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.

Anonymous Referee #4

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The authors have measured the surface tensions of binary, ternary and higher order multicomponent aqueous mixtures, containing both organic and inorganic constituents. Related to this, the authors test several different approaches to model the surface tensions of these mixtures. Finally, the authors demonstrate the sensitivity of the critical saturation ratio to the surface tension of the solutions. One of the main conclusions of the manuscript is that data on binary aqueous systems is needed to accurately model
the surface tensions of higher order mixtures. The results and conclusions of this manuscript are unique and certainly valuable for the aerosol modeling community. I recommend the manuscript to be published in ACP, after the minor comments below have been properly addressed:

General comments: The manuscript is in some parts difficult to follow because 1) some slight structural inconsistencies and 2) all the relevant parameters and concepts are not explained / consistently referred to. These problems will be particularly pronounced, if the reader does not have a sufficient background in thermodynamics of mixtures. The authors should carefully check that all the relevant variables and concepts as well as the used models are first explained comprehensively and referred to consistently later on in the text.

Specific comments/technical corrections:

Abstract, p. 12058, line 16; (also Sect. 3.2.2, p. 12073, line 19): The authors state that ‘...two predictive models found in the literature provided a range of values...’ Can you speak of a range when you actually have two point results?

Introduction, p. 12060, line 22: $R$ and $T$ should be explained as well.

Sect. 2.1: The authors explain how the binary and multi-component (number of constituents higher than 3) samples were chosen and prepared. Why do they skip the ternary systems in this context? In my opinion it would be logical to say a couple of words about those as well.

Sect. 2.1, p. 12063, line 13: The reference Svenningsson et al. (2006) should be in parentheses, i.e. (Svenningsson et al., 2006).

Sect. 2.1, p. 12063, line 20: In the end of the section 2.1, the authors state ‘The ability of models chosen in this study to reproduce the behaviour of such (multicomponent) systems are discussed in Sects. 4.3 and 4.4’ Why do the authors only refer to the comparisons made for the multicomponent systems, as the same kind of comparison
was made for binary and ternary systems as well? I think it would be consistent to refer to these comparisons (Sects. 4.1 and 4.2) as well.

Sect. 3.1, p. 12064, lines 23-25: The concepts 'Gibbs dividing surface' and 'Langmuir adsorption isotherm' should either be shortly explained or comprehensive references should be given.

Equations 2 (p. 12065, line 2), 6 (p. 12067, line 23), 9 (p. 12069, line 27), 11 (p. 12070, line 11), 15 (p. 12074, line 19), 16 (p. 12075, line 2), 27-30 (p. 12079, lines 2-5 and 7): The notation of the natural logarithm should be consistent in all the equations. Now varies between ‘Ln’ and ‘ln’. I would suggest ‘ln’.

Equations 2 (p. 12065, line 2), 10 (p. 12070, line 7), 15 (p. 12074, line 19), 16 (p. 12075, line 2), 27-30 (p. 12079, lines 2-5 and 7): The notation of the adsorption equilibrium constant should be consistent in all the equations. Now varies between ‘K’, ‘k’ and ‘k’. I would suggest ‘K’.

Equation 2 is exactly the same as Eq. 11. Why do they both need to be presented?

Sect. 3.1, p. 12065, lines 19-22: The authors should explain what 'competing adsorption' means.

Equation 6, p. 12067, line 23: Should be denominator in the logarithm be ‘x_i gamma_i’? Also, the variable N should be explained.

Several places in the manuscript, for instance, p. 12068, line 24; p.12073 lines 7, 8, 10, 17; p. 12097, Table 2: The correct reference for ‘Yens-Woods/Yens-Wood/Yens and Woods’ method should be checked. In my opinion the correct way to cite this method would be ‘Yen-Woods’.

Sect. 3.2.2, p. 12071, line 15: Should ‘sudgen’ be replaced by ‘Sudgen’?

Sect. 3.2., second para: Why is the temperature dependence of glutaric acid surface tension presented here? Is it used in the work, and if it is, what about the other com-
pounds? I find this para a little unconnected to the rest of the manuscript.

Sect. 3.2.2, p. 12073, line 20: Should you refer to Table 2 instead of Table 1?


Sect. 3.3, p. 12078, line 16: Table 4 should be referred to before Table 5, or the order of the tables should be changed accordingly.

Sect. 4.1, Tables 1a, 2 and 3, Figs. 3: In Tables 1a, 2 and 3 citric acid is mentioned. Why is not the results related to it presented in Figs. 3/ mentioned at all in the text?

Sect. 4.2, p. 12081, line 6: Why was the mass ratio 45:55 chosen in one of the ternary systems, whereas in the other ones the ratio was 1:1?

Sect. 4.2, p. 12081, lines 8 and 27: Should Table 5 be cited instead of Table 4?

Sect. 4.3, p. 12082, lines 16 and 19: Should Table 6 be cited instead of Table 5?

Sect. 4.4, p. 12083, line 18: Should Table 7 be cited instead of Table 6?

Sect. 5: In several places the authors state that as the dry size increases, the aqueous solution in the droplets gets more dilute. The physical background of this should be explained. Sect 5, p. 12087, line 12: I assume that the authors mean Table 7 instead of Table 6.

Sect 5, p. 12088, line 6: ‘Figure 8’ should probably be replaced by ‘Figure 9’, and I assume that you mean ‘Figs 5-8’ instead of ‘Figs. 5.1-5.4’

Sect 5, p. 12088, line 15: A space should be entered between ‘40’ and ‘nm’.

Table 2, p. 12097: The reference ‘Marrero and Gani (2001)’ is not in the reference list. In the text (Sect. 3.2.1, p. 12068, line 27) the authors cite Topping et al. 2005b in the corresponding context. Also the meaning of all the variables in the Table should be explained. The same applies for all the tables.
Tables 3a-3d, p. 12098-12101, and Figures 1a-1c, p. 12106-12108: Why are the systems not presented in the same order in the tables and figures? It would be easier to follow the results if the order of the systems was the same. And: what happened to citric acid?

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 12057, 2006.