

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2021-561

Anonymous Referee #1

Referee comment on "Droplet activation of moderately surface active organic aerosol predicted with six approaches to surface activity" by Sampo Vepsäläinen et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-561-RC1>, 2021

General comments

The paper focuses on cloud droplet activation models with different approaches to account for surfactant effects, which is relevant topic for ACP. The models are presented and used accurately. The language is fluent and precise, and the overall presentation quality is good. However, the paper is mostly based on previously developed approaches that has been tested elsewhere. Partly for this reason the main aim and the conclusions are also somewhat unclear, so they need to be emphasized (details below) before the manuscript can be accepted.

Specific comments

Introduction: the extensive review of previous research shows that this is not a new topic, so it would be important to point out the open research questions which will be addressed in this study. Specifically, what is the added value of comparing multiple models here when they have been examined elsewhere sometimes with greater details and often with larger amount experimental data? Is it important that the models predict different partitioning equilibrium when the predicted critical supersaturations are similar? Regarding the previous studies mentioned in the introduction, are there clear issues with the models that can be fixed here?

A separate companion study is mentioned in the introduction (line 116), but could these be merged? It looks like introduction and theory & methods sections already contain relevant information for the strong surfactants. Combining the results could show which models are suitable for both strong and moderately strong surfactants. In addition, this would expand the range of modelled systems currently focusing on malonic acid-ammonium sulfate.

Line 106-113: "We quantify surfactant ... dicarboxylic acids (e.g. Booth et al., 2009; Hyvärinen et al., 2006; Ruehl et al., 2016)." seem to be misplaced; Sect. 2 could be more suitable for the details.

Line 162: What do you mean by the "method of solving the Gibbs adsorption equation"? Numerical method (this should not have noticeable effects on results)?

Lines 181 and 231: The layer is insoluble, but it doesn't stop water vapor, right? This should be clarified because there are studies where such layer is impermeable.

Line 187: "Predictions with the comprehensive..." indicates that surface tension depends on surface concentration, but should it depend on bulk concentration?

Line 241: Why is the minimum surface thickness dependent on droplet size in your simulations (Ovadnevaite et al. (2017) used two different constant values)?

Theory & Methods: Why only some models use parametrized solution density and water activity (Table 1)? Is this causing differences seen in Fig. 1?

Line 315: "This is most likely an effect..." seems vague. You could confirm this by swapping parameterizations. There are a few other cases where "likely" (lines 321, 354, 373, etc.) could be confirmed.

Line 322-323: For me it doesn't look like the partial organic film model predicts a "considerably" lower SSc (it is about the same as for the four other models) and neither is the dc "considerably" smaller (it is the second smallest).

Line 327: "falls between the monolayer and simple model predictions" could be true, but it looks like all other SSc values except that from the simple partitioning model are similar.

Lines 334-336: "The sensitivity analysis shows ... predictions are quite robust ... respect to relatively large variation ..." is a vague statement.

Lines 344-347: This sentence is one example which shows that these models have been examined in previous studies, so emphasizing new findings is especially important. The same comment applies to sections describing surface tension and organic bulk-surface partitioning.

Section 3.1.2: Is the shape of droplet growth curve important for other than SSc? This is not clear based on the introduction. Also, is the first part of the Köhler curve (before the film breaks) from the compressed and partial film models physically meaningful, or is it based on assumptions, parameters or extrapolation? Regarding the model parameters, the supplement shows sensitivity analysis for the compressed film model, but this is not done for the partial film model (or the other models); why and what would the results show?

Section 3.2: The same comment as above. Surface tension from the compressed film model is lower than the "lowest physically realistic value for the droplet surface tension" (line 398), what does this mean? Related to the first part of the Köhler curves, it is written in the manuscript that the compressed film model uses extrapolation (line 413: "...extrapolation of the model parameters outside of their validity region..."), so should this part of the curve be considered when determining the SSc?

Section 3.3: The same comment as for Sects 3.1.2 and 3.2. The authors write that the compressed film model predicts surface tensions that "has no physical meaning for the Köhler curve (Fig. 1) until the surface tension (Fig. 2) starts increasing" (lines 457-458), so what happens if SSc is seen before this point (Fig. S8(d))?

Conclusions: Are the "large differences between the different models in the predicted degree of surface partitioning of the organic component" mostly related to the model assumptions (this question applies to surface tension as well)? It would be useful to point out possible unexpected findings. Previous studies, which are mentioned in the previous sections, have made similar findings, so what is new in this study? In addition to listing

the differences between model predicted surface tensions and partitioning factors, you could mention which of these predictions are closer to reality and what do they mean from microscopic point of view. You could also mention if current findings have any large-scale effects for cloud activation or clouds in general.

The final conclusions (e.g., lines 507-510) should be clarified and made stronger. Do you recommend using partitioning models (or two of the models; "either" in line 509)? What kind of experimental data do you need? There is at least experimental SSc data available (e.g., Abbatt et al., *Atmospheric Environment*, 39, 4767-4778, 2005) and direct observations at least for Malonic Acid (Blower et al., *The Journal of Physical Chemistry A*, 117, 2529-2542, 2013).

Technical corrections

Below are few quotes from the text that seem to have something wrong or could just be improved for clarity.

Line 26: "and inducing concentration gradients"

Line 27: "compartments"

Lines 50-51: "They presented ... uptake (Sorjamaa et al., 2004)."

Line 56: "and systematically across their"

Line 79: "aerosol: Enhancement of"

Line 85: "by Sorjamaa et al. (2004); Prisle et al. (2008, 2010),"

Line 96: "natrium"

Line 105: "their immediate atmospheric"

Line 109: "to be about"

Line 118: "Theory & Methods"

Line 141: "particle, with the"

Lines 147 and elsewhere: "supplement section"

Line 165: "whence"

Line 174: "inside"

Line 189: "the very surface area-to-bulk"

Line 248: "For phase the bulk phase"

Line 276: "be even distributed"

Table 1: "'when they differ from the pure surfactant"

Line 312 (also 328 and 338): "0.19% supersaturation"

Line 243: "predictions will bias SSc values too high"

Line 377: "The pure surface tension of malonic acid"

Line 388-389: "fraction of 0.965 the ammonium"

Line 405: "(Eq. (S9) in"

Line 458: "(Eq. (6))."

Line 469: "approximately 0.65."

References: please reorder references with the same first author according to the ACP standards