

Authors' response to comments by anonymous referee #2:

This study presents an attempt to distinguish the role of particle phase state on the hygroscopicity of biomass burning surrogates and mixtures with ammonium sulfate. I consider the addition of such studies to the literature worthwhile. However, this paper requires a number of changes and clarifications before being accepted for publication. Before these are clarified, I found it difficult to provide further critique on a number of results presented. After reading the first review, which I tend to agree with on specific points raised, I present a number of different factors the authors need to address below:

Response: We are grateful to referee #2 for her/his comments and suggestions to improve our manuscript. We have implemented changes based on these precious specific comments in the revised manuscript of the article. We repeat the specific points raised by the reviewer in italic font, followed by our response. The pages numbers and lines mentioned are with respect to the Atmospheric Chemistry and Physics Discussions (ACPD) paper (original version).

Specific comments and author response:

(1): Abstract: I would recommend removing reliance on the word 'slightly'. Please quantify 'slightly' or remove entirely. This paper often feels a little too qualitative in nature, and I would recommend checking all instances like this. The authors also comment on 'similarity of hygroscopic parameter k '. Please quantify this. What range do you consider to be similar? line 35: Presume the authors mean sub-saturated 'RH'. line 35: 'at' the same environment? This is unclear. I would recommend checking all grammar throughout the document, as also noted by the other referee.

Reply: As this referee pointed out some grammar deficiencies, we rephrased several statements to improve grammar, wording and sentence structure. Further rephrasing will also be considered in preparation of the revised version of this article.

First, the word “slightly” has the meaning “very small in degree or amount”, which is not quantitative, but implies a small change relative to the magnitude of the quantity it is referring to.

As such, it is not untypical to be found it in scientific literature. However, we agree that the sentences concerned can be improved by better wording.

Second, concerning the statement “A similarity of the hygroscopicity parameter κ for organic surrogate compounds mixed with ammonium sulfate for different mass fractions during the different seasonal periods in the Amazon is observed.”, figure 6a shows a small difference in the hygroscopicity parameter κ for mixtures of organic surrogate compounds and ammonium sulfate representing biomass burning particles during the dry and wet periods in the central Amazon Basin. For example, measured κ values for bio-mix-dry aerosol particles were determined to be between 0.16 and 0.18 in the range from 90 to 40 % RH using the HTDMA technique, which is slightly higher than that of κ determined for the bio-mix-wet aerosol particles ($\kappa \approx 0.12 - 0.15$). We have revised these sentences in the manuscript.

Third, “the RH-dependent hygroscopicity parameter κ for organic surrogate compounds....at the same environment,....field observation conditions” at the same environment refer to the RH condition, Here, hygroscopicity parameter κ at 90 % RH in the laboratory compared with Kappa κ at the same RH.

Related additions and changes included in the revised manuscript were made for the following sentences:

Page 1 line 17: “We observed that levoglucosan and humic acid aerosol particles release water from upon dehumidification in the range from 90 % – 5 % relative humidity (RH).”

Page 1 line 18: “4-Hydroxybenzoic acid aerosol particles, however, remain in the solid state without diameter growth both upon humidification or dehumidification and exhibit a small shrinking in size at higher RH compared to the dry size. For example, the measured growth factor of 4-hydroxybenzoic acid aerosol particles is ~ 0.96 at 90 % RH.”

Page 13 line 345: “The E-AIM prediction is in relatively good agreement with results from the HTDMA measurement but typically overestimates the water content of particles consisting of mixtures at the RH range close to the ERH of AS.”

Page 2 line 32-37, we revise: “Lastly, two distinct mixtures of organic surrogate compounds, including levoglucosan, 4-hydroxybenzoic acid, and humic acid were used to represent the average water-soluble organic carbon (WSOC) fractions observed during the wet and dry seasons in central Amazon Basin. A comparison of the organic hygroscopicity parameter for the simple mixtures, e.g.

$\kappa \approx 0.12$ to 0.15 for the wet-season mixture in the 90 % to 40 % RH range, shows good agreement with field data for the wet season in Amazon (WSOC $\kappa \approx 0.14 \pm 0.06$ at 90 % RH). This suggests that laboratory-generated mixtures containing organic surrogate compounds and ammonium sulfate can be used to mimic, in a simplified manner, the chemical composition of ambient aerosols from the Amazon for the purpose of RH-dependent hygroscopicity studies.”

Page 5 line 124-126, we add: “The three organic compounds levoglucosan, 4-hydroxybenzoic acid and humic acid were used as surrogates for the rich-class of water-soluble organic components in biomass burning aerosols. The influence of the distinct chemical structure of these compounds was studied with regard to the water uptake and evaporation of the pure organic compounds as well as for mixed organic-AS-containing particles. Furthermore, a comparison with field data from the Amazon was performed to quantify the ability of mixtures of these three organic compounds to mimicking the hygroscopic behavior of complex ambient organic particles originating from biomass burning emissions.”

Page 10 line 284-287, we add: “Also, no ERH of 4-hydroxybenzoic acid in the dehydration mode was observed during the experiments; the likely reason is that the highest RH reached in the humidifier was approximately 98 %, which may be below the ERH of 4-hydroxybenzoic acid, reported as possibly above 95 % RH in another study (Mochida and Kawamura, 2004).”

(2) Page 7, line 176: 'Here the AIOMFAC-based thermodynamic equilibrium model is used to calculate the DRH...in the multicomponent system based on the known solubility of AS in the organic-free system.? I'm not sure why you have chosen to do this when the benefit of the AIOMFAC activity coefficient model is to account for inorganic-organic interactions. Please justify this as the proceeding equations do not necessarily correlate with this statement.

Reply: The reviewer is right, that an advantage of the AIOMFAC-based model is to account for inorganic-organic interactions in liquid mixtures. Such interactions will change the activity coefficient and thus we determined ERH and DRH not based on the soluteconcentration but the activities calculated by AIOMFAC. The given statements in the text are correct and consistent with Eq. (1); however, the sentence is perhaps not clear enough in the given form. We have therefore made a few revisions to clarify that knowledge of the solubility of AS in the organic-free system can be used to calculate its solubility in organic-inorganic mixtures with the help of a thermodynamic model.

Related additions and changes included in the revised manuscript:

Page 7 line 169-178, we add: “For example, in the case of a ternary liquid mixture of levoglucosan + AS + water in solid-liquid equilibrium (SLE) with a crystalline AS phase at a certain temperature T , a constant molal ion activity product $IAP_{AS} = IAP_{AS}^{(sat)}(T)$ is established (necessary SLE condition). In this case the liquid mixture is a so-called saturated solution with respect to AS. While the molar amount of AS in a saturated solution depends on the other mixture constituents, the value of $IAP_{AS}^{(sat)}(T)$ is a function of temperature only, since it is derived from the fixed chemical composition and associated chemical potential of the crystalline phase. A reference value for $IAP_{AS}^{(sat)}(T)$ can therefore be calculated with the AIOMFAC model from an experimentally determined solubility limit of AS in a known mixture, such as the molality of AS at the point of saturation in the binary aqueous system (water + AS). The RH at which full dissolution of a solid phase upon humidification is just reached, the DRH, is directly related to the conditions at which a saturated solution becomes subsaturated upon addition of water. Here the degree of saturation with AS can be determined unambiguously by the computed value of IAP_{AS} as a function of mixture composition and temperature. Making use of these thermodynamic relationships, the AIOMFAC-based equilibrium model is used to calculate the DRH and ERH of AS in the multicomponent system, as outlined below.”

Page 7 line 188, we add: “The RH at which this $IAP_{AS}^{(sat)}$ value is just reached in a certain bulk solution at equilibrium with its environment (in contrast to $IAP_{AS} < IAP_{AS}^{(sat)}$ at higher RH), is the (bulk) DRH of AS.”

(3) Page 8: line 209 'Differences in the density models are expected to lead to relatively small differences.' This needs a qualifying reference or a demonstration of sensitivity. What do you mean by relatively small?

Reply: Fig. 1 shows the impact of different density treatment in the E-AIM and the AIOMFAC-based models. In the case of predicted mass growth factors of ammonium sulfate, both models agree well with each other, especially at $RH > 70\%$ RH, indicating that the corresponding differences in predicted diameter growth factors must be due to the different way the conversion

from particle mass to particle volume is done by the two models. The expected difference is on the order of the HTDMA measurement error or less.

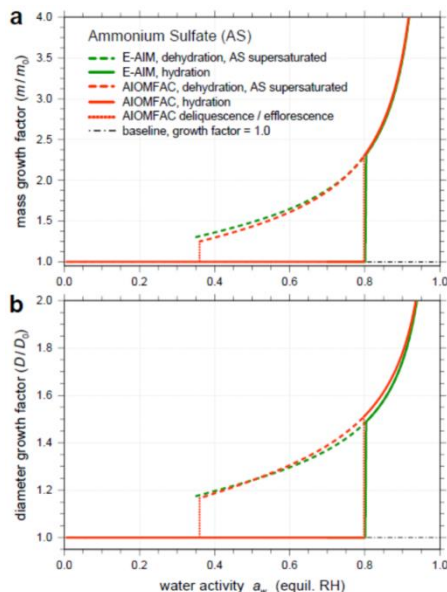


Fig. 1. Comparison of E-AIM and AIOMFAC-based mass growth factors (a) and growth diameter growth factors (b) for the binary ammonium sulfate + water system at 298.15 K (Fig. 4 of Lei et al., 2014).

Related additions and changes included in the revised manuscript:

Manuscript revision. We revise the corresponding sentence (page 8, line 209) to include the **reference**: “Differences in the density models are expected to lead to relatively small difference, typically on the order of the HTDMA measurement error or less (e.g. Fig. 2a), in the application to HGF predictions, as demonstrated by Lei et al. (2014) for the case of diameter vs. mass-based HGF of AS droplets.”

(4) Page 10, line 272: ‘standard UNIFAC..’. Which set of interaction parameters are you using? A reader should be able to replicate these results.

Reply: We use the set of UNIFAC interaction parameters included by default in the E-AIM model and in AIOMFAC. To clarify a bit further, we add an additional statement in the manuscript.

Related additions and changes included in the revised manuscript:

On page 10, line 273, we add: “The UNIFAC models within E-AIM and AIOMFAC are based on the original model expressions by Fredenslund et al. (1975) and both include the extensive parameter set by Hansen et al. (1991) as well as revised parameters for certain group interactions of water with carboxyl and hydroxyl groups by Peng et al. (2001). Of relevance for levoglucosan and other sugar-like compounds, the AIOMFAC model also contains certain revised group parameters for hydroxyl groups and special alkyl groups for their interactions with water, introduced by Marcolli and Peter (2005) for polyols, as further detailed in Zuend et al. (2011).”

(5) Page 10, line 276: 'intra molecular interactions are fully considered by these models..'. What is UNIFAC based on?

Reply: The referee probably misread the sentence. The sentence in the manuscript reads: “However, the molecular structure of levoglucosan with several polar functional groups in close vicinity may account for a small deviation between models and measured HGFs at RH below 70 %, because intramolecular interactions are **not** fully considered by these models.”

UNIFAC is an abbreviation that stands for Universal quasichemical Functional Group Activity Coefficients. It is based on the quasichemical theory of liquid solutions proposed by Guggenheim (1950), generalized by Abrams and Prausnitz (1975), and applied to functional groups within the molecules (group-contribution concept) by Fredenslund et al. (1975). The UNIFAC method allows to calculate/predicts liquid-phase activity coefficients of non-electrolyte solutions.

(6) Page 18, line 482: 'at RH > 95% the water content of hygroscopic particles increases dramatically with a small increase in RH, leading to the predicted change in the mixtures k parameter that is best representing the hygroscopic growth under such high-RH conditions'. This statement is not clear. Are you suggesting that variable 'k' values are required? Please rephrase and clarify.

Reply: Yes, if it is attempted to represent the hygroscopic behavior at high RH accurately, in particular for CCN activity predictions vs. hygroscopicity at subsaturation - it needs to be considered that κ parameters are not constant and their values depend on the RH range of measurements or models from which they were determined; see, e.g. the discussion by Rastak et

al. (2017) or Wang et al. (2017). The field data from the Amazon as well as the prediction with Eq. (4) based on the laboratory measurements indicate a marked difference in the determined appropriate hygroscopicity parameter κ between subsaturated and supersaturated water vapor conditions, with a transition predominantly in the RH from 90 % RH to ~100 % (Rastak et al., 2017). For example, the κ parameter obtained from field data is $\sim 0.15 \pm 0.06$ at 90 % RH, while its value reaches $\sim 0.18 \pm 0.04$ at RH > 100 % (just prior to CCN activation). The reason for the difference is that hygroscopic particles uptake water dramatically above 95 % RH when approaching 100 % RH, which is clear from model predictions, as demonstrated in Fig. 6 by application of Eq. (4).

Related additions and changes included in the revised manuscript:

Page 18 line 481-485, we add: “For example, the κ parameter obtained from field data is $\sim 0.15 \pm 0.06$ at 90 % RH, while its value reaches $\sim 0.18 \pm 0.04$ at RH > 100 % (just prior to CCN activation). A likely reason for the difference is that hygroscopic particles, especially those containing sparingly soluble organics like 4-hydroxybenzoic acid, take up water dramatically above 95 % RH when approaching 100 % RH (Huff Hartz et al., 2006; Chan et al., 2008; Rastak et al., 2017), which is clear from model predictions, as demonstrated in Fig. 6 by application of Eq. (4). The predicted curve in the mixture’s effective κ parameter may well capture the change in hygroscopicity under such high RH conditions.”

On page 18, line 485, we add: “Consequently, for a precise representation of the hygroscopic growth behavior (e.g. HGF) at high RH (> 95 %) by the κ -Köhler model, the value of κ would need to be varied. While a variable κ value is contrary to the attempted simplicity of the single-parameter κ -Köhler model, it is at least advised to consider that κ values derived from HGF data at 80 % or 90 % RH may not apply accurately for the calculation of CCN activation properties of such biomass burning aerosols.”

General comments:

(1) Please add a brief discussion on the expected performance of activity models for 4-hydroxybenzoic acid. It would help the reader understand where sensitivities might lie.

Replay: According to the reviewer's suggestion, we have added some additional discussion to the revised manuscript to highlight that the difference between models and measurements are not due to model error, but due to assumptions about the physical state, as mentioned on page 11, line 293.

Related additions and changes included in the revised manuscript:

On page 11, line 291, we add the following statements after “obvious in Fig. 2c”: “These deviations surpass by far the expected error in model performance, which is typically less than 0.05 in HGF units for $RH < 85\%$, as indicated also by an intercomparison of the AIOMFAC and E-AIM predictions in Fig. 2c and the much improved model-measurement agreement for the case of mixed 4-hydroxybenzoic acid + AS particles shown in Fig. 4 (discussed in Section 3.2.2). However, note that the validity of the shown model predictions in Fig. 2c depends on whether the assumption of a liquid solution droplet is plausible.”

(2) Have the authors considered how a variable morphology would influence results? Is there no literature data on studies using AIOMFAC on this?

Response: We add some discussion on potential particle morphology effects. To our knowledge, there are no studies where morphology (non-sphericity) of solid particles is explicitly considered with AIOMFAC. In the case of phase-separated particles, overall sphericity is still assumed in AIOMFAC-based HGF predictions.

Related additions and changes included in the revised manuscript:

Page 11, line 296, we add: “Morphology effects, such as the restructuring of non-spherical polycrystalline particles over a certain RH range or liquid-liquid phase-separated particles of non-spherical shapes, have been discussed by several groups (Sjogren et al., 2007; Reid et al., 2011; Lei et al., 2014). In the case of hygroscopic growth and deliquescence under hydration conditions for 4-hydroxybenzoic acid particles and mixtures of 4-hydroxybenzoic acid with ammonium sulfate. An offset between measurement and model predictions was observed both in the RH range below the deliquescence of the particles and above it, i.e. above 80 % RH, (Lei et al., 2014). It is suggested that deviations are primarily caused by a change in solid-state particle morphology during hydration, leading to a restructuring of the polycrystalline particle shape towards more compact, near-spherical shape as the RH approaches the particle deliquescence point. This would explain rather uncommon HGF values of less than 1.0 at elevated RH also shown in Fig. 2c. Similar

behavior was found for experimental growth factors of mixtures containing adipic acid and AS and systematic deviations between the associated ZSR predictions and observations by Sjogren et al. (2007). Thus, while experimental data hint to the possible influence of non-spherical particles and their humidity-induced restructuring as a source of uncertainty, model predictions of HGF, such as those with the AIOMFAC model, assume by default a spherical particle shape even for solid phases and/or in cases where LLPS is present.”

(3) What is the residence time of particles in the HTDMA? If there were a phase state change from which kinetic mass transfer limitations might apply, how might this change your conclusions?

Reply: We add further discussion on the effect of potential mass transfer limitations for aqueous aerosol HGF measurements and for the solid-liquid phase transitions during deliquescence.

Related additions and changes included in the revised manuscript:

Page 13 line 335-343, we add: “the rather high viscosity of solutions containing levoglucosan is expected to increase considerably toward RH (Marshall et al., 2016). This increase in viscosity might impede the crystallization of AS in the mixed systems on the time scale of the experiment. Mass transfer limitation effects on the deliquescence process of crystalline organic particles and the water uptake or evaporation have been investigated in several experimental studies (Peng et al., 2001; Choi and Chan., 2002; Chan and Chan., 2005; Sjogren et al., 2007; Zardini et al., 2008; Ciobanu et al., 2010; Smith et al., 2012; Mikhailov et al., 2013; Hodas et al., 2015). Mass transfer limitations may impact the outcome of experiments significantly if the characteristic time scales for equilibration is similar to or larger than the residence time of particles in the experimental setup. In this study, the total residence time of the aerosol sample during the equilibration phase before entering the DMA2 is about 8 s. In order to improve the probability that the particles reach equilibrium with the target RH during this residence time, the monodisperse aerosol selected by DMA1 is first humidified to 98 % RH. The aerosol particles are then exposed to a lower target RH by a two-step process using double Nafion tubes. Kerminen (1997) estimated the necessary residence time for achievement of water equilibrium of aqueous droplets to be between 8×10^{-6} s and 0.1 s for 100 nm and 500 nm particles, respectively. Therefore, the typical residence time of a few seconds in the humidification or dehumidification section in a HTDMA measurement is assumed to be sufficient for most equilibrium hygroscopicity measurements (Brooks et al., 2004;

Mikhailov et al., 2004). Moreover, our HGF results for the three pure organic components are in good agreement with respective data by Mochida and Kawamura, (2004), Brooks et al., (2004) and Chan and Chan (2005), conducted with different techniques and/or residence times. However, there are cases where water equilibration could be impeded substantially in the presence of highly viscous or glassy particles at low RH, e.g. for ternary sucrose + NaCl + water particles of $> 6 \mu\text{m}$ in diameter studied by Bones et al. (2012), who report an equilibration time scale $> 1000 \text{ s}$ for such particles. Note that, aside from viscosity, there is an important size-dependence of the particles on the equilibration time scale (e.g. Koop et al. 2011). For aqueous 100 nm particles used in HTDMA experiments at room temperature, Bones et al. (2012) indicate that the equilibration time scale for water is likely only of concern for $\text{RH} < 10 \%$ in such an instrument. We therefore conclude that the residence time of 8 s is very likely sufficient to allow for equilibrium HGF measurements in dehydration mode, at least down to 10 % RH (when starting with aqueous solution droplets).

Mass transfer effects in hygroscopicity measurements of aerosol particles during hydration conditions have been encountered previously, particularly when a solid-liquid phase transition (deliquescence) is involved (Peng et al., 2001; Chan and Chan., 2005; Sjogren et al., 2006). For example, Peng et al. (2001) observed in electrodynamic balance (EDB) experiments under conditions of very slow humidification that glutaric acid aerosol particles showed a deliquescence phase transition in the RH range from 83 to 85 % over the course of several hours. This is a much longer time span than that of $\sim 40 \text{ min}$ for the deliquescence of other super-micron sized dicarboxylic acid particles (e.g., malonic acid) in EDB experiments. This observation indicates that the solid-liquid phase transition of glutaric acid particles may likely be mass transfer limited during the hydration process. In this context, it is possible that the deliquescence of initially solid, pure 4-hydroxybenzoic acid particles at $\text{RH} > 97 \%$ is further impeded by slow dissolution, which could have led to the absence of deliquesced particles (Fig. 2c) on experimental time scale.

(4) I would appreciate more discussion on how the reliance on 3 organic surrogates influences conclusions for a SOA class that is likely much more complex. Are you studies sensitive to complexity, influenced by a discrete range of solubilities and 'step-like' transitions? How would you test this?

Reply: According to the referee's suggestions, we will added more discussions on the reliance of organic surrogate compounds on hygroscopic behavior of mixtures of biomass burning in the revised manuscript.

In this study, we focus on two distinct aspects of hygroscopicity-related research: (1) well-defined hygroscopic growth factor measurements and modeling for simple organic-inorganic mixtures. (2) Comparison of the effective hygroscopicity parameter of relatively simple mixtures of representative organic biomass burning surrogates to hygroscopicity values (κ) obtained from field data. The experimental observations with well-defined organic-inorganic laboratory systems suggests that such systems exhibit a considerable variability with regard to liquid-liquid and solid-liquid phase transition during humidification/dehumidification. These phase transitions may be absent or different in other systems of highly complex biomass burning organic aerosols from the field. Moreover, whether typical field particles show step-like phase transitions cannot answered without direct measurements in the field or with carefully sampled ambient particles. Our experimental hygroscopicity data for model systems representing biomass burning particles tend to show that distinct step-like features in the hygroscopicity curves are at least less noticeable in the RH range from 90 to 40 % when several organics are mixed, which is in agreement with findings by Marcolli et al. (2004). Marcolli et al. (2004) suggest that mixtures containing many different organic compounds tend to thermodynamically favor the liquid state and suppress crystallization of individual organic compounds.

Related additions and changes included in the revised manuscript:

Page 16 line 434-443: "To represent of biomass burning organic compounds.....compounds and ammonium sulfate" was revised to

"According to Decesari et al, (2006), sampling of aerosol particles, including the WSOC fraction, was conducted from September 9 to November 14, 2002 in their field study, the sampling time was subdivided into different periods. Despite of significant changes in the chemical composition of tracer compounds from the dry to the wet period, the functional groups and general chemical classes of WSOC changed only to a small extent in the Amazon basin near Rondônia, Brazil. Model compounds represent semi-quantitatively (presence/abundance of functional groups) and the chemical structure of WSOC can be used as surrogates in microphysical models involving organic aerosol particles over tropical areas affected by biomass burning scenarios (Andreae et al., 2002; Artaxo et al., 2002; Rissler et al., 2006; Decesari et al., 2006). Here, we focus on experimental

observations and model calculations for relatively simple mixtures of inorganic-organic surrogate components reflecting mixtures of aerosol components found during different seasons during biomass burning events. However, we are fully aware of that fact that actual biomass burning aerosols are typically much more complex in terms of particle chemical composition. Aerosol particle properties from biomass burning events depend on the types of sources, external/internal population mixing state, water-solubilities, and phase state of the diversity of organic compounds and their mixing with inorganic constituents during different time periods in the field (e.g. Decesari et al., 2006).”

Reference

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