

***Interactive comment on “Surface tensions of multi-component mixed inorganic/organic aqueous systems of atmospheric significance: measurements, model predictions and importance for cloud activation predictions” by D. O. Topping et al.***

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Topping et al. have compared measured surface tensions of multicomponent inorganic/organic aqueous solutions to those calculated using surface tension models that use either one-component or binary surface tension data as input. In general, it is found that models using binary data perform better in predicting multicomponent surface tensions. The different surface tension models are then used in calculating critical

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supersaturations for particles of different compositions, and it is found that the critical supersaturation is sensitive to the surface tension model especially at small particle sizes.

I believe that the work done on measuring multicomponent surface tensions and evaluating different models against the measurements is very valuable and should definitely be published in ACP, and I believe that referees 2-4 have commented adequately regarding the surface tension work. However, I am not entirely happy with the critical supersaturation calculations. First of all, the title of section 5 (Activation predictions) is misleading. That a particle exhibits a Köhler maximum at a given supersaturation does not necessarily mean that it is able to activate to a cloud droplet at that SS. Cloud drop formation is a complex process, and dynamic factors as well as the particle size distribution have an effect on the activation. I suggest that the title of section 5 is changed to "Critical supersaturations" (activation is mentioned in several other places, a similar change should be made where appropriate.)

A more important concern, however, is the neglect of bulk-to-surface partitioning of the organics. The authors state on p. 12086 that "Whilst some theoretical studies suggest the effect of highly surfactant compounds may alter the water activity by decreasing the effective number of molecules in the Raoult term, experimental studies for atmospherically relevant species have not been carried out nor has the effect of non-ideality been explored, and as such is not considered here (Sorjamaa et al., 2004)."

First, not only is the Raoult term affected: the depletion of surfactants from the bulk of the droplet decreases the bulk concentration used in determining the surface tension, which is therefore increased. Secondly, the bulk-to-surface partitioning is calculated using well established Gibbs surface thermodynamics, which is not dependent on whether the surface tension depressing organic is an atmospherically relevant compound or not, and therefore experimental support of the theory for sodium dodecyl sulfate (Sorjamaa et al., 2004) certainly suggests that it is better to account for surfactant partitioning than not in Köhler calculations, regardless of the type of the surfactant.

Thirdly, we have recently shown that the partitioning affects Köhler maxima (and cloud drop activation) also with clearly less surface active compounds than SDS (Sorjamaa and Laaksonen, 2006, Kokkola et al., 2006). Finally, concerning the non-ideality: it is true that the activity coefficient terms in the Gibbs adsorption equation have been approximated as unity in the partitioning calculations. However, the same approximation is done also in evaluation of the Raoult term in any case, so I do not think that the fact that "non-ideality has not been explored" is a good argument for completely ignoring the partitioning, especially as the available experimental support for the partitioning theory (Sorjamaa 2004) suggests that the non-ideality is a minor issue.

I would also like to point out that as the bulk-to-surface partitioning depends on the gradient of the surface tension, the relative differences of Köhler maxima (and not just their absolute values) obtained using the different surface tension models may change somewhat when the partitioning is accounted for.

In conclusion, I do not find good reasons for ignoring the partitioning in the Köhler calculations, and recommend that the authors should repeat the calculations before the paper is accepted to ACP.

#### References:

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