

Interactive comment on “Deliquescence, efflorescence, and phase miscibility of mixed particles of ammonium sulfate and isoprene-derived secondary organic material” by M. L. Smith et al.

Anonymous Referee #1

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General comments

This paper presents a detailed evaluation of the hygroscopicity of isoprene-derived secondary organic material condensed onto ammonium sulfate (AS) seed particles. An elaborate hygroscopicity measuring protocol was applied to the aerosol continuously produced in a flow reactor. To obtain deliquescence relative humidity (DRH) and efflorescence relative humidity (ERH) a sophisticated modeling procedure was developed. The manuscript is well and carefully written but readability suffers in some parts from the many abbreviations that are used. This paper is well suited for ACP

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since phases and phase transitions of mixed organic/inorganic aerosol particles are an important factor influencing aerosol properties. I recommend publication after the following points have been considered for revisions.

A concept of experiments section or paragraph at the beginning of the “Results and discussion section” might improve the readability of the detailed evaluation protocols and procedures. This section or paragraph should summarize in words how DRH and ERH as a function of organic volume fraction are derived from the applied measuring and evaluation procedures. A table that lists all the abbreviations might also be helpful.

The O:C ratio of the organic material is stated as 0.67-0.74 in many parts of the manuscript including the abstract. Only on page 9923, line 6, it is mentioned that it is connected to a measuring uncertainty of +/- 30%. The range of O:C should be extended to include this uncertainty or the uncertainty should be stated together with the numbers.

The authors exclude a liquid-liquid phase separation into an organic-rich and AS-rich phase because the low DRH values at high organic volume fractions suggest miscibility. However, a miscibility gap does not need to cover the whole composition range of a phase diagram and might still be present at low organic volume fractions. This should be mentioned in the revised manuscript.

The uncertainties connected with the DRH and ERH values extracted from the data are not given in the manuscript. Considering the complex evaluation procedure, uncertainties in the organic volume fraction but also in the DRH and ERH values could be notable. They should be added to Figures 4 and 5 or at least be discussed in the text.

Specific and technical comments

Abstract: Page 9904, line 4: It should be added that experiments have also been carried out at 60% RH.

Page 9904, lines 15-18: The full parameterization does not need to be given in the

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abstract.

Introduction: Page 9905, line 12, multiple phases: is there any example of more than two liquid phases in the context of atmospheric aerosols?

Results and discussion:

Page 9920, line19: shouldn't it be Figure 3c instead of Figure 3a?

Page 9921, lines 1-2: It is not clear which points are meant here. They should be marked by a different color in Figs. 4 and 5.

Conclusions and implications:

Page 9923, lines 24-25. This sentence is confusing. It should be improved or deleted.

Page 9924, lines 10-11. The kinetically driven decrease in ERH on the basis of homogeneous nucleation theory should be explained more explicitly.

Page 9925, lines 10-13: what is meant by heterogeneous morphology? The solid and/or liquid phases that are meant to occur should be explicitly stated.

Appendix A, Hygroscopic growth:

The equation on page 9928 is not numbered. It should be numbered as Eq. A1 and the following equations need to be renumbered.

Figure 1:

The writing in this figure is difficult to read even on the screen. The font should be increased.

Figure 2:

The green point in panels a and b of Fig. 2 is difficult to understand: it has $P(\epsilon) = 0$ in panel b but corresponds to a fraction 0.2 of nondeliquesceable particles. Can you clarify this?

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Figure 5:

This Figure would be easier to interpret when the lines and the gray shaded area giving the percentage error in O:C ratio were replaced by lines that directly state the O:C ratio for O:C from 0.7 – 0.9. If the Bertram et al. (2011) DRH and ERH parameterization is accurate, O:C of the investigated samples should be rather 0.9 than 0.67 – 0.74. Is this discrepancy due to an inaccurate DRH/ERH parameterization or too low O:C ratios of the organic material measured by the AMS?

Supplementary material:

Caption to Figure S4: Eq. (3A) should be replaced by Eq. (A3).

Figure S5:

The inset in panel A should also be explained in the figure caption. Shouldn't the y-axis label of the inset read $P(\epsilon)$ instead of $f(\epsilon)$?

The black dashed line in panel B should be explained in the figure caption.

The measured curve should also be shown in panel B.

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