

Editor comment to the manuscript entitled “Modeling the surface tension of complex, reactive organic-inorganic mixtures” by Schwier et al.

In my function as an editor of this manuscript I would like to highlight/rise two major comments that are to some extent already addressed in the formal reviews.

- 1) This paper deals with the surface tension of mixed (reactive) organic-inorganic solutions. The authors state in the introduction that their main motivation to investigate the surface tension of aqueous solutions is: “Surface-active organics can impact the cloud nucleating ability of atmospheric aerosols by lowering the surface tension, thus influencing the supersaturation required for cloud droplet activation as described by Köhler Theory”. This study focuses on near-saturated inorganic solutions with variable amounts organics (and some purely organic mixtures), motivated by the fact that “Currently, few datasets exist with surface tension information for organics in near-saturated salt solutions, typical of atmospheric aerosols”. 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.
- 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.
 - 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?
 - 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)!?

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.
- 4) Surface tension measurement (p. 555, l. 23):

“Briefly, droplets of the bulk solutions formed at the tip of a 100 μ L 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?

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