

Atmos. Chem. Phys. Discuss., referee comment RC3
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Comment on acp-2021-763

Anonymous Referee #3

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-RC3>, 2021

This study used the FTIR spectroscopy and two-D correlation analysis to investigate the hygroscopicity of several single-component aerosols and their mixtures. The sequential order during the hydration process was discussed with the 2D-IR spectroscopic analysis. The method is new. However, as a technical note, validation of the performance of current method should be conducted in a more thorough way. I have several comments regarding current version of the manuscript and hope the authors could carefully address those before being considered for publication.

Major comments

Since submitted as a technical note, the authors should provide a solid and sound validation of the accuracy and stability of their technique. For instance, was your DMA calibrated? How much did you dry your aerosols? How many particles did you deposit onto the substrate before further measurements, would the concentration of the particles deposited influence your results? How was the RH measured, what was its uncertainty? Was the cell well-insulated that the RH could be maintained? What parameters did you directly obtain from the technique and how did you interpret them? Each single part of your instrument should be described clearly. Second, the authors used AS to calibrate the instrument. However, the figures the authors plotted are lack of critical data points, particularly between 74% RH and 81% RH.

The authors deposited 100 nm particles onto a substrate. These material could not represent nanoparticles anymore. The condensation of water molecules on a curved particle could be much different from that on a flat surface with deposited substances on. The author should clarify these differences and give proper discussion, emphasizing its own atmospheric relevance. A section discussing its atmospheric implications is suggested.

The authors stated in their abstract as well as in the intro part that: "current techniques are also difficult to identify the intermolecular chemical interactions of phase transition micro-dynamics during nanoparticle deliquescence process because their limited temporal resolutions are unable to capture the complex femtosecond-level intermediate states". I am curious from current results you presented did not show a higher temporal resolutions and femtosecond-level intermediate states were not displayed, either. Otherwise, please clarify.

Based on my third comment, the author should emphasis the novelty of current technique. What are the advantages compared to traditional hygroscopicity measurements? I assume Sect 3.3 and 3.4 are quite important, contributing new insights to current understandings. However, the discussion in these part was kind of weak and plain. Proper comparisons with other studies may help.

Minor comments:

Line 90: AN or SN? Similar in line 96.

Line 102: Current work is of great significance, not only for the haze control across China, but also for the whole atmospheric community. There remains huge gaps and requires significant efforts for the haze control across China using current results. Consider rephrase the sentence or extend the content.

Line 108: How much you dried your aerosols? For instance, to which RH condition?

Line 110: How many particles you deposited onto the substrate? Will they overlap on each other?

Line 151-160: Even though you described our instrumentation, I am still confused how you measure or derive D_{wet} .

Line 193: What do you mean by "area of OH"?

Line 211: Your DRH value for small particles is similar to that of large ones from other studies. But this is not enough to prove the accuracy of your method, as GF value of AS at a certain RH is also a necessity and it varies between small particles and large ones. Rephrase the sentence and make thorough comparison.

Line 211, also for Fig.2 and Fig. 3: Moreover, I observed that you performed the measurements at 74% RH and at 81% RH. At 81% RH, you observed deliquescence and you reported this value as DRH. This is not sound, it could be any values between 74% and 81% RH, please consider add more data points between these two RHs, specifically for your Fig.2 and Fig. 3. Before you add more data points at those RHs, I cannot be convinced.

Line 218: What do you mean by 'nanoparticle volume increases but its mass keeps constant'? Please clarify.

Line 223: When you referred the calculation to previous section, for instance, M_{water} and M_0 , you should specify all of these quantities in previous part. I am quite confused that how you obtained M_{water} , M_{wet-Mo} ?

Line 228: Please add comparison with other studies, especially 100 nm particles of oxalic acid. Boreddy et al. (2018) measured the GF of oxalic acid using a HTDMA and they found that oxalic acid started to absorb water at $RH < 45\%$ and the GF of oxalic acid was 1.47 at 90% RH. Please consider giving proper discussion.

Line 236: I suggest you add the results for sodium nitrate (SN) particles before you discuss the results for mixtures. Again, it should be SN but not AN, isn't it? Also in Fig. 5, what does NN stand for?

Line 254-259: Similarly, add more data points for GF at RH between 74% and 81% to complete your validation. And the content of this paragraph was well known by hygroscopicity community, you don't have to repeat it, but give detailed description of your DRH value as well as corresponding GF and compare with other studies.

Line 260-265, also for Fig. 6: At RH between 70% to 85%, your results for mixtures deviate from the predictions. Give proper explanations. You cannot just state that they are in good agreement with other studies without presenting their data points.

Line 269: What does absorption peak mean? Please clarify.

Line 271: Since 2D-IR spectroscopic technique is a major instrument for your results, you should describe it in a more detailed way. For instance, in Sect2.1. The description was

too simplified and should be extended. Give possible examples and emphasize its significance. I do think line 267-277 should go to the experiment section. And please further explain "Synchronous correlation maps" and "asynchronous correlation maps", give examples and how we use these results, what do these results indicate or how we interpret them.

Line 310: So all the auto-peaks were all the same during all the RH conditions? Similar questions to the asynchronous correlation maps.

Line 341-345: Too long sentence, please rephrase it.