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Interactive comment on “Amorphous and crystalline aerosol particles interacting with water vapor – Part 1: Microstructure, phase transitions, hygroscopic growth and kinetic limitations” by E. Mikhailov et al.

Anonymous Referee #2

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General remark:

This article is a mix of review paper and a research paper. The Appendix summarizes again well known applications of Köhler theory and derivatives on monography level.

We have to make a decision, before we analyze the contents in detail: do we want to have such a conglomerate of review and research paper ?

The difficulty: on hand there is nothing really new in the presented manuscript, with exception of a some experimental details. On the other hand it may well be that nobody

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before wrote it down the way it is presented here in this manuscript. It reads well and is entertaining, although it could be shortened.

But in any case the measured data base is to scarce for the generality of conclusions. In this context, the literature data appears to be somewhat selected to support the (interesting) speculations about and interpretations of the experimental findings. Of course the authors try to make their case. But how representative is the reviewed literature ?

From this point of view I would reject the manuscript, and ask the authors to split the manuscript in a paper where they present their data and a sound, observation based interpretation. And in a review about semi-solids, gels and their importance in atmospheric science, wherein the results from the independently refereed research paper of course maybe quoted.

Research part:

The research part presents some new observation for Oxalic acid (OA) and Levoglucosan (LG), both systems well investigated in the literature. It uses a kind of review of literature mostly regarding polymeric systems (gels, microgels, rubbers, viscuous liquids, glassy state etc.) to set up a new interpretation of water uptake of OA and LG, and organic particles in general. I see some interesting aspects, but the paper needs major revisions before it can be published in ACP.

These are my main concerns:

Ammonium sulfate data were also presented. They demonstrate positively the good performance of HTDMA. However, the notation,

"However, readers interested only in amorphous particle behavior might want to skip Sect. 3.2 and proceed to Sect. 3.3.",

section 3.2 dealing with the ammonium sulfate, indicates the minor importance in the context of the manuscript. Section 3.2. should therefore be shortened to HTDMA

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calibration issues.

Oxalic Acid:

The jumping between the oxalic acid dihydrate (no evaporation, Köhler calculations) and amorphous structure (early deliquescence, wiggles - stepwise deliquescence) is inconclusive.

You assumed that high charging during generation is a possible reason for forming the amorphous solid particles with a highly porous structure made out of oxalic acid dihydrate. Did you try to equilibrate charges by using a delay tube? Did that produce more compact particles ? That would support positively your assumption. (Dinar et al. ACP 2006, p5213, reported on such an effect for ammonium sulfate.)

I can hardly detect wiggles on the OA growth curves. Are they reproducible and do they appear always at the same RH? How often did you repeat the humidograms. I think from the data presented the stepwise deliquescence is not supported.

Many interpretations of the measured data is based on analogy to literature data. I don't understand, why observations for polymers and proteins are a suited analogy for oxalic acid, an almost inorganic, small compact highly polar molecule (throughout sections 3.3.2. and 3.3.3.)

Is oxalic acid the typical representative for organic acids or organic molecules in general? I believe that observation for oxalic acid cannot be generalized in terms of "typical behavior of organic compounds".

The discussion of ideality of OA solution on page 7368, line 18 ff, neglects the formation of hydrogen bonded networks, you draw on in explaining the gradual deliquescence and gel like structures above page 7356, line 6 ff.

Levoglucosan:

Why did highly charging during the spray process not lead to porous particles in the

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levoglucosan case? I expect levoglucosan to be susceptible to high charging.

On page 7363, line 9-13 again you refer per analogy to references which deal mainly with polymers, not with levoglucosan.

Köhler Calculations:

Section 3.5.1 should be strictly reduced to the fact that AS served as working reference case for the later Köhler calculation for the organics.

Misunderstandings in the CCN community, p. 7366, line 25, should not be discussed in this form in this manuscript. Moreover, the work by Rose et al. alone is a insufficient reference to underline the great impact and importance of AIM and other ion interaction models in physical chemistry and geochemistry.

Summary and Conclusions:

Generalized conclusions on behavior of organics, organic acids or even sugars cannot be drawn from the presented material. The data base is to small. OA and LG are small and compact molecules compared to e.g. carbohydrates in general.

Generalized conclusion on behavior of organic mixtures should not be drawn from the given material. The companion paper is not published yet.

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