

Interactive comment on “Relative humidity-dependent viscosity of secondary organic material from toluene photo-oxidation and possible implications for organic particulate matter over megacities” by M. Song et al.

Anonymous Referee #2

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Review of Song et al., ACPD, 2016

The authors have measured relative humidity-dependent viscosity of toluene SOA after a series of publications on viscosity measurements of α -pinene and isoprene SOA. This group is currently one of the only groups that can achieve such challenging measurements by combining two different unique experimental methods of a bead-mobility technique and a poke-flow technique. Based on viscosity measurements and RH observations in major urban cities in the world, they estimated whether anthropogenic SOA particles in these cities would be well-mixed or not. The experiments seem to

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be conducted well and the manuscript is written clearly, but some analysis/discussions should be deepened/expanded as below. I have several major comments that should be addressed and implemented in the revised manuscript before publication in ACP.

Major comments:

- Toluene is assumed to be a good proxy of anthropogenic precursors (P10, L28, without references). While this assumption might be fine, more justifications would be necessary: emission and ambient concentrations of toluene and other anthropogenic precursors are missing and not discussed sufficiently. Thus, general conclusion of phase state of OA in megacities (last sentence of abstract) sounds not convincing to me. The authors could look into emission inventories to check whether toluene is indeed a major precursor in selected major cities. Alkane, alkene and other aromatic compounds may be also important anthropogenic precursors and SOA generated by these precursors may have different viscosities. For example, a recent study showed that dodecane SOA may be less viscous and naphthalene SOA may be more viscous, compared to pinene and isoprene SOA (Berkemeier et al., ACP, 14, 12513, 2014). Please expand discussions on this issue.

- Moreover, as the authors also mentioned in P12, L15-20, I think that the temperature effect on viscosity is so important and probably as large as the RH effect (e.g., Koop et al., PCCP, 2011) that it is not easy to draw general conclusion of anthropogenic SOA phase state by measurements of toluene SOA only at room temperature. As shown in Fig. 6, most cities (except tropics) have lower temperature than 20 C in winter.

- As pointed out in the text, organic aerosols are likely to be internally mixed with inorganics such as sulfate and nitrate (P11, L2). This and also other groups have shown by experimental and modeling studies that a liquid-liquid phase separation is expected if an O:C ratio of an organic-rich phase is low, whereas a mixed one phase may be likely for high O:C. Have you measured an O:C ratio of toluene SOA? Fig. 7 in Lambe et al. AMT, 4, 445, 2011 showed that O:C ratio of OFR-generated toluene SOA

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can be as high as 1.0. This issue should be discussed.

- What was RH in an OFR chamber during SOA formation? Was it all at the same RH? A recent study has shown that as the RH at which the α -pinene SOA is formed increases, there is a decrease in viscosity, and SOA that is formed dry and subsequently humidified remains solid to high RH (Kidd et al., PNAS, 111, 7552, 2014). This potential RH effect needs to be discussed.

- Water acts as plasticizer and plays a key role in determination of phase state. It would be interesting to plot viscosity against water mass fraction in SOA, which can be estimated using a hygroscopicity parameter kappa (e.g., Lambe et al., AMT, 4, 445, 2011; Hildebrandt Ruiz et al., ACP, 15, 8301, 2015).

Minor comments:

- P3, L11: The model used in Riipinen et al., 2011 did not treat bulk diffusion and they have rather emphasized an importance of gas diffusion and condensation. Thus, this reference seems not to be appropriate here. Instead, the authors could cite Shiraiwa et al., PNAS, 110, 11746, 2013.

- P4, L16: Loza et al., EST, 47, 6173, 2013 conducted also very similar experiments that may be worth included.

- P10, L7, L10: "However" is used twice in a row. Please refine.

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