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Atmospheric Chemistry and Physics Discussions

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 #2

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The authors have measured surface tensions of several solutions relevant to atmospheric aerosols. The investigated solutions contain various organic compounds and (in some cases) additionally inorganic salts. The authors have also attempted to model the experimental results using several different predictive methods and their combinations. The work shows that entirely predictive methods may fail to reproduce accurately the surface tension of atmospheric aqueous aerosols which typically contain

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complex mixture of various compounds, whereas methods utilizing experimental data for binary mixtures give more accurate results for complex solutions. The dataset provided is novel and so is the modeling approach. Given that surface tension is a key quantity when modeling cloud droplet activation, aerosol water uptake or properties of nanometer-sized particles, the results are not only valuable for researchers working with aerosol thermodynamics but to a large part of the aerosol modeling community. I recommend the publication of the manuscript in Atmospheric Chemistry and Physics after the authors have addressed the following relatively minor issues.

General comment:

Since the results presented in the study have potentially a large range of applications, the authors should make the work more accessible to a reader lacking strong background in thermodynamics. To this end, several improvements to the manuscript should be made. First, the authors do not explicitly define several key terms in the text (such as competitive and non-competitive adsorption, “coupled inorganic/organic approach” and “LiLu full model”) nor explicitly state the corresponding equations. Also the authors should clarify the definitions of some other terms (e.g. ideality and semi-ideality) and, when discussing about the results, remind the reader about the meaning of the key terms. Appropriate revisions should be thus made to Sections 3.3 and 4. Second, the authors test several combinations of different modeling approaches (the authors call them “permutations”, I would prefer term “combination”) but do not give the corresponding sets of equations. Therefore the authors should make sure that the reader does not have to guess about applied equations. Also, the authors should use consistent terminology when referring to different models and their combinations.

Specific comments:

1. Abstract. The sentence ending with phrase "...and this was composition dependent" is a little bit unclear (line 20, p. 12058). The abstract contains also non-scientific/qualitative terms: “realistic” (line 21, p. 12058) and “significant differences”

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(line 6, p. 12059).

2. Introduction, p. 12059. The parameters R and T should be defined in connection with eq. 1.

3. Introduction, p. 12060, line 12. It should be made clear to which term in equation 1 the Raoult effect refers to.

4. Section 3.1., p. 12064-12065. The discussion about three groups of calculation(?) proposed by Hu and Lee (2004) is a little bit vague. Also, “ln” not “Ln” in eq. (2) and in subsequent equations as well. Finally, the reader is forced to guess what is meant by two different schemes of Li and Lu (2004). Does the difference lie in treating adsorption - competitive or non-competitive? If so, material in the last paragraph of the section should be presented earlier.

5. Section 3.2, p. 12066. What does “linear dependence” (line 9) mean?

6. Section 3.2.1., p. 12066-12068. The term “N” is not defined in eq. 6. The parameter “A” is lacking indices (p. 12068, line 12). Also, the authors should give at least references discussing the validity of eq. 7 (p. 12068, lines 19-20).

7. Section 3.2.1., p. 12069-12070. Equations 11 and 2 are identical. Are they derived in similar fashion? If so, why the derivation is not presented in connection with eq. 2 which would be the logical order?

8. Section 3.2.2, p. 12072. The second paragraph is out of the context of the section (lines 5-20). It remains unclear if eq. 13 is used at all in the work. Moreover, why results are presented only for glutaric acid? Finally, can temperature dependence be incorporated into the “LiLu model” and if so, how?

9. Section 3.3., p. 12073-. Tables 3 and 4 seem to have changed places in the text - the authors should make sure that the contents of the tables are presented in the right order throughout the manuscript.

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10. Page 12074, line 6. What is “n”?

11. Page 12075, lines 12-13. Two typos appearing here should be corrected (“von Szyszkowski” and “Laungmuir”).

12. Page 12075, line 19. Word “of” is missing between the words “coupling” and “intermolecular”.

13. Page 12077, equation 22. Should the left-hand side of the equation contain omega with an overbar?

14. Page 12078. Line 16: What is “full” LiLu model? Moreover, the step from (23) to (24) needs justification since the result does not follow simply by taking logarithms of both sides of (23). Also, index “sol” is missing from the left-hand side of (24).

15. Page 12079. Line 2: Here eq. 11 is used, not 13. Equation 27: one overbar seem to be missing.

16. Section 4.1. Page 12080, lines 2-3. Actually many of the studied solutions do not display notable decrease in the surface tension compared to pure water over the studied mole fraction range (e.g. oxalic and malic acids, levoglucosan). Referring to my general comment (see above), the authors should make sure that the reader understands which model combinations and which equations are used when referring to e.g. “Tamura mixing rule” or “Suarez method”. This applies to Section 4 in general.

17. Section 4.4., p. 12083-12084. The results for “Multi 1” are not explicitly discussed in contrast to other higher-order solutions.

18. Page 12086, lines 1-2. “highly surface-active”, not “highly surfactant”.

19. Fifth and sixth paragraphs of Section 5 (p. 12086-12088). The differences should be indicated in percentages using reasonable accuracy.

20. Sixth paragraph of Section 5 (p. 12087-12088). The result that methods based on utilizing binary data may yield unphysical predictions for more complex systems is

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important since it may limit the applicability of such methods. This point should be discussed in more detail and brought out in conclusions and in abstract.

21. Last paragraph of Section 5 (p. 12088). The paragraph contain some qualitative and vague sentences. These are the first sentence and sentence “Calculations carried out here...in a consistent theoretical framework”. Also, critical supersaturations predicted by different models converge with increasing particle size and, on the other hand, only particles having dry diameters $>$50 nm (roughly) are able to activate under tropospheric conditions. Therefore it would be interesting to get a rough estimate on the relevance of these differences to the lower troposphere, i.e. how large differences in predicted CCN concentrations are induced by using different modeling schemes in the particle size range 50-100 nm?

22. Tables. The authors should define all symbols appearing in the tables (e.g. for Table 3). Also, referring to the general comment given above, the authors should make sure that the reader understands what is meant by terms appearing in Tables 4, 6 and 7.

23. Figures. The various red lines in Figures 5-8 should be introduced separately.

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