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Surface tensions of multi-component mixed inorganic/organic aqueous systems of atmospheric significance: measurements, model predictions and importance for cloud activation predictions

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Abstract. In order to predict the physical properties of aerosol particles, it is necessary to adequately capture the behaviour of the ubiquitous complex organic components. One of the key properties which may affect this behaviour is the contribution of the organic components to the surface tension of aqueous particles in the moist atmosphere. Whilst the qualitative effect of organic compounds on solution surface tensions has been widely reported, our quantitative understanding on mixed organic and mixed inorganic/organic systems is limited. Furthermore, it is unclear whether models that exist in the literature can reproduce the surface tension variability for binary and higher order multi-component organic and mixed inorganic/organic systems of atmospheric significance. The current study aims to resolve both issues to some extent. Surface tensions of single and multiple solute aqueous solutions were measured and compared with predictions from a number of model treatments. On comparison with binary organic systems, two predictive models found in the literature provided a range of values resulting from sensitivity to calculations of pure component surface tensions. Results indicate that a fitted model can capture the variability of the measured data very well, producing the lowest average percentage deviation for all compounds studied. The performance of the other models varies with compound and choice of model parameters. The behaviour of ternary mixed inorganic/organic systems was unreliably captured by using a predictive scheme and this was dependent on the composition of the solutes present. For more atmospherically representative higher order systems, entirely predictive schemes performed poorly. It was found that use of the binary data in a relatively simple mixing rule, or modification of an existing thermodynamic model with parameters derived from binary data, was able to accurately capture the surface tension variation with concentration. Thus, it would appear that in order to model multi-component surface tensions involving compounds used in this study one requires the use of

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appropriate binary data. However, results indicate that the use of theoretical frameworks which contain parameters derived from binary data may predict unphysical behaviour when taken beyond the concentration ranges used to fit such parameters. The effect of deviations between predicted and measured surface tensions on predicted critical saturation ratios was quantified, by incorporating the surface tension models into an existing thermodynamic framework whilst firstly neglecting bulk to surface partitioning. Critical saturation ratios as a function of dry size for all of the multi-component systems were computed and it was found that deviations between predictions increased with decreasing particle dry size. As expected, use of the surface tension of pure water, rather than calculate the influence of the solutes explicitly, led to a consistently higher value of the critical saturation ratio indicating that neglect of the compositional effects will lead to significant differences in predicted activation behaviour even at large particle dry sizes. Following this two case studies were used to study the possible effect of bulk to surface partitioning on critical saturation ratios. By employing various assumptions it was possible to perform calculations not only for a binary system but also for a mixed organic system. In both cases this effect led to a significant increase in the predicted critical supersaturation ratio compared to the above treatment. Further analysis of this effect will form the focus of future work.

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