

# ***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 #1**

Received and published: 8 March 2012

This manuscript describes modeling techniques for partitioning of organic species between the gas phase and particle phase. The product output of MCM for gas-phase oxidation of  $\alpha$ -pinene is coupled with a combination of the thermodynamic model AIOM-FAC and compound vapor pressure estimation from the EVAPORATION model to estimate the formation of liquid-liquid phase separation in the particle phase, as well as the mass and composition of the oxidation products. The coupled models incorporate the effect of inorganic species, water content of aerosol, RH, temperature and organic loading levels on the liquid-liquid phase separation and organic/inorganic partitioning. Due to the computational expense, several simplified scenarios are compared to the benchmark thermodynamic calculation. It is concluded from the simplifying assump-

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tions, that proper treatment of organic gas/particle partitioning must include consideration of non-ideality in the condensed phase and the non-ideality of mixing in solutions containing organic and inorganic ions. This manuscript is well-suited for publication in ACP following minor corrections and some further discussion points.

Comments:

1. Page 2224. Holding the ammonium sulfate mass concentration constant in the simulations of high, moderate, and low SOA loading has the effect of varying the fraction of organic:inorganic (which is stated by the authors, P. 2224 lines 17-18). The higher ratio of organic: inorganic in the high loading experiments lessens the importance of non-ideal interaction between the organic and inorganic phases (also stated on P. 2224: “the influence of organic – ion interaction is diminished”). Could the authors comment on the effects of varying the organic:inorganic fractions at one mass loading (i.e., the loading and organic:inorganic fractions are co-varied in present study)? If the organic:inorganic were held constant, would the accuracy of the forced 1-phase case be highest for the low loading system (highest O:C modeled), as phase mixing is expected to increase with increasing O:C?

2. Page 2212 line 1 - 18: The parameterization used by Bertram et al., 2011 is based on a purely empirical relationship (with discussion of organic molecule polarity as a factor in phase separation or mixing). Can your benchmark model say anything additional regarding the thermodynamics that would support their parameterization?

3. Page 2217 line 6 – 10: This system is at 40% RH and the authors describe this as salt-free aqueous organic mixture, yet in Section 4.2, page 11 – 17, the authors state that the RH range 30% to 99% can be assumed to be a liquid PM phase containing inorganic ions. These two statements should be clarified. This is due to the solid seed used in the 40% RH chamber experiments, but should be clarified.

4. Page 2217-2218: Good agreement is found between the model and experimental chamber data for mass yield, but because dry seed was used, no treatment of phase

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separation was needed, and consideration of ideal vs. non-ideal behavior was also not a large influence on the model results. (P. 2217 line 10 “For such a salt-free aqueous organic mixture, the model predicts a one-phase state as the equilibrium PM-state for all models”; P. 2218 line 21: “non-ideality is not an important factor in the salt-free case”). The main conclusions of the manuscript are that both non-ideality in the condensed phase and phase separation as a consequence of non-ideal behavior need to be considered for accurate modeling of SOA composition and mass (P. 2200 lines 23-25), while the main result of the comparison to chamber data seems to be validation of the accuracy of the MCM v3.1 and EVAPORATION, with no treatment of phase separation. The purpose of the comparison to chamber data (it demonstrates excellent prediction of PM mass for a 1-phase organic system) should be made clearer to the reader.

5. Page 2221 line 18 – 19: Provide a reference for the statement “probably lens-shaped phase on a largely aqueous organic-inorganic droplet.”

6. Page 2233 line 6 – 8: Provide reference from atmospheric aerosols.

Minor Comments:

Page 2200 line 7: AIOMFAC and EVAPORATION acronyms should be defined.

Page 2200 line 16: replace “different” with “several”

Page 2200 line 18 - 20: Awkward sentence, perhaps replace “Both” with “By”

Page 2200 line 21: Replace “at high RH by more than 200%” with “by more than 200% at RH values of X%”

Page 2202 line 16: Reference Koop et al., 2011 and Poschl et al., 2011

Page 2202 line 28: Remove Lienhard et al., 2012 as it has not yet been published

Page 2211 line 19: Insert comma after “computations”; Insert comma after “allowed”

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Page 2214 line 21: replace “of” with “from”

Page 2218 line 13: define “these conditions” are you referring to low PM mass concentration or high PM mass concentrations

Page 2222 line 11-13: Awkward sentence - “For example, the bar graphs in the bottom panels of Fig. 4 show how PINIC partitions predominantly to the PM phase at high loading, is mostly found in the PM phase at moderate loading (80 %), but only to about 40% in the PM at low loading.”

Consider changing to - “For example, the bar graphs in the bottom panels of Fig. 4 show how PINIC partitions predominantly to the PM phase at high loading, is mostly found in the PM phase at moderate loading (80 %), and is found in substantial amounts in both the PM and gas phases at low loading (40% in PM phase).”

Page 2229 line 2 – 3: Use a better phrase than “highly super-linearly”

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

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