

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 #4: *"This manuscript describes measurements of the sensitivity of an ACSM to OA for three oxidant/VOC SOA systems produced in a mid-sized Teflon chamber using an ACSM, SMPS, DMA-APMA. Relationships of sensitivity and effective density to f_{44} are explored. Experiments were conducted in attempt to separate effects on sensitivity due to collection efficiency (CE) and relative ionization efficiency (RIE) of OA. Conclusions are made on trends of CE, RIE, phase state, HOA vs OOA, effective density, and oxidation state.*

The manuscript is generally poorly written and makes a range of conclusions using flawed logic and speculation based on incomplete experimental evidence and lack of context from the existing literature. Due to some critical flaws (discussed below), I do not believe the manuscript is publishable in its present form. If it was published, it would "pollute" the literature with trends that are not well supported and may be due to experimental factors completely unrelated to those that are discussed."

Response: As suggested by the reviewer, we have thoroughly revised the manuscript, which has been proofread by a native English speaker. Since HOA was not specifically studied in this work, we have limited our conclusions on chamber-generated SOA only to avoid possible overstatements. Therefore, Figure 8 has been removed from the manuscript. Additional discussion of the inter-comparison between this work and previous AMS calibrations have been added in section 3.4. Moreover, additional laboratory calibrations of instruments used in this work have been conducted to fully address the uncertainties associated with our measurements.

All the efforts above were meant to constrain the measurements uncertainty of a Q-ACSM. Our results strongly indicate that measurements of OA with various oxygen contents by a Q-ACSM using constant conversion factors may induce significant errors in mass concentration measurements for laboratory generated SOA. The results of this work appeared to be consistent with the results of previous work using different experimental settings and reconfirmed the reliability of the AMS/ACSM techniques. Our work demonstrated the capability of the low-resolution Q-ACSM in doing SOA chamber studies and the necessity to calibrate RF in laboratory experiments. Although the results of this work may not be directly applicable to ambient Q-ACSM measurements, it is reasonable to believe that the RRF of a Q-ACSM could be highly variable under different ambient conditions and thus similar comprehensive calibrations of Q-ACSM for complicated ambient conditions is critically needed in the future work.

"With substantial additional work (including additional experimental evidence, analysis, and calibrations) and documentation, greatly scaled back conclusions, more thorough context, and, the dataset may eventually be publishable. Even in that case, the authors would need to present a convincing case of what parameters they are truly quantifying, including quantifying their uncertainties, and why the results are useful and better inform the analytical study of OA in the laboratory. In its current form, the manuscript is misleading and does not advance findings that are adequately constrained, new, or useful to the literature. Therefore, I strongly recommend that the paper is rejected, and the authors are encouraged to do the additional work and resubmit a much improved paper at a later time. See details below."

Response: As suggested by the reviewer, we have revised the manuscript completely. The major revisions of the manuscript include:

1. The special terms (e.g., RF, CE, RIE) were precisely defined in the "introduction" and their mathematical correlations were also clearly stated. The term "sensitivity" was replaced with "relative response factor (RRF)" and its calculation method was given in the new section 2.5 *Calculation and Uncertainty Evaluation*.
2. We performed additional experiments to calibrate the ACSM lens transmission efficiency (E_L) and the specifically calibrated E_L was used in PM_{SMPS} calculations; Both SMPS and APM were calibrated

before the experiment and the results have been used to precisely evaluate their measurement uncertainties, which were used to constrain the possible errors of measured CE, RIE, and RRF. Therefore, the overall measurement uncertainties of this research were well defined.

3. More information on instrument calibrations, uncertainty evaluations were given in the supplementary information. Average number and mass distributions in all of the experiments were also plotted in the supplementary information (Figs. S4-6).
4. The manuscript was reorganized and fully revised. Inter-comparisons between our experiments and previous calibration work using AMS were added in a new section 3.4 *Comparison with AMS Calibrations*.
5. The SOA coating thickness for the AS seed particles were calculated (18-25 nm), which was found to be enough to fully cover the 70 - 80 nm (in mobility diameter) AS seed particles. AS seeds were fully covered with SOA in most of our experiments. This support our claim of possible morphology change during AS seed experiments.
6. Although RIE_{org} was not directly measured in our experiments, it appeared that the variation of CE_{org} with f_{44} could largely explain the observed anti-correlation between RRF and f_{44} , indicating that RIE_{org} may not vary significantly with f_{44} within the studied SOA range.
7. Figure 8 is removed from the manuscript to limit our discussion within the experiment setting only.

Specific comments:

1. “The paper really only quantifies an empirically-defined response factor, which is the ratio of the mass concentrations reported by the ACSM and calculated from the SMPS volume and the measured particle density. As thoroughly documented in the AMS literature, that response factor can have trends due to several effects:

$$RF = (RIE_{real} / RIE_{assumed}) * (Eb_{real} / Eb_{assumed}) * (EL_{real} / EL_{assumed}) * (ES_{real} / ES_{assumed}) * (SMPS_{calc} * SMPS_{volreal}) * (p_{real} / p_{assumed})$$

where $CE = Eb * EL * ES$ shows the 3 components of CE, due to particle bounce in the vaporizer, incomplete transmission by the aerodynamic lens, and non-spherical particle effects, respectively as defined by Huffman et al. (2005). It is also clear that errors in the estimation of particle volume from the SMPS number distribution and/or the density will also appear in RF.

Response: We agree with the reviewer that many factors (e.g., E_L , E_S , E_b , SMPS size-distribution, and ρ_{eff}) would affect the response factor of the Q-ACSM. Typically, the response factor of the Q-ACSM is defined as: (line 84)

$$RF_{org} = IE_{NO_3} \cdot RIE_{org} \cdot CE_{org} \quad (1)$$

In our study, a term “relative response factor (RRF)” was defined to express how Q-ACSM measured mass concentrations using default $RIE_{org} = 1.4$ and $CE_{org} = 0.5$ deviate from the SMPS-APM measured mass concentration, where RIE, E_b , E_L , and ρ_{eff} were all considered in the calculation: (line 182)

$$RRF = \frac{PM_{ACSM}(assuming\ RIE=1.4, CE=0.5)}{PM_{SMPS}(amended\ by\ \rho_{eff}, ES, EL)} = \frac{RIE_{org} \cdot CE_{org}}{1.4 \cdot 0.5} \quad (2)$$

The detailed calculation procedures have been given in section 2.4 (Lines 177-195).

2. “When one is trying to characterize a widely used instrument and publishing the observed results as if they are representative of all instruments of the same type, great care needs to be taken to examine each of these terms and their uncertainties. The present paper has some speculative interpretations of RF based on RIE and E_b , even though it completely ignores all the other effects! My best guess is that E_L explains a substantial fraction, and potentially most of the trend in RF, as when the particles grow in the SOA experiments, the upper end of the distribution may fall outside of the ACSM lens transmission, thus creating

an experiment and time-dependent trend on RF. It is possible that some of the other factors that are ignored in this paper may also contribute to the observed trend. For this reason, the paper is not publishable.”

Response: The E_L of the aerodynamic lens system of the Q-ACSM was measured and applied to the particle size distributions of all the experiments. For the experiments without SA seed particles, a small portion of smaller particles (<100 nm) were lost in the lens (Fig. S4). For the experiments with SA seed particles, more than 95% particles were transported through the lens (Fig. S6).

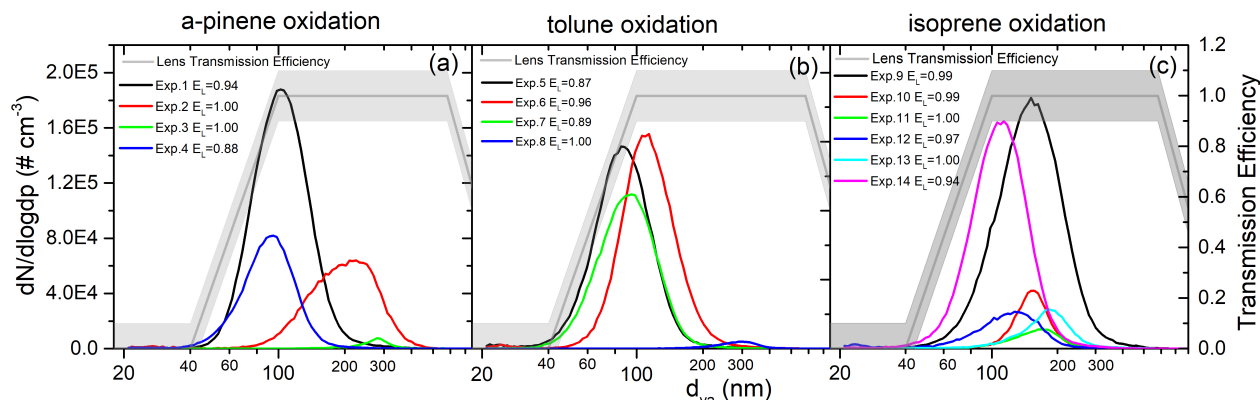


Figure S4. Average number size distributions measured with the SMPS as a function of d_{va} (nm) during the relative stable state for experiments using (a) α -pinene (b) toluene, and (c) isoprene, respectively. d_m was converted to d_{va} via $d_{va} = d_m \cdot \rho_{eff}/\rho_0$. Gray lines represent the Q-ACSM aerodynamic transmission efficiency (E_L) as a function of d_{va} (nm). For most of the experiments, more than 94% of the total particle mass was transported into the Q-ACSM through aerodynamic lens. While for three of them (Exp. 4, 5, and 7), a portion of particles were too small to pass the lens and only 87-89% of the total mass was transported.

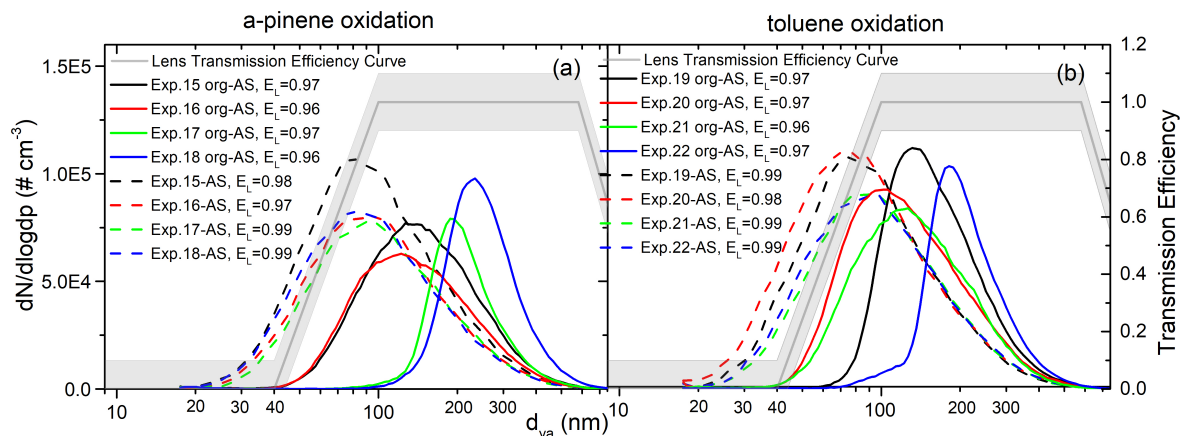


Figure S6. Average number size distributions (d_{va}) measured before (dashed lines) and after (solid lines) SOA (panel a: α -pinene; panel b: toluene) was coated. Gray lines represent the Q-ACSM E_L as a function of d_{va} (nm). Gray lines represent the Q-ACSM aerodynamic transmission efficiency (E_L) as a function of d_{va} (nm). For all the experiments, more than 95% particles were transported through the lens.

3. “The authors need to go back to the lab and present multiple additional pieces of information, including:

1) A detailed characterization of E_L vs. particle size for their own instrument. It is well-known that E_L varies for different instruments (and sometimes in time for a given instrument), especially at the upper end of the particle size. However, Figure 2 suggests that losses at the small end may also be a problem. These are challenging experiments, but they are doable. If the authors cannot accomplish this, they should not

resubmit this paper elsewhere, as the results would be ambiguous, and it would be very misleading to report them as trends in E_b and RIE_{org} while ignoring E_L . Using the transmission curves from the literature is not acceptable, given substantial variability in E_L across instruments. See below for literature references.

Response: We have fully characterized the E_L as a function of particle size in our laboratory. (SI Lines 32-51)

“The lens transmission efficient (E_L) of the Q-ACSM was calibrated using the same setup as in Figure S1. The only difference was that the DMA was set to select a variety of particles with diameters ranging from 200 nm to 600 nm instead of a fixed single diameter of 300 nm. To minimize multiple charge problem for larger particles, we conducted E_L calibration under two sets of comparably low NH_4NO_3 solution concentrations (~ 2 mM and ~ 0.5 mM). However, for the set of experiment with extremely low NH_4NO_3 concentration (~ 0.5 mM), the Q-ACSM sensitivity was not high enough to detect NH_4NO_3 particles, which therefore was not shown here. Consequently, the calibration result of the lens transmission efficiency was shown in Figure S2.

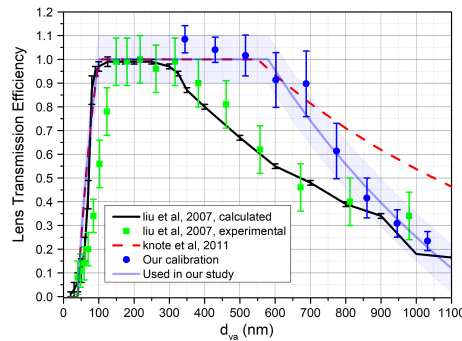


Figure S2. Lens transmission efficiency as a function of particle vacuum aerodynamic diameter (d_{va}).

Lens transmission efficiencies for 300-600 nm particle were nearly unity, which was in reasonable accordance with some previous studies (Jayne et al., 2000; Knoté et al., 2011; Hu et al., 2018), but the diameter range with a unity lens transmission efficiency was much broader than that reported by Liu et al. (2007). The blue line in Figure S2 was used to modify size distribution in our experiments (i.e., 0% transmission for d_{va} below 40 nm; linear increase in transmission vs $\log(d_{va})$, from 0% to 100% at $d_{va} = 100$ nm; 100% transmission efficiency from $d_{va} = 100$ nm up to $d_{va} = 580$ nm; linear decrease in transmission vs $\log(d_{va})$ from 100% at 580 nm to 0% at 1200 nm). For d_p below 300 nm, we used the results reported by Knoté et al. (2011) indicated by the red dashed line in Figure S2. About 10% of uncertainty (shaded area) was applied to the E_L used in this study as indicated by the blue curve in Figure S2.”

2) Using the measured SMPS volume distributions, the measured density, and the measured ACSM transmission vs particle size (in d_{va} space, see DeCarlo et al., 2004), the authors can “trim” the SMPS volume to correspond to the volume actually sampled by the ACSM.

Response: The SMPS-APM measured mass concentration was obtained as following:

E_L as a function of particle diameter in the aerodynamic lens was applied to SMPS-APM measured mass concentration. E_s was assumed to be 1, the possible non-spherical situation was considered by adding an error of 5% for E_s (Lines 182-195):

$$PM_{SMPS} = V_{SMPS} \cdot \rho_{eff} \cdot E_L \cdot E_s = \sum_{i=1}^n \left\{ \frac{\pi}{6} d_{m,i}^3 \cdot dN_i \cdot \rho_{eff,i} \cdot E_{L,i} \cdot E_{s,i} \right\} \quad (3)$$

Eq. (3) is used to calculate PM_{SMPS} , where V_{SMPS} is the volume concentration calculated with SMPS measured size distribution; n is the total number of bins for SMPS measurements; $d_{m,i}$ and dN_i are

respectively the geometric mean diameter and the number of counts in size bin i ; $E_{L,i}$ and $E_{S,i}$ are the aerodynamic lens transmission efficiency and shape factor for particles of $d_{m,i}$. After applying $E_{L,i}$ and $E_{S,i}$, any deviation in CE_{org} should be caused only by particle bounce on the vaporizer surface (E_b). The measured E_L (Fig. S2) is in reasonable accordance with previous studies (Jayne et al., 2000; Knote et al., 2011; Hu et al., 2017), while larger than other reports (Liu et al., 2007) as for 300 nm~1000 nm (d_{va}) particles. E_s was assumed to be $1\pm5\%$. d_m and d_{va} were convertible via $d_{va} = d_m \cdot \rho_{eff} / \rho_0$ for a reference density ρ_0 of 1000 kg m⁻³ (Jayne et al., 2000; DeCarlo et al., 2004). The uncertainties of $d_{m,i}$ ($\pm4\%$), dN_i ($\pm10\%$ for 20-200 nm and $\pm20\%$ for 200-800 nm particles) (Wiedensohler et al., 2012; Wiedensohler et al., 2018), ρ_{eff} ($\pm3\%$), E_L ($\pm10\%$) and E_s ($\pm5\%$) were well propagated to the calculation. As a result, the relative uncertainty of PM_{SMPS} was between 19.4~26.0%, depending on the actual size distribution. Details of uncertainty evaluation can be found in SI.”

3) It does not seem acceptable to present density measurements at only one size, especially if it is the peak of the number distribution. A few points across the distribution, and importantly the peak of the volume distribution, should be included for each system.

Response: In this work, we consider the chamber-generated SOA were mostly unimodal distributed (as shown in Fig. S4), especially for the AS seeded experiments, when only a single mode of AS particles was present in the chamber. The number-size distribution measured by the SMPS was used to deduce the aerosol total volume. The peak size of the distribution was considered the most representative size of the SOA population. The APM was used to measure the true mass of a certain size (mobility diameter) of particle and thus APM is usually used with a DMA for size selection first (McMurry et al., 2002). The mass distribution obtained by the APM was determined by the resolution of the APM, i.e., its rod rotational speed and its physical radius. The magnitude of the APM peak was only used to determine the exact mass of a certain size particle not the actual number concentration of that size of particle in the chamber. Accordingly, the effective density of the SOA population was calculated, which was then used to calculate the total mass of the SOA. As SOA continued to grow as the experiment proceeded, it was important to track the SOA changes with high time-resolution. Typically, one SMPS-APM scan can be completed within 10 min, which cannot be achieved by the traditional filter-based aerosol measurement technique. We have tested the APM with a certain size of PSL, slight change in peak selection of the DMA setting by a few nm, the mass measured by the APM was not significantly affected but the magnitude of the peak was substantially reduced. Therefore, we believe slightly off in the peak size selection would not affect the APM mass measurement. In fact, our results showed that the average effective density from each experiment did not show any dependence on the particle size. It has been stated in the revised paper that “Before applying this measured ρ_{eff} to the whole size distribution, ρ_{eff} of particles in all sizes in the same moment were assumed to be the same.” (Lines 168-170).

4) Evidence of good alignment of the aerodynamic lens in the ACSM should be presented. The particle beam width depends on particle size and shape, and if the lens is not well-aligned, then there can be particle losses that are strongly size-dependent, giving rise to another source of variation of EL that would show up in RF.

Response: The lens alignment was done right before the RIE calibration using NH_4NO_3 particles (Line 155). The setup of lens alignment was the same as that of NH_4NO_3 RIE calibration, except that CPC was not used. 300 nm NH_4NO_3 dry particles were atomized, dried, and size-selected by DMA. The NO_3 signal intensity was monitored with the “Lens alignment tool” while the particle lens was moved across one of the planes (horizontal or vertical axis) to the limit where no NO_3 signal was seen. The center position of the four edges was assigned for the lens plate.

5) Evidence of calibration of the SMPS size (using PSLs) and especially, volume concentration, needs to be presented. It is typical in field studies when multiple SMPSs are present (all operated by reputable

groups) that the reported volume concentrations range a factor of 4-5. That's the typical accuracy that can be expected for an SMPS that is running on a lab and that has not been thoroughly calibrated and quality-checked. Even after careful work, discrepancies often remain, see for example Figure 8 in Wiedensohler et al. (2012). There are a number of papers that specify the checks that one has to do in order to ensure quantitative SMPS measurements, and it was especially glaring that this topic was not even mentioned in the present paper. See for example Wiedensohler et al. (2018) and Stolzenburg and McMurry (2018). An intercomparison to other instruments such as additional SMPSs and OPCs (for example UHSAS) would be useful to gain confidence in the measurements. Evidence of quantitative response of the CPC, ideally by comparison to an electrometer, but otherwise by comparison to several other CPCs (across the size range) is also needed.

6) Evidence of accurate density measurements using known materials, and across the particle size range contributing to the measurements shown, needs to be presented.

Response: The SMPS-APM system was fully calibrated before the experiments. Detailed operation procedures have been given in lines 170-175:

“The flow rates and the voltages applied to the DMAs were well calibrated before the experiments. Three sizes of polystyrene latex (PSL) spheres (81 ± 3 nm, 147 ± 3 nm, 269 ± 5 nm) with a density of 1.05 g cm^{-3} were nebulized with an atomizer (TSI, Model 3076) and then dried by a diffusion dryer filled with silica gel for size calibration of the SMPS system. The same PSL spheres were also used for the calibration of APM (see Fig. S3). The bias of SMPS size and APM effective density measurements were within $\pm 4\%$ and $\pm 3\%$, respectively.”

The uncertainty analysis of the SMPS measurements was as following (SI Lines 71-90):

“ PM_{SMPS} was calculated using Eq. (3) in the main text. Theoretically, the uncertainty of PM_{SMPS} ($\sigma_{PM-SMPS}$) was associated with $d_{m,i}$, dN_i , $\rho_{eff,i}$, $E_L(d_{m,i})$, and $E_{s,i}$. As discussed in the instrument calibration in Section 1, uncertainties of $d_{m,i}$ (σ_{dm}) and ρ_{eff} (σ_ρ) were within $\pm 4\%$ and $\pm 3\%$, respectively. The uncertainty of E_L (σ_{EL}) in the range of 40-600 nm (d_{va}) was estimated to be $\pm 10\%$ according to Figure S2. The uncertainty of dN_i (σ_N), which was the combination of the uncertainties of particle charging efficiency (σ_{char}), diffusion loss (σ_{diff}), and CPC counting efficiency (σ_{count}) (Buonanno et al., 2009), was assigned to be $\pm 10\%$ for particles with a diameter of 20-200 nm and $\pm 20\%$ for 200-800 nm (Wiedensohler et al., 2012; Wiedensohler et al., 2018). The uncertainty of E_s (σ_{ES}) was estimated to be $\pm 5\%$. The uncertainty of PM_{SMPS} was hence estimated by Eq. (S3) using the averaged sized distribution (as shown in Figures S4 and S6) and ρ_{eff} in each experiment:

$\sigma_{PM-SMPS}$

$$= \sqrt{\sum_{i=1}^n PM_{SMPS,i}^2(d_{m,i}) + \sum_{i=1}^n PM_{SMPS,i}^2(dN_i) + \sum_{i=1}^n PM_{SMPS,i}^2(\rho_{eff,i}) + \sum_{i=1}^n PM_{SMPS,i}^2(E_L(d_{m,i})) + \sum_{i=1}^n PM_{SMPS,i}^2(E_{s,i})} \quad (S3)$$

where $PM_{SMPS,i}(x_i)$ is the product of the uncertainties of each bin x_i ($\sigma_{x,i}$), and the corresponding sensitivity

coefficient $\frac{\partial PM_{SMPS,i}}{\partial x_i}$, as shown in Eq. (S4):

$$PM_{SMPS,i}(x_i) = \frac{\partial PM_{SMPS,i}}{\partial x_i} \cdot \sigma_{x,i} \quad (S4)$$

Then, Eq (S3) can be simplified as Eq. (S5):

$$\sigma_{PM-SMPS} = \sqrt{\sum_{i=1}^n (9\sigma_{dm,i}^2 + \sigma_{dN,i}^2 + \sigma_{\rho_{eff},i}^2 + \sigma_{EL,i}^2 + \sigma_{ES,i}^2) PM_{SMPS,i}^2} = \sqrt{\sum_{i=1}^{n^*} 0.0378 PM_{SMPS,i}^2 + \sum_{i=n^*}^n 0.0678 PM_{SMPS,i}^2} \quad (S5)$$

where n^* corresponds to the bins with d_m larger than 200 nm. Overall, the uncertainty of PM_{SMPS} was estimated to be between $\pm 19.4\%$ and $\pm 26.0\%$ with the exact value depending on the size distribution (Figures S4 and S6)."

7) The uncertainty in each term of the equation for RF, as well as in 1-6 above, needs to be quantified, and the uncertainty needs to be propagated. Only then we can know if any of the reported trends for (for example) Eb have any meaning.

Response: The uncertainty analysis is detailed in the supplementary information section 2 (SI Lines 63-110):

"2 Uncertainty Analysis

In this work, the reported quantity was considered as a function (y) of measured variables ($x_1, x_2, x_3 \dots x_n$) in the form of $f(x_1, x_2, \dots, x_n)$. The uncertainties associated with measured $x_1, x_2, x_3 \dots x_n$ were $\sigma_1, \sigma_2, \dots \sigma_n$, respectively. Accordingly, the mean value of y (\bar{y}) and its uncertainty (σ_y) were calculated using Eq. (S1) and Eq. (S2) as following:

$$\bar{y} = f(\bar{x}_1, \bar{x}_2, \dots, \bar{x}_n) \quad (S1)$$

$$\sigma_y = \sqrt{\left(\frac{\partial y}{\partial x_1}\right)^2 \sigma_1^2 + \left(\frac{\partial y}{\partial x_2}\right)^2 \sigma_2^2 + \dots + \left(\frac{\partial y}{\partial x_n}\right)^2 \sigma_n^2} \quad (S2)$$

2.1 PM_{SMPS} uncertainty

PM_{SMPS} was calculated using Eq. (3) in the main text. Theoretically, the uncertainty of PM_{SMPS} ($\sigma_{PM-SMPS}$) was associated with $d_{m,i}$, dN_i , $\rho_{eff,i}$, $E_L(d_{m,i})$, and $E_{s,i}$. As discussed in the instrument calibration in Section 1, uncertainties of $d_{m,i}$ (σ_{dm}) and ρ_{eff} (σ_ρ) were within $\pm 4\%$ and $\pm 3\%$, respectively. The uncertainty of E_L (σ_{EL}) in the range of 40-600 nm (d_{va}) was estimated to be $\pm 10\%$ according to Figure S2. The uncertainty of dN_i (σ_N), which was the combination of the uncertainties of particle charging efficiency (σ_{char}), diffusion loss (σ_{diff}), and CPC counting efficiency (σ_{count}) (Buonanno et al., 2009), was assigned to be $\pm 10\%$ for particles with a diameter of 20-200 nm and $\pm 20\%$ for 200-800 nm (Wiedensohler et al., 2012; Wiedensohler et al., 2018). The uncertainty of E_s (σ_{ES}) was estimated to be $\pm 5\%$. The uncertainty of PM_{SMPS} was hence estimated by Eq. (S3) using the averaged sized distribution (as shown in Figures S4 and S6) and ρ_{eff} in each experiment:

$$\sigma_{PM-SMPS} = \sqrt{\sum_{i=1}^n PM_{SMPS,i}^2 (\sigma_{dm,i}^2 + \sigma_{dN,i}^2 + \sigma_{\rho_{eff},i}^2 + \sigma_{EL,i}^2 + \sigma_{ES,i}^2)} \quad (S3)$$

where $PM_{SMPS}(x_i)$ is the product of the uncertainties of each bin x_i ($\sigma_{x,i}$), and the corresponding sensitivity coefficient $\frac{\partial PM_{SMPS,i}}{\partial x_i}$, as shown in Eq. (S4):

$$PM_{SMPS}(x_i) = \frac{\partial PM_{SMPS,i}}{\partial x_i} \cdot \sigma_{x,i} \quad (S4)$$

Then, Eq (S3) can be simplified as Eq. (S5):

$$\sigma_{PM-SMPS} = \sqrt{\sum_{i=1}^n (9\sigma_{dm,i}^2 + \sigma_{dN,i}^2 + \sigma_{\rho_{eff},i}^2 + \sigma_{EL,i}^2 + \sigma_{ES,i}^2) PM_{SMPS,i}^2} = \sqrt{\sum_{i=1}^{n^*} 0.0378 PM_{SMPS,i}^2 + \sum_{i=n^*}^n 0.0678 PM_{SMPS,i}^2} \quad (S5)$$

where n^* corresponds to the bins with d_m larger than 200 nm. Overall, the uncertainty of PM_{SMPS} was estimated to be between $\pm 19.4\%$ and $\pm 26.0\%$ with the exact value depending on the size distribution (Figures S4 and S6).

2.2 RRF uncertainty

RRF was calculated with Eq. (2) in the main text. The uncertainty of RRF (σ_{RRF}) was related to PM_{ACSM} and PM_{SMPS} and was calculated using Eq. (S6). PM_{SMPS} uncertainty obtained from Section 2.1 of the SI was used to calculate the RRF uncertainty, instead of using the standard deviations of the PM_{SMPS} measured in the “relative stable state” of chamber studies. While for the case of Q-ACSM, the standard deviation in the stable state was used to evaluate the RRF uncertainty.

$$\sigma_{RRF} = \sqrt{\left(\frac{\partial RRF}{\partial PM_{ACSM}}\right)^2 \sigma_{PM-ACSM}^2 + \left(\frac{\partial RRF}{\partial PM_{SMPS}}\right)^2 \sigma_{PM-SMPS}^2} = \sqrt{\left(\frac{\sigma_{PM-ACSM}}{PM_{SMPS}}\right)^2 + \left(\frac{PM_{ACSM} \sigma_{PM-SMPS}}{PM_{SMPS}^2}\right)^2} \quad (S6)$$

2.3 CE uncertainty

For the experiments using AS seeds, the collection efficiencies before and after SOA coating were calculated using Eq. (S7) and Eq. (S8), respectively:

$$CE_{AS} = \frac{0.5 PM_{ACSM-AS1}}{PM_{SMPS-AS}} \quad (S7)$$

$$CE_{org} = \frac{0.5 PM_{ACSM-AS2}}{PM_{SMPS-AS}} \quad (S8)$$

where, $PM_{ACSM-AS1}$ and $PM_{ACSM-AS2}$ represents average Q-ACSM measured mass concentrations before and after SOA coating, respectively. $PM_{SMPS-AS}$ was the mass concentration of AS measured by SMPS before SOA coating. The default CE is 0.5. The uncertainties of CE_{AS} and CE_{org} were calculated with the Eq. (S9) and Eq. (S10), respectively.

$$\sigma_{CE-AS} = 0.5 \sqrt{\left(\frac{\sigma_{PM-ACSM-AS1}}{PM_{SMPS-AS}}\right)^2 + \left(\frac{PM_{ACSM-AS1} \sigma_{PM-SMPS-AS}}{PM_{SMPS-AS}^2}\right)^2} \quad (S9)$$

$$\sigma_{CE-org} = 0.5 \sqrt{\left(\frac{\sigma_{PM-ACSM-AS2}}{PM_{SMPS-AS}}\right)^2 + \left(\frac{PM_{ACSM-AS2}\sigma_{PM-SMPS-AS}}{PM_{SMPS-AS}^2}\right)^2}$$

(S10)”

4. “For Also it should be noted that for internally mixed particles such as those generated here Eb will be a property of the particle that is influenced by its sulfate and OA content. So it is nonsensical to write Eb,SO₄ and Eb,OA (as the authors do in their notation) as if those were different quantities in the seeded experiments, or as if they were a property of the species and not the mixture.

Moreover, it is not standard practice to use ACSM or AMS measurements for quantitative yield experiments for chamber experiments in the first place, due to the complexities outlined above. Rather, researchers typically have used SMPS volume together with density estimates (or comparisons with filters as well as other methods), and OA/SO₄ ratios, to quantify yields. Jimenez et al. 2016 (which is referenced in this manuscript) states “Finally, we reiterate the need for direct calibration in laboratory experiments utilizing specific organic compounds or mixtures.” So, this issue is not new and nor do the authors offer an advance in addressing the issue.

As discussed on that reference, the trends of AMS response observed for field and laboratory experiments are often different, due to the wider range of particle materials that can be made in the lab. Lab experiments typically have trouble replicating the OA composition observed in the atmosphere. Therefore, no implication should be made that the results from a lab study apply to field measurements. Any statements about the performance of the AMS / ACSM in field measurements should be derived from the analysis of field measurements, of which there is an extensive record in the published literature. Comparisons of AMS with SMPS in field studies are not consistent with the trends shown here, see for example Jimenez et al. (2016) and references therein.

Response: The SOA particles generated during the experiments using AS seeds should be assume a core-shell morphology. We have calculated the coating thickness and found that 18-25 nm should be thick enough to fully cover the AS seeds. AS in most of the experiments were fully covered (Lines 278-282). After the AS particles are fully covered by organic shell, the collection efficiency should reflect the property of the SOA shell. Thus, the CE measured with/without SOA coating were considered as the CE_{AS} and CE_{org}, respectively. We agree with reviewer that filter-based aerosol measurement can achieve higher precision but it also requires longer analysis time and pretreatment of the filter. SMPS-APM technique can achieve the same goal within 10 min and thus can be used to track the changes of SOA insider the chamber with higher time resolution and high precision (+/-3%).

Due to the low-cost and durability, Q-ACSMs have been widely used in both ambient and laboratory experiments to quantify aerosol chemical compositions, especially for long-term observations in highly polluted regions (such as in China). However, the performance of Q-ACSM in organics measurement was rarely evaluated because of its low time and m/z resolution and the lack of particle size selection. To the best of our knowledge, this study represents the first comprehensive chamber calibration of CE_{org} and RRF_{org} for a Q-ACSM. Our results of RRF and CE_{org} calibration were generally in line with previous studies conducted with the AMS.

5. “Other Important Comments:

As was extensively detailed by Reviewer 2, the manuscript is full of grammatical errors, confusing text, logical flaws and overstatements of conclusions. I agree with the comments of Rev. 2, and will thus focus on the most important additional points, and a few similar points with additional context or emphasis. However, the list is not comprehensive as it would be too time consuming to point out all the issues and any effort to revise and resubmit the paper should go beyond just the issues detailed by the reviewers. A detailed review of the Abstract is below, followed by other issues organized by topic.

Response: We have revised the manuscript thoroughly as suggested by all reviewers. The revised manuscript has been proofread by a native English speaker. Point-to-point responses to reviewer 2's comments have been uploaded separately.

6. *“Abstract: L21: “comprehensively” is an extreme overstatement.*

Response: It has been deleted.

L25/L31/L32: “oxidation states” were not measured in this study.

Response: “oxidation states” have been replaced with “ f_{44} ” in the revised manuscript.

L27: “exact mass” doesn’t make sense.

Response: It has been removed.

L35: The cause of decreasing RF was not “pinpointed” or even significantly constrained.

L36: The CE was not determined quantitatively.

L37-39: “Our experiment results along with previous literature reports strongly implied that as the SOA oxidation state increases, SOA will transform gradually from a liquid state ($CE \gg 1$) into a solid (or glassy) state with a CE of 0.2~0.5.” This is a gross overstatement of what the evidence presented in this manuscript supports. The trends observed may have been any combination of lens transmission effects, Eb effects of mixed dry ammonium sulfate / OA particles, OA RIE effects, OA CE effects, or variable errors on the SMPS or CPC used for comparison. Neither CE effects nor phase state effects were isolated in this study.

Response: After extensive calibrations and uncertainty analysis, the statement related to RF and CE has been revised as (lines 30-40): “Our results showed that RRF for a specific type of SOA anti-correlated with f_{44} regardless of the VOC precursors. RRF decreased from 2.16 to 0.81 when f_{44} increased from 0.057 to 0.191. Meanwhile, ρ_{eff} increased from 1.09 to 1.34 g cm⁻³, implying more compact structure for more oxidized SOA. The anti-correlation between RRF and f_{44} might be due to the decreased RIE_{org} or CE_{org} for more oxidized SOA. To further explore the actual cause, ammonium sulfate (AS) seed particles were injected into the chamber before SOA were produced. After the 80 nm AS particles were fully coated with SOA (coating thickness > 18~25 nm), CE of the aerosol changed from CE_{AS} (0.285±0.067) to CE_{org} of the SOA shell. The experiment showed that CE_{org} decreased from 0.882 to 0.313~0.475 when f_{44} increased from 0.127 to 0.209, most likely caused by a phase change of SOA from liquid to solid/glassy.

7. *“L39-41: “Meanwhile, the RIE of OA decreased substantially when SOA transformed from hydrocarbon-like OA (HOA) into more oxygenated OA (OOA) and may further decrease as O/C continued to increase.” Like with Eb, the effects of RIE of OA were NOT isolated in this study. Moreover, the results presented here have no bearing on HOA since only chamber SOA was studied. Also, no data on O/C was presented here (inferring O/C from f_{44} for chamber SOA is not justified – see details below)”*

Response: Since HOA particles were not measured in this study, we have limited our conclusion to be based on chamber-generated SOA only. Therefore, Fig. 8 has been removed from the manuscript.

8. *“L41-44: “Our results indicated that the current Q-ACSM calibration procedure using a constant RIE may lead to somewhat underestimation of more oxidized OOA but overestimation of less oxidized HOA, i.e., a variable RIE shall be applied, most likely as a function of the SOA oxidation state.” As stated above (and further explained below), RIE effects were not isolated.”*

Response: The sentences have been revised as (Lines 37-41): “The variation of CE_{org} with f_{44} could explain a large fraction of the observed decrease in RRF, while the influence of RIE_{org} cannot be excluded. The trends in RRF and CE_{org} for Q-ACSM were in accordance with those done with the high-resolution AMS, demonstrating the capability of the low-resolution Q-ACSM in doing SOA laboratory studies and the necessity to calibrate RF when conducting laboratory SOA experiments.”

9. *“More Detail on Lens Transmission Effects: The ACSM lens transmission declines to zero for both small and large particles. In particular the upper size cut depends on the specific lens being used (Hu et al., 2017), and can even change in time after an instrument is moved to a different location. The transmission also depends strongly on the pressure inside the AMS aerodynamic lens, which can decline dramatically if the pressure in the lens is reduced (e.g. Bahreini et al., 2003), for example due to a partially clogged critical orifice, or to just being set at the wrong pressure by using an improperly sized critical orifice. This value should be documented here for all experiments, and care should be taken to ensure that all SOA experiments are done with the same lens pressure used in characterizing El vs. size. The trends of the response factor shown here may be significantly influenced, or may even be dominated, by differences in the particle size ranges analyzed by the ACSM and SMPS. It is not acceptable to publish a paper that attempts to quantify CE and RIE and the ACSM without a detailed and careful quantitative calibration of the transmission of the ACSM vs size, and applying that transmission curve to all SMPS measurements. Using transmission curves from the literature is not acceptable, as there is considerable variation in those, and it is not known which literature curve (if any) may apply to the ACSM used here. I would expect to see several figures and pages of text devoted to calibrating the AMS size transmission before any conclusions about CE or RIE could be credible.”*

Response: The E_L calibration procedure and results (Fig. S2) have been described in the supplementary information (SI Lines 32-51). A 100 μm critical orifice was used in front of ACSM, the pressure in the chamber was 1.22 ± 0.02 torr among all the experiments (Lines 157-158). When the pressure gets lower than 1.20 torr, the critical orifice would be cleaned. As a result, E_L should be relatively constant during the experiments. More details regarding E_L calibration has also been given in the response to comment #3.

No volume size distributions are shown here for the different experiments, so it is impossible for the reader to even estimate the extent to which differences in particle size transmission could be a problem here.

Response: The size distributions of particles during different experiments and the changes of size distribution within one experiments have been provided into the supplementary information (Figs. S4 and S6). The supplementary information also provided the detailed procedure of PM_{SMPS} mass concentration calculation.

As Reviewer 2 points out, the decreasing trend in RF vs f_{44} (Fig. 5a) could be simply due to an artifact of increasing particle size extending beyond the upper size limit of 100% aerodynamic lens transmission efficiency. While a well-performing lens can transmit particles with 100% efficiency up to ~550 nm vacuum aerodynamic diameter (~400-450 nm mobility diameter; Knote et al., 2011; Hu et al., 2017), lenses with reduced transmission at larger sizes is not uncommon (Liu et al., 2007; Takegawa et al., 2009; Hu et al., 2017; Campuzano-Jost et al. 2017), resulting in large losses for sizes as small as 450 nm vacuum aerodynamic diameter (~300-350 nm mobility diameter). Such effect may also play a role in the results presented in Fig. 6 and Fig. 7, as suggested by Rev. 2. Only then should the trends in RF can be considered reliable.

Response: According to our calibration, “Lens transmission efficiencies for 300-600 nm particle were nearly unity, which was in reasonable accordance with some previous studies (Jayne et al., 2000; Knote et al., 2011; Hu et al., 2018), but the diameter range with a unity lens transmission efficiency was much broader than that reported by Liu et al. (2007). The blue line in Figure S2 was used to modify size

distribution in our experiments (i.e., 0% transmission for d_{va} below 40 nm; linear increase in transmission vs $\log(d_{va})$, from 0% to 100% at $d_{va} = 100$ nm; 100% transmission efficiency from $d_{va} = 100$ nm up to $d_{va} = 580$ nm; linear decrease in transmission vs $\log(d_{va})$ from 100% at 580 nm to 0% at 1200 nm). For d_p below 300 nm, we used the results reported by Knote et al. (2011) indicated by the red dashed line in Figure S2. About 10% of uncertainty (shaded area) was applied to the E_L used in this study as indicated by the blue curve in Figure S2.” (SI Lines 42-51)

10. *“Collection Efficiency and Relative Ionization Efficiency Sections: The sections on CE (sect. 3.3) and RIE (sect. 3.4) are extremely under-supported, misleading, and highly speculative. Given the experiments conducted and methods used, separation of the effects of CE and RIE does not seem possible. This applies to both relative trends and absolute values. Those sections should be completely eliminated unless unambiguous new evidence can be provided. The RIE section provides no experimental evidence and only an incomplete discussion of the published literature on this topic. The CE section discusses experiments where SOA was coated on top of dry ammonium sulfate seed and the “Response Factor (RF)” was calculated. However interpretation of the results are flawed. The RIE of sulfate was not calibrated, even though it may vary substantially (e.g., Zhang et al., 2017). Despite the lack of sulfate calibration, the authors assume an RIE of sulfate and calculate a CE of ammonium sulfate that depends proportionally on that value. While it is stated that the absolute value is not important but rather just trends matter, in the same paragraph the argument that when a lot of SOA is added to the ammonium sulfate seed, a CE of unity is observed which supports that the aerosol is now in a liquid state. Not only does that line of logic require that ammonium sulfate is calibrated, it also requires that the RIE of the OA is known. So, generally the main figure of that section (Fig. 7) is very problematic since the RIE of SO₄ and OA are not known. Also, note that calculation of a CE for dry ammonium sulfate of 0.28 in this work does not narrow the range of uncertainty from lack of calibration for ammonium sulfate RIE since a range of a factor of 2 (0.2-0.4) has been observed for dry ammonium sulfate (Hu et al., 2017 and references therein). Moreover, equating the trends in the RF to changes in the RIE of the SOA is not necessarily justified since the effects may be the “results of a combination of a AS core and organic shell” as the authors state in Line 305. Thus, any trends in Fig. 7 may be dominated by the SOA coating thicknesses. However, changes in CE and RIE of the SOA may also be factors. Consequently, any quantitative conclusions that can be drawn about CE of OA is extremely under-constrained here. In order to separate RIE and CE effects, a direct, unambiguous measurement of CE needs to be made (e.g., see Xu et al., 2018).”*

Response: CE_{AS} was measured during the AS seed experiments. Our results were a little higher than the reported value. The manuscript has been revised as (Lines 268-277) : “ CE_{AS} for dry AS particles varied from 0.265 to 0.298 in different experiments, with an average value of 0.285 ± 0.067 which was slightly higher than the reported 0.24 ± 0.03 (Matthew et al., 2008). The large uncertainty mainly originated from SMPS measurements and the bias might come from the using of default RIE value. In the case of RIE_{SO_4} , a value of 1.15 was used here as suggested by previous studies (Canagaratna et al., 2007; Ng et al., 2011; Petit et al., 2015), which would not likely change during the organic coating experiments (Matthew et al., 2008). However, recent studies have shown that RIE_{SO_4} might vary with different instruments (Budisulistiorini et al., 2014; Crenn et al., 2015). For most of the experiments, all condensable organic vapor seemed to condense directly onto seed particles instead of forming new particles. While in one experiment (Exp. 22), there were small amounts of new particles formed, which was indicated by a smaller mode on the size distribution (Fig. S6) and further confirmed by a sudden increase in total particle number concentration. However, the newly formed particles only contribute slightly to the total particulate mass for their much smaller sizes. In addition, to avoid the influence of new particle formation on CE_{org} or RIE_{org} deduction, only PM_{ACSM} and PM_{SMPS} associated with AS signals were used to deduce the actual CE_{org} in our experiments”

11. *“Incomplete Literature Context / Major Omissions: Several instances stand out as having a major lack of literature review/support or simply omitting fundamental references on topics discussed. For*

example, the Kuwata et al. (AS&T 2012) paper which formulates the relationship between SOA density and O/C and H/C is never referenced or discussed (or any other paper on such relationships). A reader might get the impression that this manuscript was the first to show a correlation between SOA density and oxidation. Also, the paper Xu et al. (AS&T 2018), which extensively investigates RIE of OA for a large range of compounds and SOA (unambiguously separating CE and RIE), is completely missing. Another example is the section “Effects of f44” (3.2), where only a few peripheral references are provided in discussion of these results; whereas, there is a large body of work discussing trends in f’s and elemental ratios for SOA chamber studies. Even the papers introducing the f44 vs f43 diagram (Ng et al., 2010; 2011) are not referenced! Without such context, it is impossible to determine if anything new was learned from the present studies.”

Response: The related studies on SOA densities were referenced in lines 225-231 (George and Abbatt, 2010; Kuwata et al., 2012). The most recent studies about RIE_{org}/CE_{org} were cited in the revised manuscript (line 107 and lines 307-328) (Robinson et al., 2017; Xu et al., 2018). The papers introducing f_{44} vs f_{43} diagram was referenced in line 68 (Ng et al., 2010; Ng et al., 2011). A specific section (see 3.4 *Comparison with AMS Calibrations*) was added to compare with all the related works we know that was conducted by AMS. One of the strength of our calibration compared to the former calibrations was that we related CE_{org} variation with f_{44} to effective density variations, which possibly indicated a phase change. This was the first attempt to precisely evaluate the CE_{org} of a low-resolution Q-ACSM for various chamber-generated SOAs, which indicated the critical need for more complicated calibrations before deploying a Q-ACSM for long-term field measurements.

12. “f44, O/C, Oxidation State: The use of f44 is used throughout the manuscript to be equivalent to O/C and “oxidation state,” which is an unjustified step for chamber data. The authors state in Lines 200-203: “Since m/z 44 signal basically reflected the oxygen content in OA, the O/C ratio can be deduced directly from f44, both of which have been widely used to represent the oxidation state of OA (Canagaratna et al., 2015).” While this may be true for ambient OA, this does not necessarily apply to chamber data. Canagaratna et al. (2015) (Fig. 8a) shows that relationship for ambient, standards, and laboratory produced SOA (α-pinene, toluene, isoprene, sesquiterpenes). Ambient OA and some standards follow a well-established relationship. However, taken together the chamber SOA shows no correlation, which is not surprising since the presence of acids (which produce CO₂⁺ in the AMS/ACSM) and other oxidized functional groups would not necessarily be expected to track for specific oxidant/VOC systems. Only sesquiterpenes and α-pinene fall on the ambient parameterization line and are clustered within a narrow region.

Moreover, if the RIEs of OA indeed vary a lot for the systems studied then it would seem very possible that changes in f44 could be controlled as much by changes in the overall OA sensitivity (the denominator of $f_{44} = m/z\ 44 / OA$) as by the actual relative abundance of m/z 44 in the SOA. CO₂⁺ in the AMS is expected to be largely formed from the decarboxylation of organic acid groups on the vaporizer, forming CO₂ gas, which would have a constant RIE, and thus would not track changes in overall OA RIE.

Therefore, the use of the terms O/C and oxidation state should be avoided beyond an explanation that it might be roughly indicative of oxygen content and possibly of acid content.”

Response: As suggested by the reviewer, O/C and “oxidation state” have been replaced with the term “f44”.

13. “Clarity in Scope: All conclusions and interpretation should be scaled back to clearly state to apply to only RF (CE x RIE), effective density, f44 (not O/C or oxidation state), three oxidant/VOC SOA systems, dry conditions, and laboratory SOA. No extrapolation to ambient OA should be given or implied, since no evidence is presented and current evidence in literature suggests that such an extrapolation is not warranted – see e.g., Xu et al., AS&T 2018, Jimenez et al., AS&T 2016.”

Response: Extrapolation to HOA or ambient OA were removed from the revised manuscript.

14. *“Figure 8: This figure should be removed: no axis are given, and the data for several of the trends have not been derived in this paper, so this is really a cartoon from the author’s imagination, which has no place in a paper. If the authors do all the extra work to make this a serious study, then the figure should be included with quantitative axes and with error bars for both X and Y.*

In addition, the trend in density is not new and was published at least half a decade ago (Kuwata et al., 2012). The trends in CE and RIE of OA are not demonstrated in this paper and are especially misleading. The trends in RF may be dominated by lens transmission effects, and may have a contribution from the instrument that is being compared with, and can only be supported with further evidence. HOA is not studied here at all.

Also note that a published study (Pajunoja et al., 2016) reported the opposite trend for bounce vs. O/C, in that study bounce decreased as O/C increased.”

Response: Figure 8 has been removed.

15. *Other Miscellaneous Comments (substantial and minor):*

L75: Real time composition measurements is the key development that AMS made possible/routine (not sized-resolved).

Response: It has been revised as (Line 57): “real time size-resolved aerosol measurements”.

*L93-103: In this paragraph where CE and other effects on AMS sensitivity are introduced, it would be useful to instead use the more precise terms of each of the components of CE: lens, shape-related, and bounce effects ($CE = E_L * E_s * E_b$; Huffman et al., AS&T 2005).*

Response: These terms have been described as (Lines 88-93): “CE is mainly composed of three parts: E_L (the transmission efficiency of particles as a function of vacuum aerodynamic diameter (d_{va}) through the aerodynamic lens), E_s (to account for the disperse loss of non-spherical particles from the main air stream), E_b (to account for the bouncing of particles from the vaporizer surface). Among these, E_L can be explicitly taken into account by applying transmission efficiency in the aerodynamic lens, E_s is usually near unit for most particle types according to previous beam width measurements, and E_b is usually the most variable and uncertain part (Huffman et al., 2005; Salcedo et al., 2007).”

L101-103: This statement about the high uncertainty in CE and IE is unclear whether it applies to ambient or laboratory data and should be clarified (since it is not true for ambient data, per the Jimenez et al. 2016 reference cited).

Response: It has been revised as (Line 82): “laboratory generated SOA”.

L124-125. AMS light scattering does not employ laser-based vaporization.

Response: It has been changed into (Line 108): “light scattering-based detections”.

L126-130. This statement claiming that the increased fragmentation in the capture vaporizer will “highly complicate the AMS quantification process” does not reflect conclusions of the Hu et al., 2017 paper cited here, nor any of the several other papers investigating the capture vaporizer. For example Hu et al. (2018a) state that “tracer ion marker fractions, which are used to characterize the impact of different sources are still present and usable in the CV.” and Hu et al. (2018b) state “Consistent time series of positive matrix factorization (PMF) factors and their fractions of total OA were found across the CV and SV in the three very different ambient data sets ranging from biogenic- to anthropogenic-dominated, indicating limited loss of source determination information despite the increased fragmentation.” It appears that this

statement was added to dismiss this method as a practical way to reduce CE uncertainties. In fact, in the recent paper by Xu et al., AS&T, 2018, it was shown that the capture vaporizer can be used as a method to isolate changes in OA RIE. Given the other limitations of the ACSM, the authors may want to consider performing a similar study using a CV, which would make Eb~1 and thus substantially simplify the interpretation of the trends on RF.

Response: It has been revised as “Most recently, a new type of capture vaporizer has been developed to achieve a unit CE (Hu et al., 2017). RIE_{org} can be directly measured with this capture vaporizer (Xu et al., 2018). Nevertheless, the determination of RIE_{org} and CE_{org} appears to be a complicated but essential issue for accurate AMS/ACSM OA measurements that necessitate further investigations, including to explore new ways of SOA standard generation and to constrain the measurement uncertainty with various measurement techniques.” (Lines 110-114)

L162: “self-synthesized” is confusing. Consider alternative phrasing such as “synthesized in house”.

Response: The phrase “self-synthesized” has been deleted.

L177: “m/z calibration”, not “mass calibration”

Response: The phrase “mass calibration” has been replaced with “m/z calibration”. (Line 155)

L195: add “a” before “strong”

Response: It has been revised accordingly. (Line 199)

L235-238: “Especially, laboratory studies have shown that the ozonolysis of α -pinene can lead to considerably higher SOA yield than that from OH initiated reactions (Yao et al., 2014). Therefore, in this work we were focusing on the isoprene-OH and α -pinene-O₃ reactions only”.

This does not seem like a good reason to omit these data from the analysis. OH oxidation is an important loss for α -pinene in the atmosphere and makes substantial SOA. This data should be shown and discussed. Also, Yao et al. should not be the primary reference here.

Response: Both OH and O₃ initiated oxidations of α -pinene were included in the study and the reaction conditions were listed in Table 1. The results in Yao et al. (2014) only showed that for the same amount of α -pinene, ozonolysis reaction can lead to more SOA yield, which may not necessarily imply the actual relative importance of α -pinene oxidation channels under ambient conditions. These sentences have been removed.

L243: grammar.

Response: It has been revised as (Line 283): “For α -pinene initiated SOA,”

L291: grammar

Response: It has been revised as (Line 264): “Eight experiments using AS seeds were conducted in total (Table 2)”.

L293: grammar

Response: The sentence has been deleted.

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