

Response to Reviewer 1:

The technical note summarizes a set of interesting points to be clarified regarding the role of surface active organic aerosol in cloud activation. In light of several recent works, including those of the authors, this topic is very timely and their call for a discussion on the topic is commendable. While it is useful on occasion to summarize the knowledge and open questions in the field, my main concern is that the note brings very little new to the table. Most or all of the points have been made previously, but little references or perspective is given to the substantial body of work already existing on the subject.

We thank the reviewer for his or her comments. As a technical note, the intention of this work is to clarify a key technical point that is often obscured – namely that the evolution of surface tension, in addition to its absolute value at any given droplet size, is an important factor in regulating the activation of CCN. We acknowledge that the theory behind this is established, but it is only in recent experimental efforts that direct observations of modification to Kohler curves have been possible. In these works, the evolving surface tension is shown to change the size of the activating particles, with activation occurring at much larger wet diameters. However, the effect on the activation efficiency (through S_{crit}) is more subtle and can easily be missed when performing typical CCNC experiments. Thus, we aim to clarify why surface tension affects Kohler curves in this manner, and to convey the situations where surface tension effects might be notable and detectable using standard CCN instrumentation. We do not seek to provide a complete or exhaustive review of the literature in the very broad science of CCN activation, instead our focus is on two main points:

1. The evolution of surface tension is the main driver for changes in CCN activation relative to assumptions of fixed surface tension.

2. Current models struggle to explain and predict experimentally observed data across a wide range of parameter space (size, organic fraction and inorganic composition), but experiments can be performed that clearly reveal surface effects which are needed to help to drive the development of new theories and practical, predictive models of CCN growth and activation by the community.

p.2 l. 36-38: “In this note, we focus on the role of surface tension, and discuss the limitations of current approximations in light of recently published works that reveal how it is primarily the evolution of surface tension that impacts the activation process.” Only two papers are discussed in detail, the works of Ruehl et al. (2016) and Ovadnevaite et al. (2017). The discussion is not sufficiently substantiated independently of these works.

To our knowledge, it is only in recent papers that experimental observations have been able to corroborate the assertion that surface tension influences hygroscopic growth and CCN activation in organic and mixed inorganic-organic aerosol (Forestieri et al., 2018; Ruehl et al., 2016). These recent studies have done this via direct measurements of droplet size at known saturation ratios (i.e Kohler curves) that show the shape of the curve as activation is approached. While much previous work has discussed the role of surface tension, as noted in our manuscript, there remain only very few studies that directly measure the effects. We wish to emphasize that now the goal is

to find a model description to best describe the data over all observable space, based on established physical parameters, rather than fitting an empirical or semi-empirical model to individual datasets. This is challenging, as in many cases surface tension effects are obscured by the way data is presented or analyzed. Indeed, we show that the common practice of showing S_{crit} versus D_{dry} is not effective at highlighting differences that may be attributed to a variable surface tension, due primarily to the larger role that the particle size has in the Köhler equation.

p.3 l.72-75: Effects of inorganic co-solutes is discussed by e.g. Svenningsson et al. (2006), Prisle et al. (2010,2011), Frosch et al. (2011), Kristensen et al. (2014), and Hansen et al. (2015). Effects of dissolution is discussed by e.g. Shulman et al. (1996) and Bilde and Svenningsson (2004).

We thank the reviewer for their recommended additional literature to include in our work, and these have been added to this discussion, indicated below. We acknowledge that there are significant bodies of work outside of those cited in our Technical Note, though we have chosen to avoid a comprehensive review of the field in light of the focus of the manuscript.

“Furthermore, surface partitioning may be influenced by non-surface active components in the system, such as the presence of inorganic material and co-solutes (Asa-Awuku et al., 2008; Boyer et al., 2016; Boyer and Dutcher, 2017; Frosch et al., 2011; Petters and Petters, 2016; Prisle et al., 2011; Svenningsson et al., 2006; Wang et al., 2014). Other factors that have been shown to influence the shape of Köhler curves are: (1) solute dissolution, encompassing both water-solubility and solubility kinetics (Asa-Awuku and Nenes, 2007; Bilde and Svenningsson, 2017; McFiggans et al., 2006; Petters and Kreidenweis, 2008; Shulman et al., 1996), (2) liquid–liquid phase separation (i.e. limited liquid–liquid solubility) (Rastak et al., 2017; Renbaum-Wolff et al., 2016), and (3) the dynamic condensation (or gas–particle partitioning) of organic vapors (Topping et al., 2013; Topping and McFiggans, 2012) (Topping et al., 2013; Topping and McFiggans, 2012).

p.4. l.81: “. . .there are many observations that cannot be explained in such simple terms”. Please be specific, which observations cannot be explained and what is the argument that they are unexplained.

The studies cited apply κ -Köhler theory without fully considering the potential influence of surface tension. Collins et al., 2016 note that the method of sea-spray production from bubble bursting likely leads to aerosol more highly concentrated in surface active species. Good et al., 2010 suggest surface tension as a potential source of the deviation in their CCN concentrations. Yakobi-Hancock et al., 2014 briefly discuss data at high organic fraction that shows a much higher κ_{org} than would be expected (albeit with some large uncertainty). These papers demonstrate the conditions where surface tension effects are likely to be significant – the presence of organic molecules with rather low water-solubility present in relatively high concentrations in small particles, as concluded in our manuscript. Thus, these observations cannot be fully explained without considering surface effects. We have modified our language to better reflect the intended meaning:

“While measured cloud droplet number concentrations in the atmosphere have been explained in several cases with simple parameterizations that neglect dynamic surface effects (Nguyen et al., 2017; Petters et al., 2016), there are many observations that are not **fully** explained in such simple terms **and in those cases a substantial population of CCN may exhibit behavior characteristic**

of surface effects (Collins et al., 2016; Good et al., 2010; Ovadnevaite et al., 2011; Yakobi-Hancock et al., 2014)”

p. 5-6 and Fig. 1(A): A substantial part of the manuscript is devoted to explaining basic Köhler theory, as done by many authors previously, including basic textbook material. The presentation could benefit from a similarly detailed presentation of the surface tension and partitioning models used.

We carefully state the factors that go into the Köhler equations in order to be thorough and precise in our message. Without fully describing the basic equations, it can be hard to see how water activity and the Kelvin effect affect the results, and how these must be considered separately. A detailed description of the partitioning models of Ruehl et al. (2016) and Ovadnevaite et al. (2017) can be found in their respective manuscripts. Additional information about the AIOMFAC-based models was added to the manuscript on line 236 (line numbers of original manuscript); see our reply to referee #2. However, it is clear that a complete, accurate and predictive description of CCN activation applicable in the general case is not yet available. Rather, our intention is to promote additional laboratory studies and analysis to identify more cases where surface tension has real and measurable effects, in an effort to gain the insight required for producing a predictive theory that captures the full complexity of CCN activation. Thus, we take a step back and focus on the fundamentals of Köhler theory that necessitate the development of the partitioning / surface tension models.

p.7 l. 171-72: “This also seemingly supports previous assertions that surface tension does not impact activation, since it is generally argued that at the point of activation the droplet is sufficiently dilute and essentially exhibits a surface tension like pure water.” Please reference a sufficient amount of such previous assertions to support the claim of a general argument.

Earlier in the text, we discuss the references that point to this assertion (lines 44+) and we have amended the text to refer the reader back to this section:

“Traditionally, however, surface tension has been reduced to a fixed term in the Köhler equation (Abdul-Razzak and Ghan, 2000; Facchini et al., 2000; Petters and Kreidenweis, 2007) and is usually given a temperature-independent value equal to that of pure water at 25 °C. This is because for any decrease in surface tension due to bulk–surface partitioning and surface adsorption, it is assumed that there is an increase in the solution water activity because adsorbed material, previously acting as a hygroscopic solute, is removed from the droplet (bulk) solution (Fuentes et al., 2011; Prisle et al., 2008; Sorjamaa et al., 2004). Thus, the effects approximately cancel out in the calculation of a droplet’s equilibrium saturation ratio via the Köhler equation and so are often neglected. Furthermore, it has been shown in some cases that there is insufficient material in a droplet at the sizes approaching activation to sustain a surface tension depression (Asa-Awuku et al., 2009; Prisle et al., 2010).”

p.8 l.180: “. . . knowing the surface tension only at the point of activation is insufficient to determine the critical properties at activation. . .” Please provide references.

We have rephrased this statement to clarify our meaning, as this reflects an important point. The text here now reads:

“Clearly, an evolving surface tension prior to activation can matter in such systems and, consequently, knowing the surface tension only at the point of activation is in a general case insufficient for determining the critical properties at activation (absent any droplet size measurement), because the position of the maximum in the Köhler curve and, thus, the droplet size at activation, will depend on the trajectory of surface tension evolution. Moreover, knowing the surface tension at the activation point only, may not allow for an accurate prediction of whether a droplet of given dry diameter will activate at a given environmental supersaturation (compare the green curve with the *iso- σ* curve of 72 mN m⁻¹ in Fig. 1B, both having the same surface tension at their points of CCN activation, yet different critical supersaturations and wet diameters). Similar conclusions have been drawn previously (e.g. Prisle et al., 2008), although the measurements by Ruehl et al. (Ruehl et al., 2016) were the first to verify this experimentally.”

p.11 l.258-64: “This predicted size effect indicates that it is not generally valid to assume that all activating CCN will have a surface tension equivalent to or close to that of pure water –nor is it appropriate to use a single measurement of the surface tension of a multicomponent CCN of known dry composition at its activation size (only) to determine its Köhler curve. Furthermore, these model predictions also suggest that measurements of the surface tension of larger CCN particles (e.g. > 150 nm dry diameter) may not allow for conclusions about the surface tension of much smaller CCN, e.g. of 50 nm dry diameter.” These points have certainly been made by other studies than the one specifically described, including those mentioned above, and Wittbom et al. (2018). A broader perspective should be provided.

Studies that do not explicitly account for surface partitioning, such as Wittbom et al., indicate there is a size-dependence to their values of κ , and implying to us that there might be surface tension effects at play. The studies of e.g. Prisle et al. (2008), Prisle et al. (2010) and Sorjamaa et al. (2004), now cited in the text at this point, discuss size-dependent surface tension but do not appear to show clear deviations from an assumption that surface tension is size-independent. Indeed, Sorjamaa et al. conclude the opposite of the observations reported here – that surface effects become more significant with larger particles. As we show in Figure 2, we note the opposite, that smaller particles of the same composition are more CCN active and thus suggests greater surface tension effects. We have now cited additional papers in this context that discuss size dependencies relating to surface partitioning.

p.12 l. 292-92: “. . .in contrast to other studies that allow both coated particle size and organic volume fraction to vary simultaneously.” Although in many studies particles are produced by other methods than coating (e.g. nebulization), again many other works have indeed recognized the importance of independently varying the particle size and organic fraction, for studying the effects of organic surface activity and mixing effects, including those listed above.

We acknowledge that other studies have performed measurements in this manner and have now included references to Prisle et al. (2010) and Wittbom et al. (2018). Many studies do however

only show data where f_{org} is varied in conjunction with D_{dry} , which we argue conceals subtle surface effects that might influence the system.

p.13 l.310-12: “One could ignore the physical meaning of K and simply use it as an all-encompassing parameter to describe activation efficiency.” This was already done by several previous studies, e.g. Kristensen et al. (2014) and Hansen et al. (2015). I do understand that the authors caution against it, but see also my points below. p.13 l.312-13: “In this case, the generality of the parameter to interpret observations in different conditions is lost.” Please explain what is meant by this statement, and how that would not be the case by separating out surface tension from the K parameter. Please give references to previous work debating this, as well as to the original work of Petters and Kreidenweis (2007) and following articles to support why this is not the proper way to interpret their presented framework.

When comparing the formulation of κ -Köhler theory by Petters and Kreidenweis (2007) to the complete Köhler theory, the κ parameter is equal to the ratio of the molar volume of water to the molar volume of a non-electrolytic solute for an ideal solution – therefore representing a simplified model for water activity. Thus, deviations from this exact value can be attributed to non-ideality. Water activity, however, is a function of the bulk concentration, and thus should be decoupled from surface effects (and be size invariant). By using κ to account for both water activity and surface tension, a size dependence is introduced due to bulk–surface partitioning and surface-to-volume ratio, and thus its generality is lost. In the general Köhler equation, water activity is a factor distinct from the Kelvin factor and therefore one distinguishes between surface tension effects and bulk solution non-ideality (which relates to hygroscopicity also at $RH < 100\%$). By using κ as an adjustable fit-parameter, one may be able to fit experimental data, but by doing so, the retrieved κ will be system-specific and size-specific if the actual surface tension of the droplets varied with size. Such system- and size-specific fitted models will not serve the purpose of establishing a predictive theory and model. This is why we caution against it.

p.13 l.318-19: “. . . a specific suberic acid hygroscopicity alone cannot explain the observations across a range of particle sizes and compositions” That was also found for other well-defined organic aerosol by e.g. Kristensen et al. (2014), Hansen et al. (2015), and Wittbom et al. (2018). Indications of similar relations have been seen for ambient aerosol by e.g. Hong et al. (2014,2015).

The mentioned studies report agreement when looking at critical supersaturation vs D_{dry} (D_{coated} in our manuscript). Indeed, Kristensen et al. conclude that a fixed surface tension of water and a fitted κ value for the organic fraction gives best agreement. Hansen et al. are not definitive in their conclusions of a size dependent κ , although they do indicate that smaller particles exhibit a greater surface tension reduction, and the focus of Wittbom et al. is on solubility, which may show different size dependencies than surface tension and is thus not directly relevant for the discussion here. The data and choice of coordinates we report in Figure 2 is the most clear demonstration that a single value of κ cannot reproduce the data for this system across different sizes. Decoupling size from composition allows for a more rigorous assessment of the effectiveness of a model in account for each parameter. D_{dry} is not a sensitive metric for observing deviations due to surface tension because both size and composition vary, and their effect on the extent of surface tension

effects is opposite. That is to say, as D_{dry} gets smaller (likely to increase role of surface tension), the organic fraction decreases (likely to decrease role of surface tension). Thus, the influence of surface tension is masked by the manner in which the data is presented.

p.15 l.349-50: “. . .all organic material is assumed to reside in a water-free organic shell-phase (an organic film) at the surface of the aqueous droplet.” This assumption was also used by e.g. Prisle et al. (2011).

This assumption, as we discuss, is the most simple to apply, and yields the best results for the sample case discussed. Given that it is not well-supported by the applied partitioning theory for the case of suberic acid, it is interesting that this is the case and suggests we have a long way to go to quantitatively understand what is happening during the CCN activation process. In Prisle et al. (2011), this assumption is applied for the case of “classic” surfactants for which conceptually such an assumption might seem more appropriate. In our case, this assumption still yields good results, despite the molecules not being true surfactants. In general, this organic film assumption represents a limiting case, and in reality a balance between surface partitioning and water activity suppression should explain the results. We have added the following sentence to acknowledge these previous studies:

“This observation is consistent with the results of (Prisle et al., 2011) who applied a similar simple model to droplets containing ionic surfactants and hints at a significant suppression of surface tension by suberic acid, which is likely highly enriched at the droplet surface”

p.15 l.349-50: “. . .neither of the models fully captures the observed behavior at all for and size regimes.” Previous studies have made similar conclusions for other aerosol and other models, please bring these into perspective to present a broader perspective of the issue.

[The referee refers to line 359] The discussion in this section of the manuscript concerns the system and models shown in Fig. 2. To discuss previous literature which makes similar conclusions for other aerosol systems – yet not presented in terms of organic volume fraction at constant size cases – does, in our opinion, not add to the purpose of the discussion at this point in the article.

p.17 l.399-400: “Surface tension effects can lead to significant differences from classic, hygroscopicity mixing rule mechanisms for CCN activation.” This was also seen by Kristensen et al. (2014) and Hansen et al. (2015), among others.

We have added additional citations here to refer readers to the works the reviewer suggests.

p.30: For example Shulman et al. (1996), Sorjamaa et al. (2004), and Prisle et al. (2008) present similar Köhler curves. Please discuss Fig. A2 in context of previous results.

This type of Köhler curve is produced when additional factors that determine the water activity or surface tension vary with the changing particle size. Solubility limitations and surface tension suppression both produce plots that may exhibit discontinuities. The point of Fig. A1 and associated text in the manuscript is to illustrate the different outcomes of the employed AIOMFAC-based model variants for a specific case of the aqueous suberic acid + ammonium

sulfate system including accounting for liquid–liquid solubility limitations. We do agree with the referee that Köhler curves similar in shape have been produced by other models for other systems in the past, including in the work by Shulman et al. (1996) but we note also that the shapes of such variable- σ Köhler curves were only recently verified experimentally.

The most significant novelty of this work lies in the measurements presented in Fig. 3, but these results lack a comprehensive discussion.

These results were included to highlight the other factors that influence observations and demonstrate that co-solute effects are important to consider. These observations must also be accounted for in any predictive overall model, and are thus included to illustrate the need for a comprehensive theory applicable over the relevant parameter space.

Significantly more novel content should be added to the work before publication. For example, no explanation is given for the variation between the salt seeds and no modeling is done to provide interpretation beyond general speculation. Would it not be possible and facilitating to the discussion to make model predictions using exactly the outlined framework, for these aerosol? CCN activation experiments could be made with the systematic variation in particle size and organic fraction which this technical note specifically calls for. How are the impacts on water activity from the different salts treated in the model? Could intrinsic salt hygroscopicity/solubility explain the variation with mass fraction, and if not, why? More generally, a discussion could be added on the atmospheric/organic aerosol process relevance of suberic acid and mixtures with each of the salts.

As a technical note focused on conveying a single point, the role of evolving surface tension on CCN activation, a broader discussion is beyond the scope of this note. Moreover, we emphasize that the goal for showing the data of Fig. 3 is not in fitting a specific (simple or sophisticated) CCN activation model to the data presented in Fig. 3; rather, the point is made that additional complexity is observed with respect to organic–inorganic interactions stemming from different types of solute species. We agree that these data are particularly interesting, and hope further studies will be able to learn from what we present here and offer more tangible theoretical frameworks that allow the relevant physical parameters to be accurately included.

For the model part, I commend the effort to use a more general activity model for calculating the Köhler curves. This is indeed one of the major sources of discussion related to identifying surface tension effects in Köhler theory. However, the use data presented in Table 1 is not sufficiently justified. For example, why was the 20 C values used? What is the basis for assuming similar surface tension based on structural similarity? For other compounds, small changes in molecular length can lead to greatly enhanced surface activity. How did Riiipinen et al. make the determination of the pure compound value? How is pure ammonium sulfate (a solid at 20 C) surface tension justified? How is the minimum shell thickness estimated? .

Adipic acid and suberic acid are the closest in size of the dicarboxylic acid series, and both contain even numbers of carbons. It is indeed true that smaller changes can result in large differences of surface tension, but this is typical seen when going from even to odd, and that adjacent even or odd compounds are more similar (Ruehl and Wilson, 2014). The tabulated value for suberic acid

is that estimated for the pure liquid (subcooled) component; its value is likely similar to that of pure liquid (subcooled) adipic acid and other dicarboxylic acids. Adipic acid's pure value was determined theoretically using the Macleod-Sudgen method, as described in Riipinen et al. A pure component surface tension is a physicochemical property that is distinct from a molecule's surface affinity in a solution droplet (however, when such a substance contributes a substantial area fraction of the surface, its surface tension value will affect that of the droplet). A temperature of 20°C was used because this was the ambient temperature in which the experiments were conducted. The ammonium sulfate was not considered to influence surface tension at dilute aqueous conditions, as noted in the footnote to Table 1. We have amended the text in the table to state "surface tension of aqueous AS (at T)". The minimum shell thickness is one of the assumptions applied in the predictive AIOMFAC-based model and is established based on the approximate size of the molecules and related monolayer thickness.

What would be the impact on Köhler curves using more mixture specific input data? Can the authors rule out that any of the effects described are not caused by solution non-ideality and misrepresentation of the experimental system by use of incompatible thermodynamic data? It would strengthen the arguments significantly to use accurate reference input data if such may be obtained from a thorough literature search, or from performing new measurements for the specific systems in question. The lack of proper input data for different Köhler models is a major limitation of obtaining a comprehensive molecular framework, as called for in this technical note – this would be one of my first additions to the discussion.

We agree with the reviewer that any model should use input data which are directly measured rather than fit to the experimental data in question. We are not sure what misrepresentation the reviewer might be referring to – the main conclusion of the paper, as supported by the experimental results, is that we do not have a correct model, and perhaps lack the correct inputs or physicochemical details to fully understand what is happening. However, we do consider bulk phase non-ideality in the models explicitly, including organic–inorganic interactions in the AIOMFAC model, aspects often ignored in other models. We are confident that the thermodynamic data used for model inputs are correct (or reasonable estimates where direct measurements are lacking). The application of a spectrum of models in this work is to demonstrate that surface effects can reasonably explain and constrain the observations, but that a robust, accurate and fully predictive model for such coupled hygroscopicity and surface tension effects is yet to be developed.

The discussion about discontinuities in the activation curves is very interesting. It would be useful to see Fig. 1(B) developed for somewhat more realistic conditions. Specifically, do similar discontinuities persist in Köhler curves when concentration dependent partitioning is taken into account? What are the relation between the curves in Fig. 1(B) and A2? Why do they look different?

Figure 1(B) is a schematic using exaggerated surface tension dependencies to highlight the differences that may be encountered based on different surface tension evolutions. Those shown in A2 result from the AIOMFAC-based model variants for the suberic acid + AS system, as described in the manuscript. Not all possible schematic surface tension evolutions are exhibited by

a single system. Discontinuities in Köhler curves, similar to the schematic in Fig. 1B, have been observed e.g. by Ruehl et al. (2016) for droplets containing ammonium sulfate and various dicarboxylic acids. Those observations and discontinuities were reproduced by a (fitted) compressed film model of CCN activation.

The present technical note draws primarily on the previous work of Ruehl et al. (2014,2016). It should be made clearer which results are summarized from there, and which are new for this work. The presentation and arguments would benefit greatly from an independent presentation of the model framework and main assumptions used. In particular, it is not entirely clear what partitioning framework is used and how the surface composition is actually evaluated. It is not clear how mutually different the employed frameworks are, that is, how wide a range of possible representations of the aerosol are mapped with the three different models used. As the majority of previous work uses a Gibbs-based partitioning model, would it not make sense to include such calculations in the comparison? The results in Fig. A2 could be discussed in greater depth, as could the statement “. . . the simplified organic film model provides the best description of these experimental data” p. 16 l.374. Why was the “best framework” not verified by e.g. measuring the droplets size at various equilibrated RH?

The main intention of this paper is not to present a “best” model in the sense of a recommendation to the community. The main goal is to show that current models are insufficient in capturing the details and the dynamics across a range of parameter space relevant to CCN activation. While different from classical Szyszkowski-Langmuir isotherm model for bulk-surface partitioning and surface tension, the compressed film model by Ruehl et al. (2016) employed in this work is also a model that includes a thermodynamically consistent isotherm model for relating surface composition to bulk concentration. A more detailed description of the models is therefore unwarranted.

References

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