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Comment on acp-2021-763

Anonymous Referee #2

Referee comment on "Technical note: Real-time diagnosis of the hygroscopic growth micro-dynamics of nanoparticles with Fourier transform infrared spectroscopy" by Xiuli Wei et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-763-RC1, 2021

In this manuscript, the authors present the hygroscopic study of 100 nm ammonium sulfate and ammonium sulfate mixed with sodium nitrate/oxalic acid nanoparticles using Fourier transform infrared (FTIR). The aerosol liquid water content was obtained using the FTIR spectral, and further the hygroscopic growth factor was calculated. The GF measurements are neither better nor worse than those of previous studies. The sequential order during the deliquescence process was also discussed with the 2D-IR spectroscopic analysis. From this point, the method is quite meaningful to better understand the intermolecular interaction within the phase transition.

The work is laboratory-based but has relevance to modeling. The topic of the manuscript fits into the scope of ACP. However, there are still some small mistakes in the manuscript. More discussion could be addressed before publication. I have several comments and suggestions for the authors below.

Major comments:

1. The authors mentioned the nanoparticle with approximately 100 nm is the electrical mobility diameter (line 25). Later the diameter of 100 nm was described as volume equivalent diameter in the experiment description section. The authors better describe clearly. For spherical particles, assuming that the particle and its mobility equivalent sphere have the same charge, then Dem=Dve. For non-spherical particles, the shape factor and slip correction factor should be considered.

2. The authors stressed several times that the nanoparticle shape is one of the large uncertainties for the hygroscopicity study. Because normally the nanoparticle is assumed to have a spherical shape. Actually, the authors also supposed spherical particles when calculating the growth factor in this study, even the particles were deposited on the

substrate. Since this is not a shape factor study, I would recommend do not emphasize this point.

3. About RH:

1). The authors claim the accuracy of RH measurement for the sample cell is 0.1% (Line 117). Is this 0.1% available for the whole studied RH range? Which sensor or instrument was employed? Normally the uncertainly will get larger when the RH increases (Mikhailov and Vlasenko, 2020). And 0.1% RH is extremely precise.

2). The initial RH is 45% for AS measurement and there is an OH stretching vibration peak at 3250 cm-1 for all RH below DRH in Figure 2. Can authors explain why? I am considering whether the RH downstream of diffusion dryer (or DMA) was below the ERH value of AS. If not, then the particle is a droplet at the initial 45% RH.

4. The authors mentioned the mass of nanoparticles was quantified using a simple procedure. Could authors prove more details? And it would be nice to add some discussions about the uncertainty of mass and GF. Actually, there are error bars shown in some figures. But there is no description in the main text.

5. The authors illustrated the OA does not absorb water in the RH between 40% to 90% in Figure 4. However, some previous studies demonstrated that oxalic acid dihydrate could form which will absorb water continually (Wang et al., 2017; Ma et al., 2019; Prenni et al., 2001). Since the FTIR could identify the hydration interactions, any explanations?

6. The DRH point in both AS/NA and AS/OA systems is lower than the pure AS. According to 2D-IR results, the hydrolysis reaction mechanism of AS/OA is different from AS/NA. Also considering OA does not absorb water, then why the DRH value gets smaller?

Minor comments

According to the detailed description of manuscript type in ACP (https://www.atmospheric-chemistry-and-physics.net/about/manuscript_types.html), I would recommend Research articles instead of Technical notes.

Line 38, “but only AN can change the hydrolysis reaction mechanism for AS in AS/AN and AS/OA mixtures.” Is there AS/AN and AS/OA mixture experiment?

Line 42, "… between nanoparticle and medium", medium size particle? Please rewrite this sentence. Same Line 102

Line 47, "Nanoparticles have long atmospheric lifetimes of weeks to months". Any references?

Line 80, "the hygroscopic growth process of a single aerosol with particle diameter of less than 100 nm" maybe become "the hygroscopic growth process of a single particle diameter less than 100 nm". Please rewrite this sentence.

Line 155, "room temperature is assumed to be 25 °C". What does this mean? No temperature sensor could measure the room temperature? Since the temperature has an influence on the RH, is there any temperature monitoring inside the sample cell?

Line 156, the authors described the RH varies from 50% to 95% in the present work. But in AS study, the initial RH is 45%. In the OA study, the initial RH is 40%.

Line 209, "the nanoparticle volume increases but its mass keeps constant." Quite misleading, its means sulfate not nanoparticle, right?

Line 255, "at the RH of 79.9 ± 0.10%", is this RH measurement from this study? And how do the authors obtain this value? Since the FTIR measurement is a real-time method. Why authors don't provide more data points between 74% and 81% RH?

Line 282, what does the 1097 cm-1 stand for? NH4+? It seems hard to see a peak at 1097 cm-1 from Figure 2.

Line 310, what does the 1320 cm-1 stand for?

I would recommend the authors provide all FTIR spectral figures (OA, AS/NA, and AS/OA) at least in the supplement.

Table1, I would recommend adding the reference (or data source) for density and solubility.

Technical corrections

Unified abbreviation. Line 33 & 37, Sodium nitrate (SN); Line 38 & 90, sodium nitrate (AN); In the Figure 5&6, NN.

Line 52, “is” not are

Line 68, “the” nanoscale

Line 73, “the” nanoparticle

Line 92, “in” real time, not on real time; also other places

Line 93, “the” molecular scale

Line 209, “large” size particles, not big

Line 212, “the” Kelvin effect

Line 238, “behavior” not behaviour

Line 247, “via direct measurement of the aerosol diameter”

Line 259, “increasing”

Line 263, “the” FTIR measurement

Line 349 & 359, “the” 2D-IR spectroscopic

Line 362, “a” better understanding

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