Overall:

The manuscript by Ullah et al. reports new measurements of aerosol dynamic viscosity. Viscosity was determined for particles composed of sucrose–water, a commonly used proxy for atmospheric organic aerosols, at discrete relative humidity (RH) and temperature (T) conditions using established techniques. Notably, the measurements of sucrose particle viscosity as a function of temperature are novel and represent a valuable contribution to the literature. The authors apply their experimental data within the Vogel–Fulcher–Tammann (VFT) framework and the Arrhenius mixing rule to parameterize sucrose viscosity as a function of RH and T.

This parameterization is further combined with observed tropospheric RH and T fields to estimate the viscosity (and phase state) of organic aerosol particles throughout the troposphere. Additionally, the authors use these viscosity distributions to assess N_2O_5 uptake on organic aerosols.

Overall, the manuscript addresses an important and timely topic that aligns well with the scope of the journal. However, several key assumptions are insufficiently discussed, and there are minor issues that require attention (see specific comments below). Therefore, I recommend major revisions before the manuscript can be considered for publication.

General comments:

C1: The authors use re-analysis data to calculate the average RH and T fields in the troposphere, and then use these RH and T-distributions to estimate the phase state. However, the source of these RH and T data and their representativeness for atmospheric conditions require further clarification in the main text, as this is somewhat hidden on L68-73 of the SI. It also remains unclear why a climatological dataset was not considered. A more detailed discussion of the rationale and potential implications of this choice would strengthen the manuscript.

C2: The authors provide novel data on the T-dependence of sucrose viscosity and use this data to derive a fragility parameter of 13. The way this fragility parameter is derived and how the determined fragility value compares to previous estimates of the fragility parameter is largely lacking and an in-depth discussion thereof should be added to the revised manuscript, as the estimated viscosity data is sensitive to the fragility value assumed fragility parameter. In addition, the authors claim their fragility parameter to be the “first experimentally constrained estimate of Df (L132). It could be valuable to provide a similar analysis as provided in their Fig. 3, but calculated for a fragility parameter of 10, as traditionally assumed in several studies, to clarify the role of this new, experimentally better constrained fragility parameter.

C3: The authors use sucrose as a proxy of atmospheric organic aerosols. However, the discussion as to what extent the viscosities measured for sucrose particles can be extrapolated to complex atmospheric organic aerosols is insufficient and should be extended.

C4: There are several assumptions made in estimating the effect of particle viscosity on the N2O5 uptake in Sect. 2.5 that require further explanation and/or justification (e.g., choice of numeric values for various parameters in eqs. 3 and 4). As the effect on N2O5 uptake is prominently advertised in the manuscript title, this Section will need to be much improved.

C5: I am unclear about the choice of the content of Sect. 4 on the atmospheric implications. Why are only the estimates on the N2O5 uptake included here and not also the content of the current Sect. 3.2? Combining these two Sections (3.2 and 4.1) into one “Atmospheric implications” section and providing some further discussion on possible caveats on extrapolating their measurements to atmospheric conditions and particles would be helpful.

Specific comments:

L31-33: There are plenty of more recent (review) articles for this statement and much work has been done since the publication of the cited works. Please consider adding some more recent references to this statement, such as, but not limited to, e.g. (Zhang et al., 2021).

L35: It could be meaningful to specify as “amorphous semi-solid” and “amorphous solid (or glassy)”, at least here when you first introduce the different phase states, to avoid confusion with crystalline solid phase states that OA can also exist in.

L36: Consider introducing the symbol (η) used to represent dynamic viscosity already here and then use it throughout.

L38-39: “Accurate determination of aerosol viscosity and phase state….” Here and elsewhere (e.g., L50) it is unclear to me if you use “viscosity” and “phase state” synonymously or not. Please clarify.

L45: Consider adding (Shiraiwa et al., 2017)

L50: Please add (Koop et al., 2011)

L54: Please add (Petters et al., 2019)

L57-63: You might find the following helpful: (Li et al., 2020; Li and Knopf, 2021)

L60: Please specify what you mean with “reactivity”

L66: I think the year for the Bateman study should be 2015, please check: https://pubs.acs.org/doi/10.1021/jp508521c

L82: Renbaum-Wolff et al. 2013a and Renbaum-Wolff et al. 2013b refer to the same paper, please correct.

L89: What do you mean with “thermodynamic conditioning”? Please specify. (Also on L103)

Fig. S1: Caption: Red line is not dotted, but solid, please fix. Please also add information on the goodness of fit to the figure, e.g., RMSE and R^2.

Fig. S2: The colored position labels appear very blurry and unreadable in most of the images. Please fix.

Fig. S3a/b:

- Caption: "… the uncertainty in RH from the RH sensor during calibration at each temperature…" I do not follow this. Why is the uncertainty in RH not the same across all data points? Is this uncertainty not given by the error/uncertainty in your RH-sensor? Please specify in the text how the RH uncertainty was determined more clearly by giving a reference to Section S1, where your deliquescence values are described.
- Why is there only a single data point for 273 K?

L110: Fig. S4: I cannot really see an evolution of the particle morphology in these panels. For each RH/T combination only 1 image before and 1 image after poking is shown. It could help to add one more column of images, where the particle is shown after a fixed time after poking (e.g. 10 s) for ALL RH/T combinations. Right now, each RH/T combination shows a different experimental flow time, which makes a comparison very hard.

Fig. S5: Caption. Add "." After Sellier et al. (2015).

L119: Specify: "to temperature changes"

L121: Here and in some other equations the formatting of "ln" is off, as it is italicized in some eqs. and not in others. Cf. dominator vs. denominator in eq. (2). Please fix.

L123: What is "infinite temperature"? Please also move the reference to Angell 1991 in front of the comma.

L125: Why have a "(293K)" in the nominator?

L129-133: It is unclear how you determined a fragility parameter from your Fig. S8. A detailed description of the steps how the experimental data was used to determine the fragility parameter should be added to the SI of this work. Looking over the work by Derieux et al. (2018) cited by the authors, it appears to me that they suggest a "lower limit fragility parameter of 10 ± 1.7". This does not seem too far off from the reported value of 13 ± 1. A more detailed description of what the fragility parameter means for the interpretation of your data and those from previous work should be added to better rationalize differences between the fragility parameter reported by you and others.

L134: Please replace "Simulation" with "Estimation of" here and elsewhere

L135: Here and elsewhere, please check placement of references and punctuation.

Sect. 2.5: The various assumptions made in this section should be explained in more detail. It is unclear to me how the chosen parameters fit to your experimental conditions. For example, the authors use a coefficient for the surface reaction of 2.5 x 10^{-4} bas on the "low RH conditions" by Grzinic et al. 2015. Are these low RH conditions representative for the entire RH-range probed by you? Similar, an explanation should be given, why α_b = 0.035 can be chosen.

L154: “The result shows…” This statement is misleading. You might observe viscosity changes over 9 orders of magnitude when comparing across the RH range from ~10-90%, but for any given RH value, the range of viscosities observed across the different temperatures tested is much less than 9 orders or magnitude. Please reformulate more carefully.

Fig. 1: Caption: It is unclear to me, how the colored, dashed fit-lines were established. Are these fits constrained to the viscosity of water at 100 % RH? Is this a fit to just the data points of this study or are literature values taken into account where available, i.e., at 293 K?

L164: “…, indicating a semi-solid state.” I think the reasoning or cause-effect relationship is off here. The fact that the <u>reduction</u> in viscosity at 50% RH was lager when going from 273 K 303 K as compared to at 80% RH does not allow you to make statements about the phase state. I.e., it is not the change in viscosity but the absolute viscosity values that allow you to make statement about the aerosol phase state. Please improve the description. Maybe I am misinterpreting what you want to say here?

L165: Please see my comment above. “Flow time contrast” for what? To fully recover to the initial particle morphology after poking? Please clarify.

L170: Should the viscosity threshold for a “glassy” phase state not be > 10^{12} Pa s?

L185: “Figure 2…” This sentence is incomplete, please fix.

L201: “Viscosity isopleths…” This sentence is incomplete, please fix.

L183: It could be helpful to add a reference to your Section S2 and/or your eq. (S2.1) here, to clarify how the RH and T-dependent viscosity values were derived.

L188: “…the contours are nearly vertical”. While true, it could be more appropriate to talk about a slope here, as you do further down. To be more quantitative, you could consider discussing the slope of the black, dashed line (semi-solid/liquid transition) across the 3 temperature ranges that you discuss between L187-193.

L207: Add “… we applied the viscosity parameterization (eq. S2.1)…”

L207-208: Do you refer to the RH and T fields shown in Fig. S9? If yes, a reference would be appropriate. The authors should also add further information about where this RH and T-information was retrieved from. It is stated that this is re-analysis data. Re-analysis of what data? The authors should also clarify why only a period of 2020-2024 was used to determine the average RH and T fields and why they consider this representative?

L209-201: “… derived from monthly mean ambient RH and T …” Could this not be simplified here in the main text by saying that these are average RH and T profiles over the time period 2020 to 2024? It also seems inconsistent to write about monthly averages here and annual averages on L240.

L211: Add “fractional Stokes-Einstein…”

L218: Please quantify “extremely high RH”.

L219: Viscosity is not “suppressed”, rather does the presence of water vapor and water in the particles prevent the formation of highly viscous phase states. Please improve formulation.

L225-226: “… together these factors tend …” I am not sure I follow this sentence. What are multi-component SOA fields?

L230: Can you give some examples of these chemical transport models and provide some references for the assumption of fast mixing?

L233: Add “as a proxy for (secondary?) organic aerosol. However, real…”

L236: I would probably delete “under humid conditions”. As most atmospheric inorganic substances undergo deliquescence and efflorescence, it will dependent on the history of the particle, whether the inorganic components of an internally mixed organic-inorganic aerosol are dissolved and hence can promote water uptake or not.

Sect. 4.1: General: It is not entirely clear to me how you use the RH- and T-dependent viscosity data to calculate the N2O5 uptake. Going back to L148-149 it seems like you use the viscosities along with the fractional-Stokes Einstein relationship to calculate N2O5 diffusion coefficients and then use these calculated diffusion coefficients in eqs. (4) and (3), as described in SI L53-5. It would be helpful to make this clearer in the main text, e.g., on L148-149 and maybe also re-iterate at the beginning of Sect. 4.1.

L251: “… could extend over several shorts of the year…” It is unclear what you mean here, please rephrase.

L252: Please quantify “cold conditions”

L257: “uptake rates of” delete “of”

L260: “As shown in Fig. 3c” This sentence seems incomplete and is hard to understand, please reformulate.

L263: For better comparison of your current Fig. 3c and Fig. S11, it would be helpful to have them next to each other. Consider splitting Fig. S11 into two panels, where one panel reproduces your Fig. 3c and the other panel displays the results of your sensitivity analysis.

L266: I would not call this a climatology, which usually would contain data over several decades. Rather use the wording of “average RH- and T-fields” or similar.

L266: I would encourage the authors to tune down the wording here a little bit. There have been previous attempts to explore the phase state of organic aerosol throughout the troposphere, as evident by the studies cited in this work. Related, with this statement the authors seem to claim that the sucrose viscosity is representative for the viscosity of complex atmospheric aerosol, directly contradicting their statement on L236-237.

Supporting Information:

L32: Delete “.” In front of “(Weight, 2019)”

L36: Move “(Price et al. 2016)” in front of the “.”

L58: Add "." After Kiland et al. (2023).

L66: eq. (S2.3) what values of k was used here? It would be helpful to replace "k" by the Greek symbol "kappa" (κ) here and elsewhere, as traditionally used to describe aerosol hygroscopicity.

L75: "Petters" is misspelled.

L79: Numbering of eq. is off and should read "(S3.1)"

Fig. S7: It is unclear to me how you use eq. (S2.1) to determine a hygroscopicity value from this data. Please specify in the text.

Fig. S9: Please increase the size of this figure over the full page to allow the reader to read out values from the figure. In its current form it is extremely hard to read the figure. The authors could also consider removing the panels of the different seasons, if a season-dependent viscosity is not discussed.

Fig. S10: Please increase the size of this figure over the full page to allow the reader to read out values from the figure. In its current form it is extremely hard to read the figure.

L268: Why do the authors only mention the need of T-dependent viscosity parameterizations? What about the role of RH for organic aerosol viscosity, which according to your Fig. 2 is the key parameter in controlling the aerosol phase state in the troposphere.

References:

Koop, T., Bookhold, J., Shiraiwa, M., and Pöschl, U.: Glass transition and phase state of organic compounds: dependency on molecular properties and implications for secondary organic aerosols in the atmosphere, Physical Chemistry Chemical Physics, 13, 19238–19255, https://doi.org/10.1039/C1CP22617G, 2011.

Li, J. and Knopf, D. A.: Representation of Multiphase OH Oxidation of Amorphous Organic Aerosol for Tropospheric Conditions, Environ. Sci. Technol., https://doi.org/10.1021/acs.est.0c07668, 2021.

Li, J., Forrester, S. M., and Knopf, D. A.: Heterogeneous oxidation of amorphous organic aerosol surrogates by O_3, NO_3, and OH at typical tropospheric temperatures, Atmospheric Chemistry and Physics, 20, 6055–6080, https://doi.org/10.5194/acp-20-6055-2020, 2020.

Petters, S. S., Kreidenweis, S. M., Grieshop, A. P., Ziemann, P. J., and Petters, M. D.: Temperature- and Humidity-Dependent Phase States of Secondary Organic Aerosols, 46, 1005–1013, https://doi.org/10.1029/2018gl080563, 2019.

Shiraiwa, M., Li, Y., Tsimpidi, A. P., Karydis, V. A., Berkemeier, T., Pandis, S. N., Lelieveld, J., Koop, T., and Pöschl, U.: Global distribution of particle phase state in atmospheric secondary organic aerosols, Nature Communications, 8, 15002, https://doi.org/10.1038/ncomms15002, 2017.

Zhang, Y., Liu, P., Han, Y., Li, Y., Chen, Q., Kuwata, M., and Martin, S. T.: Aerosols in Atmospheric Chemistry, American Chemical Society, https://doi.org/10.1021/acsinfocus.7e5020, 2021.