Editor Comments

We thank the editor for his insightful comments to our manuscript. We address his points below.

1) Studying near-saturated ammonium sulphate solutions in the context of cloud condensation nuclei (CCN) activation seems to be conceptually wrong. CCN activation occurs at relative humidity (RH) > 100% while aqueous ammonium sulphate solutions are saturated at RH around 80%. A sound discussion of this fact is certainly needed, including the question whether and how any of the presented results can be applied for dilute ammonium sulphate solutions of droplets at the point of CCN activation.

We thank the editor and reviewers for bringing this point to our attention.

This study focuses on aerosol conditions (high salt content, low pH and complex organic mixtures) and provides experimental surface tension data at these conditions. While these conditions are not appropriate for Köhler Theory (which describes the condition of particle activation), this study does provide insight into the presence of organics at the gas-aerosol interface, important for both heterogeneous chemistry and ice nucleation (McNeill et al., 2013).

We have added emphasis throughout the manuscript to the importance of surfactant films on aerosols for heterogeneous chemistry and ice nucleation. We have adjusted the discussion to be clearer and better describe our recommendations for modeling aerosol surface tension and modeling aerosols at particle activation conditions. It now reads, "The compositions of the experimental systems described within this work mimic aerosol conditions near 100% RH (near-saturation salt concentrations). For aerosol systems at higher relative humidity (> 100% RH), i.e. those at the condition of particle activation, the use of salt parameters may be inappropriate due to the higher water content at this point. Based on our results for organics in aqueous systems (Fig. 1, S1-3), to describe particle activation, if the dilute salt concentration is known, the Tuckermann approach (Eq. 5 or 6) is preferred; if the ionic concentration is unknown, we recommend the use of the Henning model with water fit parameters.

For aerosol conditions (up to 100% RH), we recommend the use of salt fit parameters in the Henning model (Eq. 3) over the Tuckermann approach, which uses water fit parameters (known more readily than salt fit parameters) but also requires an additional empirically determined parameter, k. The Henning (with salt parameters) and Tuckermann models provide similar modeling fits and goodness of fit (χ^2) values, yet the Henning model is overall a simpler and more physical approach. We recommend that the Henning model, with experimentally determined salt parameters, be used to incorporate implicit salt effects on complex mixtures in future aerosol surface tension studies and predictions of surfactant film formation in aerosols."

2) I do have serious concerns about the statistical method used to determine "the standard deviation of the model fit" shown as grey dashed lines in the figures:

- Please specify the statistical definition and meaning of these curves in more detail.

We have clarified the definition in Section 3, Results: "In all figures, the black dots represent the experimental data, the black solid line is the model fit, and the gray dotted lines show the standard deviation of the model fit (determined from the error associated with the fit parameters of the S-L equation, a and b)."

- Is this a good measure of the fit uncertainty in the applications shown here, given the fact that sometimes all experimental data points are outside the range spanned by grey lines?

The error associated with the fit parameters shows the "goodness" of using the S-L equation to describe the experimental data. If experimental data points are outside the range of the standard deviation of the S-L equation, this simply indicates that the S-L equation does not describe the data well.

- How can it be that the fitted model curve is often outside the range spanned by the grey lines that represent the "standard deviation of the model fit" (Fig. S7B is probably the most extreme example)!?

We agree that the standard deviations of the model sometimes appear strange. Depending on the error associated with the fit parameters (a and b), the standard deviation of the model fit drastically changes (i.e. there can be sign changes for certain terms within the Szyszkowski-Langmuir equation). We have checked and the standard deviations shown are, indeed, correct based on our calculations.

Further remarks:
3) Treatment of temperature dependence:
The surface tension of pure water or any aqueous solution is temperature dependent, as correctly stated in the introduction. Therefore, both σ and σ_0 in equation 1 are temperature dependent (likewise for Eqs. 3-6). This temperature dependence should explicitly be expressed in these equations.

We have added the explicit temperature dependence to these equations.

4) Surface tension measurement (p. 555, l. 23):
"Briefly, droplets of the bulk solutions formed at the tip of a 100 μL syringe, where they equilibrated over time scales of 2–5 minutes …" - It needs to be clarified whether or not the exchange of water molecules between the droplet and the surround gas changes the concentration of the solution. If so, by how much? Would this introduce significant experimental uncertainty?

We have not measured the change in composition of the solution before and after measurement; however, from previous studies (see Li et al., 2011), we know that sufficient amounts of volatile species (acetaldehyde and formaldehyde) remain in the particle phase to suppress the surface tension of the mixtures over the time periods that we used for this study (either by becoming less volatile when hydrated of forming oligomers). We have not taken volatility effects of these organics or water molecule gas-liquid exchange into account in terms of the supplied organic concentrations; however this would be an interesting subject for future experiments.

5) P. 557, last line: "σ_0" will depend on the reference solution (0.05 M acetaldehyde or 0.05 M glyoxal or 0.05 M methylglyoxal).

For Eq. (1), (3) or (4), the S-L equation, and the Henning and Schwier2010 models, respectively, σ_o *indicates the surface tension of the solution in the absence of organics. For aqueous solutions, this is the surface tension of water; for ionic solutions, in our case, it is the surface tension of 3.1 M* $(NH_4)_2SO_4$*. We always use these surface tensions as our reference points, and we have added the reference surface tension in the absence of organics as a dotted line to each figure to more clearly show the surface tension depression from the presence of organics.*

6) Please start the X-axis of all figures at zero, which corresponds to the "reference solution" (e.g. 0.05 M acetaldehyde in Fig. 1) without addition of the extra organics (e.g. leucine in Fig. 1). Please add a marker for the surface tension of the reference solution (from literature) in all figures, as this is an important value for interpreting the effects of adding extra organics, and as this value is included in the fit curves too (if I got that right).

We apologize for the lack of clarity in regard to the figure plotting: Currently, the x-axis for the figures is shown as the total organic concentration (variable organic + constant organic), and the first point of each figure is that of the constant concentration organic (0.05 M acetaldehyde, methylglyoxal, or glyoxal) in the absence of the varying organic when applicable (Figure 1-4, S1-9). This "reference solution" point cannot be at zero, because then the x-axis would solely be in terms of the varying organic rather than the sum of all organics present in the solution, and we believe that plotting the data in this way would actually be more confusing to the reader. Including the reference point of no organics (either 72.5 dyn cm^{-1} *for aqueous solutions, or 78.5 dyn* cm^{-1} *for mixtures in 3.1 M* $(NH_4)_2SO_4$*) actually distorts the figures so that the model fits and experimental data are no longer the focal point. We do agree with the editor that this reference data should be included in the plots for clarity, so we have added the reference surface tension in the absence of organics as a dotted line to each figure.*

To make the above discussion clearer to the reader, we have added the following text to Section 3, Results:

"In all figures, the black dots represent the experimental data, the black solid line is the model fit, and the gray dotted lines show the standard deviation of the model fit (determined from the error associated with the fit parameters of the S-L equation, a and b). The black dashed line is the reference surface tension of each solution in the absence of all organics (72.5 dyn cm^{-1} *for aqueous solutions, 78.5 dyn* cm^{-1} *for ionic solutions). The x-axis is always given in terms of total organic concentration, so when applicable, the first point of each figure is that of the constant concentration organic (Fig. 1-4, S1-9).*

7) The number of significant digits seems to be too high for many numbers in various tables and figures.

We have decreased the number of significant digits.