

***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 #2**

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General comment

This paper gives a broad overview and a new parameterization of phase transitions in an important class of aerosol particles, namely organic/sulfate submicrometer-sized

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particles. In addition to efflorescence and deliquescence, liquid-liquid phase separation is also studied. For reliable parameterization, a series of own new measurements is presented that complements literature data. The parameterization was tested against smog chamber and field data. It exhibits a reasonable accuracy and should be simple enough for incorporation in e.g. chemical transport models. This is an excellent paper that deserves publication in ACP. I have some suggestions for changes that might improve the readability of the manuscript.

Specific comments

Page 17764, line 13: what type of RH sensor was used? What is its accuracy and precision?

Page 17766, line 21: The formulation should be improved. It should be added that this statement refers to Fig. S2.

Pages 17767 and 17768, discussion of “reduced chi squared”: Considering Fig. 5a, the large “reduced chi squared” values arise from cases where the parameterization wrongly predicts liquid-liquid phase separation. In those cases, the experimental SRH is set to 0 %, leading to large contributions to “reduced chi squared”. Therefore, it might be interesting to calculate a second “reduced chi squared” that excludes the cases where no liquid-liquid phase separation was observed.

Page 17771, line 10: point (2) is difficult to understand. It should be explicitly stated what O:C and org:sulf range should be covered.

Caption to Table 1: “We also excluded any ERH results where it could not be shown that ammonium sulfate was the first solid to crystallise in the three-component particles”: How do you show that ammonium sulfate is the first component that crystallizes?

Table 1: ERH values of polyethylene glycol-400 / ammonium sulfate values are given in more detail in “Ciobanu et al., J. Phys. Chem. A 2010, 114, 9486–9495”.

Figure 1: The contrast of the droplets in panel (b) should be improved.

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Figure 3: at what RH have the Raman spectra been measured? What is the light spot in the middle of the droplet, the illuminating laser?

Figure 4: Panels (b) and (c) might be improved when additional contour lines for ERH between 30 – 35 % and DRH between 75 – 80 % would be added.

Figure 5 might also be placed in the supplementary material.

Supplementary material:

Lines 345 – 355: The procedure to adjust the measured ERH to the parameterization is not justified. The values in the Tables and Figures should be the measured values without any corrections for particle size. The parameterization should be adjusted to describe ERH of submicrometer particles, not the other way round.

Table S1: This Table is almost the same as Table 1 of the main manuscript. These two tables might be merged to one Table in the main manuscript.

Table S2: this Table should go into the main manuscript. Some cells in the columns “ERH” and “DRH” are empty. Does this mean that no ERH (down to 2 %) and no DRH was observed or that the cycle was stopped at a higher RH?

Table S3: this table would be much more informative if columns with the SRH values were added.

Table S5: are the ERH values in this Table shifted by 4.2 %?

Table S6: DRH and ERH values for each individual data point should be given in an additional column.

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