

Review of "Effect of mixing structure on the water uptake of mixtures of ammonium sulfate and phthalic acid particles" by Weigang Wang et al.

### Summary:

Wang et al. investigated the effect of different internal mixing structures (homogeneously mixed and core-shell structure) on the water uptake of aerosols consisting of ammonium sulfate (AS) and phthalic acid (PA). In addition, they studied how the amount of PA in the particle affect the water uptake. They used specific HTDMA-instrument to select specific sized particles, add PA coating into them (when core-shell structures were studied) and humidify then following by measurement of the growth of the particles as they uptake water. To accompany the measured data, they used theory for estimating the hygroscopic growth of individual components and Zdanovskii-Stokes-Robinson (ZRS) relation to calculate the hygroscopic growth of mixed particles. For homogeneously mixed particles (also referred as well-mixed particles) they observed, for example, that a decrease in the hygroscopic growth factor (GF) with increasing mass fractions of PA at above RH 80% level. They state that these results also agreed with previous studies, and the predictions from ZRS also agreed rather well for the well-mixed particles. For the core-shell structured particles, Wang et al. observed an increase in the GF as the size of the AS core decreased (below 80% RH level). At above 80% RH level, they observed a decrease in the GF with decreasing size of the AS core. For the core-shell structured particles ZRS predictions underestimated the hygroscopic growth. As a general comment, the methods and experimental procedures are adequately described, and they seem valid for this type of study. My main criticism concentrates on to the relevance of the study, and what new information it brings to the scientific community.

However, I do recommend this paper for publication if the issues raised below are adequately addressed.

### Major comments:

Why liquid well mixed AS-PA would have different hygroscopicity compared to the AS particle with PA coating? Or was this the research question of the study?

The introduction of the draft is strongly focused on the water uptake of the aerosols which is the main theme of the paper. The atmospheric relevance of phthalic acid is discussed in the introduction (lines 86-98) into some extent, and shortly mentioned in the conclusions (lines 347-349). However, as published studies about the hygroscopicity of organic coatings with inorganic core do exist could you provide more explanation what new this study brings and how PA is relevant to the atmosphere in larger scale and thereby justify its use in this specific study? Can the results acquired for PA be generalised for other organic compounds too? For example, the authors mention that PA is common in rural mountains and marine atmospheres in Asia, so is it specific to those areas only? Could you, for example, give an estimate how much of PA there is in the atmosphere compared to other organic compounds? In addition, even though the use of ammonium sulfate is very common for this type of studies, its relevance/why it was used could also be shortly mentioned.

As you note in the introduction (lines ), with  $0.5 < O:C < 0.8$  the LL-phase separation is possible (and was always observed with O:C smaller 0.5 based on You et al. 2014) and depends on the organic used. As for phthalic acid (C<sub>8</sub>H<sub>6</sub>O<sub>4</sub>), O:C is 0.5, how do you know that the particles actually are well mixed in section 2.1.1 after humidification? This is critical for the whole study and this assumption should be justified.

Regarding section 2.1.1. As solid crystalline ammonium sulfate is not spherical, how much this affects the estimation of the coating thickness? Further, could the difference between measured GFs and modelled ones be explained by this uncertainty in the coating thickness and further in the calculated organic mass

fraction? Even though the coating thickness is calculated to give equivalent mass fraction of PA compared to the well mixed case, I would assume that even small uncertainty in the HTDMA measurements and thus in the coating thickness affects the amount of calculated PA mass fraction. The mass fraction should be marked also to the figure 3.

There are numerous English language mistakes, especially in the last half of the paper. I have marked some of them for specific comments below. However, I suggest careful reading and re-writing, especially for the section 4, as there were some paragraphs that I could not understand at all. Related to section 4 (Summary and conclusions), I find that the main results mentioned in the abstract (e.g. at high RH GF decreases as thickness of PA shell increases) and conclusions derived from those are completely missing.

**Specific/minor comments:**

The acronym HTDMA is never explained, even though HTDMA instrument/setup is major part of the study.

Lines 54-56: terms homogeneously mixed and well mixed are both used later in the paper and figures, which is confusing (I understood they mean the same thing). I would select one of them and use it throughout.

Line 55: ...aerosol particles may be divided...

Line 59: ...of the earlier studies...

Lines 69-71: verb is missing, I assume you mean something like ...few laboratory studies have investigated the influence...

Line 76: change difference to different?

Line 93: correct typo in hygroscopicity

Line 104: ...aerosol particles have an average...

Section 2.1: Temperature to what the particles are exposed to is not mentioned, please add it. Ambient temperature was mentioned later at some point, but please be more specific as the experiments should be repeatable by others.

Line 166: ...aerosols were pre-humidified...

Line 168: I am not sure if I understand what you mean by "Finally the conditioning core-shell particle" here (referring to "the conditioning"). Maybe just the core-shell particle or remaining core-shell particle?

Line 174: Is the equation (1) now from the AIOMFAC model mentioned in lines 117-122? If it is, I would also mention that here as by first reading time I was wondering from where Eq. (1) comes from.

Line 211: ...AS in the...particles dissolves...

Lines 231 and in some other places: when reporting numbers, I believe the correct way is to use "and" before the last number (i.e. 100, 150 and 200nm instead of 100, 150, 200nm)

Line 246: literatures -> literature

Line 247: ...observed a strong higher water uptake... -> unclear, possibly "observed higher water uptake"

Line 248: correct typo: mass rations -> mass ratios

Lines 259-260: very long sentence, hard to follow. Could you reformulate/have two sentences instead?

Line 276: change “it assumes” into “it is assumed that”

Line 276: please remind the reader about the morphology effect also here with a short sentence in addition to referring section 3.2.

Line 289: correct “is” to “are” as you have plural

Line 294: AP most likely typo, please correct

Line 298-299: The sentence is unclear, maybe remove “the”?

Lines 301-304: very long sentence, so what was actually observed?

Line 309: repetition (observation was observed). Maybe change to ...a contracting observation was made...

Line 332-333: I do not understand the sentence starting “they found that...”, please elaborate/correct sentence structure

Lines 338-342: Again, very long and complicated sentence, hard to follow. Please simplify.

Line 352: change “estimation of” into “estimator for”

Lines 357-359: It seems there is main verb missing?

Lines 360-361: “According to filed studies...a variety of organic aerosol particles were characterised in the atmosphere...” This seems a little off from the context. Or do you mean that also in other studies various particle properties have been characterised? By first reading it seems you are writing about identifying particles, which is not related to your study in that sense.

Lines 366-367: ...to depend on the difference of influence of kinetic limitations”. Please reduce the use of “of” for clarity.

Line 369: change “significant” to “important”?

Line 370: change “combining” to “combined”

Line 371: change “organics coating” to “organic coatings”

Line 370-375: starting from “understanding...”. Very long sentence, please break into smaller sections & elaborate, as now it is really hard to understand

Line 377: are the humidity cycles or particles depending on the ambient RH history? Make this clear in the text.

## References

You, Y. et al, International Reviews in Physical Chemistry, 2014, Vol. 33, No. 1, 43–77,  
<http://dx.doi.org/10.1080/0144235X.2014.890786>