

[Comment from Anonymous Referee 2]

General comment:

Authors presented a very-well thought approach to deal with a very complex problem, the CCN activation of surfactant-enriched aerosol particles. The model is presented in a very fluent and clear way, easy to understand for possible users. The sensitivity analysis is performed carefully to assess dependencies to particle size and composition. However, the discussion in section 5 **could benefit from a comparison of model outputs to laboratory measurements of critical supersaturation for similar particle-systems** (e.g. [8-11]).

Even when I understand that this task is challenging due to the scarce data along the size-composition range of atmospheric relevant systems, the statements related to underestimation of CCN activity of ultrafine SSA particles in climate models are too strong without a proper validation of model results. Even if particles with 50 nm-diameter activate at 0.5% supersaturation level in updrafts, they could also deactivate in downdrafts leading to non significant changes in cloud droplet number concentrations at cloud base.

Nonetheless, I recommend the manuscript for publication after addressing the comments due to the completeness of the modelling approach.

Its future implementation in cloud models could bring valuable information about the formation of marine stratocumulus in pristine areas where sea spray emissions from leads can be richer in organic compared to those from open oceans. If statements in this study become proven, it would be necessary to reformulate how SSA emissions are depicted in the marine boundary layer. Even if statements do not hold, being nonactivated, surfactant-enriched SSA could be transported vertically promoting the formation of mixed-phase clouds at higher altitudes in pristine atmospheres. This is particularly important to improve our understanding of the Arctic amplification phenomenon.

[Our answers] We thank the anonymous reviewer for their comments and careful reading.

To better explain why a **validation against critical supersaturation data** from literature has not been included in our study, we added the following text at the end of the section “Uncertainties in the modelling approach”:

“As an additional validation step, future work should be directed at comparing SS_{crit} predicted with the combined model to measurements of SS_{crit} of lab-generated surfactant containing aerosol particles. A comparison to data from literature was not included in this study for two reasons. First, such literature data is very limited, as can be seen from the study by Lin et al. (2018), where the experimental data was not sufficient to draw a conclusion about which of their two models was more accurate. Second, in previous studies, the exact composition of the aerosol particles was not confirmed by a measurement, but taken as the composition of the solution filled into the atomizer. We suggest that a verification of the particle composition after atomization by e.g. an aerosol mass spectrometer is urgently needed for a reliable comparison to modelled SS_{crit} values. To our knowledge, no study has yet investigated potential composition changes when surfactant-containing particles are produced with atomizers.”

Minor comments :

1. Line 86 : It is important to include here more information about the pure component surface tension required to perform calculations with the model. This parameter is crucial for the model implementation. Although this is explained in detail in Kleinheins et al. (2024), the model user would benefit from a short summary of the different assumptions related to this variable. It could be useful to explore correlations based on a hypothetical supercooled liquid state for substances that are solid at atmospheric temperatures (e.g.[1-3])

We agree that the pure component surface tension is an important parameter in both the Eberhart and the Monolayer model. However, a general discussion of the choice of pure component surface tension values is beyond the scope of this study. The Eberhart model has been analyzed previously in Kleinheins et al. 2023 and 2024 showing a good performance. Also, the Monolayer model has been presented and analyzed elsewhere. Here, the focus is the application of the models to sea spray aerosol particles and less so a discussion of the physical properties of organic substances in general.

2. Line 142 and 305: The molecular volume of each substance is also crucial in the estimation of the monolayer thickness. As before, the model user would benefit from a short summary of the possible data sources and assumptions related to this variable, especially in the case of surfactants (e.g. [4])

With respect to estimating the monolayer thickness, we think that the exact value of the molecular volume is less a problem than the shape of the surfactant molecule and how they arrange at the surface. Therefore, the Monolayer model could potentially be improved by addressing these geometrical assumptions. However, this study focuses on applying the Monolayer model as is, and therefore a general discussion about the molecular volume and the assumptions behind the monolayer thickness is beyond the scope of this study. However, future work should address this topic.

3. Line 300 : Salting-out effects modify the CMC in SDS-NaCl aqueous solutions leading to minimum surface tension values below the levels observed in aqueous SDS solutions (e.g. [5-7]). This effect should be explored more if SSA model particles are going to be represented in the same way in future studies. The Eberhart model assumes that the surface tension is the linear combination of the pure compound surface tension, and even using the salting-out parameters can fail representing cloud droplet solutions along the Köhler curve, especially at low relative humidity values (e.g. RH = 95 %).

The effect of a lower surface tension at the CMC of water(1)-surfactant(2)-salt(3) solutions compared to that of water(1)-surfactant(2) solutions can indeed be captured by the Eberhart model. The higher the value of the “surface non-ideality factor for a salting-out system” A_{23}^{SO} , the stronger the surface tension lowering at the CMC. As shown in the supplement Section “Influence of salting-out”, even a value of $A_{23}^{SO} = 22.63$, which is comparably high (Kleinheins et al. 2024) had no influence on the critical supersaturation. We agree that at lower RH, the salting out parameter in the surface tension term could have an effect on the hygroscopic growth of aerosol particles, yet, when the focus is on the critical supersaturation of aerosol particles, it is not relevant.

4. Figure S2 : About the mass closure. The tolerance value for the function makes use of the absolute value. I am sure that you check this, but just in case I kindly ask... have you check the sign of the individual number of moles at the droplet bulk and surface compartments?

Thanks for pointing this out. We double-checked our calculations and concluded that as long as the droplet radius is larger than one monolayer thickness δ , the bulk and surface volumes give positive numbers. As a consequence, the number of moles in the bulk and at the surface are positive, too.

Would it be possible to have cases when the numerator is negative but the condition is still satisfied?

When searching numerically for a solution via iteration, the mass conservation is tried to be fulfilled as closely as possible. It is correct that in that process, both negative and positive values can appear in the numerator. However, both a negative and a positive numerator would be equally unphysical. In reality, $n_i^{bulk} + n_i^{surf}$ must be strictly equal to n_i^{tot} so that the numerator is equal to zero. A small deviation from the perfect solution where mass is strictly conserved is inherent to all numerical solvers and as such not a problem of the presented solver approach.

At very small droplet sizes for pure surfactant dry particles, it is impossible to accommodate surfactant molecules in a monomolecular layer at the droplet surface compartment. The model, being just a model, tends to give negative solutions.

The limit of very small particles is surely a question of theoretical interest. Yet, such particles are too small to activate to cloud droplets and therefore not the focus of this study.