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 f44, 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 f44 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 < \textit{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$_{\text{org}} = 1.4$). Clearly, a higher than default RIE$_{\text{org}}$ 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, f44 vs. f43 could be different. Q-ACSM might reports much higher f44 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, these is 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$_{\text{org}}$ 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$_{\text{org}}$. For example, CE$_{\text{org}}$ of liquid squalane (CE = 1) decreased with SOA condensation (Robinson et al., 2015). Most recent CE$_{\text{org}}$ calibration with light-scattering single-particle (LSSP) module also suggested that CE$_{\text{org}}$ (0.25 ~ 0.4) could be much less than 1 (Robinson et al., 2017). An inverse relationship between CE$_{\text{org}}$ and $f_{\text{44}}$/f$_{\text{57}}$ 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 $f_{\text{44}}$ than the AMS (Fröhlich et al., 2015).”

References:


