Response to Reviewer 2:

While it is not entirely clear to me why this is being framed as a "technical note," and not just a paper, I find the results to be generally interesting. There is effectively a very long introduction in which the authors lament that others continue to neglect the role that surface tension can play, followed by some interesting discussion of some new experiments. I suggest that the discussion about the new experiments is expanded.

We thank the reviewer for his/her time in reviewing our manuscript. We present this work as a technical note in order to convey the key point to the work – that it is the evolution of surface tension, in addition to the absolute value at any given droplet size, that is important in regulating the activation of CCN. The introduction serves to ensure the reader has sufficient context in order to appropriately apply and considerer surface tension effects when assessing CCN data.

L150: I find the terminology "schematic dependences" to be a bit unclear. It would be good to see this explained a bit further. Do these come from anywhere in particular, or are they totally made up? What sort of cases are they meant to illustrate, i.e. why these three?

These are schematic in the sense that they do not conform to the behavior of any particular solute/solvent system. However, they represent the types of dependencies that are encountered and the resulting implications on the Kohler curves. The text now reads:

"Distinct schematic dependences of the surface tension on the droplet size, representative of the types of dependence that might be encountered in real aerosol, are imposed for the purpose of illustration, shown in the lower panel of Figure 1B"

L170: It is not clear to me how the statement that "This. . .supports assertions that surface tension does not impact activation. . ." follows from the previous sentence. In the examples, the activation diameter for all three non-constant surface tension curves is lower than that obtained assuming a constant surface tension equal to that of pure water. In fact, the authors go on to say just this. So, it would seem that the activation diameter is impacted. It would be helpful if the authors could clarify their particular point here.

Figure 1B shows the opposite of what is being asserted by the reviewer – each of the curves associated with a variable surface tension exhibits an activation diameter that is larger (not lower) than the constant surface tension curve. While surface tension at the point of activation may be equal to that of pure water, the simulations here indicate that the point of activation is modified by the evolution of the surface tension. The point in the mentioned sentence on L170 is that in most CCN measurements, only the activation supersaturation is measured for a given dry size, but not the activation size nor the surface tension. Thus, one might assume that the surface tension at the activation point is equal to that of pure water, which may frequently be the case. However, as the examples in Fig. 1B show, assuming that the surface tension is constantly that of pure water prior to reaching those dilute conditions and using a determined hygroscopicity parameter to predict the Köhler curve and CCN activation point would lead to a substantially different critical supersaturation and activation diameter (that of the peak in the 72 mN/m Köhler curve). Or in case of a measurement of critical supersaturation in order to determine a particle's true hygroscopicity parameter at CCN activation, an incorrect parameter would be determined (one that would accidentally combine surface tension evolution effects and actual hygroscopicity in a non-trivial and incorrect way).

L182: The "discontinuity" that the authors refer to here is not entirely clear to me. Do the authors mean that each of the three variable curves shown suddenly changes relatively to assuming a constant surface tension of 30 mN/m? This is not clear from the words, as the discontinuity does not occur "at its minimum surface tension" but instead when there is a (in the model used here) discontinuous increase in the minimum surface tension.

The term "discontinuity" was used because the gradient of the Kohler curve changes discontinuously when realistic surface tension profiles are included. To better reflect the changes in the function itself, we now refer to these as abrupt changes in the text. The "second discontinuity" comments have been omitted in the revised manuscript as they distract from the more important facets of the discussion.

L235: It would be helpful if the "relatively simple, phase composition- and morphology specific surface tension model" were described here, in brief. If I understand correctly, a linear mixing rule is used.

At this line in the manuscript, we now add the following brief description of the AIOMFAC-based model and mixing rules for surface tension used:

"A detailed description of this AIOMFAC-based model, its variants and sensitivities to model parameters and assumptions is given in the supplementary information of Ovadnevaite et al (2017). Briefly, the equilibrium gas–particle and liquid–liquid partitioning model is used to predict the phases and their compositions for a bulk mixture "particle", here of known dry composition, at given RH and temperature. For other applications with given total gas + particle input concentrations, the equilibrium condensed-phase concentrations can be computed as a function of RH (accounting for partitioning of semivolatiles). Assuming spherical particles of a certain dry diameter with a core-shell morphology of liquid phases in the case of LLPS, density information from all constituents is used to compute volume contributions and the size of the particle at elevated RH. In addition, the surface tension of each individual liquid phase is computed as a volume-fraction-weighted mean of the pure-component surface tension values. In the case of LLPS, the surface coverage of the organic-rich shell phase is evaluated by considering that it must be greater than or equal to a minimum film thickness (monolayer as a lower limit); this determines whether complete or partial surface coverage applies for a certain wet diameter. The effective surface tension of the whole particle is then computed as the surface-area-weighted mean of the surface tensions of contributing phases. This way the surface tension evolves in a physically reasonable manner as a droplet grows, including the possibility for abrupt transitions from a low surface tension, established due to full organic droplet coverage under LLPS, to partial organic film coverage after monolayer film break-up, and further to complete dissolution of organics in the aqueous inorganic-rich phase (single aqueous phase). This equilibrium model is here referred to as AIOMFAC-EQUIL. We also employed two AIOMFAC-CLLPS model variants, in which the organic constituents are assumed to reside constantly in a separate phase from ammonium sulfate (complete LLPS, an organic film), either with or without water present in that phase, discussed in Section 4.”

L235: It would also be helpful if the authors were to clarify whether the model predicted the surface activation effect completely independent of the observations, or whether some amount of model tuning was included (perhaps associated with exactly what species were chosen to represent the organic phase).

In the study by Ovadnevaite et al. (2017) a predictive model based on AIOMFAC was employed. The predicted surface tension evolution by that model is a result of the calculations when liquid–liquid phase separation is included; no model tuning was performed concerning the model physics. However, the reviewer is correct that the selection of surrogate species, used to represent a particular “ambient” organic-inorganic aerosol system, will affect the predictions. In the case of the Ovadnevaite et al. study, the selection of surrogate species and their concentrations were constrained by available observations on the chemical composition of the aerosols. Obviously, no molecular-level composition information was available and as such, a range of options may be considered. Hence, the choice of system components and compositions may affect predicted CCN properties, however, beyond that, the surface tension and particle size as a function of water activity was purely a result of the computations. Extensive results and sensitivity calculations on composition and physical parameters are documented in the SI of Ovadnevaite et al. (2017) and not of relevance for the discussion in this technical note.

Concerning the modelling results described in this work, the organic to salt ratio, hence dry composition, is known and no tuning of input parameters was performed. Instead, we rely on physical parameters established independently of this work to assign feasible input parameters and compare with our experimental observations. Fitting a model to these data would not yield insight into whether our understanding of several key processes is complete. Indeed, our conclusions are that we do not have an accurate model description of the process over a range in dry particle sizes, and so we are still some way off from a predictive framework.

L244: I suggest that the authors modify the sentence ending “process” to say “process, for these systems.” It is not clear that this is the case for all systems. If it were, we might expect to see a lack of closure more generally in ambient measurements.

We have amended the manuscript to reflect this clause.

L251: What is stated does not appear to be the case for the example shown in Fig. S3.7.4 in the Ovadnevaite paper.

The figure panels corresponding to the equilibrium AIOMFAC model with LLPS in that study show activation occurring at the point when surface tension returns to the value for pure water (Fig. S3.7.4a by Ovadnevaite et al.). This is in contrast, for example, to the prediction by the same model for the same system and conditions by dry particles of 41 nm diameter, which is why we point to a size dependence when the surface tension at the activation point is discussed. Other models, including the organic film variant, predict a reduced surface tension at the activation point even for larger dry diameter, yet also that variant points to a size dependence of the surface tension at activation.

L305: I do not find it clear what the authors mean when they state that the compressed film model "more closely predict[s] the observations." More closely than what? The simple model assuming a constant kappa_org = 0.35 seems to do quite well. I do get that the authors are working to say that kappa_org = 0.35 is not the correct answer and thus the good agreement is not for the right reason. Yet this discussion requires clarification.

We agree this is confusing, and that statement has been removed, replaced with:

"The compressed film model decouples the water activity component from the surface tension component, and thus should better represent the physical processes at work. However, the agreement for all these models breaks down further when looking at different sized particles."

as a concluding remark prior to our discussion of size dependencies.

L316: Is the compressed film model here simply inadequate, even with determination of new parameters? Or can it work if it were returned to this dataset? If the latter, this would give some indication of how uncertainty in the derived parameters for the model impacts subsequent calculations.

In the current compressed film model framework, no combination of parameters was able to reproduce the trends for all size regimes considered. Some imposed additional size dependencies on the input parameters might produce good agreement, but at that point the model becomes a multivariable fit rather than a predictive physical framework.

L328: This is only true for the smallest particles.

This is stated on line 327 referring to the 40 nm particles.

L330: I am not seeing entirely what information is lost by choice of procedure or data representation. Both ways of representing the data seem to drive one to similar conclusions: for a given representation the assumption of a constant kappa_org can allow for model-measurement agreement, but the kappa_org values are not necessarily realistic. I do not understand why the authors couldn't similarly apply the models shown in Fig. 2A/B to the data in Fig. 2C? There need not be loss of information.

We are trying to show in these figures that by including size and composition in a single axis (as in the case of Figure 2C with D_{coated}), it becomes much harder to discern whether the model is doing a good job or not. The smallest size particles, which show the biggest surface effects when predominantly organic in nature, are at low organic volume fractions in this representation (fixed inorganic seed size), and the larger particles that do contain more organic show less surface sensitivity. Hence, for the same data, it might be reasonably concluded that a fixed value of kappa_org results in good agreement across the dataset, when in reality it is not fully capturing all the dependencies. There is no true loss of data, and we have amended our comments in the manuscript to better reflect our meaning.

“Thus, the choice of experimental procedure or data presentation can impact the interpretation of experimental observations and we caution care when presenting such data. When composition and particle size are coupled in D_{coated}, the sensitivity to surface tension effects is diminished, as smaller particles will contain a lower volume fraction of organic material”

L348: It is not clear to me how these are AIOMFAC calculations any longer, if the organic and salt are forced into separate phases. Does one even need AIOMFAC to do this?

The non-ideal mixing among the compounds within each phase is still treated with AIOMFAC, necessary to determine the water content as a function of RH (of the inorganic core phase or of both phases, depending on the model variant). The AIOMFAC-CLLPS (w/ org film) model is a simplified implementation of a possible, yet extreme case of phase partitioning and serves as a starting point. We are not presenting this model as the answer; indeed more questions are raised by the success of such a simple implementation in predicting surface effects in this case study.

L361: How is it that the essentially made up model (assuming a complete surface film) does better than the model that explicitly accounts for the system thermodynamics? Is this just saying that the thermodynamic model is wrong (e.g. bad parameterization or parameterization used outside the RH range where it is valid)? Or is there perhaps some kinetic effect that must be accounted for? The discussion that comes a few sentences later seems to suggest perhaps the former. But, some additional and more explicit discussion is necessary, rather than a statement (line 375) that this finding is “intriguing”.

This is indeed an interesting point, and the answer is we don’t know. Clearly the more rigorous thermodynamic models miss something, such as how interfacial energies between liquid phases (not considered explicitly) might drive the system towards enrichment of the organic phase. The AIOMFAC-CLLPS (w/ org film) presented here is a limiting case in this treatment of the system and is an educated first guess at the solution. Kinetic effects may be in play, but typically these would make activation harder (dissolution kinetics, water uptake kinetics etc.).

Minor Comments:

Fig. 1: It would be good to change from the red/green color scheme to one that is more friendly to color blind people.

Done.