reduction from total aerosol surfactant concentration, or disregarding surfactant properties altogether.

[23] The organic surfactant representation using only the effects of reduced droplet surface tension yields large differences from the base case in many parameters studied, especially in the predicted CDNC. Differences are less pronounced for the resulting radiative fluxes, but still large. The parametrizations based on detailed considerations of surfactant properties generally give quite similar predictions of CDNC to each other, and these simulations show much less significant differences from the base case than the representation based on surface tension only. Since these parametrizations are considered to yield the most consistent representations of organic surface activity in CCN activation, using a representation accounting only for effects on surface tension according to total organic concentration therefore cannot be recommended. In particular, if the aim is to avoid chemical complexity of cloud microphysics in global climate modelling, it appears to be better to omit surfactant effects altogether than employing the approach based on total droplet organic concentrations.

[24] The representation of *Prisle et al.* [2011] is chemically less specific, yet more versatile, and computationally easier to implement for large scale modelling, than that of *Topping* [2010]. Predicted cloud droplet numbers and the magnitude of the shortwave cloud radiative forcing are markedly similar in both of these experiments. The similarity of results from these parametrizations to the results obtained using traditional Köhler representation is partly explained by cancellation effects in cloud microphysics.

However, whether this is a general feature of surface active organic aerosol mixtures needs to be investigated further.

[25] The present results demonstrate that taking only the surface tension effects of surfactants into account may lead to erroneous results. Assessment of the actual influence of surfactants on aerosol indirect climate forcing nevertheless awaits further constraints by observations of global cloud droplet numbers and surface active aerosol properties. Measurements to constrain surface tensions for atmospheric organic aerosol mixtures are by no means rendered futile, as complex surfactant effects may influence other aerosol and cloud properties in addition to the immediate droplet activation [*Romakkaniemi et al.*, 2011].

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