

We are very grateful for the reviewer's comments. A point-to-point response to reviewers' comments is provided below, and the manuscript has been revised accordingly. The line numbers in the response refer to those in the revised version of the manuscript.

Reviewer #1: *“The manuscript by Li et al. investigated the sensitivity of a Q-ACSM for the measurements of chamber secondary organic aerosol (SOA) from three volatile organic compounds. The authors found that the sensitivity of the Q-ACSM to SOA was found to be anti-correlated with the aerosol oxidation state regardless of the VOC precursors. This study also shows the applicability of Q-ACSM in chamber studies although rare studies use Q-ACSM for chamber studies because of low sensitivity and time resolution. This manuscript is generally well written and fits within the scope of AMT. However, some conclusions are overstated and need more evidence.”*

Major comments:

1. *“The authors concluded CE as a function of particle phase states, which needs to be reconsidered. First, the RH was maintained at 10 – 15% during the experiments, we could not expect the changes in phase states of SOA particles. Second, Docherty et al. (2013) also showed the variations of CE as a function of f_{44}/f_{57} , which is primarily caused by particle bounce at the vaporizer.”*

Response: We agree with the reviewer that the particle bounce at the vaporizer can explain most of the observed anti-correlation between CE_{org} and f_{44} . The “phase states” here were referring to the phase state of dry SOA particles without the uptake of water. For example, some organics (e.g., oleic acid and nonylaldehyde) with a CE of near unit are intrinsically liquid (Docherty et al., 2013), while smog chamber-generated SOA particles formed via the oxidation of biogenic emission from scots pine appear to be in amorphous semisolid or amorphous solid (glassy) states, rather than being liquid (Virtanen et al., 2010). Particle morphology research based on glass transition temperature (T_g) indicated that during oxidation of liquid pinene, T_g increased from much lower than room temperature (175K) to near room temperature (290K), indicating that the SOA products might vary between liquid state, semi-solid state and solid (glassy) state resulting from a combined effect of increasing molecular weight and O/C (Koop et al., 2011). Accordingly, we believed that the anti-correlation between CE_{org} and f_{44} was actually caused by the phase change with f_{44} . This was supported by the measured effective density that increased from 1.07 to 1.34 with increasing f_{44} . The explanation could be that SOA particles with higher f_{44} would have higher density and would act more like a “bouncing” solid ball instead of a “sticky” droplet. This has been discussed in lines 318-322.

2. *“It is difficult for this manuscript to conclude that RIE changes as SOA evolves from HOA to more oxygenated OOA. This is an SOA experiment, which cannot give you any information on HOA. Many factors can affect the anti-correlation between RF and f_{44} , but RIE would not be the important one. As shown in the latest research by Xu et al. (2018), organic aerosol with OS below -1.5 can have much high RIE, but in this study, the f_{44} does not support this.”*

Response: This work did not cover HOA particles and thus we removed all statements related to HOA. In the case of SOA, we have reevaluated the role of CE and RIE on the observed anti-correlation between RRF (relative response factor) and f_{44} . Within the studied SOA range, we found that the variation of CE_{org} can largely explain the observed RRF changes with f_{44} , indicating that variations of RIE_{org} with f_{44} may be limited in our experiments. Recently, Xu et al. (2018) showed that RIE_{org} for organic with $-1.0 < \overline{OSc}$ (average carbon oxidation states) < 0.5 varied between 1.6 ± 0.5 (2σ), which is relatively higher than

the widely used default value ($RIE_{org} = 1.4$). Clearly, a higher than default RIE_{org} value is consistent with the overall positive deviations of RRF observed in our experiments (Lines 327-332).

3. “I would also suggest the authors comparing the result in this study with those previously reported by HR-ToF-AMS. For example, f_{44} vs. f_{43} could be different. Q-ACSM might reports much higher f_{44} than that of HR-ToF-AMS (Fröhlich et al., 2015).”

Response: As suggested by the reviewer, we have inserted *Section 3.4 Comparison with AMS Calibrations* (Line 299) into the revised manuscript. As indicated by the reviewer, there is an intrinsic difference between a Q-ACSM and an AMS. For example, ammonium nitrate in the ambient aerosol sample may affect the m/z 44 signal in Q-ACSM due to the low mass resolution of Q-ACSM (Fröhlich et al., 2015). Nevertheless, the calibration results obtained from this work are expected to be comparable with those done with the AMS. The following statement has been inserted into section 3.4 (Lines 303-317):

“In previous studies, CE was demonstrated to be variable for particles with different chemical composition, phase state and under different RH, with CE_{org} ranging from less than 0.15 to ~ 1 (Docherty et al., 2013;Alfarra, 2004;Robinson et al., 2015;Matthew et al., 2008). CE for biogenic SOA has been reported to be close to 1 based on both chamber experiments (Kiendler-Scharr et al., 2009) and field measurements conducted in Amazon, where aerosols were dominated by liquid SOA (Allan et al., 2014;Chen et al., 2009). However, it has been proposed in theoretical, chamber, and field studies that besides liquid state, organic particles can exist in semi-solid or solid state (Virtanen et al., 2010;Vaden et al., 2010;Shiraiwa et al., 2011), which might lead to lower CE_{org} . For example, CE_{org} of liquid squalane ($CE = 1$) decreased with SOA condensation (Robinson et al., 2015). Most recent CE_{org} calibration with light-scattering single-particle (LSSP) module also suggested that CE_{org} (0.25 \sim 0.4) could be much less than 1 (Robinson et al., 2017). An inverse relationship between CE_{org} and f_{44}/f_{57} ratio in chamber SOA generation experiments (Docherty et al., 2013) was very similar to our results. However, our results could not be quantitatively compared with the previous ones because the Q-ACSM might report comparably higher f_{44} than the AMS (Fröhlich et al., 2015).”

References:

Alfarra, M.: Insights into atmospheric organic aerosols using an aerosol mass spectrometer, University of Manchester, 2004.

Allan, J. D., Morgan, W. T., Darbyshire, E., Flynn, M. J., Williams, P. I., Oram, D. E., Artaxo, P., Brito, J., Lee, J. D., and Coe, H.: Airborne observations of IEPOX-derived isoprene SOA in the Amazon during SAMBBA, *Atmos Chem Phys*, 14, 11393-11407, 2014.

Chen, Q., Farmer, D. K., Schneider, J., Zorn, S. R., Heald, C. L., Karl, T. G., Guenther, A., Allan, J. D., Robinson, N., Coe, H., Kimmel, J. R., Pauliquevis, T., Borrmann, S., Poeschl, U., Andreae, M. O., Artaxo, P., Jimenez, J. L., and Martin, S. T.: Mass spectral characterization of submicron biogenic organic particles in the Amazon Basin, *Geophysical Research Letters*, 36, 10.1029/2009gl039880, 2009.

Docherty, K. S., Jaoui, M., Corse, E., Jimenez, J. L., Offenberg, J. H., Lewandowski, M., and Kleindienst, T. E.: Collection Efficiency of the Aerosol Mass Spectrometer for Chamber-Generated Secondary Organic Aerosols, *Aerosol Sci Tech*, 47, 294-309, 10.1080/02786826.2012.752572, 2013.

Fröhlich, R., Crenn, V., Setyan, A., Belis, C. A., Canonaco, F., Favez, O., Riffault, V., Slowik, J. G., Aas, W., and Aijälä, M.: ACTRIS ACSM intercomparison-Part 2: Intercomparison of ME-2 organic source apportionment results from 15 individual, co-located aerosol mass spectrometers, 2015.

Kiendler-Scharr, A., Zhang, Q., Hohaus, T., Kleist, E., Mensah, A., Mentel, T. F., Spindler, C., Uerlings, R., Tillmann, R., and Wildt, J.: Aerosol Mass Spectrometric Features of Biogenic SOA: Observations from a Plant Chamber and in Rural Atmospheric Environments, *Environ Sci Technol*, 43, 8166-8172, 10.1021/es901420b, 2009.

Koop, T., Bookhold, J., Shiraiwa, M., and Pöschl, U.: Glass transition and phase state of organic compounds: dependency on molecular properties and implications for secondary organic aerosols in the atmosphere, *Physical Chemistry Chemical Physics*, 13, 19238-19255, 2011.

Matthew, B. M., Middlebrook, A. M., and Onasch, T. B.: Collection efficiencies in an Aerodyne Aerosol Mass Spectrometer as a function of particle phase for laboratory generated aerosols, *Aerosol Sci Tech*, 42, 884-898, 10.1080/02786820802356797, 2008.

Mikhailov, E., Vlasenko, S., Martin, S., Koop, T., and Pöschl, U.: Amorphous and crystalline aerosol particles interacting with water vapor: conceptual framework and experimental evidence for restructuring, phase transitions and kinetic limitations, *Atmos Chem Phys*, 9, 9491-9522, 2009.

Robinson, E. S., Saleh, R., and Donahue, N. M.: Probing the evaporation dynamics of mixed SOA/squalane particles using size-resolved composition and single-particle measurements, *Environ Sci Technol*, 49, 9724-9732, 2015.

Robinson, E. S., Onasch, T. B., Worsnop, D., and Donahue, N. M.: Collection efficiency of [alpha]-pinene secondary organic aerosol particles explored via light-scattering single-particle aerosol mass spectrometry, *Atmos Meas Tech*, 10, 1139, 2017.

Shiraiwa, M., Ammann, M., Koop, T., and Pöschl, U.: Gas uptake and chemical aging of semisolid organic aerosol particles, *Proceedings of the National Academy of Sciences*, 108, 11003-11008, 2011.

Vaden, T. D., Song, C., Zaveri, R. A., Imre, D., and Zelenyuk, A.: Morphology of mixed primary and secondary organic particles and the adsorption of spectator organic gases during aerosol formation, *Proceedings of the National Academy of Sciences*, 107, 6658-6663, 2010.

Virtanen, A., Joutsensaari, J., Koop, T., Kannosto, J., Yli-Pirilä, P., Leskinen, J., Mäkelä, J. M., Holopainen, J. K., Pöschl, U., and Kulmala, M.: An amorphous solid state of biogenic secondary organic aerosol particles, *Nature*, 467, 824, 2010.

Xu, W., Lambe, A., Silva, P., Hu, W., Onasch, T., Williams, L., Croteau, P., Zhang, X., Renbaum-Wolff, L., Fortner, E., Jimenez, J. L., Jayne, J., Worsnop, D., and Canagaratna, M.: Laboratory evaluation of species-dependent relative ionization efficiencies in the Aerodyne Aerosol Mass Spectrometer, *Aerosol Science and Technology*, 52, 626-641, 10.1080/02786826.2018.1439570, 2018.

Young, K. D., and LeBoeuf, E. J.: Glass transition behavior in a peat humic acid and an aquatic fulvic acid, *Environ Sci Technol*, 34, 4549-4553, 2000.