

***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.***

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Response to Referee #2

We thank anonymous Referee #2 for the time and effort invested in the review of our manuscript. With many of the referee's statements, however, we do not agree. Responses to individual comments are given below.

Referee Comment 1:

This article is a mix of review paper and a research paper. The Appendix summarizes  
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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?

Response:

We consider it necessary and good scientific practice to specify exactly which methods and models have been used in a scientific study, so that fellow researchers can reproduce experiments and model calculations without guessing or spending much effort and time on recollecting the necessary information from different sources. As outlined in our manuscript and in related earlier papers we consider this particularly important for the application of Köhler models of aerosol-water interaction.

In our understanding of scientific publishing, appendices are well suited for compact and exact description of the tools applied in a study. We like to have such a description at hand for all readers of our manuscript (including ourselves and our co-workers) and we are not aware of an alternative publication that would compactly and exactly summarize the various modelling approaches used in our study (equations, parameters, software tools etc.). Moreover, we do not think that an equation equivalent to Eq. A18 has been published before. Beyond that, we consider it appropriate not only for review articles but also for research articles to synthesize and refer to the results of relevant related studies.

Referee Comment 2:

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

Response:

We disagree with regard to the insinuated lack of novelty. In the following we point out

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some aspects that we consider as new scientific results and innovative messages of our manuscript:

1) H-TDMA measurement data describing the surface and bulk water uptake and hygroscopic growth of aerosol particles composed of three abundant atmospheric aerosol components (ammonium sulfate, oxalic acid and levoglucosan) with higher precision than any earlier (H-TDMA) study we are aware of (at least for the investigated particle size range and for low RH). Besides enhanced precision, our data sets also provide new information from hydration&dehydration experiments and from the analysis of the width and broadening of particle size distributions.

2) New experimental evidence, detailed discussion of the new and earlier experimental results, and new conclusions concerning the kinetic limitation of water uptake by levoglucosan particles, including the retention of residual water under “dry” conditions and its effect on the determination of Köhler model parameters.

3) New experimental evidence, detailed discussion of the new and earlier experimental results, and new conclusions concerning the hydration state, the microstructure (xerogel, swollen/collapsed network structures) and the volatility of oxalic acid particles - especially under “dry” conditions and at low/medium relative humidity.

4) New terms and concepts for the description and first evidence for the potential influence of amorphous and semi-solid substances for the interaction of aerosol particles and water vapour in atmospheric research (laboratory and field measurements) and atmospheric processes. We are not aware of any publication presenting a similar conceptual framework as outlined in Fig. 3 of our manuscript. To our knowledge, the following terms have not at all or hardly been used and have not been proposed for general use in atmospheric (aerosol) science: semi-solid, gel-like/rubbery/ultraviscous, xerogel/aerogel, moisture-induced glass transition, . . . We agree that most (but not all) of these terms are or have been more widely used in other scientific disciplines. With regard to scientific research and progress, however, we think that the transferring, ap-

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plying and building upon concepts from different scientific disciplines is an important part of innovation.

5) Other recent studies have suggested that solid amorphous phases (glasses) may be important for aerosol-water interactions at low temperatures in the upper atmosphere. We suggest that (semi-)solid amorphous phases (glasses, rubbers, gels, ultra-viscous liquids) and moisture-induced glass transitions may be important also at ambient temperatures in the lower atmosphere. In particular, they might play a role in the kinetics of CCN and IN activation, thus influencing the formation and properties of clouds and precipitation. We expect that the relevance of glassy and semi-solid states will become a new paradigm of atmospheric aerosol research, and we think that the experimental evidence and theoretical concepts presented in our study are major contributions to this development.

For clarification we intend to include the first half of the preceding paragraph in the revised manuscript. Moreover, we intend to adjust the title, restructure the manuscript, and separate the general conceptual framework (new Sect. 2 after introduction) from the presentation and discussion of our experimental results (new Sect. 4 after experimental methods) and of our model calculations (new Appendix B after Köhler model description). We hope and expect that these modifications will clarify the main findings and messages of our study.

Referee Comment 3:

But in any case the measured data base is too 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?

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This comment seems to suggest that we would have been (inappropriately) selective with regard to searching and referencing the scientific literature, but no facts or references are provided to support the apparent insinuation.

We agree that the data base of our own measurements would have been insufficient to support our general conclusions. This is why we have invested several years since the completion of our experiments in 2005 for searching and synthesizing information from related studies in various disciplines (atmospheric aerosol research, food, materials and pharmaceutical science) in order to develop the conceptual framework outlined in Fig. 3. Only within this framework it became possible for us to consistently explain our own experimental findings and to relate them to the results of earlier atmospheric aerosol studies (Sects. 3.3.4 and 3.4.3).

Based on the extensive literature search, on carefully comparing our results to those of related earlier studies, and on recollecting some basic knowledge of chemistry students and practitioners (difficulties of crystallizing organic substances) we have come to and are able to support the general conceptual framework and conclusions of our manuscript.

If our conclusions were based on selective perception and not sustainable, it should have been easy for the referee to come up with suitable references proving our conclusions wrong or exaggerated.

We found and cited numerous references supporting our most general conclusion that “Many organic substances (including carboxylic acids, carbohydrates and proteins) tend to form amorphous rather than crystalline phases upon drying of aqueous solution droplets.”, but we found no reference indicating the opposite (i.e. that only few organic substances would exhibit this behaviour).

Similarly, we found and cited numerous references supporting our second most general conclusion that “Amorphous organic substances tend to absorb water vapor and undergo gradual deliquescence and hygroscopic growth at much lower relative humidity

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than their crystalline counterparts.“ but we found no references indicating the opposite (i.e., that amorphous organics tend to deliquesce at the same RH as crystalline organics).

#### Referee Comment 4:

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.

#### Response:

This statement seems to imply that research articles should not build upon and reference many other studies. As explained above, we found it necessary to consult a wide range of scientific literature in order to explain our findings, and the results are presented in our manuscript.

We are not aware of any other manuscripts addressing the importance of semi-solids and gels in atmospheric science. Accordingly, it has not been and still is not our intention to write a review article on the topic proposed by Referee #2. Instead it has been and remains our intention to publish a research article that reports new experimental results on aerosol-water interactions of atmospherically relevant substances together with a conceptual framework that supports a detailed understanding and efficient description of the observed phenomena and their implications.

#### Referee Comment 5:

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, viscous liquids, glassy state etc.) to set up a new interpretation of water uptake of OA and LG,

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and organic particles in general. I see some interesting aspects, but the paper needs major revisions before it can be published in ACP.

Response:

With regard to the statement that the studied systems are “well investigated in the literature” we would like to point out the following:

1) As discussed in our manuscript, earlier EDB, AFT-FTIR, and HTDMA studies of OA and LG particles exhibited unexplained differences and presented no consistent picture of phase states and transitions.

2) HTDMA results for OA particles were published only in one paper reporting no dehydration experiments and no data at relative humidities lower than 45% (Prenni et al. 2001).

As demonstrated by the numerous references cited in our paper, amorphous and semi-solid states are best known for polymeric systems but are also widespread and well known for substances of low molecular mass. Examples are citric acid, glucose, lactose and other mono- or disaccharides, etc. (e.g., Corrigan et al., 1984; Abdallah and Weiss, 2000; He et al., 2006; Murray, 2008; Zobrist et al., 2008). Please note that we had also explicitly pointed this out in our manuscript (p. 7344, l. 20-26): “The rubbery state usually occurs in polymeric substances, in which the individual macromolecules are entangled or cross-linked by covalent or hydrogen bonds, van der Waals forces, or other types of interaction (Erman and Mark, 1997; Cowie and Arrighi, 2007; Sperling, 2006). In food science and pharmaceutical research, however, the term rubber is also used for semi-solid amorphous substances with low molecular mass such as monosaccharides (Roos and Karel, 1991, 1992; Roos et al., 1999; Hancock and Zografi, 1997; Bhandari and Howes, 1999; Burnett et al., 2004).”

Upon revision of our manuscript we will add an additional clarifying statement in the introduction section (p. 7337, l. 19): “The formation of amorphous solid particles

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upon spray-drying of aqueous solutions of organic substances is a well-known standard process in the food and pharmaceutical industries. It applies to organic compounds with a wide range of molecular masses (e.g., mono-, di- and polysaccharides), and the amorphous state of spray-dried organic materials was confirmed by X-ray diffraction, differential scanning calorimetry and dynamic vapor sorption methods (Corrigan, 1984; Bhandari and Howes, 1999; Roberts and Debenedetti, 2002; Price and Young, 2003; Burnett et al., 2004; Langrish and Wang, 2006, and references therein)."

#### Referee Comment 6:

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 calibration issues.

#### Response:

The ammonium sulfate data are not only relevant for H-TDMA calibration. They are important also for other aspects of our study:

- 1) They exemplify the typical hygroscopic behaviour of crystalline inorganic salts (including phase transitions and microstructural rearrangements), and provide a benchmark against which the hygroscopic behaviour of the investigated amorphous organic particles can be directly compared (measurement precision, broadening effects, surface adsorption vs. bulk absorption, etc.).
- 2) We use them to investigate the surface adsorption of water vapour on crystalline surfaces through combination of different modes of H-TDMA operation (in comparison to bulk absorption of water vapour into amorphous substances).
- 3) Last but not least, they provide a benchmark for different Köhler models, simplifying

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assumptions (volume additivity), and the determination and conversion of different hygroscopicity parameters, some of which are also relevant for the investigated organic substances and for the mixed systems investigated in the companion paper (soon to be submitted). We are not aware of any other publication presenting similarly precise measurements of the hygroscopic growth of AS aerosol particles in the investigated size range and similarly good agreement with different Köhler models (Figs. 6a and 7a), illustrating also the concentration dependence of various parameters describing hygroscopicity and non-ideality (Fig. 7b).

Please note that the relevance of this aspect is confirmed by Referee #1: “In the final section of the manuscript, the growth factor data are modeled using a variety of currently used techniques, providing interesting insights into the non-ideal behavior of aqueous ammonium sulfate, oxalic acid, and levoglucosan solutions.” [www.atmos-chem-phys-discuss.net/9/C943/2009/](http://www.atmos-chem-phys-discuss.net/9/C943/2009/)

Referee Comment 7:

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

Response:

We are not sure what Referee #2 means with “jumping”. It would have been helpful to get some more feedback than just the above statement. We have tried to explain the complex interplay of hydration state and microstructure of the oxalic acid (OA) particles in Sect. 3.3 in a systematic and logically coherent way. In Sect. 3.3.1 we provide evidence that the oxalic acid particles investigated in our system were stable, i.e., that oxalic acid did not evaporate on the time scale of our experiment (which is not self-evident with regard to glutaric acid and related earlier studies). In Sect. 3.3.2 we present and discuss the evidence that the oxalic acid particles are amorphous, because they exhibit hygroscopic growth at relative humidities well below the deliquescence threshold

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of crystalline oxalic acid. In Sect. 3.3.3 we discuss details in the observed hygroscopic growth curves and their implications for particle microstructure and phase transitions. In Sect. 3.5.2 we present Köhler model calculations confirming that the “dry” oxalic acid particles most likely consisted of oxalic acid dihydrate.

As indicated above, we have invested substantial effort in developing a consistent explanation and detailed understanding of the observations and underlying processes, and we have tried our best to convey the reasoning and line of arguments to the readers of our manuscript. We will try to clarify and improve this upon manuscript revision. In particular, we will add references to earlier studies suggesting that the tendency of aqueous acids to form glasses is particularly pronounced at compositions that correspond to stable hydrates.

#### Referee Comment 8:

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.)

#### Response:

We appreciate the constructive suggestion, intend to follow up on it in future studies, and agree that further investigations are desirable to unambiguously identify the reasons for the formation of highly porous structures upon spray-drying of aerosol particles. Within this study we have not performed the suggested experiments. Based on earlier investigations, however, we are confident that charge effects are the most plausible and likely explanation why particles generated by spray-drying tend to be more porous when they contain electrolytic substances (Mikhailov et al., 2004). We intend to add a clarifying statement including references to Dinar et al. (2006) and other related studies in the revised manuscript.

## Referee Comment 9:

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.

### Response:

The wiggles are best visible in Figs. 4c (varying offset between blue and red symbols) and Fig 5d (scatter of neighbouring data points) around  $\sim 45\%$  and  $\sim 55\%$  RH, respectively. Comparison with the hydration and dehydration curves of AS and LG shows that wiggles (diameter variations exceeding measurement precision) were indicative of phase transitions and did generally not occur upon or continuous hygroscopic growth of fully deliquesced aqueous droplets.

As explained in the manuscript, we interpret these wiggles and - more importantly - the concurrently observed broadening of the measured particle size distributions as indicative for kinetically limited transformation processes. Thus, we do not expect every individual wiggle to be particularly well reproducible, but we do expect similar wiggles (irregularities) to occur under similar experimental conditions.

Each of the humidograms/curves presented in Figs. 4-7 is the result of 2-3 repeated experiments (RH scans in H-TDMA operation modes 1, 2 or 3), and the measurement uncertainty of the data points was generally less than 0.4% (standard deviation of five repeated measurements) as specified in our manuscript (Sect. 2.3). Please note that we have also explicitly addressed and discussed the (limited) significance of the observed wiggles in our manuscript (Sect. 3.2.2, p. 7355, l. 28): "With regard to the growth curves, the observed deviations are at the edge of experimental precision, which is limited primarily with regard to RH, but for  $\sigma_g$  they are clearly larger. Only above  $\sim 80\%$  RH did the growth factors and geometric standard deviations agree within experimental uncertainty, indicating that the particles were fully deliquesced liquid aqueous solution droplets."

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We did not intend to suggest that the amorphous OA particles underwent stepwise deliquescence, and that this would have been indicated by wiggles in the growth curves. As detailed in the manuscript, we suggest that the discernible stepwise diameter increase at  $\sim 43\%$  RH (Fig. 5d) is indicative of a stepwise onset of the deliquescence transition.

Referee Comment 10:

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.)

Response:

As outlined in Sect. 3.1.2, p. 7344, gels are generally defined as “two-phase mixtures of liquids dispersed in (semi-)solid amorphous matrices (supramolecular networks; Flory, 1974; Keller, 1995; Abdallah and Wise, 2000; Sangeetha and Maitra, 2005; He et al., 2007).” Obviously, and as detailed in the referenced papers of Abdallah and Wise (2000) and He et al., (2007), supramolecular networks can be formed not only by large molecules but also by small molecules.

As explained in Sect. 3.3.2, p.7356, we suggest that the polar oxalic acid molecules form supramolecular networks via hydrogen bonding in highly concentrated aqueous droplets. From a physical chemistry perspective, this is more than plausible and fully consistent with the tendency of oxalic acid to form hydrates, which are indeed hydrogen bonded supramolecular networks as detailed in the reference papers of Hermida-Ramon et al. (2004), Chitra et al., (2004), and Wenger and Bernstein (2007).

As outlined in Sect. 3.1.2, p. 7345, the uptake of water into a gel can involve gradual swelling as well as stepwise volume increases related to thermodynamically well-defined phase transitions (Dusek and Patterson, 1968; Tanaka, 1978; Erman and

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Mark, 1997). Upon drying, gels can form highly porous structures (xerogels/aerogels; Kistler et al., 1935; McNaught and Wilkinson, 1997). As explained in Sects. 3.3.2 and 3.3.3, this is the most plausible explanation for the observed behaviour of OA particles.

Referee Comment 11:

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".

Response:

We do not think that oxalic acid is "the typical representative for organic acids or organic molecules in general" and we agree that our observations for oxalic cannot be generalized in terms of "typical behaviour of organic compounds". We are not aware of having suggested anything like that.

Referee Comment 12:

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.

Response:

We see no contradictions in this context. Based on the relatively high acidity of OA one might expect van't Hoff factors larger than unity also in concentrated solutions and not just in dilute solutions as observed in our study in agreement with earlier studies (Varga et al., 2007). The reported observation that OA does not reduce water activity more than an ideal solute molecule may well be due to a compensation of the dissociation of OA molecules by hydrogen bonding between OA molecules. We agree that this is one of the many interesting questions that arise from considering intermolecular interactions and the formation supramolecular networks in aerosol particles consisting of highly concentrated aqueous solutions of organic molecules. However, we see no

need and no opportunity for addressing and clarifying this aspect further in our study.

Referee Comment 13:

Levogluconan: Why did highly charging during the spray process not lead to porous particles in the levogluconan case? I expect levogluconan to be susceptible to high charging.

Response:

As explained in our manuscript, levogluconan is a non-electrolyte, and based on earlier studies we expect charge effects upon spray-drying primarily for electrolytes. Our observations reported here and in an earlier study (Mikhailov et al., 2004) are in line with this expectation. If the referee had contrary evidence or alternative suggestions, it would have been helpful to provide references.

Referee Comment 14:

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

Response:

Again the references deal with compounds of high as well as low molecular mass. In the revised manuscript we will add the following sentence for clarification: Good examples for comparison with the sugar anhydride levogluconan are sugars like glucose, lactose, sucrose and other mono- or disaccharides (Bhandari and Howes, 1999; Burnett et al., 2004; Zobrist et al., 2008).

Referee Comment 15:

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 an insufficient reference to

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underline the great impact and importance of AIM and other ion interaction models in physical chemistry and geochemistry.

Response:

We agree that the AS model calculations serve primarily as a reference for comparison with the organics substances. Beyond that, however, they also provide a benchmark for different Köhler models, simplifying assumptions (volume additivity), and the determination and conversion of different hygroscopicity parameters, some of which are also relevant for the investigated organic substances and for the mixed systems investigated in the companion paper (soon to be submitted). We are not aware of any other publication presenting similarly precise measurements of the hygroscopic growth of AS aerosol particles in the investigated size range and similarly good agreement with different Köhler models (Figs. 6a and 7a), illustrating also the concentration dependence of various parameters describing hygroscopicity and non-ideality (Fig. 7b). Please note that the relevance of this aspect is confirmed by Referee #1: "In the final section of the manuscript, the growth factor data are modeled using a variety of currently used techniques, providing interesting insights into the non-ideal behavior of aqueous ammonium sulfate, oxalic acid, and levoglucosan solutions." [www.atmos-chem-phys-discuss.net/9/C943/2009/](http://www.atmos-chem-phys-discuss.net/9/C943/2009/)

With regard to the AIM-based Köhler models we simply consider it relevant and appropriate to explain why we regard and use these as accurate reference models. We know that interested members of the atmospheric aerosol science community have/had not yet fully recognized that the AIM can and should indeed be regarded and used as an accurate reference for both CCN and HTDMA experiments with ammonium sulfate. Our results and conclusions are consistent with earlier studies, but earlier HTDMA studies we are aware of had not yet demonstrated similarly good agreement between experimental data and (AIM-based) Köhler model calculations. To our knowledge, Rose et al. (2008) was the first study systematically exploring and discussing the accuracy of AIM-based Köhler models in comparison to other Köhler models. Since the publication

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of our discussion paper, a related study was published by Kuwata and Kondo (2009). We will include additional references to this study as well as to other studies dealing with the precision of HTDMA and CCN experiments.

Unlike suggested by Referee #2, we had not intended to “underline the great impact and importance of AIM and other ion interaction models in physical chemistry and geochemistry”. Nevertheless, we intend to follow up on the referees comment by including further general references on ion interaction models (Pitzer 1973; Pitzer and Mayorga, 1973; Pitzer 1991) in addition to the four AIM references of Clegg et al. that we had already cited in our discussion paper. If Referee #2 had provided any specific suggestions, we would have been happy to include them.

Following the recommendation to focus on key messages and reduce the length of the main manuscript, we will move the entire Köhler model section including Figs. 6 and 7 into the appendix. Moreover, we will shorten the discussion of AIM-based reference models.

Referee Comment 16:

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 too small. OA and LG are small and compact molecules compared to e.g. carbohydrates in general.

Response:

As specified above (Response to Referee Comment 3), we agree that the data base of our own measurements would have been insufficient to support our general conclusions. As indicated by the many citations and long list of references in our manuscript, however, we have synthesized information from related studies in various disciplines (atmospheric aerosol research, food, materials and pharmaceutical science) dealing with both small molecules like OA, LG and various sugars (mono- and disaccharides)

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as well as large polymers (polysaccharides, proteins, etc.). Based on this information we have developed the conceptual framework outlined in Fig. 3, which explains our own experimental findings as well as related phenomena observed in earlier studies and general features of water interaction with amorphous and semi-solid organic substances.

Referee Comment 17:

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

Response:

We did not intend to draw generalized conclusions on the behaviour of organic mixtures. For clarification we intend to reformulate the last paragraph of our manuscript upon revision.

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 7333, 2009.

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