

We thank reviewer #1 for providing a positive review and the suggestions to improve the manuscript. We answer the reviewers' questions and comments below (our reply in blue, reviewer's comments in black).

Liviana et al. used three experimental measurement techniques and various theoretical calculation methods to quantify the viscosity of the ammonium nitrate-sucrose-water ternary aerosol systems and estimate the characteristic internal mixing times of such systems. Overall, the study data is comprehensive, the research methods are abundant, the discussions are detailed, and the scientific reliability is high. However, some discussion can be added:

1. Lines 249-253: The discussion in the manuscript attributed the differences in viscosity measurement to the volatilization of NH_4NO_3 . While this is a possible reason, the volatilization of ammonium nitrate at room temperature causing changes in the OIR is unlikely to generate such a significant viscosity error (the OIR may increase from 1:1 to close to 2:1, but is unlikely to increase to 4:1 as in the experimental conditions in Figure 4). It is suggested to add a discussion on systematic errors between optical tweezers and the measurement methods in the manuscript, as well as the influences of suspension droplets and bulk phase solutions on viscosity measurements in this part of the discussion.

As we point out in the manuscript (line 251), the most reliable and best-established viscosity measurements are those of bulk viscometry (which are limited towards large viscosity). As shown in Fig. 4 and pointed out in the text these clearly deviate from the tweezer's data of Tong et al. (2022). Therefore, we conclude that the data of Tong et al. (2022) most likely are erroneous. As we are no experts to judge possible systematic errors of optical tweezer viscosity measurements, we refrain from discussing possible errors of the experiment of Tong et al. (2022) in detail. We agree with the reviewer that a significant volatilization is needed to explain the data of Tong et al. and such a volatilization should have been noticed by Tong et al. as they are tracking radius of the particles in their experiments. Alternatively, the actual OIR was not the reported 1:1 ratio. We therefore will delete the last sentence of the paragraph (lines 252- 253) and just end the paragraph with the statement that the Tong et al. data are questionable.

2. Line 365-371: Based on the data presented in the manuscript, the method of estimating the viscosity of mixed particles using a mole-fraction-based mixing rule is indeed more reliable. Furthermore, it is expected that the authors will add a discussion on the following topics in the conclusion section: The current measurement and estimation methods for aerosol viscosity are actually showing quite large uncertainty (especially the poke-flow method), as shown in Fig. 3 and Fig. 5, where the differences under the same water activity conditions can reach two orders of magnitude. So, does the authors have any recommended measurement and prediction methods in the conclusion section? Or discuss the tolerance levels for quantifying aerosol viscosity?

We thank the reviewer for this comment. We tried to discuss these points in the paragraph starting on line 373 in the conclusion section. We will rephrase this paragraph following the reviewers' suggestions as follows; it will read:

“The available experimental techniques are limited in their applicable viscosity range, particularly at viscosities above 10^8 Pa s and carry uncertainties of orders of magnitude in this range (see Fig. 4). Hence, aqueous phase viscosities of some organic compounds or related mixtures at low RH cannot be measured precisely. Our results here suggest that a strategy to obtain the much-needed data to further validate predictive models such as AIOMFAC-VISC at low water activities is to mix organics with monovalent inorganic salts. The viscosities of these mixed systems at low RH will carry sufficiently low uncertainties with the techniques presently available (cp. Fig 4) to constrain mixing rules. Such measurement data may further the development of predictive models for viscosity of complex, atmospherically relevant aerosol.”

3. Line 14-17: The authors emphasized that throughout the mid-latitude troposphere, the viscosity of inorganic-organic mixed aerosols is relatively low, and the kinetic limitations of gas-particle partitioning can be ignored. However, the work of this study seems insufficient to support this point of view: First, as the authors have mentioned, if the particle is in the form of an organic coating, the timescale of gas-particle partitioning on a high-viscosity coating is likely to be considered, and such organic-coating particles may be ubiquitous in the troposphere; Secondly, the quantification work of this study is mainly based on nitrate particles, and it can be known from previous viscosity measurement data that nitrate could significantly reduce the viscosity of the mixing system, while other major inorganic salts of aerosols, such as sulfates, may not have such large reducing effect on viscosity (it is recommended that the authors compare the viscosity of nitrate and sulfate aqueous solutions at different water activities). In a word, the results of this study may not represent the actual atmospheric aerosols of various inorganic salts and organic compounds mixtures. It is suggested to soften the statement in the abstract section, which may be too categorical.

Thank you for these comments. Regarding the effect of organic coatings: the statements in the abstract refer to internally mixed single-phase particles (containing aqueous inorganic salts) only. We refrain from making a broader statement about the case of phase-separated particles with a viscous organic coating, which as the referee suggests may show comparably slower equilibration with the gas phase.

Regarding the difference between sulfate and nitrate: consider Fig. 6 of the paper and compare panels (E and F) for ammonium sulfate mixed with sucrose for OIR= 1 and OIR=4 respectively with the corresponding ones for ammonium nitrate at for example $a_w=0.4$, the sulfate systems show a viscosity of about one order of magnitude larger compared to the nitrate ones. While this is a significant increase, mixing times will increase by the same factor

which is not sufficient to push them to a range in which the mixing times approach several minutes.

Nevertheless, we will take the advice of the reviewer and soften the last sentence in the abstract section as follows:

“Further data are needed to see whether this assumption may even hold for the entire troposphere at mid-latitudes and $RH > 35\%$.”