Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2022-787-RC2, 2023

Comment on acp-2022-787

Anonymous Referee #2

Referee comment on "Quantifying particle-to-particle heterogeneity in aerosol hygroscopicity" by Liang Yuan and Chunsheng Zhao, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-787-RC2, 2023

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.

- 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.
- Nitpicky terminology comment: The term “aerosol” already refers to a population, so there is no need to say “aerosol population”.
- 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.
- Explanation starting at line 104: This applies for one particular particle size, I suggest making this clear at the start of this section.
- Equations 2 and 4: add limits to the integral.
- Equations 8 and 9: suggest to not use ii as the counter variable. Use k or \ell, for

example.

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