

## General comments

I appreciate your revisions and responses to the referees' and my previous comments. I still have numerous comments.

As a major issue remains the lack of focus and the demonstration of the extent to which the presented framework improves CCN predictions as compared to previous models.

As the manuscript reports on 'new developments or novel aspects of experimental and theoretical methods and techniques that are relevant for scientific investigations', it will qualify after revision for a Technical Note. However, unless it is significantly revised and rewritten, I cannot see 'substantial advances and general implications for the scientific understanding of atmospheric chemistry and physics' as it would be required for a Research Article.

[https://www.atmospheric-chemistry-and-physics.net/about/manuscript\\_types.html](https://www.atmospheric-chemistry-and-physics.net/about/manuscript_types.html)

Please note that the manuscript title should start with 'Technical Note:'.

- It would have helped a lot in the author's response if you clearly stated in which lines and sections you changed text and how you addressed the referee comments.
- Many sentences are very long. While this is, of course, a personal choice and writing style, it makes reading often very difficult. It might also add to the lack of conciseness that was criticized in the previous referee reports. I do not think that the comment by Referee #3 has been sufficiently addressed "Overall, the manuscript is too long, difficult to follow, and the discussion regarding the implication of the model is overreaching... If the manuscript were shortened, and focused more on under what conditions and applications the different levels of partitioning treatment are needed, it would be a useful contribution to the field."  
The manuscript length has not changed and instead of a concise and clear discussion of the model's novelty and need, I am still left with questions such as
  - for which compounds does it need to be applied? You state in the introduction that previous models perform well for common aerosol surfactants.
  - under what conditions does it outperform previous simpler approaches? Throughout the manuscript (e.g. Figures 1 and 2) you show that previous simpler models give nearly the same results. Why do we need the full partitioning model?
  - for what mixtures is it relevant? – e.g. in Figure 1, it looks like that  $\leq 80\%$  NAFA particles are well represented by model (K) and (S). How likely is it that atmospheric particles are composed of  $> 80\%$  surface active compounds?
- In your author's response, please state why the second author has been removed from the manuscript so that this change is documented in the openly accessible files.

## Specific comments

p. 1, l. 9: 'allow for a description of chemically unresolved mixtures' – something seems to be missing here: 'the description of water uptake or surface partitioning or...?'

p. 1, l.4: What 'different mechanisms' are referred to here?

l. 17-19: Is this a finding from the current study or common knowledge?

l. 20-22: Does this mean that previous models that account for Gibbs adsorption are equally good or is the current framework the first that uses this adsorption?

p. 3, l. 16-18: This sentence is not clear. 'Köhler calculations' is not a common expression. Do you mean that 'hygroscopic growth of particles'? Is the sentence saying that commonly equations for surface/bulk partitioning are used that were derived based on bulk solutions rather than droplets?

p. 3, l.33 – p. l. 15: This part of the introduction is very exemplary for the confusion and unclear message regarding the novelty of the study throughout the whole paper:

- l. 33, you state that the CCN predictions for common aerosol surfactants work well but might be too simple as they only consider binary or ternary mixtures.

- p. 4, l. 2: you state that mixtures in atmospheric aerosol are likely much more complex.

- p. 4, l. 13: Here, you refer again to one proxy compound.

As this part of the introduction should set up the motivation of your study, it should clearly state why (i) fatty acids are not sufficiently good proxies (for which there are obviously good models), (ii) your proxy compound is more representative than fatty acids to represent aerosol surface activity.

p. 8, l. 15 (Section 2.2.1): It would be much easier to follow if all parameters in Eq-8 were presented together and not one page later. I suggest restructuring this section and only presenting the relevant information.

p. 10: Section 2.3: It would help a lot if you clarified here which of the models have been used before and which ones are new in the current study. Just reading this section, I assumed that (P) is the novel part of the current study, and (B) and (K) represent simplifications of it. Brief clarifications like this would immensely help to identify the novel parts of the study and how they are related to each other.

p. 15, l. 21: I do not understand how surfactants can enhance hygroscopicity. Hygroscopicity is a bulk property and expressed by the Raoult term, but reduced surface tension affects the Kelvin term. Do you mean 'enhanced water uptake' or do you mean that the amount of soluble mass in the bulk is reduced because of surface/bulk partitioning?

p. 17, l. 28; p. 19, l. 10: 'effects' should be singular. There is only one Raoult effect and Kelvin effect, respectively.

p. 21, l. 4: 'Gibbisan'?

p. 23, l. 35ff: 'When macroscopic composition-dependent relations for surface tension, as well as other solution properties, are connected to microscopic droplet states via a bulk/surface partitioning model, the effect of bulk/surface partitioning in droplets is to move the solution mixing state to a different point in the composition domain, as illustrated in Figs. 2 and S2 for the present case of NAFA–NaCl droplet mixtures' – please reword this sentence and reduce it to its main message

Section 3.4.2: What is the main message of this section? It seems like a text that should belong to an introduction. How does this text support or reject any of the previous or presented models? The fact that surface active organics are present in atmospheric particles had been stated before.

Section 3.4.3: How much of this information has been discussed in the previous sections of the paper?  
How would the current model improve previous predictions?

Section 3.4.4: If you want to highlight the importance of surface tension effects for cloud droplet formation, not the residence time in the CCN counter is of importance, but the time a particle spends in an updraft below cloud up to cloud base.  
During this ascent, the relative humidity (saturation) is constantly increasing and the time between a high saturation ( $> 0.9$ ) to cloud activation (i.e.  $S = 1$  or slightly above) is only a few seconds. A few words along those lines would be helpful here.

p. 30, l. 14 ff: I do not understand the relevance of surface reactions in the context of cloud processing. These topics seem very hand waving and vague.

Cloud droplets have diameters of several micrometers; they are certainly not submicron droplets. The diameter of cloud droplets is thus  $\sim 100$  times larger than that of a dry particle (e.g. 100 nm particle grows to 10 micron droplet), which means that its surface is 10,000 larger. It seems unrealistic to me that there is sufficient surface active material in CCN to affect surface tension of cloud droplets.

Therefore, extensive surface partitioning does not seem likely as the dilution of the bulk phase is completely different than in non-activated particles.

For which curvatures (i.e. droplet diameters) does the Young-Laplace equation apply? Is it relevant for cloud droplet sizes? What is known about pressure-dependence of aqueous phase reactions under atmospherically relevant parameter ranges?

Unless you can bolster any of these effects by either reasonable estimates or references, I suggest removing these speculations.