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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 #2

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The manuscript entitled, "Modeling CCN activity of chemical unresolved model HULIS. . . " by Prisle and Molgaard, details a CCN modeling study of mixtures of NaCl and Nordic Aquatic Fulvic Acid aerosol. The modeling approach utilizes 4 difference surfactant representations: (P) a comprehensive partitioning model (Prisle et al 2010) (S) Simple surface representation (Prisle et al. 2011) (B) Bulk representation and (K) Basic Kohler theory. The overall goal of the study as I see it, is to evaluate how well these different models replicate SSc vs. dry diameter for this series of organic/inorganic mixtures.

I recommend publication only after the authors have clarified and discussed the following issues:

(1) The parameterization of surface tension of NAFA/salt solutions is taken from previous pendant drop experiments. It has however been pointed out in many studies to date, that macroscopic measurements of concentration vs. surface tension in macroscopic solutions may not be relevant to the CCN size range considered here. This disconnect between droplet and macroscopic measures of surface tension is made even more stark by the arguments presented here that in CCN droplets surface partitioning depletes the bulk concentration which is not the case for macroscopic surfactant solutions. This would then indicate that relying on Szyskowski type equations for surface tension parameterizations of complex mixtures are of no use for CCN studies (see Harmon et al., Physical Chemistry Chemical Physics 15 (24), 9679-9693). The authors need to address this deficiency more clearly in their manuscript. In fact the authors should comment on whether an "insoluble" surfactant model is more appropriate for CCN studies of organic partitioning. To clarify an insoluble surfactant entirely resides at the interface, reduces surface tension and has negligible bulk concentration. Therefore there is no relationship between bulk concentration and surface tension but rather the key relationship is surface coverage vs. surface tension. This point connects to point #2. So as I see it model (S) describes as presented describes an unphysical situation where the organic is at the interface and neither reduces water activity or surface tension.

(2) In model (S), the surfactant resides entirely at the interface but does not reduce surface tension or water activity at the point of activation. This model indeed provides the best predictions of the data in Fig. 1, despite being quite physically unrealistic (see comment #1 about insoluble surfactants). The authors should show and discuss, if this is indeed the case, that although surface tension is not reduced below pure water at the point of activation, the computed Kohler curves do in fact exhibit the correct perturbations due to surface tension reduction prior to activation as was observed in

Ruehl, Davies and Wilson, Science (2016). The authors should also compare and contrast their model (S) with the delta representation described in this paper. Finally, Ovadnevaite, et al. (Nature 2017) presents compelling evidence that surface tension can be reduced at the point of activation in their liquid-liquid phase separation model. It seem reasonable to me to provide some further context about how models P, S, B, and K might be related to the LLPS model. For example, if that authors relax the assumption in (S) that the surface tension at the point of activation is that of pure water, but rather something smaller how does this change the overall conclusions of the paper?