Anonymous Referee #1

This manuscript uses poke-flow measurements to determine the viscosity of organic-rich urban $PM_{2.5}$ collected from Seoul and Beijing in the autumn. They find that the viscosities of the filter extract samples are comparable to or higher than laboratory model systems (sucrose/ammonium sulfate/water, and citric acid/ammonium sulfate/water). The two biggest drawbacks in this study are that the droplets created from filter extracts are not necessarily representative of the original aerosol morphology and the poke-flow technique was limited to an RH regime lower than during sample collection. However, these limitations are discussed and clear within the manuscript, and these measurements provide important context for the viscosities of laboratory model systems as compared to field-collected material. Overall, I believe this manuscript will be suitable for publication after the authors address the following comments.

Response: Listed below are our point-by-point responses to the comments of the reviewer of our manuscript. For clarity, the referee comments are reproduced in black with bracketed numbers (e.g., [1]), and the authors' responses are provided in red with matching numbers (e.g., [A1]). We thank both reviewers for their careful reading of the manuscript and for their constructive and helpful suggestions.

Specific Comments:

[1] Line 176, 178, 182: The language of single liquid and two-liquid I think can be confusing here since these are complex field samples, I would recommend changing to "single-phase liquid" and "two-phase liquid" or LLPS.

[A1] We thank the reviewer for this helpful comment. To improve clarity, we have replaced "single liquid" and "two-liquid" with "single-phase liquid" and "two-phase liquid (LLPS)" throughout the manuscript, including in Sects. 3.2 and 4.

[2] Figure 1: Are the scales for 60% and 20% the same as the scales for 85%? In addition, if feasible I think it would be helpful for the scale to remain consistent between samples for all of the 85% and below samples. It appears that the 95% samples are all at the same scale, so I think it would be helpful to visualize the relative changes in size across samples.

[A2] We thank the reviewer for this helpful suggestion. To make it clearer, we have included the explanation in the figure caption as:

> "Optical images obtained during RH decrease for (a) Seoul and (b) Beijing $PM_{2.5}$ droplets showing phase separation, as RH decreases from ~95 % to 85, 60, and 20 %. Upon dehydration, particles transition from a homogeneous single-phase liquid to a core-shell morphology, illustrating separation of organic and inorganic components driven by water

loss. The images at ~95 % RH are shown at the same scale, while those at ~85 % RH and lower are presented at a consistent scale to facilitate comparison between samples. The scale bar represents 20 µm. Seoul (10/15) and Beijing (10/14) samples have already been reported by Song et al. (2025) and are included here for completeness of discussion."

[3] Line 219: The particles are semi-solid under the conditions of poke-flow, but during collection the RH was generally much higher. This is touched on at later points but please also include a discussion of what that could indicate for these samples here.

[A3] We thank the reviewer for this important suggestion. We agree that the contrast between the ambient RH during sample collection and the experimentally accessible RH range in the poke-and-flow measurements deserves explicit discussion in Section 3.3. We have added the following text after the first sentence of that section (Lines 266–274):

> "Although the poke-and-flow experiments were conducted under drier conditions (RH < ~45 %) than the mean ambient RH during sampling (Seoul: 69 ± 12 %; Beijing: 54 ± 13 %), direct quantitative viscosity constraints at higher RH could not be obtained, as the droplets behaved as low-viscosity liquids above ~45 % RH and relaxed too rapidly to yield a resolvable $\tau_{(exp,\ flow)}$. Nevertheless, comparison with sucrose–AS–H_2O systems (Fig. 5) indicates that semisolid behavior may still occur during at least episodic portions of the sampling period, particularly for the Beijing samples and during lower-RH periods in Seoul. These results, therefore, suggest that semisolid behavior of urban $PM_{2.5}$ cannot be ruled out even under the RH ambient conditions observed during the sampling period, particularly during drier episodes, and that viscosity measurements at higher RH remain an important target for future work."

[4] Line 235: Please add citations for why you are assigning 10^8 as the lower limit of viscosity for these particles and indicate if this is based on any physical or chemical properties.

[A4] The assignment of ~10^8 Pa·s as a conservative lower-bound viscosity when no restorative flow is observed within the experimental timescale (> ~2 h) is a well-established convention in the aerosol viscosity literature using the poke-and-flow technique (Renbaum-Wolff et al., 2013b; Grayson et al., 2015; Jeong et al., 2022; Gerrebos et al., 2024). The relevant citations have been added to the revised manuscript (Line 291)."

References:

- Gerrebos, N. G. A., Zaks, J., Gregson, F. K. A., Walton-Raaby, M., Meeres, H., Zigg, I., Zandberg, W. F., and Bertram, A. K.: High viscosity and two phases observed over a range of relative humidities in biomass burning organic aerosol from Canadian wildfires, Environ. Sci. Technol., 58, 21716–21728, https://doi.org/10.1021/acs.est.4c09148, 2024
- Grayson, J. W., Song, M., Sellier, M., and Bertram, A. K.: Validation of the poke-flow technique combined with simulations of fluid flow for determining viscosities in samples

with small volumes and high viscosities, Atmos. Meas. Tech., 8, 2463–2472, https://doi.org/10.5194/amt-8-2463-2015, 2015.

- Jeong, R., Lilek, J., Zuend, A., Xu, R., Chan, M. N., Kim, D., Moon, H. G., and Song, M.: Viscosity and physical state of sucrose mixed with ammonium sulfate droplets, Atmos. Chem. Phys., 22, 8805–8817, https://doi.org/10.5194/acp-22-8805-2022, 2022.
- Renbaum-Wolff, L., Grayson, J. W., Bateman, A. P., Kuwata, M., Sellier, M., Murray, B. J., Shilling, J. E., Martin, S. T., and Bertram, A. K.: Viscosity of α-pinene secondary organic material and implications for particle growth and reactivity, Proc. Natl. Acad. Sci. U.S.A., 110, 8014–8019, https://doi.org/10.1073/pnas.1219548110, 2013b.

[5] Figure 3: Similar to Figure 1, please adjust these images if at all possible to make the scale bars consistent between samples.

[A5] As suggested, we have adjusted the images in Fig. 3 so that the scale bars are as consistent as possible across samples. This facilitates direct visual comparison of hole-closure dynamics and particle morphology across the different $PM_{2.5}$ samples.

[6] Figure 4: Same thing as Figures 1 and 3, please adjust the scaling. Additionally, is there a reason why the brightness is so different for Beijing 10/14 0 s vs after 1–2 hours? I think it is probably clear there is no change in morphology, but the difference in brightness makes it more challenging to tell.

[A6] To address the comment, we have adjusted the images in Fig. 4 to make the scale bars as consistent as possible between samples. Regarding the brightness difference in the Beijing 10/14 panels: this arises from slight changes in illumination and focus conditions between the needle-insertion frame (t = 0 sec) and the subsequent frames acquired during the 1–2 h waiting period.

[7] Figure 5: Please indicate what experimental techniques were used to determine viscosities for the other studies included here.

[A7] We thank the reviewer for this suggestion. We have updated the caption of Figure 5 to specify the measurement technique used in each cited study. The revised caption now reads:

> "RH-dependent viscosities of Beijing and Seoul $PM_{2.5}$ droplets compared with sucrose–AS–H_2O and citric acid (CA)–AS–H_2O systems from previous studies (Jeong et al., 2022; Tong et al., 2022; Sheldon et al., 2023). Viscosities in the previous studies were determined using the poke-and-flow technique (Jeong et al., 2022), a dual optical tweezer system (Tong et al., 2022), and the droplet coalescence method (Sheldon et al., 2023)."

Technical Corrections:

[8] Line 166: “deducing” should be changed to deducting, subtracting, or another synonym. The same applies to Line 20 in the Supplementary Information.

[A8] Thank you for the correction. “deducing” has been replaced with “subtracting” in both the main manuscript and in the Supplementary Information.