Authors' response to reviewer comments for the revised manuscript "Modeling CCN activity of chemically unresolved aerosol, including surface tension, non-ideality, and bulk/surface partitioning"

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We thank the editor and reviewers for their time and valuable additional comments and suggestions. Below, we address each of the reviewers' comments and indicate corresponding changes made in the revised manuscript.

Editor's comments

1. All three referees agree that the novelty of the current study in terms of our understanding of the role of surface partitioning on CCN is limited. The paper is very lengthy while it does not become clear what the innovative aspects of the study are at the current state of the field. The discussion and conclusions should be more concise and highlight the need of the consideration of surface partitioning and the conditions under which your findings are relevant in the atmosphere. The current discussion is rather vague and pretentious, given that the study is based on one ternary mixture and strongly dependent on the fit parameters. Some discussion of the relevance of this mixture and/or the distribution of similarly surface-active compounds throughout CCN distributions should be added to demonstrate in a more balanced way the general implications for atmospheric science rather than to limit the study to investigations that are primarily of technical interest.

   We have tried to sharpen the focus of the manuscript on the key aspects related to the introduction of the mass-based Gibbs partitioning framework and the findings related to the role of surface activity for complex aerosol mixtures. We have tried to further clarify the relation of the predictive framework and our results to atmospheric aerosols and recent developments concerning the understanding of the role of surface activity for CCN activation of complex aerosol. In particular, we here predict that a complex surface active mixture display similar CCN properties as previously found for model aerosol comprising single, strong industrial or atmospherically relevant surfactants.

Only a few studies have to date addressed the importance of surface tension and bulk–surface partitioning of surface active species for predictions of CCN activity in atmosperic modeling (Prisle et al., 2012; Lowe et al., 2019). While a common approximation has been to ignore surface activity in large-scale models, recent process studies by e.g. Ruehl et al. (2016), Ovadnevaite et al. (2017), Prisle et al. (2019), and Davies et al. (2019) have found potential impacts of droplet surface tension for predictions of CCN activity, even when bulk–surface partitioning of surface active species is accounted for. These studies highlight the importance of a full consideration of mechanisms driven by surface activity in cloud activation and need for a predictive description of the droplet surface properties. Our present work addresses exactly this need, by presenting a thermodynamic framework that enables comprehensive Köhler calculations for chemically unresolved surface active mixtures, thereby enabling benchmarking simplifying assumptions of droplet properties and representative choices of surface active proxy mixtures in various conditions.

The immediate relevance of model-HULIS reference mixtures for atmospheric aerosol has been discussed in several previous works, including that of Kristensen et al. (2014), and references herein. For this work, NAFA was considered as an excellent model system because it constitutes a chemically unresolved, strongly surface active mixture, for which a very large set of surface tension and water activity data was available, spanning a wide range of mixtures in ternary composition space, allowing us to construct fully continuous ternary composition-dependent parametrizations for these quantities, and in turn to perform thermodynamically consistent calculations of droplet growth and activation, including bulk–surface partitioning and evolving surface tension and water activity. To our knowledge, only this single comprehensive data set exist for chemically unresolved organic mixtures of any atmospheric relevance. Construction of this data set has been enabled by the commercial availability of the NAFA reference samples, which have allowed the preparation of a large amount of independent macroscopic sample solutions for both surface tension and water activity characterization. The atmospheric relevance of chemically unresolved surface active aerosol mixtures is highlighted in a range of works, including those of Nozière et al. (2014); Gérard et al. (2016); Ovadnevaite et al. (2017); Frossard et al. (2018); Kroflič et al. (2018); Frossard et al. (2019); Prisle et al. (2019).

While the present work illustrates the application of the mass-based Gibbs partitioning framework for one ternary system comprising NAFA and NaCl, the fully continuous parameterizations for both surface tension and water activity are based on measurements for a large number of individual ternary sample solutions (Kristensen et al., 2014; Lin et al., 2020). Results of CCN activation are presented for ternary mixtures covering the full organic–inorganic composition range and a wide range of particle sizes, representing a wide coverage of growing droplet states. The recent study of Davies et al. (2019) presents results for particle systems comprising four different inorganic salts, but mixed with a single organic compound and evaluated at discrete particle compositions and sizes. The work focuses on effects of surface tension in growing droplets and applies a similar volume-based mixing rule of pure component surface tensions as has been used previously, e.g. by Ovadnevaite et al. (2017) and Malila (2018), but surface tension has not been specifically determined, for either the pure organic component in question or its binary or ternary aqueous mixtures at different droplet dilution states. Instead, the pure organic surface tension is approximated by that of another compound and the mixing rule is not verified against measurements for the system in question. This underscores one of the major challenges presented to performing thermodynamically consistent, predictive Köhler modeling of surface active organic aerosol, which is the general scarcity of comprehensive data sets for the key thermodynamic properties concerned. Obtaining such data sets of sufficient quality and breadth is a non-trivial task, in particular for chemically complex, unresolved mixtures. The results presented in this work illustrate thermodynamically consistent, predictive Köhler modeling with the presented mass-based framework using independently constrained solution properties based on the single such comprehensive data set of atmospheric relevance currently available. We hope that by presenting the mass-based framework for thermodynamically consistent predictive Köhler calculations including bulk–surface partitioning and evolving surface tension and water activity, we will motivate more experiments to characterize these properties for unresolved mixtures of atmospheric relevance.

We do not consider the dependence on fit parameters for the surface tension parametrization to be particularly strong, quite the contrary, as seen in Figs. SXXX (d) of the SI (comparing pink with green trace and blue with red trace). For the water activity parametrization, the dependence on fit parameters was negligible and results are not shown. A significant dependence is seen only on the assumed (not fitted) value of the mean molecular weight of the unresolved organic mixture, with respect to an extremely conservative variation of a *full order of magnitude*, and only for the conditions where effects of bulk–surface partitioning are most prominent, i.e. the smaller particles comprising the very highest mass fractions of surface active material. The strong sensitivity is seen for very high assumed mean molecular weights of the organic mixture because these extreme conditions describe a situation where the surface adsorption of a single mass unit will lead to large changes in the mass-balance between surface and bulk phases of the droplet and resulting concentrations in each phase. Nevertheless, only for an assumed mean molecular weight of NAFA an order of magnitude smaller than the reported experimentally determined value (Mäkelä and Manninen, 2008) do we see a large deviation of model predicted CCN activity from the measurements of Kristensen et al. (2014).

We have tried to bring out each of these aspects more clearly in our revised manuscript.

2. The current study is heavily based on your previous study (Prisle et al., 2010) which is explained in detail throughout the manuscript. While there is, of course, generally no objection to apply such previously published framework in a follow-up study, the detailed description of this established framework results adds to the lack of focus.

   The general methodology is similar to both some of our own previous work (Prisle et al., 2010) and many others in the field, including recent works of Ruehl et al. (2016), Ovadnevaite et al. (2017), Forestieri et al. (2018), Malila (2018), Davies et al. (2019), and Prisle et al. (2019). The main differences between these works are in the applications of specific thermodynamic functions, for example for bulk activity, surface tension, and bulk–surface partitioning. The present framework allows thermodynamically consistent, continuous Köhler calculations with a Gibbs partitioning model for unresolved surface active organic mixtures. While the model of Malila (2018) uses a different partitioning scheme, a similar approach as presented here can also be implemented to enable predictions for chemically unresolved mixtures. We have used the framework presented here already in the recent study of Prisle et al. (2019), however, for the unresolved pollenkitt mixtures in that study, we did not have a sufficient amount of independent measurements for different mixtures to obtain fully continuous ternary parameterizations of surface tension and therefore to perform fully thermodynamically consistent Köhler calculations.We also did not have data to facilitate considerations of non-ideal solution effects on droplet water activity. In the present manuscript, a more detailed description of the model as well as reference to the previous works was added during the first revision, to highlight both similarities and differences with respect to existing frameworks. We have tried to further clarify these discussions in the revised manuscript.

3. Even though the manuscript was initially submitted more than two years ago, the referees and I have to evaluate it under the aspects of current knowledge and state-of-the-art in the field. While the idea of surface/bulk partitioning of surfactants on atmospheric aerosols might have been more innovative back then, it is much more established by now in the atmospheric chemistry community. In addition, some more advanced and refined approaches have been put forward, e.g. (Davies et al., 2019) that should be discussed to explain the observations in the current study (cf Report by Referee #1).

   We agree the concept of bulk–surface partitioning is by now fairly well established, at least in the specialized field of surface active aerosol, and in particular following the recent direct experimental demonstration of such partitioning effects on pico-liter droplet surface tension by Bzdek et al. (2020). However, as pointed out by e.g. Davies et al. (2019) and Lowe et al. (2019) and highlighted by the findings of several recent works concerning the influence of bulk–surface partitioning on surface active organic aerosol CCN activity (Ruehl et al., 2016; Ovadnevaite et al., 2017; Forestieri et al., 2018; Lin et al., 2018; Malila, 2018; Davies et al., 2019; Prisle et al., 2019; Lin et al., 2020), different surface models produce different results and while each provide the ability to describe selected systems and conditions, neither has been established as capable of broadly representing the effects of surface activity on atmospheric aerosol CCN activity.

The aim of this work has not been to introduce the idea of bulk–surface partitioning, but to present a thermodynamically consistent framework for performing predictive evaluations of these effects in growing droplets comprising chemically unresolved surface active mixtures and to use these calculations to gauge the importance of various manifestations of surface activity during droplet growth and activation. Previous works have either focused on well-defined mixtures of known composition, or assumed proxy compositions with well-known properties, but with unknown ability to globally represent the unresolved mixture in question under varying conditions. Common for these frameworks, including our own earlier work, is that the comprehensive description of solute-solute interactions, in particular between organic and inorganic components, quickly becomes very complicated and poorly constrained by measurement, when several distinct molecular species are present in droplet solutions and all mutual interactions must be explicitly considered. The need for thermodynamically consistent predictive modeling is also highlighted by Davies et al. (2019), who follow the general approach of Li et al. (1998), Sorjamaa et al. (2004), and many later works, using a single organic compound with well-known or assumed known molecular properties. Davies et al. (2019) use the models of Ruehl et al. (2016) and Ovadnevaite et al. (2017), which we discuss in the qualitative comparison of various recent approaches to including bulk–surface partitioning and evolving surface tension and water activity in Köhler calculations. Because these models are based on different assumptions and each presented for different selected systems, the necessary thermodynamic data is currently not available to perform quantitative comparison of all model predictions for the same aerosol systems. Such a comparison is the focus of ongoing work.

Reviewer #1

Prisle and Molgaard present a revised manuscript describing a modeling framework for treating the CCN activity of particles containing mixed NAFA and sodium chloride, using data from Kristensen et al. (2014). This is a well-written paper and it **addresses a real need to treat surface active mixtures of unresolved atmospheric material**. The model is mature and incorporates considerations from across different studies over the past decades, including the authors' own work and including considerations from others' work.

I do not believe that the results of the model fundamentally change the understanding of surface-active compounds in aerosols serving as CCN. Surface tension and interfacial processes in the atmosphere are an interesting frontier in atmospheric chemistry, and new techniques are needed to better constrain the problem. Nevertheless, this work contributes new modeling results incorporating updated data and approaches. I have a couple of minor comments and believe that the manuscript is worthy of publication.

1. On page 8, the incorporation of surface tension measurements is discussed. The measurement at 600 s seems very long given that nanoscale droplets are modeled. The explanation beginning on line 19 is not clear. Please clarify.

   Surface equilibration in microscopic (submicron and micron-sized) activating cloud droplets is considered to be diffusion controlled (Alvarez et al., 2012) and much faster than for the macroscopic (millimeter-sized) droplets measured with the pendant drop tensiometer (Alvarez et al., 2010). Activation in the CCNC instrument takes place during exposure times of about 1 s, depending on the particle size and required critical supersaturation (Kristensen et al., 2014). Following the considerations of Prisle et al. (2008), the ratio of the measurement time scales between the pendant drop surface tension measurements corresponding to 600 s and the cloud droplet activation measurements is estimated to be of the same order of magnitude as the ratio of diffusion distances in the droplet systems involved in the two types of measurement, given by the diameter ratio of the droplets, i.e. (pendant drop/activating droplet)$\sim$(millimeter/few microns)$\sim$(600 s/1 s)$\sim 10^2$–10^3. Because the surface tension data set for NAFA mixtures corresponding to measurement times of 600 s is much more comprehensive than those corresponding to later measurement times (Kristensen et al., 2014; Lin et al., 2020), we here use the data set for measurement times of 600 s, corresponding to the middle or lower end of this dimensionless ratio. With this simple argument, any potential dynamic effects of surfactant solute diffusing to the droplet surface, which could leading to incomplete (non-equilibrium) NAFA partitioning, are assumed to be at least comparable between the surface tension parametrizations and the droplet activation measurements, if present at all. In the pendant drop measurements, the measurement times of 600 s for macroscopic solutions are seen to correspond to times where the steepest and the majority of the total dynamic surface tension decrease has already occurred. We therefore do not consider such dynamic effects to significantly impact either the cloud droplet activation measurements or the comparison with our presented model predictions.

   We have tried to clarify these considerations of molecular diffusion in relation to the relative measurement time scales between surface tension and cloud droplet activation. We have furthermore added to the discussion references to Alvarez Langmuir and Alvarez soft matter provided by Reviewer #2.

2. Page 17 – “sharp infliction points” - “sharp inflection points”

   Corrected.

3. Page 24 – “signaficantly” - significantly

   Corrected.

Reviewer #2

The authors have done a reasonable job addressing the suggested revisions from the prior review two years ago. I have only a few remaining comments:

1. Page 3 lines 8-10: The claim that Bzdek et al. presented the ”first” measurements of concentrations dependent surface tensions for finite-sized droplets. . .” is an unnecessary overstatement since it neglects the substantial work and thinking on these kinds of topics from the non-atmospheric chemistry communities. Some illustrative examples:

   Jin et al. (2004)

   Alvarez et al. (2010)

   Alvarez et al. (2012)

   We thank the reviewer for bringing these references to our attention. While the experimental works of Jin et al. (2004); Alvarez et al. (2010) concern droplets or bubbles suspended in an infinite surfactant solution reservoir and therefore somewhat different conditions than finite-sized surfactant solution droplets suspended in air measured in CCN experiments, we found the works most interesting and have added the references to the discussion in our revised manuscript. The work of Alvarez et al. (2012) involves considerations of surface area to bulk volume ratio in combination with a Langmuir isotherm model for micro-liter droplets, which are several orders of magnitude larger than the sub-micron sized droplets involved in cloud droplet activation calculations in our present work. The aim of these considerations is mainly to establish the conditions for which the depletion of a bulk surfactant solution is sufficient to alter the equilibrium and dynamic surface properties in order to avoid the depletion effects of finite confined volumes. This is the opposite situation of our own overall aim to establish the conditions where these depletion effects are significant for growing and activating cloud droplets.

2. The authors, whether intentional or not, leave the reader with the strong impression (e.g. page 24 lines 32-35) that properly accounting for surface-bulk partitioning resolves a major source of error in modeling CCN. This has been discussed in many prior papers from this group and other as cited in the current manuscript. While I generally agree with this, I am also struck by Figure 1, where the most accurate Model (P) doesn’t satisfactorily account for the entire range of compositions most notably NAFA rich particles. This is a similar deficiency noted in Davies, et al. Atmos. Chem. Phys., 19, 2933–2946, 2019 for alternative models. Additionally, there are

a number of other examples of complex organic rich (or purely organic) aerosol systems created in the laboratory that show clear evidence of reduced surface tension, despite expectations to the contrary when surface-bulk partitioning is properly accounted for. George et al., Atmospheric Environment, Volume 43, Issue 32, 2009,Pages 5038-5045 and Harmon et al. Phys. Chem. Chem. Phys., 2013,15, 9679-9693. These studies along with the discrepancy between model and measurement for 80-100% NAFA imply more complex behavior than can be explained by accurately accounting for surface-bulk partitioning. I don't feel as if the authors have provided this literature context or adequately discussed the 80-100% NAFA model/measurement discrepancies in their revised manuscript.

We have added the references provided to the discussion. We have elaborated on the comparison of experimental and model results for particles with the highest NAFA mass fractions, highlighting the challenges for the model in general and specifically for modeling the NAFA system with a relatively high average molecular mass. We already discuss the behavior observed for several complex aerosol systems, indicating different impact of surface activity on droplet activation properties than previously for simple strong model surfactants. These results motivated us to attempt thermodynamically consistent, predictive modeling of such a complex surface active mixture, to more closely examine the underlying dynamics between partitioning, surface tension, and water activity, and to include also an independently constrained account of concentration-dependent non-ideal solution effects on droplet water activity, which in addition to surface activity could contribute to the observed differences droplet activation behavior.

3. Page 17 line 5: "infliction" should read "inflection"

   Corrected.

Reviewer #3

This work presents a thermodynamic framework for droplet growth, which combines treatment of surface tension, water activity, and surfactant partitioning representation. Results are presented for a NaCl + Nordic Aquatic Fulvic Acid (NAFA) systems with the whole range of mixing ratios, and five surface/bulk partitioning representations are considered. While the paper comprehensive and focuses on a topic of interest to the field, there are a couple of major issues that need to be addressed before final publication.

1. Four parameters from the Szyszkowski equation (2 parameters for qst1 and 2 parameters for qst2) was fit to pendant drop equilibrium surface tension measurements. No goodness of fit statistics or confidence intervals of the four fit parameters are presented. 2 parameters were used for the relationship between water activity and composition, though the source of the data nor the goodness of fit are apparent. All fit parameter should have uncertainties reported for equations (9), (10), (11), and (12).

Both surface tension and water activity data for ternary parametrizations were obtained from Kristensen et al. (2014) and Lin et al. (2020). We have added the goodness of fit statistics for the parametrizations used in the calculations. We emphasize that these parametrizations contrain the aqueous solution behavior of NAFA–NaCl mixtures independently of the CCN measurements modeled with our framework. The exact forms of the fits are not a main focus of the Köhler model and very similar model results were obtained for a range of functional forms and values of the surface tension and water activity fit parameters.

2. Overall, the manuscript is too long, difficult to follow, and the discussion regarding the implication of the model is overreaching. Only one ternary mixture at varied mixing ratios is examined, with many fit parameters. Still, the systematic examination of the 5 partitioning treatments is certainly of interest to the field. The sensitivity analysis of the model input parameters such as molecular weight of the NAFA is appreciated. 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 model is predictive and itself does not contain any fit parameters, it can be used with any surface tension and water activity parameterizations, containing any number of fit parameters. The main benefit of the model is that it enables thermodynamically consistent predictive modeling for unresolved surface active mixtures, provided the parameterizations used represent fully continuous variations with respect to the resolved component fractions. We have moved some of the results which are important for the conclusions, but may unnecessarily complicate the presentation, to the Supporting Information, and tried to focus the discussion of the various key implications of the model results in relation to both reported experimental CCN activity and simpler predictive frameworks, while maintaining the additions made in response to previous reviewer comments. We have further clarified the discussion points with use of subheaders.

Similarly to other studies focusing on detailed thermodynamic modeling of surface active aerosol CCN activity, our work concerns a limited number of mixtures covering a selected span of aerosol and droplet states. As such, we do not consider that our results should be used to make too generalized conclusions regarding the conditions for which the different levels of partitioning considerations should be used. Rather, we intend to use the comparison between different model approaches as a means to highlight the interplay of different interrelated underlying mechanisms. We also wish to highlight that the NAFA–NaCl aqueous droplet systems exhibit CCN activation behavior which is more in line with what has previously been seen for much simpler surface active components and less resembling the behaviors observed recently by Ruehl et al. (2016), Ovadnevaite et al. (2017), and Prisle et al. (2019) for other complex aerosol systems. Our point is that the manifestation of bulk–surface partitioning behavior in CCN activity for complex surface active aerosol is likely to be highly dependent on conditions and in general not well represented by a single model for all atmospherically relevant systems. We have tried to emphasize this point in the revised manuscript.

3. Page 17, Line 17: should be 'leading to a simultaneous' instead of 'leading a to a simultaneous'

   Corrected.

4. Page 19, Line 28: should be 'except for' instead of 'expect for'

   Corrected.

5. Page 22, Line 1: should be 'on' instead of 'om'

   Corrected.

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