Reviewer #2,

Comment**:** This paper presents a method of how to derive metrics of diversity of a particle population with respect to hygroscopicity using H-TDMA measurements. The authors also demonstrate the use of their method by applying it to a dataset from the ambient atmosphere. This work fulfills an important need in our community, and I commend the authors on their contribution. So far, it has been very challenging to quantitatively derive these metrics from measurements since they rely on the quantitative knowledge of per-particle composition which have been challenging to obtain. Given that H-TDMA datasets have been collected in many different environments and could all be analyzed using the method described in this paper, this work has a great potential for deepening our understanding of aerosol mixing state in the ambient atmosphere and for providing much needed data to validate mixing-state-aware models.

The paper is concise and well-structured. It fits within the scope of ACP and I recommend publication after a few minor comments are taken into account.

Reply**:** Thanks for the comments and suggestions. The point-by-point responses are listed below.

Comment**:** Specific comments:

(1) line 21: The statement that heterogeneity in hygroscopicity is not considered in models is a little strong. Sectional models do capture the dependence in (average) hygroscopicity with size and modal model capture the variation in hygroscopicity for different sub-population. I suggest saying "not fully considered" or "not adequately considered". In fact, it is the case that many modeling approaches do provide some information about how hygroscopic and non-hygroscopic species are mixed (e.g., MAM4 in CESM) but so far, suitable measurement data has been lacking to validate these predictions. Developing a method to provide this kind of data is the contribution of this study.

Reply**:** Thanks for the comment. We have revised the expression. (Lines 21-22, Page 1 in the revised manuscript)

(2) Nitpicky terminology comment: The term "aerosol" already refers to a population, so there is no need to say "aerosol population".

Reply**:** Thanks for the comment. We have revised the expression for the whole manuscript.

(3) Line 32: Where the kappa-pdf is introduced would be a good place to cite Su et al., 2010, Atmos. Chem. Phys., 10, 7489–7503, 2010, where a general concept and mathematical framework of particle hygroscopicity distribution for the analysis and modeling of aerosol hygroscopic growth and CCN activity is presented.

Reply**:** Thanks for the comment. We have added this reference. These literatures are crucial for perfecting this paper. They help to deepening and highlighting the significance of this study. (Lines 28-33, Page 2 in the revised manuscript)

(4) Explanation starting at line 104: This applies for one particular particle size, I suggest making this clear at the start of this section.

Reply**:** Thanks for the comment. We revised this sentence to make the expression clearer. (Line 104, Page 4 in the revised manuscript)

(5) Equations 2 and 4: add limits to the integral.

Reply**:** Thanks for the comment. These equations have been revised. (Equations 2 and 4 in the manuscript)

(6) Equations 8 and 9: suggest to not use ii as the counter variable. Use k or \ell, for example.

Reply**:** Thanks for the comment. We have revised the counter variable and used "*j*" instead. (Lines 113-116, Page 5 in the revised manuscript)

(7) Line 141: Can you explain a bit more why kappa for the coarse mode is assumed to be 0? Couldn't you have non-hygroscopic primary particles in the coarse mode that have aged and acquired some coating materials that make the more hygroscopic (or at least increase their kappa to > 0)?

Reply**:** Thanks for the comment. In this study, κ for the coarse mode is assumed to be 0 by considering that the primary chemical composition in this mode is nearly hydrophobic without aging and coating. The same assumption is applied in Chen et al. (2012). κ for larger particles is hard to be observed by H-TDMA technique so far, due to the technical bottleneck. Shen et al. (2021)

extended the H-TDMA measurement of aerosol hygroscopic properties to 600 nm in the urban environment, but for aerosol larger than 1 μm, κ can only be derived from the chemical composition according to the method used by Liu et al. (2014) and Gysel et al. (2007). As the reviewer mentioned, studies showed that aerosols larger than 1 μm are actually hygroscopic (Hegg et al., 2006; Massling et al., 2009; Liu et al., 2014), with κ slightly greater than 0.1 (Liu et al., 2014), due to the coating in atmospheric aging processes.

However, considering that aerosol particles larger than 1 μm generally have short lifetime and low number concentration except for the dusty weather, the influence of these particles on radiative forcing and cloud is limited. Also, the assumption that κ for the coarse mode is 0 can only have limited impact on the result of the size-resolved heterogeneity. More importantly, this section focuses on the application of the proposed algorithm. If κ-PDF for aerosols larger than 1 μm could be obtained in the future, it could be directly applied in the framework provided by our study to discuss the size-resolved heterogeneity in aerosol hygroscopicity and its impact on aerosol climatical and environmental effects.

This discussion is added in the revised manuscript. (Lines 143-148, Page 6 in the revised manuscript)

(8) Line 170-173: These sentences are unclear, can you please rephrase?

***Reply*:** Thanks for the comment. We have revised this paragraph in the manuscript. (Lines 173-178, Pages 7-8 in the revised manuscript)

(9) In the introduction/conclusion, you could stress more explicitly that existing H-TDMA datasets could be analyzed using this algorithm. This could have a large impact on how we use and think about these datasets and will help providing data for constraining models.

***Reply*:** Thanks for the comment. This suggestion deepens the significance of our work, and we added this discussion in the results and conclusions section to further show the importance of this work. (Lines 184-185, Page 8 and Lines 283-287, Page 11 in the revised manuscript)

References

Chen, J., Zhao, C., Ma, N., Liu, P., Göbel, T., Hallbauer, E., Deng, Z., Ran, L., Xu, W., Liang, Z., et al.: A parameterization of low visibilities for hazy days in the North China Plain, Atmospheric Chemistry and Physics, 12, 4935–4950, https://doi.org/10.5194/acp-12-4935-2012, 2012.

Gysel, M., Crosier, J., Topping, D., Whitehead, J., Bower, K., Cubison, M., Williams, P., Flynn, M., McFiggans, G., and Coe, H.: Closure study between chemical composition and hygroscopic growth of aerosol particles during TORCH2, Atmospheric Chemistry and Physics, 7, 6131–6144, https://doi.org/10.5194/acp-7-6131-2007, 2007.

Hegg, D., Covert, D. S., Crahan, K., Jonsson, H., and Liu, Y.: Measurements of aerosol size-resolved hygroscopicity at sub and supermicron sizes, Geophysical research letters, 33, L21808, https://doi.org/10.1029/2006GL026747, 2006.

Liu, H., Zhao, C., Nekat, B., Ma, N., Wiedensohler, A., Van Pinxteren, D., Spindler, G., Müller, K., and Herrmann, H.: Aerosol hygroscopicity derived from size-segregated chemical composition and its parameterization in the North China Plain, Atmospheric Chemistry and Physics, 14, 2525–2539, https://doi.org/10.5194/acp-14-2525-2014, 2014.

Massling, A., Stock, M., Wehner, B., Wu, Z., Hu, M., Brüggemann, E., Gnauk, T., Herrmann, H., and Wiedensohler, A.: Size segregated water uptake of the urban submicrometer aerosol in Beijing, Atmospheric Environment, 43, 1578–1589, https://doi.org/10.1016/j.atmosenv.2008.06.003, 2009.

Shen, C., Zhao, G., Zhao, W., Tian, P., and Zhao, C.: Measurement report: aerosol hygroscopic properties extended to 600 nm in the urban environment, Atmospheric Chemistry and Physics, 21, 1375–1388, https://doi.org/10.5194/acp-21-1375-2021, 2021.