Overall:

The manuscript by Ullah et al. presents measurements of the dynamic viscosity of aerosol particles generated from washing off filter samples of urban particulate matter ($PM_{2.5}$) collected in two megacities in Asia. Aerosol viscosities were determined using a combination of poke-and-flow experiments and fluid dynamic simulations. In addition, the authors report observations of liquid–liquid phase separation (LLPS) and other phase transitions during controlled dehumidification cycles from approximately 100% RH to 0% RH.

The dataset is valuable, particularly given the scarcity of viscosity measurements for real atmospheric particles. In my opinion, this novelty outweighs the relatively small number of filter samples and the limited number of droplets analyzed per sample, even though this limitation should be more clearly discussed. Overall, the subject of the paper aligns well with the scope of the journal, and the manuscript is clearly written.

I recommend consideration for publication after the points below have been addressed.

General comments:

- Sect. 2.1: It would be helpful to add some more information on the method used to wash off the filters. How did you ensure that all the organic and inorganic material was washed off the filter and was nebulized onto your glass slides for LLPS and viscosity analysis?
- Sect. 2.3: The explanation of how the microscope images were combined with fluid dynamic simulations deserves a more detailed discussion in the main text, as this is the focus of the results presented in this manuscript. Some important details are covered in the Supporting Information (Section S3), but flipping back and forth between the main text and the SI makes the reading difficult. See my specific comments below.
- The study contains a very limited number of samples, which make generalizations of observed trends on atmospheric PM2.5 difficult. While this limitation is acknowledged in parts of the manuscript (Sect. 3.3), it would be good to emphasis this in both abstract and conclusion. It could be helpful to indicate the number of droplets analyzed in terms of viscosity and LLPS for each filter samples and add this information to Table 1. E.g., for the Beijing filter sample of 10/03 I can only find 2 viscosity data points in Fig. 2, which renders a comparison with the other samples very challenging.
- Sect. 3.2: Can the authors provide more quantitative information on the separation relative humidity, i.e., the RH at which liquid-liquid phase separation was observed in your particles? Are the observed 1- vs. 2 liquid phases consistent with the O:C ratio of the organic material and the OIR of the samples and trends discussed in literature that the occurrence if LLPS is impacted by these parameters, i.e., O:C ratio and OIR

Specific comments:

L24: Change “to non-flowing behavior” to “phase state”.

L24: I would tune down “compositionall constrained” a bit, and or explain more clearly that you provide overall mass fraction of organic material and inorganic material (your Fig. S2), but no detailed chemical composition.

L34: Replace “cloud nucleation” by “cloud formation”

L36: Delete “within the regional and global”.

L35-37: Can the authors provide a specific example of which parameterizations are currently inaccurate?

L41: Replace “inorganic species” by “inorganic salts” and delete “sea salt”. Alternatively delete “trace metals”, as this would be part of the inorganic material.

L50: Change to: “… of PM2.5, particle phase state, described in terms of dynamic viscosity is particularly important for controlling particle reactivity. Viscosity describes…”

L51: Write as “to flow or deform”

L52: Write as “aerosols with dynamic viscosities”

L53: Add “are considered as amorphous semisolids”

L54: Add “are considered as amorphous solids” and change to: “Viscosity dictates how rapidly molecules…”

L56: Change to “liquid-like”

L59: The cited reference of Freedman 2017 discusses liquid-liquid phase separation and should be removed here.

L64: Add reference: (Baboomian et al., 2022)

L84: “using optical microscopy”. Please globally change “optical microscopy” to “light microscopy” throughout manuscript.

L100: Please add details how collected filters were stored.

L110: When the droplets are equilibrated at 100% RH, do you not condense a lot of water onto your glass slide, as you are essentially at bulk water saturation?

L114: Please add information on model and manufacturer of the RH sensor used.

L116: Please change to: “N2 and H2O-saturated vapor”

L119: Do not capitalize “flow”, consistent with use on e.g., L120.

L121: Please also add the reference to Renbaum-Wolf et al. (2013)

L122: Are your droplets really 20-40 µm in diameter? Based in your scale bar in Fig. 1 it appears that some of your droplets were considerably larger than 40 µm.

L123: “… in the flow-cell.” After this statement it would be good to provide some brief explanation of the poke-flow technique. You have some of this on L130-132, but I feel that it would be helpful to get some of this description earlier to better follow the description in your Sect. 2.3.

L128: “to identify the critical RH for particle cracking.” Does this mean that the particle is glassy? Please clarify.

L134: “in the absence of…” Do you mean that the flow time could not be experimentally measured? Please elaborate and explain.

L135: delete “high”

L136: Please indicate version number of COMSOL and the modules used for your analysis.

L139: Please add: “In the simulations, the dynamic viscosity...”

L140: Please provide details within what uncertainty the experimental and simulated flow times had to match along with a reasoning for it.

L141: Pleas add details to the text how this “lower-bound viscosity” was determined.

L182: Can the authors quantify what fraction of their phase separated samples adopted a core-shell morphology and what fraction had other morphologies?

L199: Add: “viscous amorphous semisolids”

Fig.1:

- consider deleting “likely” from labels of red arrows
- Caption and figure: Please consistently provide spacing between numeric value and percentage symbol or not, e.g., “90%” vs. “90 %”. Please check ACP house-style.

L216: Based on your Fig. 2, “RH < 45%” would be more appropriate, also on L19.

Fig. 2:

- Y-axis label: Please fix units to read “Pa·s”

Fig. 3:

- What is the difference between “after poking” and “post poking”? Would it not make sense to use one label for this, as all images were captured after retracting the needle from the particle.
- Why is there a “T” symbol in the “after poking” and a “τ” symbol in the “post poking” column. Also, for the former units of “s” are missing.

Fig. 4:

- See comments of Fig. 4.
- Fig. 5 appears blurry, please replace with a high-resolution version of that figure.

L267: Can you clarify what you mean with “phase-resolved chemical imaging”?

L279: How often have 1 vs. 2 vs. 3 phases been observed?

Comments to supporting information:

L11: Change to 2 tables.

L13: Add “measured by an β-ray continuous ambient”

L19: delete blank line

L29: Please provide information what a 1:1 methanol-water solution was used for filter extraction.

L39: Please provide a brief explanation of what the slip length is and how it affects your COMSOL simulations. The upper and lower limit of the slip length of 5 nm and 10 µm denote a wide range of possible values. What was the final slip length value chosen for your simulations, so that the simulated and modeled flow times matched. Was the slip length value the same for all simulations or is this a free parameter and why?

Could the range of possible slip length values be narrowed down? How? According to your statement on L56-59, the wide range of possible slip length values leads to the large uncertainty of the viscosity estimates.

L44-46: For the lower bound of viscosity the lower bounds of material density and contact angle were used. What was the slip length value and the contact angle value used to determine the lower bound viscosity values?

L46-48: For the upper bound of viscosity the upper bounds of material density and contact angle were used. What was the slip length value and the contact angle value used to determine the upper bound viscosity values?

L48: Add “highly concentrated aqueous solutions”

L53: Please provide details how the contact angle was determined.

L54-55: How good is the assumption of a perfect torus geometry and what is the uncertainty in determining the size of the hole from your microscopy images? For example, when I look at your Fig. 3a at 33% RH, the inner hole after poking has a highly aspherical shape. How is this taken into account in your COMSOL simulations? How does the shape of the hole of affect your COMSOL simulations and ultimately your viscosity estimates? Some of this information should be discussed in the main text.

L69: Remove “,” after than.

L70: Add “forming the OA matrix”

L75: Add “surrounding OA matrix”

L77: How is the radius of the matrix molecules determined?

Fig. S1:

- Turn blue font in caption to black font.

- Looking at the blue pentagon symbols at ~30 % RH, there is an enormous spread between the two data points, i.e., different droplets from the same filter (10/15). Can the authors provide some reasoning for this spread in flow time and hence also viscosity (Fig. 2)? Is this due to different composition of the individual droplets generated from the same filter extract? This should be discussed in the main text.

References:

Baboomian, V. J., Crescenzo, G. V., Huang, Y., Mahrt, F., Shiraiwa, M., Bertram, A. K., and Nizkorodov, S. A.: Sunlight can convert atmospheric aerosols into a glassy solid state and modify their environmental impacts, Proceedings of the National Academy of Sciences, 119, e2208121119, https://doi.org/10.1073/pnas.2208121119, 2022.