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Interactive comment on "Modeling CCN activity of chemically unresolved model HULIS, including surface tension, non-ideality, and surface partitioning" *by* Nonne L. Prisle and Bjarke Molgaard

Anonymous Referee #1

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Prisle and Molgaard present a modeling study of the partitioning behavior and CCN activity of mixtures of Nordic Aquatic Fulvic Acid and sodium chloride. Both the experimental data and the models used are published elsewhere (Prisle et al., 2010; Prisle et al., 2011; and Kristensen et al., 2014). The novelty of the work is the modeling results for these particular compounds. The authors use water activity, surface tension, and CCN activation measurements (Kristensen et al., 2014) along with new modeling results to conclude that surfactant partitioning to the droplet surface reduces CCN activity of mixed NAFA/NaCl particles. Although this finding is consistent with the majority of

experimental studies of mixed surfactant/salt CCN activity or water uptake (including Kristensen et al.), it is at odds with several high profile papers. The study is interesting and could contribute to the literature if the following issues are addressed.

Issues:

[1] The modeling results are interesting and informative. However, the major issue with this work is that the novelty is limited to the application of the models to the NAFA + NaCl data. I hesitate to recommend publication without extending the model framework and discussion to other mixtures. This would tend to better highlight, and shift the focus to, the context of this work with regard to atmospheric aerosols.

[2] How do the models of the present study compare to the model used by Kristensen et al., 2014? The original model was simple and should be discussed.

[3] Page 10. It should be noted here that experiments by other groups have also found that using bulk surface tension exaggerates the CCN activity of surfactants, some of whom are already cited here (e.g., Rood and Williams, 2001; Alshawa et al, 2009; Ruehl et al 2010; Harmon et al., 2010; Zamora et al., 2013; Petters and Petters 2016; Forestieri et al. 2018).

[4] Page 18 line 15, 16: "This may prove to be a signature feature of surface activity impact on CCN activity for relatively strong surfactants." This statement and the statements before could be better developed and discussed in context of past (and future) findings for simple and complex mixtures. See past comment [3].

Other comments:

Page 20 line 20-27. This paragraph makes an important point about equilibration timescales that could be featured more prominently at the beginning of the paragraph. The paragraph states that the surface tensions measured after 10 minutes using the macroscopic pendant drop method should be applicable to the microscopic droplets, based on their much smaller diameters (and therefore smaller equilibration times). It

has been said elsewhere that CCN experiments with surfactants are invalid due to long partitioning times (measured for macroscopic droplets). The present work makes a good explanation why this is not the case. This is informative and could be highlighted.

Figure 1 is difficult to read. Could SS be plotted on a log scale as in the earlier papers (Prisle et al., 2010 and Kristensen et al. 2014), or could 0%, 20% 50% 80% and 100% each get their own panel with four models? Please clarify.

Page 9, line 20-22: other studies have also reported this trend for strong surfactants mixed with NaCl and for marine biopolymer mixed with NaCl (Petters and Petters 2016; Dawson et al. 2016). Similar to Kristensen et al. 2014, marine organic aerosols were the motivation of Dawson et al., thus it would be relevant to point out that the trend holds for more complex aquatic molecules.

Should figure S1 be called, model P at different temperatures? Please clarify. Also, some mention and explanation of the data points would be appreciated in the caption.

P5 line 15-16: "the NAFA bulk–to–surface partitioning equilibrium is iterated to determine the droplet bulk phase composition" – this means it was solved iteratively, no? Please rephrase.

Page 7, equation 4: Please provide a reference for eq. 4 and for the surface tension parameterization of pure water.

Page 7, equation 4: Is there a need to define so many q's (qst1, qst2, qa1, qa2) in the manuscript? If they are not mentioned more than once in the paper, could they have local names c1, c2, ...?

Page 9, line 18: what is meant here by "non-ideality" ? is this non-unity activity coefficient?

Page 9, line 30: does "the latter" refer to models (S) and (K)? Please clarify.

Figure 2a and 2b: The traces in 2b are the contributions of Kelvin and Raoult effects to

the traces in 2a, no? Please clarify.

Figure 2b. Please clarify why there is an inflection in the Raoult term.

Page 11, line 17: The (P) and (S) models are indeed similar for supersaturation calculations, but they do not trace each other. As stated later, the (P) and (K) models are more similar, particularly for growth factor at activation.

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