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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Hygroscopic and phase separation properties of ammonium sulfate/organic/water ternary solutions" *by* M. A. Zawadowicz et al.

Anonymous Referee #3

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This manuscript addresses hygroscopic properties of ammonium sulfate particles mixed with five different organic molecules investigated in a flow tube using FTIR spectroscopy. The topic is timely and I find in particular the results using deuterium substituted water vapor interesting. I have however several concerns which should be addressed before the manuscript can be recommended for publication: The manuscript should be better structured. The literature studies referred to are in several places not adequately summarized which make the text difficult to follow. Uncertainties should be more thoroughly discussed. Several conclusions are drawn in relation to particle size, but the size distribution of the particles used are not shown. Size selected experiments

would have formed a stronger basis for making such conclusions. Also conclusions made in relation to phase separation are in several places difficult to follow. My concerns and comments are detailed below and I hope the authors will find them useful in revising their manuscript.

Some of the important conclusions are based on "personal communications". The authors should provide a more detailed explanation for these.

Page 6539: "...Are overwhelmingly faster in the aqueous phase "– that in which other phases ? – should be explained.

The references given on how organics alter CCN properties are rather old, a lot of work has been done on this topic since 2001. More recent references should be provided.

Page 6540: The discussion of previous work and methods is mixed with methods from this work. It is not clear from the description what the different techniques mentioned do. Which ones requires deposition on slides, which ones not, which ones can have a continuous flow, which ones not, what are the sizes studied in the different techniques ?. I would suggest a table summarizing these characteristics.

Page 6541 A lot of theories and equations are mentioned but not explained. E.g. what is the consequence of the salting out mentioned, what does it mean for the systems studied by the authors? Is the Setchenow equation important here – then it should be written as an equation, Page 6542: The authors could refer to Table 2 already in the introduction.

Some of the text on O:C ratio was already mentioned on page 6541.

Experimental: The authors state that the atomizer aerosol output was centered around 120 nm determined with a DMA. Size is highlighted as a very important parameter in the discussion and thus more information on particle size should be given. Where was this measured, before or after drying? It must have been measured with a system containing also a neutralizer and a CPC – not only a DMA? What was the width of the

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size distribution? How many particles per cc? Was the size of super-micrometer sized particles measured?

The authors should show one or more of the measured size distributions in a Figure. Did it vary with the presence of organics in the atomizer bottle?

Some of the speculations in the discussion and conclusion section could be confirmed or vice versa if the authors did some size –selected experiments.

It is said that the flow tube used was of glass in the majority of experiments – what was the material in the rest of the experiments? Why were different materials used?

Results and discussion

It says throughout the text that for pure ammonium sulfate the effloresecence observed in this work was 35 % and the deliquescence was ∼80 %. This is not what the figures show as far as I can see – they show efflorescence around 40% and deliquesence below 80 % - more like 75% ?. Both should perhaps rather be reported as intervals that takes account of the uncertainties – when is the amount of condensed water significantly different from zero (in terms of uncertainties) and likewise for efflorescence. In some figures the efflorescence band is below the deliquescence band – why – this tells something about uncertainties?

Page 6547: For the mixed particles containing 1,4 butanediol the authors state: “Our results indicate some inhibition of ERH due to the presence of organics”. This I cannot see from Figure 5 - within the order of magnitude uncertainties indicated by the bars on the datapoint to the left it seems to me that there is no difference between the efflorescence points between the mixed and pure ammonium sulfate particles ?

It says that effloresence is completely suppressed in e.g. panel d of Figure 7 – I do not quite understand how that is observed from the figure.

What are the definitions of “complete suppression” of deliquesence and efflorescence points respectively? How is that defined in terms of change in water content per change

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in RH? (and taking into account uncertainties)

Page 6548: The study by Veghte et al. (2013) should be explained briefly in the introduction since several conclusions in the manuscript are based on results from this study.

It says that the size- distribution spanned from sub 200 nanometer size to super micrometer diameter as described in the experimental section – this was not described in the experimental section . . . it is a very broad interval – if the center was at 120 nm I would assume there were very few micrometer sized particles ? again it would be very useful for the reader to see the size distribution of the particles studied.

Page 6549: "exposing particles to an increasing flow of D2O" – should be rephrased – does this mean increasing concentrations of D2O in the gas phase?

"Assuming a relative constant aerosol content" – how good is this assumption – how much did the size and number concentration of particles vary during an experiment?

Page 6550: the background for the conclusions regarding rapid phase separation and no limitation of mass accommodation must be better explained. "on this time scale" – it is not clear what the time scale was – and would any effects be expected from theory on such a time scale?

Conclusions: Several of the issues above should be addressed also in the way that the conclusion is formulated. What was the reason that size-selected experiments were not performed – do I understand the last sentence in the conclusion right that this is currently not possible because there are two few particles to get enough intensity in the IR-spectra for the size-intervals that can be selected with a DMA?

Table 2: Results from this study should be in separate lines. The table could be extended to also include details about the method used (parameters important e.g. size of particles studied).

Abstract The first part is mostly like an introduction. The abstract could be rewritten

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and focus on the methods and results of this work.

“the results of this work tend to be in agreement with previous microscopy experiments, with several key differences which possibly reveal a size-dependent effect on phase separation in organic/inorganic aerosol particles” . This statement I find confusing and suggest that it is rephrased.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 6537, 2015.