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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Inferring thermodynamic properties from CCN activation experiments: a) single-component and binary aerosols" *by* L. T. Padró et al.

L. T. Padró et al.

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Reply to Reviewer # 2

General comment:

Padró et al. report on a new method to estimate molar volume and solubility that relies on measurements of surface tension, chemical composition, and CCN activity coupled with Köhler Theory. While difficult to determine for ambient aerosols, molar volume and solubility are required for predicting the impacts of aerosols on clouds. Therefore an appropriate estimation method for these thermodynamic properties is desirable. The authors conclude that the newly developed method predicts molar volume to within 18% for single-component

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and binary mixtures when the organic mass fractions are between 50 and 100%, and is thus best applied to ambient aerosols with high organic mass fractions.

The paper is generally well written and relevant for ACP. There are no fundamental reasons to preclude its publication. Comments are made below regarding references and units to improve readability and traceability in the Köhler Theory Analysis (KTA) section; comments also are made regarding the applicability of KTA to ambient aerosols. Upon those comments being addressed, it is recommended that this paper be published in ACP.

Specific comments:

pp. 3808-3809: Seinfeld and Pandis (1997) is cited in reference to Eqs. (2) and (3), which describe Köhler theory for the single component case; the equivalent expression for the Raoult's term (Eq (3)) in the multi-component case, Eq (5), has no citation. It is suggested that the appropriate citation be added in order that details related to the modifications required for the multi-component case can be easily referenced.

Point is well taken; we have added a few references (e.g., Raymond and Pandis, 2003; Bilde and Svenningsson, 2004).

The use of appropriate units would help further describe the added terms (e.g., volume fraction), which are relevant to the derivation and application of KTA. The use of appropriate units throughout sections 2.1-2.4 would improve the readability and traceability of the derivation of KTA and the assumptions relevant to its application.

Excellent point. We've further clarified the KTA assumptions discussion.

p. 3810: The parameter omega (Eqs. (7) and (8)) is not defined (i.e., named).

Good point. "omega" is now called "CCN activity factor" (CAF).

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Abstract and Conclusions: The authors state that this method is "a powerful and ideal method for determining the CCN characteristic of ambient water soluble organic carbon" and "is a powerful tool for characterizing the droplet formation potential of ambient water soluble organic carbon". This conclusion seems a bit overstated based on the results and analysis presented. The KTA method is an important step forward in estimating the described thermodynamic properties; however its applicability to ambient aerosols remains to be determined. While the uncertainty in estimated molar volume was evaluated for uncertainty in the parameters .... For ambient aerosols, identification and quantification of constituents remains difficult and incomplete. Thus assumptions must be made as to the density of aerosol particles, as well as to the chemical characterization of particles. The effects of such assumptions on estimated parameters, and therefore estimated molar volumes, are undefined.

The reviewer raises an important issue: to apply KTA to ambient aerosol, one needs to know particle density, surfactant characteristics and composition, but this does not undermine the power and potential of the method. The fact remains that KTA provides a method (one of very few in fact) to characterize the CCN-relevant properties of ambient aerosol. We therefore stand by our statement, slightly modified as "a potentially powerful tool", to accommodate the reviewer's concerns.

As a side comment, we have addressed many of the issues raised above. Complementary measurements (measurement of water-soluble organic carbon, inorganic ion composition and surfactant characteristics) provide the constraints required for applying KTA to ambient aerosol; we have done this to characterize the water-soluble fraction of biomass burning aerosol (Asa-Awuku et al., ACPDa) and secondary organic aerosol from alkene ozonolysis (Asa-Awuku et al., ACPDb). In the current manuscript, we do not present sensitivity analysis for volume fraction and particle density, because the composition is known; Asa-Awuku et al. (ACPDa) however have done this in their analysis of a biomass burning sample; extending the uncertainty analysis to include

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particle density is not difficult and is left for future work.

References:

Asa-Awuku, A., Nenes, A., Sullivan, A. P., Hennigan, C. and Weber, R. J.: Investigation of Molar Volume and Surfactant Characteristics of Water-Soluble Organic Compounds in Biomass Burning Aerosol Atmos. Chem. Phys.,in review - a.

A. Asa-Awuku, A. Nenes, S. Gao, R.C. Flagan, and J.H. Seinfeld Alkene ozonolysis SOA: inferences of composition and droplet growth kinetics from Köhler theory analysis, Atmos. Chem. Phys.,in review - b.

Bilde, M. and Svenningsson, B.: CCN activation of slightly soluble organics: the importance of small amounts of inorganic salt and particle phase, Tellus Series B-Chemical and Physical Meteorology, 56(2), 128-134,2004.

Raymond, T. M. and Pandis, S. N.: Formation of cloud droplets by multicomponent organic particles, J. Geophys. Res.-A., 108(D15), 2003.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 3805, 2007.