

In their manuscript the authors, A.N. Schwier, G.A. Viglione, Z. Li, and V.F. McNeill, present experimental results including an appropriate theoretical description for the surface tension of organic-inorganic mixtures containing 2-6 reactive species. The surface tension is a crucial property for aerosol particle hygroscopic growth and CCN activation modeling purposes. Furthermore, parameterizations for complex mixtures are usually rare. Therefore, the results are interesting and contribute to the knowledge about the processes describing the growth of atmospherically relevant aerosol particles. I recommend a publication in ACP after few minor revisions and some technical corrections:

- The authors present experimental results and model calculations for a temperature of 25°C, only. As the surface tension is a function of temperature, it would be useful to see at least selected experimental data also for different conditions. The authors show state/discuss, whether the given fit parameters (which were derived for a temperature of 25°C) together with the applied model equations can be used to accurately reproduce the surface tension at different (lower) temperatures or not?
- In the conclusions, the authors recommend to use the salt fit parameters together with the Henning model (Eq. 3) for saturated solutions (with respect to $(\text{NH}_4)_2\text{SO}_4$). However, in many cases the solutions are more diluted (e.g., particle activation). The authors should give suggestions how to proceed with such solutions. Or in other words, up to which $(\text{NH}_4)_2\text{SO}_4$ concentration is it suitable to apply the water fit parameters? It seems that for diluted solutions the modified Tuckermann approach (Eq. 6) gives better results and should be preferred.
- In the discussion/conclusion part the authors should explain in more detail, why it can be stated that the reaction products of two structurally different organic species have a surface tension behavior, which is different from those of the reactants, while this is not the case for

two structurally similar species. The whole paragraph is not very conclusive in its current version.

- p. 554, l. 12 and also p. 555, l. 10: It might be not necessary, but I missed the conditions (temperature), at which the samples were stored for equilibration?
- p. 554, l. 21: "...known presence in atmospheric aerosols and ice nuclei..." - I guess, the authors mean "atmospheric (cloud) condensation nuclei (CCN) and ice nuclei (IN)..." because IN are part of the aerosols.
- p. 555, l. 21: "Our approach for surface tension measurements has previously been described (Li et al., 2011; Schwier et al., 2012)" - The given reference is a bit misleading because the method is not explain therein (at least not in more detail than in the current manuscript). Therefore, I would suggest to refer the older paper of Shapiro et al., 2009 (ACP).
- p. 555, l. 24: "measurements... at room temperature (approximately 25°C)." - What does this statement mean? What is the uncertainty of the given temperature value? The surface tension is largely influenced by the temperature, and with respect to the number of significant digits given in Tab. S1, the exact temperature has to be given. Even small temperature fluctuations of only ± 0.1 K do not allow to determine the surface tension with such a high precision (given are 5 digits).
- p. 557, l. 3: "although most aerosols are saturated with respect to the inorganic" - This statement holds for low relative humidities. At high rHs (e.g. at the point of CCN activation) this is not the case. At high rH, the solution is (usually) highly diluted. Furthermore, at low rH (and consequently saturated solution) solution effects, characterized by activity coefficients, are more crucial parameters for modeling than the surface tension. Thus, the authors should put their statement into the perspective that the considered (almost) saturated solutions represent somehow the upper limit of the conditions relevant for the atmosphere.
- p. 556-557: Is it necessary to use two different variables describing the same term (σ_0 and σ_{H_2O})? σ_{H_2O} can be also applied for Eq. 3 and Eq. 4.
- I recommend to use the same x-axis for all diagrams. There is no need to vary the scales for each diagram. For a better readability of the

labels on the x-axis it would be also helpful to slightly increase the blank space between the diagrams, respectively.

- What was the reason to do the model calculations only at a very small number of data points? I suppose, the calculations are not very computationally expensive. Due to the few data points, the black and gray lines have very sharp edges. The lines could be much smoother for smaller calculation steps.
- p. 558, l. 3: I would add a comma after "(W)"
- p. 558, l. 20: this sentence should be rewritten.
- p. 562, l. 4-5: "Washburn, 2003" - I recommend to refer to the corresponding page or number of the table.
- p. 563, l. 14: "Mannich reaction products may have significantly different surface-bulk partitioning and surface-activity behavior from the reactants..." - than the reactants?
- p. 563, l. 27-29: "determined salt parameters be used..." - a word is missing here
- The authors are not consistent using "Figure" and "Fig.", e.g.: p. 559, l. 19 and p. 561, l. 10
- I recommend to show Tab. S1 in the manuscript and not in the supplemental part. The given experimental fit parameters are, at least for my point of view, essential results of the manuscript, although some of these data have been already published elsewhere (Li et al., 2011; Sareen et al., 2010). The authors also should add the reference to these data (the data, which have been already published). It should be also explained why several values given in Tab. S1 differ from the older ones (e.g., Acetaldehyde). Are these data newly fitted values? The authors should state more precisely the origin of the fit parameters given in Tab. S1. Applying fit parameters, derived from measurements of the same binary solutions, which were also used to prepare the more complex mixtures, would increase the confidence of the conclusions given in manuscript.
- Tab. S1: The authors should comment the standard deviations given for the determined experimental fit parameters. The values are (partly) quite high. Would it be possible to reduce the standard deviations by fitting a larger number of experiments?

- In Fig. S1-S5 the standard deviations of the model calculations (gray lines) seem to be wrong.
- Several entries in Tab. S1 are missing but are necessary for the model calculations (Glyoxal). The authors should add these fit parameters.
- Tab. S2: Results of the Tuckermann (2007) approach (Eq. 5 in the manuscript) are not shown in the manuscript. Tab. S2 can be therefore omitted or calculation results should be added to Tab. 3.
- Concerning the inorganic term ($\frac{\Delta\sigma}{\Delta c_{salt}}$) in the (modified) Tuckermann approach (Eq. 5 and Eq. 6): Was the value for this term taken from literature or was it determined numerically? The applied values should be given in the manuscript.
- References:
 - Several doi-numbers are missing. The authors should complete the list. (e.g., Booth et al. - doi: 10.1039/B906849J)
 - Hyvärinen et al.: "Journal of Chemical and Engineering Data" → J. Chem. Eng. Data
 - Washburn et al.: "1926-1930;2003" → a blank is missing
 - Setschenow, J.Z.: The correct titel is "Ueber die Constitution der Salzlösungen auf Grund ihres Verhaltens zur Kohlensäure"