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Title: Rapid measurement of RH-dependent aerosol hygroscopic growth using a humidity-controlled fast integrated mobility spectrometer (HFIMS)

We thank the anonymous referees for their valuable and constructive comments/suggestions on our manuscript. We have revised the manuscript accordingly and please find our point-to-point responses below.

Comments by Anonymous Referee #1:

General Comments:

In this study, Zhang et al. developed a humidity-controlled fast integrated mobility spectrometer (HFIMS) for fast measurements of aerosol hygroscopic growth. Based on their previous work, a dual-channel humidifier and an optimized measurements sequence for different size particles were employed to achieve fast measurements. Compared to the H-TDMA techniques, the measurements time needed for a complete RH range cycle (20~85%) were largely shortened for the HFIMS, demonstrating its good feasibility in hygroscopic growth measurements for size-resolved ambient aerosols under different RHs. The manuscript is well written and easy to follow, I have several minor suggestions for authors' consideration.

Detailed Comments:

Line 35: should be "an HTDMA".

Responses: We have corrected it in the revised manuscript.

Line 76: Please clarify the residence time of aerosols in the humidification section.

Responses: The average transport time of particles from the inlet of the RH conditioner to the inlet of the WFIMS is ~ 3.2 s, and we have clarified this point in the revised manuscript in lines 82 – 83 as:

"The average transport time of aerosols in the humidification section, including the Nafion exchanger and the tubing leading to the WFIMS separator inlet, is ~ 3.2 s."

Line 95-96: Please clarify how to control the WFIMS sheath flow rate. A proportional solenoid valve coupled to a PID controller used here?

Responses: The total flow of WFIMS, i.e., the sum of the sample flow and sheath flow, is maintained by a critical orifice at a flow rate of 15.2 LPM. With the constant total flow, the desired aerosol flow rate of 0.3 LPM is achieved by PID control of sheath flow using a proportional solenoid valve. We have added a reference to detailed previous work in the revised manuscript in line 102.

Section 2.2: RH in both aerosol and sheath flow was rather sensitive to the temperature fluctuation, please state how to maintain the stability of temperature during measurements.

Responses: Yes, both the WFIMS separator and the growth section are temperature controlled, as well as the bubble humidifier. The temperature differences among different sections of aerosol and sheath flow passage, including WFIMS separator, are less than ~0.1°C, which corresponds to an RH variation of less than 0.6 % (relative change). Note that there were some variations of room temperature due to the activity in the lab, However, such variations are very slow in comparison, and they do not influence the growth factor measurements which take place at a much shorter time scale.

Line 159-160: An optimized measuring sequence was introduced for six particle sizes; however, have the authors evaluated the effect of multi-charge on particle size determination, due to a large proportion of multi-charge particles (especially for 100-300 nm) selected by differential mobility analyzer (Shen et al., 2021, https://amt.copernicus.org/articles/14/1293/2021/)?

Responses: We thank the reviewer for this constructive suggestion. The multi-charging effect is not taken into consideration in the current data inversion routine, and it will be a subject of our future work. We have clarified this in the revised manuscript in lines 229-231 as:

"The probability density function of the hygroscopic growth factor (GF-PDF) is retrieved from the HFIMS measurements using an inversion routine described in Wang et al. (2019). We note that particles carrying multiple charges could contribute substantially to the aerosols classified by the DMA, especially for particles larger than 100 nm (Shen et al. 2021). This multiple-charge effect is not currently accounted for in the inversion routine and will be a subject of future study."

Line 212-214: Have the ambient aerosols been dried before measurement by a SMPS?

Responses: Yes, the ambient aerosols were dried to an RH below 30%. It is now clarified in the revised manuscript in lines 219-220 as:

"Prior to the SMPS measurement, the aerosol sample was dried to an RH below 30% using a diffusion dryer."

Section 3.2.2: I would like to draw the authors' attention to a recent review paper on the tropospheric aerosol hygroscopicity in China (Peng et al., 2020). It may be beneficial for the authors to discuss the RH and size dependence of ambient aerosol hygroscopicity. (https://acp.copernicus.org/articles/20/13877/2020/)

Responses: We thank for the reviewer's suggestion. We have revised the text accordingly in lines 43-46:

"Particle hygroscopic growth is a function of RH and the hygroscopicity parameter (Petters and Kreidenweis, 2007), which is a function of particle composition and often varies strongly with particle size (e.g., Peng et al. 2020). Both RH and aerosol composition can exhibit strong temporal and spatial variabilities (Tang et al., 2019). In addition, the hygroscopicity parameter can vary substantially with RH (Pajunoja et al., 2015; Rastak et al., 2017; Liu et al., 2018; Peng et al., 2020)."

Line 269-270: Please add references to support this claim.

Responses: Following the reviewer's suggestion, we have added a reference, and the sentence now reads:

"The mean κ values for 35 nm and 50 nm particles then exhibit a slight decrease following the NPF event, possibly due to the depletion of sulfuric acid and increased contribution of secondary organics to the growth of nucleation mode particles in the late stage of the NPF event (Dusek et al., 2010; Zheng et al., 2020)."

References

Dusek, U., Frank, G. P., Curtius, J., Drewnick, F., Schneider, J., Kürten, A., Rose, D., Andreae, M. O., Borrmann, S., and Pöschl, U.: Enhanced organic mass fraction and decreased hygroscopicity of cloud condensation nuclei (CCN) during new particle formation events, Geophysical Research Letters, 37, https://doi.org/10.1029/2009GL040930, 2010.

Zheng, G., Kuang, C., Uin, J., Watson, T., and Wang, J.: Large contribution of organics to condensational growth and formation of cloud condensation nuclei (CCN) in the remote marine boundary layer, Atmospheric Chemistry and Physics, 20, 12515-12525, 2020.