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Interactive comment on “Changing shapes and implied viscosities of suspended submicron particles” by Y. Zhang et al.

Anonymous Referee #1

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The manuscript “Changing shapes and implied viscosities of suspended submicron particles” by Y. Zhang et al., [2015] introduces a new approach for deriving in-situ the viscosity of suspended aerosol particles using α -pinene-derived secondary organic material (SOM) as model aerosol. The SOM viscosity is determined as a function of relative humidity (RH). Due to the inherently significant role viscosity plays in particle morphology, mixing state, and heterogeneous processes, and the potential applicability of the new approach to field particles, the article addresses a timely and important topic that would make a nice addition to the current scientific literature. The key difference compared to previous studies on this topic is the application of suspended aerosol particles instead of particles adsorbed onto substrates, which may avoid potential caveats. The paper (including supplement) is thorough and well-written. I have

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only a couple of comments regarding the approach that I hope the authors can address before publication in ACP.

Specific comments

1. The authors should elaborate more why they chose to dry the particles following RH-conditioning in their flow setup and discuss potential caveats since the parameters used for deriving viscosity are based on DMA and APM measurements of the particle's properties at a relative humidity of $\leq 5\%$. How would the dynamic shape factors and thus particle viscosity differ if the DMA and APM were maintained at the same RH as in the RH-conditioning region of the setup? I am mainly concerned that this extra drying step removes sufficient liquid water from the particles, suggesting that the derived viscosities for the high RH experiments may be overestimated compared to reality. There is only one sentence on the reasoning for not including the drying step, which is in regards to hygroscopic growth factors (Page 6830, lines 6-8), but very little discussion of the water partitioning behavior over the different RH regimes in the setup that I think would be crucial here. Please provide clarification or more discussion of this potential caveat.

2. Viscosity as a function of RH is determined based on a fit of χ as a function of RH, such that when $\chi=1.00$ (defining a spherical particle), viscosity as a function of RH breaks down, which places a systematic limitation on the applicable RH range for a given particle system. Could the author's comment a bit more the inherent limitations of the approach? Since this paper introduces a new technique for determining the viscosity of suspended aerosol particles, the authors should at least comment on its applicability to other particle systems as a function of RH, ones that possess a different viscosity compared to α -pinene SOM.

Technical comments

1. Page 6823 lines 13-14: It is implied in the abstract that the particles transition from 'solid' to 'semisolid' over the RH range, which is a bit exaggerated. The particles

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become less-viscous over the applied RH range, but largely remain in a semi-solid phase state, e.g. [Koop et al., 2011].

2. Page 6824 lines 16-17: Regarding the discussion of particle phase and reactivity, references should be updated to include recent publications by Slade and Knopf [2014] and Arangio et al. [2015] regarding the impacts of viscosity, bulk diffusion, and OH multiphase kinetics.

3. Page 6827 lines 3 and 6: DMA and APM should be defined here.

4. On page 6836, the last paragraph (lines 17-28) is a repeat of the end of the previous paragraph.

5. Page 6848 (Fig. 4): It's confusing that in the figure description for panel (b), particles are exposed to an RH up to 80%, yet in the actual figure the data points extend up to RH~100%. Furthermore, the viscosities reported in Fig. 6 only go up to RH=58%. Please clarify in the manuscript the reasons for these differences.

References

Arangio, A. M., J. H. Slade, T. Berkemeier, U. Poschl, D. A. Knopf, and M. Shiraiwa (2015), Multiphase chemical kinetics of OH radical uptake by molecular organic markers of biomass burning aerosols: humidity and temperature dependence, surface reaction, and bulk diffusion, *J. Phys. Chem. A*, doi:10.1021/jp510489z.

Koop, T., J. Bookhold, M. Shiraiwa, and U. Poschl (2011), Glass transition and phase state of organic compounds: dependency on molecular properties and implications for secondary organic aerosols in the atmosphere, *Phys Chem Chem Phys*, 13(43), 19238-19255, doi:Doi 10.1039/C1cp22617g.

Slade, J. H., and D. A. Knopf (2014), Multiphase OH oxidation kinetics of organic aerosol: The role of particle phase state and relative humidity, *Geophys. Res. Lett.*, 41(14), 5297-5306, doi:10.1002/2014GL060582.

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