

[A0] For clarity and visual distinction, the referee comments or questions are listed here in black and are preceded by bracketed, italicized numbers (e.g. **[1]**). Authors' responses are in red below each referee statement with matching numbers (e.g. **[A1]**). Page and line numbers refer to online AMTD version.

Reviewer 1

The experimental method developed is elegant and well backed up by simulation, albeit not entirely unique due to their previous demonstration of the method in their Renbaum-Wolff, 2013 paper (1). The explanation of the technique is clear, hence it is an appropriate technical paper and I believe should be published. The errors on the measurements are large, but future work has been suggested that hopes to constrain the upper and lower viscosity limits.

We thank the referee for their helpful comments.

[1] The methods and results sections are concise. It seems that the required detail needed for reproducing the experiments is stated. I do not think any more figures are needed to make the technique clearer. However could a couple more measurements be made of the polybutene standards at different temperatures to see if the trend continues to match the company's measurements as shown in figure 6? The errors in the measurements at ~295 °C plotted in figure 6 mean that the viscosity limits overlaps with the values stated by the company at the higher temperature of ~335 °C. It would be good to see the changing viscosity with temperature measured with this technique as figure 6 looks a bit incomplete.

[A1] This is an excellent suggestion and something that would really add to the paper not only in terms of measuring standard compounds, but also for much needed data on the temperature dependence of the viscosity of atmospheric material. Unfortunately, however, the experimental setup employed does not allow for temperature control.

[2] The limitations of the technique are clearly outlined, such as the problems with lower and higher RH measurements for sucrose. I believe these limitations prevent this technique being widely applicable to atmospheric applications. For example due to the limited viscosity range that can be measured discussed on page 888. Can you expand on how you 'expect this approach to also be valid at higher viscosities'? How convinced are you that the viscosity range that can be measured is atmospherically relevant?

[A2] To illustrate that this approach is also valid at higher viscosities, we have carried out additional measurements using sucrose-water particles of higher viscosity. To carry out these experiments, we used needles coated with a hydrophobic material to prevent particles sticking to the needles, which occurred with uncoated needles at higher viscosities.

These additional measurements illustrate that this approach can measure viscosities ranging from $\sim 5 \times 10^2$ to $\sim 3 \times 10^6$ Pa s. This covers an important part of the range of viscosities of secondary organic material generated in environmental chambers. For example, the viscosity of the water-soluble component of SOM from the ozonolysis of α -pinene ranges from 4×10^2 to 5×10^5 Pa s between RHs of 50-70 % (1) and the viscosity of SOM derived from isoprene ranges from 2×10^2 to 5×10^6 Pa s at RH's $\leq 30\%$ (2). In addition, this technique has several advantages including a low cost and a compatibility with cascade impactors for particle collection.

In the revised manuscript Figure 4 will be updated and the discussion above on viscosities of SOM generated in environmental chambers and advantages of the technique will be included. (from P.888, L26).

References

- (1) Renbaum-Wolff, L, Grayson, J.W., Bateman, A. P., Kuwata, M., Sellier, M., Murray, B. J., Martin, S. T., Bertram, A. K.: Viscosity of α -pinene secondary organic material and implications for particle growth and reactivity, Proc. Nat. Acad. Sci. USA, 110, 8014-8019, doi:10.1073/pnas.1219548110, 2013
- (2) Song, M., Liu, P. F., Hanna, S. J., Martin, S. T., Bertram, A. K., Relative humidity-dependent viscosities of isoprene-derived secondary organic material and atmospheric implications for isoprene-dominant forests, Atmos. Chem. Phys. Disc., 15, 1131-1169, doi:10.5194/acpd-15-1131-2015, 2015

Reviewer 2

General comments:

This paper describes a method for determining the viscosity of small amounts of materials aimed at studying the viscosity of secondary organic material (SOM) derived from the oxidation of volatile organic compounds. The properties of SOM in general and the viscosity in particular have become the topic of intense research recently. For example, viscosity influences such important processes such as gas-to-particle partitioning, heterogeneous chemistry and water uptake and cloud condensation nuclei or ice nuclei activation. The technique and, thus, the manuscript is in principle suitable for publication in Atmos. Meas. Tech., because of the current research in the area of atmospheric aerosols described above, although I could also see the manuscript in a general physical chemistry or analytical chemistry journal.

Overall, the paper text and length, the figures and tables are all appropriate. However, given that a preliminary validation of the technique has been described already in a previous publication, the additional scientific information and the calibration procedure described in the current manuscript is somewhat limited. For

example, the data in Fig.4 show that the error bars with the new technique are larger and the range of measured viscosity is smaller than previous techniques. I think the authors should present more data at higher viscosity for a better validation and a more convincing statement of the usefulness of the technique.

We thank the referee for their helpful comments.

Major scientific comments:

[1] (1) The viscosity range measured in this paper to show the suitability of the technique is 10^3 - 10^5 Pa s, which I think is pretty limited, and the subsequent sentence stating 'we expect this approach to also be valid at higher viscosities' seems a bit too optimistic and not convincing to me. (P.881, L22-23 and P.888, L26-27)

[A1] To address the referee's comment we have performed additional measurements at higher viscosities using needles coated with a hydrophobic material (thank you in advance for comment [3], below). With these additional measurements, we show that the technique can measure viscosities of compounds of literature viscosities ranging from $\sim 5 \times 10^2$ - $\sim 3 \times 10^6$ Pa s. Figure 4 will be updated to incorporate these new experiments. In addition, to address the referee's comments we will remove from the manuscript the comment "we expect this approach to also be valid at higher viscosities" (P.881, L22-23 & P.888, L26-27).

As discussed above, the viscosity range this approach is able to measure, 5×10^2 to 3×10^6 , covers an important part of the range of viscosities of secondary organic material generated in environmental chambers. In addition, this technique has several advantages including a low cost and a compatibility with cascade impactors for particle collection.

[2] The investigated viscosity range of two orders of magnitude seems even smaller when the uncertainty in the individual measurements of the same magnitude is considered. It did not become clear to me, what limiting factors are responsible for this small viscosity range. At some point, it is stated that it is due to the lack of reference materials in the high viscosity range (p.881). It is not the uncertainty of the required parameters as given in tables 1 and 2, they are responsible for the error bars shown in Fig.4b, correct?

[A2] The uncertainty in the required parameters is indeed the cause of the large error bars in Fig. 4b. The lack of the availability of reference materials doesn't constrain the measurements, rather restricts the number of compounds that can be used to validate the poke-flow technique combined with simulations of fluid flow. The phrase "due to uncertainty in the value of the physical properties used during simulations" has been added to the end of the sentence at P.886, L.24 for clarity.

[3] The lower limit is due to the time resolution of the digital camera, but I guess this is not so important as other techniques also work in this area, e.g. the moving-bead technique developed by the same group. However, at the upper limit it appears to be the sticking of the viscous material to the needle, at least for the sucrose-water particles. I am not convinced that this will be much different for water-soluble SOM. What are the arguments for the expectation of the authors that it should not occur? If this were really the latter case, maybe a different needle material would be helpful. For example, a needle with a hydrophobic coating may be better suited for studies with hydrophilic materials such as sucrose-water; and an amphiphobic coating, i.e. a coating that is simultaneously hydrophobic and oleophobic, may be suitable for both hydrophilic mixtures and oils.

[A3] As outlined in comment [A1], we had needles coated by Silcotek Corporation (Belfonte, USA) with a hydrophobic coating named Dursan ©. These coated needles were able to poke particles at a lower RH without resulting in the particles sticking to the needle surface and being removed from the substrate. As such, particles of a higher viscosity have been studied and the data added to the paper. The sentence from P.882, L 25 onwards has been altered to reflect the use of the two different types of needles during studies.

[4] (2) P.879, L.27: remove comma after 'milligram scale'

(3) P.880, L.15-17: this sentence is somewhat confusing. Please rewrite, so that the seemingly contradicting threshold of 10^8 Pa s is removed.

(4) P.882, L.25: remove comma after 'sharp'

[A4] The text has been amended to address (2), (3), and (4).

[5] (5) P.884, L.7-13: I did not understand how particles with a diameter of 40-70 micrometer can be produced using a pipette. Please provide more details so that others can reproduce the procedure.

[A5] We agree that this is a useful piece of information to report and apologise for the original oversight. The heated material was picked up using the tip of the pipette, and the pipette 'flicked' in the direction of the substrate from a close distance. The material that was deposited on the slide surface ranged in size, with particles of diameter 40-70 μ m being used in experiments. A sentence has been added to Section 2.1 detailing this procedure P.884, L.10).

[6] (6) P.885, L.22-26: Do I understand it correctly that the jagged and wavy geometry only appeared in the simulations, but NOT in the actual experiments? If yes, please state this more explicitly. Could this have to do with concentration gradients within the particles?

[A6] The jagged and wavy geometry does appear only in simulations. The result is due to stretching of the mesh elements at the moving front of the particles. Even when a finer mesh

is used the problem remains. Two sentences, from P.885, L.22 onwards, have been altered/added to clarify this.

[7] (7) P.889, L.4-7: Can you do sensitivity studies with different support materials with different surface properties? These materials may produce different contact angles, which may affect their slip length, so that maybe the corresponding uncertainty can be reduced (just an idea).

[A7] This is an interesting idea, but we are not exactly sure how this would help reduce the uncertainties. We are happy to hear additional thoughts on the topic though!