Denjean et al. characterized chemical composition, hygroscopic growth, and refractive indices of α-pinene ozonolysis SOA generated in their CESAM chamber and aged over ~20 hours. They report the following results:

- The bulk O/C ratio of the SOA was approximately 0.68 near the beginning of the experiment and decreased to 0.55 with condensation and growth of less-oxidized, semivolatile oxidation products. The surface O/C ratio of the SOA was lower and was constant over time.

- The SOA showed negligible absorption in the UV/Vis region. The real refractive index of the SOA ranged from about 1.3 to 1.6 and was positively correlated with the O/C ratio.

- The hygroscopic growth factor measured RH = 90% remained constant over the course of an experiment and ranged from 1.02 to 1.07 across experiments.

- As a function of RH, the mobility diameter of size-selected, aged alpha-Pinene SOA decreases and then increases during HTDMA scans. The scattering coefficient increases continuously with increasing RH. The authors interpret these observations as a change in viscosity.

Comments

1. Overall, this manuscript has the potential for eventual publication in ACP. However, I suggest a significant rewrite before reconsideration. In my opinion, a more convincing case needs to be made as to how combining simultaneous hygroscopic and optical measurements helps characterization of SOA properties relative to separate measurements, particularly given the authors' claim that "it is critical to simultaneously determine the hygroscopic behaviour of its size distribution and optical properties as well as their dependence on the chemical composition" (P10547, L16-17).

2. Figures and quantitative analyses focusing on how optical measurements enhance the hygroscopicity measurements (and vice versa) could significantly improve the manuscript. Because many of the results obtained by the authors have already been measured previously – in some cases by multiple researchers – at present it isn't clear to me what new information is gained by combining the measurement techniques that were used, aside from perhaps the XPS measurements of SOA surface chemical composition.

3. I think that some of the discussion about SOA yields and functional group composition detracts from the main focus of the paper. Shortening or removing some of this discussion, along with a more comprehensive

discussion relating optical and hygroscopicity measurements, should further improve the manuscript.

4. There is very little discussion directly relating the calculated scattering growth factor values, f(RH), to hygroscopic growth factor values at the same RH. In my opinion this should be a significant component of the manuscript to support the authors' claim that "it is critical to simultaneously determine the hygroscopic behaviour of its size distribution and optical properties" (P10547, L15-L16). In the current manuscript the size growth factors (Figures 5-6) and scattering growth factors (Figure 7) are presented and discussed separately for the most part.

5. One application/advantage of combined f(RH) and GF measurements seems to be the ability to assess possible residence time limitations (or lack thereof) in the HTDMA. On P10553 near the end of Section 2.3, the authors set up this discussion with the statement: "the two approaches of hygroscopicity measurements could lead to different results, which carry information on water transfer dynamics, possible particles reorganisation or phase transfer equilibrium establishment." However, this is not evident from the data presented in the current manuscript.

6. The authors could use measured f(RH), size distribution, real refractive indices of SOA and of water to calculate a growth factor from the optical measurements. This calculated growth factor could be plotted along with the HTDMA-derived GF as a function of RH for "fresh" and "aged" SOA. In my opinion this is a logical extension of the data presented in Figures 5-7 and I think could more evidently show if there are residence time limitations in the HTDMA-derived GF's. Does the "scattering growth factor" increase with aging? This was not clear in the current manuscript. If the authors are aware of other ways to combine the scattering and size GF data, those should be added to the discussion as well.

7. There are many typos and grammatical errors that should be addressed. I also found two incorrect citations. Additional proofreading is required throughout the manuscript.

8. P10548, L21-L22: There are a few more recent papers that should be cited here:

> Lambe, A. T.; Onasch, T. B.; Massoli, P.; Croasdale, D. R.; Wright, J. P.; Ahern, A. T.; Williams, L. R.; Worsnop, D. R.; Brune, W. H.; Davidovits, P. Laboratory Studies of the Chemical Composition and Cloud Condensation Nuclei (CCN) Activity of Secondary Organic Aerosol (SOA) and Oxidized Primary Organic Aerosol (OPOA). Atmos. Chem. Phys., 11, 8913–8928, 2011.

Wong, J. P. S.; Lee, A. K. Y.; Slowik, J. G.; Cziczo, D. J.; Leaitch, W. R.; Macdonald, A., and Abbatt, J. P. D., Oxidation of ambient biogenic secondary organic aerosolby hydroxyl radicals: Effects on cloud condensation nuclei activity. GEOPHYSICAL RESEARCH LETTERS, VOL. 38, L22805, doi:10.1029/2011GL049351, 2011.

Mei, F.; Setyan, A.; Zhang, Q.; and Wang, J.. CCN activity of organic aerosols observed downwind of urban emissions during CARES. Atmos. Chem. Phys., 13, 12155–12169, 2013.

Rickards, A. M. J.; Miles, R. E. H.; Davies, J. F.; Marshall, F. H.; and Reid, J. P. Measurements of the Sensitivity of Aerosol Hygroscopicity and the κ Parameter to the O/C Ratio. J. Phys. Chem. A, 117, 14120–14131, 2013.

9. P10550-10551: It is not clear to me why it is necessary to go into this level of detail about aethalometer operation when the data are barely used except to confirm that the SOA doesn't absorb?

10. P10557-10558, S3.1: This section could be shortened or moved to the Supplement because the results do not seem critical to the subsequent discussion of hygroscopicity and optical properties.

11. P10562-10563, S3.4.2: This section could be shortened or removed (or moved to the Supplement) because most of this is already well established in the literature. The novel result that the "surface" and "bulk" O/C ratios are different and vary differently with aging – is already presented in sufficient detail in S3.4.1.

12. P10567, S5: It might be useful to compare the extinction cross sections of α-pinene SOA with other aerosol species known to contribute to the direct effect, such as black carbon and biomass smoke.

13. P10573, L28: reference is incorrect.

14. P10577, L24: reference is incorrect.

15. P10589, F3: Is it necessary to show these results graphically if they are all zero within the uncertainty of the Aethalometer measurement?

16. P10592, F6: Can the authors clarify how the GF(90%) values in Figure 5 are calculated when this figure suggests that GF(90%) is approximately 175 / 190 = 0.92, rather than 1.02 – 1.07?

17. P10593, F7: This figure is confusing. Panels (a) and (b) correspond to calculated f(RH) values after 1hr and 16 hr aging, but panel (a) shows different experiment dates than panel (b). Given that, it is not clear to me how to interpret trends in f(RH) over the course of an experiment because it doesn't seem possible to directly relate (a) to (b). Can the authors clarify this? Are there no experiments with nephthelometer measurements at the beginning and at the end on the same date from which there is a clear trend in σ_{scat} with aging?

18. P10593, F7: I am confused by this statement in the figure caption: "The scattering growth factors are calculated as the ratio of σ_{scat} at a specific RH to σ_{scat} at 30 % RH to avoid values lower than 1". Figure 7b shows clearly that f(RH) < 1 below ~25 % RH, down to a minimum f(RH) ~ 0.7. Is this a real trend? Please discuss or clarify.

19. P10595, F9: This figure could be removed (or moved to the Supplement) for reasons mentioned in previous comment #9.

20. P10596, F10: Some combination of the size growth factor, scattering growth factor, and k_{ext} could also be plotted as a function of O/C to illustrate the relationships between the different parameters as a function of chemical composition.

21. P10596, F10: It might be useful to add literature data to this figure from the Nakayama et al. (2012), Lambe et al. (2013), and Liu et al. (2013) studies cited in the manuscript.