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Comment

Interactive comment on “Observation of viscosity transition in α -pinene secondary organic aerosol” by E. Järvinen et al.

Anonymous Referee #1

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This manuscript reports the viscosity transition of α -pinene SOA generated during the CLOUD experiments at CERN at different temperatures using an optical method with a depolarization instrument. The method based on the different depolarizing properties of spherical and non-spherical shape of particles. The manuscript provides information of transition, which was defined as viscosity transition, from non-spherical to spherical shape when increasing RH (particles taking up water vapor). This study demonstrated an optical method that could be applied for the chamber studies on the phase transition of particles. The experimental methods are valid and the scientific approach and discussion are sound. The paper is well written and organized. I recommend it for publication with minor revision. Please see the following comments that the authors may want to consider in the revision.

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General comment:

1, Previous studies as mentioned by the authors often have a definition based on physical properties, such as viscosity, for different phases, for example, liquid, viscosity < 100 Pa s. The definition of viscosity transition here in this manuscript is not very clear, the transition from non-spherical shape to spherical means transition from solid/semi-solid to liquid or solid to semi-solid? Spherical shape does not mean the particle is in liquid state. It is suggested to provide the clear definition of the viscosity-transition. The definition of viscosity-transition-RH is placed in the later part of the manuscript, P28594 line 16. The way it is defined is not really quantitatively; can “decrease significantly” and “reached a constant level” be quantitatively defined? I suggested make these definitions at the early part of the paper and indicate that these are instrumental defined, unless physical means/values are provided associated with this viscosity-transition for definition.

2, P28595, Line 8-23, as mentioned above the definition of viscosity transition is not clear, RHg indicates the transition from solid to semisolid, caution is needed when compare the viscosity-transition RH with RHg as authors also mentioned that the viscosity-transition RH is different from RHg. If I understand it correctly, most likely the viscosity-transition RH will be higher than RHg (how much higher depending on the definition of viscosity-transition RH). Line 18-19, it is lacking of logic here for reasoning the comparison with RHg, the transition timescale and atmospheric processes are the dynamic factors, they may affect the viscosity-transition RH measured in the chamber, but not the reasons for the comparison. Why the viscosity-transition RH is more relevant for atmospheric processes? P 28597, line 11-12, why it is directly relevant for GLASSY Transition of SOA, isn't this conflict with the statement in P28595, line 8-11.

3, It is recommended to calculate or estimation the viscosity based on the transition time, particle size changes with additional assumptions. Based on these data it may be possible to estimate the viscosity of the particles or at least the changes of viscosity during the transition. There are a few more studies that maybe useful. Kidd et al (2014)

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provided viscosity information for α -pinene SOA generated at different RH, Wang et al (2015) estimated the viscosity of α -pinene SOA as a function of temperature and RH, Song et al (2015) summarized a nice set of the viscosity data for α -pinene SOA from previous studies for comparison.

Technical comments:

1, P28577, line 6, please provide full description of “CLOUD” and “CERN”

2, P28578, line 25-27, please provide references for this statement

3, P28582, line 10-12, please justify the assumption or provide references

4, P28585-28586, provide the full description of SMPS, CPC, ApicT etc.

5, P28586, What is the pressure in chamber during the experiments?

6, P28593, line 19-23, what do you mean “particle hardness”?

7, P28595, line 1, as mentioned above, the viscosity-transition RH is not quantitatively defined, is it possible that the observed wider transition RH is due to the arbitrarily defined gray area as shown in Figure 4? If consider the uncertainty of RH as described in P28586 (7-13%), there is no significantly wider RH ranges as shown in Table 1. There may be one only considerable larger range is for -38 degree C (~10%) as compared to 5-7% for higher temp.

8, P28595, line 3, in Table 1, the viscosity-transition RH values are shown in ranges. it is not convincing that viscosity-transition RH has linear dependency of temp. Please provide fitting data or supporting information for this statement.

9, P28596, line 5-7, SOA from difference precursors may behave differently. It is suggested to rephrase this statement and limited to α -pinene SOA as the manuscript only showed α -pinene SOA data.

10, P28597, line 22, “viscosity transition temperature RH at temperatures..” typo?

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11, Fig.4, unit of particle concentration is missing.

12, Fig.8 Caption, two “is” in last sentence.

Kidd, C., Perraud, V., Wingen, L. M., and Finlayson-Pitts, B. J.: Integrating phase and composition of secondary organic aerosol from the ozonolysis of α -pinene, *P. Natl. Acad. Sci. USA*, 111, 7552–7557, doi:10.1073/pnas.1322558111, 2014.

Wang, B. B.; O'Brien, R. E.; Kelly, S. T.; Shilling, J. E.; Moffet, R. C.; Gilles, M. K.; Laskin, A. Reactivity of liquid and semisolid secondary organic carbon with chloride and nitrate in atmospheric aerosols. *J. Phys. Chem. A* 2015, 119, 4498–4508.

Song, M., Liu, P. F., Hanna, S. J., Li, Y. J., Martin, S. T., and Bertram, A. K.: Relative humidity-dependent viscosities of isoprene-derived secondary organic material and atmospheric implications for isoprene-dominant forests, *Atmos. Chem. Phys.*, 15, 5145-5159, doi:10.5194/acp-15-5145-2015, 2015.

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