

Interactive comment on “The sensitivity of Secondary Organic Aerosol component partitioning to the predictions of component properties: part 2; determination of particle hygroscopicity and its dependence on “apparent” volatility” by D. O. Topping et al.

D. O. Topping et al.

david.topping@manchester.ac.uk

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The authors would like to thank the referee for their helpful comments and questions, which we have addressed below:

‘1. Introduction/Methodology, page 4. The authors based their equilibrium calculations on equation (1) which contains an implicit according to which semi-volatile organics are absorbed by the organic phase present in aerosols. However, many atmospheric

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organics are highly-water soluble and may therefore condense preferably on the aqueous phase (if present). Partially due to this, to my understanding, the absorbing mechanisms of aerosols in atmospheric conditions are still uncertain. Can the authors assess, even on a qualitative level, how sensitive the conclusions of the study are to the assumed absorbing phase?’

Response: In the general absorptive partitioning methodology used in this study, the partitioning function, as defined by equation 1 in the paper, depends on the concentration of existing condensed phase material, be it an aqueous or organic phase. In our calculations we have assumed that all components are completely miscible, such that water-soluble and water insoluble compounds condense to the same particle. If we were to assume the presence of distinct solvent phases, such as water and a hydrophobic organic phase, combined with appropriate thermodynamic models, we could assess the impact of phase separation. It should be noted however that we did probe the influence of non-ideality, which does provide insight into the role of salting in/salting out even assuming the presence of one solvent phase. Unfortunately, accounting for more than one phase is not a trivial task, only ternary systems studied in detail so far (Zuend et al 2010). The sensitivity of the conclusions to this additional effect would depend on the relative concentration of each phase and the effective volatility of compounds partitioning to both. With this in mind, it is still reasonable to assume that semi-volatile partitioning would occur in a multiphase droplet as the RH and temperature are altered within an instrument. The predicted increase or decrease in hygroscopicity relative to the one phase model would depend on being able to predict the changing stable thermodynamic state as the condensed phase composition changes.

‘2. Methodology, pages 6 and 7. The authors outline their approach quite nicely in 9 separate steps. However, I'd wish that the authors describe explicitly how the particle water content is calculated. This is certainly central to the paper and at the current form, this is hard to grasp because only a reference to a previous study is given.’

Response: The authors would like to apologise for the lack of clarity. Water is treated in

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the same way as every other semi-volatile component within the absorptive partitioning framework, its gas phase abundance calculated as a function of RH and temperature. In all cases, the equilibrium water activity is equal to the ambient RH using this approach, the absorptive partitioning framework simply a re-expression of the general equilibrium condition.

‘3. Conclusions and future work, pages 18-21. Are the authors aware any previous studies that investigate the timescale of re-equilibration of semi-volatile organics when the particle water content changes suddenly? The authors could make a literature survey and report briefly their findings (even negative ones).’

Response: As included in the response to referee #1, there is direct evidence that semi-volatile re-partitioning can take place provided by use of the TDMA to derive pure component vapour pressures from measurement of the shrinkage of particles of known composition (Bilde et al 2003). Similarly, there is the work of Prilse et al (2010) who studied the humidity influence on gas-particle phase partitioning of α -pinene + O₃ secondary organic aerosol who, conversely, reported a minimal effect of water content on SOA partitioning.

Minor comments 1. Page 9, last paragraph of the section. “. . .Hence on growth factor“, not “. . .thus growth factor”.

Response: The text has been changed

2. Equation 4, page 10. Please define S(D). 3. Page 11. Why 90% RH was chosen as a reference case?

Response: This has been changed in the text and 90%RH was chosen because it is at this RH that growth factors are commonly reported.

4. Page 16, last paragraph of 3.2. Please define S_{crit}.

Response: The text has been changed, this refers to the critical point on the Kohler curve at which particles activate into cloud droplet.

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5. Figure 5. The authors should consider if imposing results for levoglucosan and fulvic acid is really needed here. Right now the figure is rather cluttered which makes it difficult to grasp the essential information from it.

Response: We feel that providing reference lines for organics of differing hygroscopicity help the average reader to assess the general impacts of hygroscopicity changes resulting from semi-volatile partitioning.

References: Bilde, M., Svenningsson, B., Monster, J. and Rosenorn, T.: Even – Odd Alternation of Evaporation Rates and Vapor Pressures of C₃-C₉ Dicarboxylic Acid Aerosols, *Environ. Sci. Technol.*, 37, 1371–1378, 2003. N. L. Prisle, G. J. Engelhart, M. Bilde, and N. M. Donahue. Humidity influence on gas-particle phase partitioning of α -pinene + O₃ secondary organic aerosol. *Geophys. Res. Lett.*, 37, L01802, doi:10.1029/2009GL041402, 2010 A. Zuend, C. Marcolli, T. Peter, and J. H. Seinfeld. Computation of liquid-liquid equilibria and phase stabilities: implications for RH-dependent gas/particle partitioning of organic-inorganic aerosols. *Atmos. Chem. Phys.*, 10, 7795-7820, 2010

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