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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Predicting the relative humidities of liquid-liquid phase separation, efflorescence, and deliquescence of mixed particles of ammonium sulfate, organic material, and water using the organic-to-sulfate mass ratio of the particle and the oxygen-to-carbon elemental ratio of the organic component" *by* A. K. Bertram et al.

Anonymous Referee #1

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Bertram et al. present a comprehensive study on the phase transitions of particles consisting of organics, ammonium sulfate, and water. They experimentally observe the relative humidities (RH) at which small droplets undergo liquid-liquid phase separation

(SRH), the RH at which ammonium sulfate crystallizes (efflorescence) (ERH), and where it deliquesces (DRH). From their numerous experiments and from literature data, the authors develop new parameterizations to predict the RH of such phase transitions as a function of organic-to-sulfate (org:sulf) and O:C elemental ratios. They compare these derived parameterizations to data from aerosol chamber experiments and from field studies.

This is an interesting and well written paper of importance for the field of atmospheric chemistry and physics.
A great advantage of the developed parameterizations is their simplicity. They require only knowledge of two measurable properties of aerosol particles: the O:C ratio and the org:sulf ratio. The amount of new experimental data covers a wide range of O:C and org:sulf ratios from high to low RH, allowing a characterization of relevant particle phase transitions for an important class of particles found in the atmosphere. The computationally efficient and reasonably accurate parameterizations seem very useful for the improvement of the representation of phase transitions and phase states of aerosols in chemical transport models.

I therefore highly recommend acceptance of this work and publication in ACP after consideration of the general and specific comments given below.

General comments

The authors did a good job in compiling their work into a concise main article. However, the Supplementary material related to the article contains a lot of interesting information, some of which would be better presented within the main article or in an appendix. Since this is an electronic journal, I recommend the authors to move parts of the experimental descriptions and the Figures S1 and S2 of the Supplement into the main article.

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The experiments done in this study, as well as the parameterizations derived using the data, correspond to a certain temperature range. Furthermore, RH of phase transitions are at least weak functions of temperature. In most captions of the tables and figures, a statement on the temperature would therefore be appropriate. For what temperature range are the parameterizations considered applicable?

Specific comments

- Abstract, first sentence: In Section 3 it is mentioned that the authors distinguish between HOA- and OOA-like organics. Therefore it should be mentioned in the abstract that only oxygen-containing organic compounds are considered in this study, e.g., by writing "oxygenated organic material" in the second line.
- Abstract, lines 7-9: This sentence could be written better using a more precise wording. Currently the first part for example reads: "These transitions include liquid-liquid phase separation (SRH),...", however, I guess "SRH" does not literally mean "liquid-liquid phase separation", rather it means the RH where liquid-liquid phase separation is present – or better: its upper RH limit (onset). Hence, I recommend writing this sentence differently in order to distinguish between the processes and the RH they are observed or related to.
- Abstract, lines 14, 15: "..., ERH within 5 % for 86 % of the measurements, and DRH within 5 % for 95 % of the measurements." I guess here it is meant "within 5 % RH". Without the "RH", the meaning could be different.
- p. 17765, l. 11: "The morphology is an organic coating surrounding an aqueous ammonium sulfate core,...". Is this morphology found to be the same for all exper-

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iments at all org:sulf ratios? This should be made clear. Also, what is the mixture and org:sulf of the particle in Fig. 3, at what temperature?

- p. 17766, l. 2: "ERH has greater sensitivity than DRH to increases in org:sulf". This statement can be misleading. For high O:C and high org:sulf in a single mixed phase, the DRH of ammonium sulfate is expected to also span quite a large range of RH (from $\sim$80 % RH down to very low RH at org:sulf $>$ 10).
- p. 17766, l. 7: "Salting out is described by Setchenov equation...". As the Setchenov equation is not the definition of salting-out nor a perfect description, a better wording is: "Salting out may be described by the Setchenov equation..." On line 11: ", S_0 is the solubility without the salt,...". The solubility in what?
- p. 17767, l. 18: "to be consistent with the ERH of pure ammonium sulfate." The ERH at what temperature and from what references? Contrary to the DRH of pure ammonium sulfate, the ERH is not a thermodynamically fixed function of temperature only.
- p. 17768, l. 1: "... DRH of three-component organic-ammonium sulfate particles". The term "three-component" can be misleading in this context, as I guess the authors do not mean ternary mixtures of only three individual components (substances).
- p. 17769, Section 4: I think this Section would benefit from including additional information on the chamber studies and the field study now only given in the Supplement.
- p. 17782, Table 2: What is the applicable temperature range of the expressions? The parameterizations for ERH at low and high O:C ratios should be spaced more apart vertically for better distinction. Now it looks like the lower equality summarizes the upper expression, which is of course not the intention. Same issue in case of the DRH expressions.

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Technical Corrections

- p. 17763, l. 6: “and they dominant relative to...”. Check wording.
- p. 17764, l. 1: “... water/methanol mixture was than evaporated...”; “than” should be “then”.
- p. 17767, l. 10: “a decreases”. Spelling.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 17759, 2011.