Response to Anonymous Referee #2

This is a nice piece of work, I enjoy reading it. This work provides a new method to measure single particle hygroscopicity using optical tweezer. This new method is well validated against E-AIM theoretical values and other observations from previous studies. Better measurements of single particle hygroscopicity could not only help us improve aerosol-cloud-climate interactions in the climate models, can also help us measure PM pollution more precisely (Chen 2025). The manuscript is well written and structured. I only have a few minor comments to help further improve the article, detailed below.

Response: Thanks for your valuable comments, which really helped improve the manuscript. Below, we will provide a detailed and point-by-point response to your comments. All the changes have been included in the latest manuscript.

A few minor comments may help improve the discussion.

1) suggest dropping the abbreviation of AOT, could just simply call aerosol optical tweezer (refer to only tweezer later for simplicity), because AOT is commonly referred to as aerosol optical thickness in the community, therefore, to avoid confusion.

Response: Thank you for the suggestion. In the revised manuscript, we have dropped the abbreviation "AOT" and consistently refer to the optical tweezers as "optical tweezers" to denote the aerosol optical tweezers.

2) L32: I guess you want to say "diameter" growth factor (CF)?

Response: Thank you for the comment. Yes, we are referring to the "diameter growth factor (GF)" in this context, and we have clarified the terminology in the revised manuscript to avoid confusion.

"During the measurement and characterization process, Hygroscopic growth is commonly quantified by the diameter growth factor (GF), mass growth factor (GF_{mass}), and the hygroscopicity parameter (κ), which link dry and humidified particle properties (Petters and Kreidenweis, 2007; Tang et al., 2019)."

3) Please provide explanation of error bars in the figure captions.

Response: Thanks for your suggestion. We have provided explanation of error bars in the figure captions and main text in the revised manuscript.

"Additionally, the error bars for the optical tweezers measurements represent the standard deviation, obtained either directly from the statistics of measurements or calculated through error propagation."

"Figure 2. Measurement of ammonium sulfate hygroscopicity. (a) Apparent molar volume of ammonium sulfate as a function of ionic strength. (b) Measured refractive index and corresponding values derived from the constrained solute mass. The error bars for the optical tweezers measurements represent the standard deviation.”

4) I think adding some discussion of the limitation of this new method, some forward looking of how to improve it in future studies, and some perspective of potential applications in future, these would help further strengthen the article.

Response: Thank you for this valuable suggestion. We have added a discussion in the revised manuscript (mainly in section 3.2 and 3.3) to highlight the limitations, potential improvements, and future applications of the method. Specifically:

“However, our method is currently applicable only to internally mixed particles. This is because the optical tweezers can trap only liquid droplets, and the retrieval framework requires the particle to be homogeneous. For externally mixed aerosols, insoluble inclusions may be present, leading to a heterogeneous refractive-index distribution. In such cases, both optical trapping stability and the spherical, homogeneous Mie scattering assumption may break down. For these types of particles, techniques such as HTDMA, or the development of Bessel-beam optical tweezers capable of trapping solid particles, would be more suitable for hygroscopicity measurements (Zhao et al., 2020). For particles containing substantial organic material or surfactants, liquid–liquid phase separation (LLPS) may occur at low RH. This would invalidate the standard Mie-fitting procedure, and additional models—such as core–shell Mie calculations—would be required to retrieve the radii and refractive indices of the individual phases before applying further thermodynamic constraints (Vennes and Preston, 2019). In contrast, if no LLPS occurs, changes in surface tension induced by organics are unlikely to affect the results, because Kelvin effects are negligible for micron-sized droplets.

Although a detailed treatment of these scenarios is beyond the scope of the present study, we suggest that the method could be extended in the future by incorporating more sophisticated optical models (e.g., core–shell Mie theory) as well as trapping techniques compatible with multiphase particles.”

References:

Vennes, B. and Preston, T. C.: Calculating and fitting morphology-dependent resonances of a spherical particle with a concentric spherical shell, J. Opt. Soc. Am. A, 36, 2089, https://doi.org/10.1364/JOSAA.36.002089, 2019.

Zhao, W., Cai, C., Zhao, G., Zhao, C.: Design of Bessel Beam Optical Tweezers for Single Particle Study, Acta Scientiarum Naturalium Universitatis Pekinensis, 56, 1031-1037, 10.13209/j.0479-8023.2020.090, 2020.