

## ***Interactive comment on “Observation of viscosity transition in $\alpha$ -pinene secondary organic aerosol” by E. Järvinen et al.***

**Anonymous Referee #2**

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Review of “observation of viscosity transition in alpha-pinene secondary organic aerosol” by Jarvinen et al.

Jarvinen et al. introduce a new method of probing the viscosity of SOA as a function of RH and temperature, and they apply this method to determine information related to the viscosity of SOA generated from the oxidation of alpha-pinene. The experiments are novel and extremely important, as they provide laboratory data related to the viscosity of SOA at cold temperatures. Currently this type of information is very limited, yet this type of information is needed to understanding ice nucleation by SOA in the atmosphere. Although the experiments are excellent and the information provided is crucial, I do have comments on the data interpretation that should be addressed before publication. Once these issues are addressed, I highly support publication in ACP.

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Comments on interpretation of experimental results:

- 1) Page 28596, line 20. Consider replacing “for the phase transition” with “for the aspherical to spherical transition” since the authors have not proven that they have measured a phase transition. For example they haven’t proven that they are measuring the relative humidity for the glass to semisolid phase transition (which occurs around  $10^{12}$  Pa s) or the semisolid to liquid transition (which occurs at 100 Pa s).
- 2) Conclusions, line 4. Would “aspherical to spherical transition” be more appropriate than “viscosity transition”? Viscosity is expected to change (i.e. transition) with RH in a continuous manner for most RH values. For example at warm temperatures and high RH values, as the RH is increased the viscosity is expected to change (i.e. transition) yet the experiments described here are not sensitive to these changes.
- 3) Page 28587 line 1. Consider removing “during a phase transition” since the authors are not necessarily probing a phase transition. Rather they could be just sensitive to when the viscosity falls below a certain value.
- 4) Abstract. The authors claim that they are measuring the transition from the amorphous viscous state to a liquid state. However, the experimental observations (non-spherical to spherical transition) may occur at viscosities much higher than a liquid state, which has been defined in the atmospheric literature as a material with a viscosities less than 100 Pa s.
- 5) Page 28595, line 9, the authors refer to the full deliquescence relative humidity. Please define what they mean by full deliquescence relative humidity. Also, how do they know that the so-called viscosity transition occurs near the full deliquescence relative humidity?
- 6) Page 28595, line 11-13. Here the authors are discussing the timescale for water diffusion within the particle bulk. The authors should estimate what this time scale is based on recent published diffusion coefficients for water in SOA and include these

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estimated time scales in the manuscript. Based on these values is water diffusion the rate limiting step in their experiments? See [Price et al., Chemical Science, 2015; Lienhard et al., ACP, 2015].

7) In previous publications the time scale for aspherical particles to transition to spherical particles has been used to estimate viscosities [Zhang, Sanchez et al. ACPD, 2015; Pajunoja et al. Aerosol Sci. Tech. 2014]. Have the authors considered estimating viscosities from their observables? I think the current paper would be clearer if the authors estimated viscosities from their results.

Experimental issues:

1) Page 28586. Line 16-17. The authors state that two thermocouples near the wall were excluded due to wall temperature effects. I assume this means that the temperature close to the wall was colder? The experiments involved adding a flow of humidified gas and mixing the flow in the chamber with a fan. Is it possible that the temperature and relative humidity was different close to the walls, and the aspherical to spherical transition occurred close to the walls, not in the middle of the chamber? If this was the case, the temperature and RH reported may be different than the temperature and RH at which the aspherical to spherical transition occurred?

Comparison with Koop et al. 2011:

1) Page 28595, Line 20-25: I do not understand the argument that the measured “viscosity transition” can be compared to the relative humidity of the glass transition (RH<sub>g</sub>) in atmospheric context. The glass transition occurs at a viscosity of roughly  $10^{12}$  Pa s. To argue that the “viscosity transition is comparable to the glass transition, I think one would need to argue that both transitions occur at roughly the same viscosity? Can the authors estimate that viscosity at which their aspherical to spherical phase transition occurred?

2) Page 28595, line 27-29. “Our results suggest the relationship between the transition

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RH and the temperature is more linear than predicted by Koop et al. 2011. This may be especially relevant at high RH, where we can expect that the transition might take place at a much higher temperature than predicted by Koop”. I suggest the authors should modify this text since all the experimental results shown here are within the estimates presented by Koop et al. 2011 (green shaded region in Figure 11).

Minor points:

2) Page 28581. Line 4. Should “isotropic” be deleted?

3) Page 28587, line 15. “the SOA particles were generated and grown in the chamber by continuous oxidation of alpha-pinene with ozone”. Should this be “. . . by continuous oxidation of alpha-pinene with ozone and OH radicals”?

4) Figure 5. In the figure caption it is stated that the linear depolarisation ratio stayed below the detection threshold during the entire experiment. However, at times between 6:00 and 7:00 it looks like the linear depolarisation reaches as high as 0.1. This seems to be well above the LOD. Please discuss.

5) Was figure 8 discussed in the main text? If not, should it be removed from the manuscript?

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 28575, 2015.

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