

Interactive comment on “Modeling the gas-particle partitioning of secondary organic aerosol: the importance of liquid-liquid phase separation” by A. Zuend and J. H. Seinfeld

Anonymous Referee #3

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Review of “Modeling the gas-particle partitioning of secondary organic aerosol: the importance of liquid-liquid phase separation” by Zuend and Seinfeld. The authors combine three models (MCM, EVAPORATION, and AIOMFAC) as a new approach to describe the partitioning of semivolatile organic compounds into particles consisting of organic and inorganic material. With this modeling approach the authors show, among other things, that alpha-pinene SOA in the presence of ammonium sulfate will liquid-liquid phase separate and ignoring liquid-liquid phase separation can lead to incorrect predictions. The combined results are insightful and offer a promising way forward for predicting liquid-liquid phase separation and gas-particle partitioning in chemical transport models. The paper is important and should be published in ACP after the authors

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have addressed adequately the following comments:

1)Page 2217, line 7-8. “hence there is no aqueous electrolyte phase present and corresponding model computations were carried out for the ammonium sulfate-free component alpha-pinene SOA and water system at 40% RH and 298K.” I could be wrong, but I assume that the solid ammonium sulfate seed will come into equilibrium with whatever aqueous phase is present. In this case, solid ammonium sulphate will be in equilibrium with an aqueous phase containing water, ammonium and sulfate ions and organics. Could the presence of dissolved ammonium and sulfate ions influence the partitioning? The authors should either indicate why the arguments above are incorrect or alternatively discuss why the presence of dissolved ammonium and sulphate ions is not important in this case.

2)Page 2220, line 5. I agree with Referee #1 that it should be made clear that the chamber experiments are mainly a test for the MCM-EVAPORATION models, not the MCM-EVAPORATION-AIOMFAC combination since in the comparison a salt-free case was assumed.

3)Throughout the document the authors assume that the equilibrium case (benchmark) has the highest level of accuracy. See for example Table 2, column 3 and Page 2228, line 18-19, “while the benchmark equilibrium case achieves the highest level of accuracy, it requires also the largest computational efforts.” In some of the previous papers using the AIOMFAC model it was pointed out that there can be considerable uncertainty when predicting liquid-liquid phase separation with AIOMFAC. For example, Zuend, Marcolli et al. [ACP, 2011] stated “there are some liquid-liquid equilibrium data sets that reveal larger deviations compared to AIOMFAC computations”. Some discussion on the accuracy of the model for predicting liquid-liquid phase separation seems appropriate here. Perhaps on page 2211, line 23.

4)The work of Song, Marcolli, et al. [ACP, 2012] showed that liquid-liquid phase separation is predicted at an O:C of 0.8 with the AIOMFAC model, but not observed in

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experiments with an O:C of 0.8 using dicarboxylic acids. From this, is it fair to conclude that AIOMFAC over predicts liquid-liquid phase separation? Is it possible that the combined models (MCM, EVAPORATION, and AIOMFAC) are also over predicting liquid-liquid phase separation for ammonium sulfate combined with alpha-pinene SOA? Please discuss.

5) In the conclusions the authors make several broad statements about SOA. Although these broad statements are likely correct they are not categorically proven in the current manuscript. For example, the authors have not proven that all SOA completely phase separate at O:C ratios lower than 0.5. The authors should re-phrase some of these conclusions slightly to indicate that these are reasonable speculations based on the conclusions from the current manuscript and also previous research.

6) Page 2218, line 4-7. Would the model results agree more closely with the lower yields of other experiments if the dimer concentrations were set to zero.

7) Page 2223 line 1-3. Please add a reference to support the comment.

8) Page 2232, line 23-25. This bullet is confusing.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 2199, 2012.