

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 #2: *"This paper reports results from very difficult experiments aimed at understanding the sensitivity of the ACSM (Aerosol Chemical Speciation Monitor) instruments to organic material from aerosol particles. The topic of varying relative ionization efficiency for organic aerosol mass with the Aerosol Mass Spectrometer (AMS) has been debated in recent literature (Murphy, 2016a; Jimenez et al., 2016; Murphy, 2016b). Because the AMS has similar characteristics to the ACSM (aerodynamic aerosol focusing lens followed by thermal vaporization, electron impact ionization, and MS detection), most of the results described in this paper are potentially applicable to the AMS.*

The findings of this paper are fascinating and the reported changes in SOA "sensitivity" are qualitatively consistent with many previous observations. A major strength of this work is that surrogates for ambient secondary organic aerosols (SOA) were produced and important details on the chamber experiments are not needed to demonstrate the overall method for accurately measuring the SOA response factors for the ACSM. Unfortunately, this work does not provide any new, useful and quantitative information for the ACSM/AMS community and it is possible that the data from this work are unable to constrain the largest uncertainties for a broader application of these results. To make a significant contribution in the field, the work presented here should show all of the relevant calibration details and significantly reduce the largest uncertainties in the reported measurements. At a minimum, the manuscript must show that the methods work for aerosols with known chemical composition (ammonium nitrate and ammonium sulfate) and the uncertainties must be fully propagated. The paper has several major flaws that need to be addressed in revising this manuscript to make it acceptable for publication in AMT."

Response: We agree with the reviewer that calibration of a Q-ACSM or an AMS is a very difficult job and we also believe AMS/ACSM is a very powerful instrument that has made great contribution to the aerosol researches. Therefore, the accuracy of AMS/ACSM measurements is a critical factor that deserves extensive studies using various methodologies. In this work, SOA were generated inside a Teflon chamber from different VOC precursors under atmospheric relevant conditions. The volume concentration and effective density of chamber-generated SOAs were quantified by SMPS and APM, respectively. The Q-ACSM, SMPS, and APM were well calibrated before or during our experiments with ammonium nitrate, ammonium sulfate, or PSL spheres. The calibration procedures and results were provided in more details in the revised manuscript and the supplementary information. In addition, we conducted some additional calibration work on Q-ACSM (the transmission efficiency of particles in the aerodynamic lens) as suggested by the reviewer. With all these efforts, we were able to present a comprehensive evaluation of the measurement uncertainties. Our results strongly indicate that measurements of OA with various f_{44} 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. Currently, the uncertainties of OA measurement ($2\sigma = \pm 38\%$) is still quite high. Although the results of this work may not be directly applicable to ambient Q-ACSM measurements, it is reasonable to suggest that similar comprehensive calibrations of Q-ACSM for complicated ambient conditions be done in the future to further constrain the uncertainties of ambient OA measurement.

Specific comments:

Writing and Presentation of Key Concepts

1. *"This paper was very difficult to read due to numerous grammatical errors and informal usage of important terminology (e.g. response factor or RF, relative ionization efficiency or RIE, collection efficiency or CE, and oxidation state). Broad claims of "accuracy," interpretation of the results, and the relevance of this work are not supported by the actual evidence presented. The manuscript would significantly benefit from more careful technical writing and editing."*

Response: As suggested by the reviewer, we have invited experienced native English speaker to fully revise the manuscript for grammatical errors. All the AMS related terminologies have been clearly defined in the revised manuscript. The definitions of “Collection Efficiency (CE)”, “Ionization Efficiency (IE)”, “Relative Ionization Efficiency (RIE)” and “Response Factor (RF)” were all described in the introduction section. All the obscure expressions, like “accuracy” and “sensitivity”, were replaced with “RF” or “Relative Response Factor (RRF)”, the latter was a new term introduced by the authors to quantitatively express the difference between Q-ACSM and SMPS-APM measurement. It has been discussed in lines 177-182: “To elucidate how Q-ACSM measured mass concentration (PM_{ACSM} , using default $RIE_{org} = 1.4$ and $CE_{org} = 0.5$) may deviate from the true mass concentration for particles entered Q-ACSM (PM_{SMPS} , the product of E_L and E_S -amended SMPS-measured volume concentration and APM-measured ρ_{eff}), relative response factor (RRF) is defined as the ratio of PM_{ACSM} to PM_{SMPS} .

$$RRF = \frac{PM_{ACSM}(\text{assuming default } RIE_{org}=1.4, CE_{org}=0.5)}{PM_{SMPS}(\text{amended by } \rho_{eff}, E_S, E_L)} = \frac{RIE_{org} \cdot CE_{org}}{1.4 \cdot 0.5} \quad (2)$$

In addition, the conclusion of this work was reevaluated and has been constrained to the laboratory-generated SOAs only. The original Figure 8 has been removed to avoid confusion.

2. *“The primary subject of this study, the response factor (RF) for the ASCM, should be clearly defined at the beginning of the manuscript with Equation 1 moved up to the end of the paragraph on the AMS/ACSM quantification (page 5 line 103). This equation shows that both RIE and CE are important factors in determining the RF, therefore any variation in RF could be due to either one or both of these factors changing. While this was described later in the manuscript (Sections 3.3 Effects of CE and 3.4 Effects of RIE), these concepts are key to understanding and interpreting the reported RFs from these studies. The abstract should state that the RF values reported here were obtained using the default RIE for organic species (1.4) along with a CE of 0.5.”*

Response: As suggested by the reviewer, the original equation together with the definition of RF and its relationship with CE and RIE have been moved to the introduction (Line 84). When introducing the calculating method of RRF (section 2.4), it was stated clearly that “any non-unit value of RRF might be caused by the deviation of RIE_{org} or CE_{org} from the default values” (Lines 180-181). It has been clearly stated in the abstract and throughout the manuscript that the default RIE (1.4) and CE (0.5) were used to calculate the RRF in this work.

3. *“Accurately quantifying RIE and CE factors is crucial in determining the AMS/ACSM sensitivity to aerosol components and they need to be studied independently in order to generally apply the findings of this work. The work made an attempt at separating these factors, but did not adequately examine them for the analysis. The current debate in the literature is about RIE for organic aerosol (Murphy, 2016a, b; Jimenez et al., 2016) and the present work appears to contradict itself in the findings on RIE. This is probably due to careless usage of the terms “RIE” and “RF,” as well as not incorporating the CE findings into calculations for the RIEs from the measured RFs. A thorough examination of the writing of the paper would presumably clarify the inconsistencies and demonstrate if the RIE varied significantly or not for these measurements.”*

Response: As suggested by the reviewer, we have clarified the definitions of RRF and RIE_{org} (Lines 177-182). Although we have attempted to separate the effects of CE_{org} and RIE_{org} , RIE_{org} cannot be quantitatively determined because of the relatively large uncertainties and different f_{44} ranges generated between experiments with and without AS seeds. Based on additional calibrations, uncertainties associated with each instrument has been well quantified. For example, we have performed extra experiments to calibrate

the ACSM lens transmission efficiency (E_L) and the calibrated E_L was used to amend the PM_{SMPS} . We have re-evaluated the uncertainties in SMPS and APM measurements. The SOA coating thickness for the AS seed particles has also been precisely determined. We clearly redefined “relative stable states” in each experiment. Based on these data analysis, we found that the variation of RRF as a function of f_{44} can be largely explained by the CE_{org} variation, which was possibly due to particle phase changes as indicated by the effective density variation. Nevertheless, we cannot exclude the possible influence of RIE_{org} . The manuscript has also been thoroughly revised.

4. *“The last sentence in the abstract states “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.” In addition to the obvious mistakes in the grammar and use of terminology, this statement implies that previous measurements from the ACSM/AMS are incorrectly reporting OA mass concentrations. This statement needs to be supported with clear evidence of such problems. The results here, while not directly showing it, imply that the variations in the response factor can be largely explained by variations in CE rather than variations in RIE. Many studies have been using a higher CE for HOA that accounts for discrepancies between independent measurements and the AMS, for example the 2002 Pittsburgh study (Zhang et al., 2005) and subsequent work. How do the authors reconcile their results with those showing numerous, good correlations of ambient ACSM/AMS data with independent organic carbon aerosol or total mass measurements?”*

Response: Since HOA was not studied in this work, the last sentence in the original abstract has been removed to avoid overstating the conclusion of this work. After conducting more instrument calibrations, uncertainty evaluation, and data analysis work, we concluded that 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. Our results of RRF and CE_{org} calibration were generally in line with previous studies conducted with the AMS in laboratory experiments. This was not in conflict with the various ambient measurements because organics are usually internally mixed with inorganics in ambient conditions and CE of the bulk aerosol can still keep around 0.5. However, the uncertainties for ambient OA measurements is still quite high ($\pm 38\%$), which have the potential to be constrained by applying more accurate RIE_{org} values according to f_{44} . It was a pity that we cannot accurately quantify RIE_{org} with our experiment, which was largely attribute to the low resolution of Q-ACSM and the large uncertainties in SMPS volume measurements. However, it is highly suggested that more comprehensive calibrations of RIE_{org} on AMS/ACSM should be done in both laboratory and ambient conditions to further lower the uncertainties of OA measurement.

5. *“Accuracy and Uncertainties for the Number Distributions and Effective Density Measurements:*

In order to calculate the SOA mass concentrations being generated in the chamber, the measured number distributions as a function of particle size need to be converted into mass distributions using the particle density. Determining the overall uncertainty in the mass concentrations from the number distribution and the particle density is important. Here, the number distributions were measured with a scanning mobility particle analyzer (SMPS). Information about calibrating this instrument was not given in the paper, so it is unclear what the accuracy is for these measurements. Also, how well did the two number distributions match between the SMPS and the differential mobility analyzer (DMA) plus condensation particle counter (CPC) systems? A simple statement about using polystyrene latex spheres to calibrate both systems would provide additional confidence in these measurements.

Knowing the accuracy and uncertainty in the particle effective density measurements is also necessary. The

particle effective density was determined by selecting the mobility diameter at the peak in the SMPS number distribution with the DMA and the peak in the mass per particle was scanned with an aerosol particle mass (APM) analyzer with a CPC. The literature reports using a similar DMA/APM/CPC system can provide accurate effective density measurements with 95% confidence intervals of 10-30%, depending on the configuration of the system (Johnson et al., 2013). The accuracy of the system used for this study was not presented as additional calibration information. For example, how accurate and precise are the effective densities for ammonium nitrate and ammonium sulfate (or other known particles)? The demonstrated uncertainties in the effective density measurements need to be propagated to the rest of the results.

The peak in the number distribution is not the same as the peak in the mass distribution. Were additional sizes of mobility diameter scanned (not just the peak in the number distribution) to ensure that the effective density was constant across all sizes in the distribution? If not, it should be clearly stated that it was assumed that the measured effective density was the same for the entire distribution. ”

Response: As suggested by this reviewer and reviewer #4, we have provided more details of calibration methods and results of the SMPS and APM system in the manuscript (Lines 170-175): “The flow rates and the voltages applied to the DMAs were well calibrated before the experiments. Three sizes of polystyrene latex spheres (PSL, 81±3nm, 147±3nm, 269nm±5nm, 1.05 g cm⁻³) were aerosolized and dried with a collision atomizer (TSI, Model 3076) followed 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 ±4% and ±3%, respectively.” Details of the calibrations can be found in the SI section 1 and 2.”

When using APM to measure the effective density of AS seeds, “ ρ_{eff} of the dry 80 nm AS particles was measured to be 1.64 ± 0.08 g cm⁻³, which was consistent with 1.65 g cm⁻³ (Zelenyuk et al., 2006) or slightly lower than 1.66-1.70 g cm⁻³ measured for 200 nm (d_m) dry AS particles in a previous study (Matthew et al., 2008)” (Lines 246-249). However, as the bias were within ±3%, it was within the evaluated uncertainties of our results.

In this work, we consider the chamber-generated SOA were mostly unimodal distributed (as shown in Fig. S4). 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 the measured effective density was the same for the entire distribution, especially for the AS seeded experiments, when only a single mode of AS particles was present in the chamber. The following discussion has been given in the manuscript (Line 184) and the supplementary information (Line 71):

“

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

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 $1 \pm 5\%$. 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 $d_{m,i} (\pm 4\%)$, $dN_i (\pm 10\%$ for 20~200 nm and $\pm 20\%$ for 200~800 nm particles) (Wiedensohler et al., 2012; Wiedensohler et al., 2018), $\rho_{eff} (\pm 3\%)$, $E_L (\pm 10\%)$ and $E_s (\pm 5\%)$ 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 supplementary information.”

“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_{diff}) (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:

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

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):

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

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).”

6. “Uncertainties in Collection Efficiency:

One of the largest issues (uncertainties) in ACSM/AMS measurements is the collection efficiency (CE). In particular, the CE factor includes lens transmission and focusing in addition to particle bounce (Canagaratna et al., 2007; Huffman et al., 2005). Lens transmission (Liu et al., 1995a, 1995b) can often significantly affect the overall (i.e., observed) CE and it was neglected in this work. Furthermore, lens transmission can vary for nominally identical lenses (Bahreini et al., 2008) and the actual lens transmission for the instrument used in this study needs to be quantified. To fairly compare an external measurement of mass with the signals from an ACSM (or AMS), a correction to the volume distribution needs to be applied that accounts for particle losses in the lens. If the actual lens transmission cannot be determined, the theoretical lens transmission must be used to determine if the ACSM lens is transmitting the majority of the mass measured by the size distribution instrumentation.

For the data presented in Figure 2, the overlap between the measured number distribution when the system was stable after 5 hours and the lens transmission might possibly be close to 100%. Thus, the effect of lens transmission losses could be minimal for that particular experiment. Figure 3 shows that the measured signals from the ACSM using the default RIE of 1.4 and CE=0.5 agree very well with the SMPS mass measurements using an effective density of 1.2; the bias is +22% which is probably within combined experimental uncertainties. For the coatings experiments, it is possible that particles grew too large to be efficiently transmitted through the lens or that the increase in sulfate mass concentration was due to smaller particles becoming more efficiently transmitted. A reader cannot estimate lens transmission losses for these and the other experiments because the mass distributions are not shown in the manuscript (or supplemental information). Therefore, the lens transmission needs to be examined for all of the experiments to ensure that the mass sampled by the AMS is the same as the mass measured with the SPMS system.

The CE for dry, ammonium sulfate particles was used as a basis for the CE results shown in Table 2 and Figures 6 and 7. Therefore, uncertainties in the CE for dry ammonium sulfate would affect the derived CE for the organic coatings. How was CE = 0.28 determined for dry ammonium sulfate? Prior work indicates that the CE for this species is 0.24±3% (Matthew et al., 2008). How stable is the ammonium sulfate seed source? The manuscript states that a “similar amount and size” of seed particles were used in the experiments, and the variability is a little more than 10% by averaging the mass of uncoated seeds in Table 2. Figure 6 shows that the sulfate mass concentrations are at least this variable (and possibly increasing) prior to (and shortly after) injection of the VOC precursors. Toward the end of the experiment, the sulfate mass concentrations appear to be decreasing while the organic mass loading increased more slowly than the sulfate at the middle of the experiment and seemed more stable than the sulfate at the end. (The data in Figure 6 do not appear to correspond with any of the experiments shown in Table 2.) Loss rates in the chamber would affect the interpretation of the changes in mass concentrations over the time of the coatings experiments. All of these factors contribute to uncertainties in the CE derived from these experiments, which should be added to the manuscript.”

Response: As suggested by the reviewer, the lens transmission efficient (E_L) of the Q-ACSM was calibrated using the same setup as in Figure S1 and the details were given in the supplementary information (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.

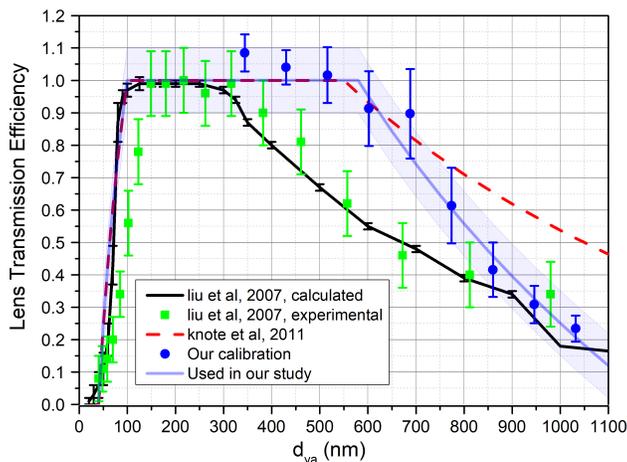


Figure S2. Lens transmission efficiency as a function of particle vacuum aerodynamic diameter (d_{va}).

Lens transmission efficiencies for 300~600nm 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 between 100 nm and 580 nm; linear decrease in transmission vs $\log(d_{va})$ from 100% at 580 nm to 0% at 1200 nm). For E_L 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.”

For the experiments without SA seed particles, a small portion of smaller particles (<100 nm) were lost in the lens. This has been discussed in SI: “For most of the experiments, more than 94% of the total particle mass was transported into ACSM through aerodynamic lens. While for three of them (Exp. 4, 5, 7), a portion of particles were too small to pass the lens, only 87-89% of the total mass was transported” (Fig. S4 caption). While For the experiments with SA seed particles, more than 95% particles transported through the lens. The statement is “For all the experiments, more than 95% particles were transported through the lens” (Fig. S6 caption)

AS seed particles were generated by an atomizer (Model 3076, TSI) with AS solution. The generated AS seed particle size distributions were very stable as the geometric mean diameter for AS in all the experiments were ~ 80 nm. The mass concentration variation is due to the variation in total number concentration, which is somehow difficult to control by injection time. However, this number concentration was not observed to affect our results.

7. *“Volatility Effects:*

One potential issue that should be addressed is the possibility of the SOA being partially volatile. Particles with low f_{44} (such as hydrocarbon-like organic aerosol or HOA) are known to be the most volatile SOA species (Paciga et al., 2016; Huffman et al., 2009). Depending on how they are operated, the SMPS and DMA/APM/CPC systems could cause evaporative losses. There should be a brief discussion on how this might affect some of the results. How might variations in volatility affect the background (filter) subtraction in these ACSM measurements? Did the background levels change as a function of f_{44} ?”

Response: Volatile HOA particles with low f_{44} values may cause higher RRF. However, during our experiments of low f_{44} , most oxidation products may remain in the gas phase and we did not observe the background signal of the Q-ACSM varying regularly with f_{44} , as the background signals were more related with the particle concentration. Hence, we did not expect volatility would affect our experiments.

8. *“Technical Corrections:*

Page 1 Lines 1-2 (Title): It is important to distinguish the term “sensitivity” from the “response factor,” here and throughout the manuscript, where the “response factor” was measured and the “sensitivity” was inferred. In addition, “different oxidation states” were not measured and should be replaced in the title with “varying average oxygen content” or similar phrasing. “Oxidation state” should be replaced throughout the manuscript with more appropriate phases, such as “ f_{44} ” or “average oxygen content” depending on the context.

Response: Throughout the manuscript the term “sensitivity” has been replaced by “RF” or “RRF”. As suggested by the reviewer, the term “ f_{44} ” or “average oxygen content” were used throughout the revised manuscript to replace the “oxidation state”.

9. *“Page 1 Lines 20-21 (Abstract): This statement is not true as written. Should delete or replace the word “comprehensively.”*

Response: The sentence has been revised as: “The response factor (RF) of a quadrupole based aerosol chemical speciation monitor (Q-ACSM) for secondary organic aerosols (SOA) has been investigated in this work.”

10. *“Page 1 Line 21 (Abstract): revise “SOA samples were generated ...” to “SOA was generated . . .”*

Response: It has been revised as: “SOA were generated under simulated photochemical oxidation conditions in a 4.5 m³ Teflon chamber from three different volatile organic compounds (VOC)”.

11. *“Page 1 Line 23 (Abstract): The “dozens of ppbv” concentrations of the precursor VOCs used in this study are not typical. Suggest deleting/revising the phrase “atmospheric relevant concentrations” at this location as well as other places in the manuscript.”*

Response: To avoid confusion, the phrase has been deleted in the manuscript.

12. *“Page 2 Lines 24- 25 (Abstract): The sentence starting with “Different SOA oxidation states were achieved by . . .” should be re-written to something along the lines of “For some experiments, varying degrees of average oxygen content in the SOA were obtained by ...”*

Response: The sentence has been revised as: “For some experiments, different degrees of average oxygen content of SOA, indicated by the ratio of m/z 44 signal over total organic ion signal intensity (f_{44}), were obtained by changing the ratio of the VOC precursors to the oxidants (O_3 or OH).”

13. *“Page 2 Lines 26-28 (Abstract): Needs to be revised – circular argument about the “exact mass” “used to deduce” the “effective density” and “mass concentration.”*

Response: The sentence has been revised as: “An aerosol particle mass analyzer (APM) and a scanning mobility particle sizer (SMPS) were used to determine SOA effective density (ρ_{eff}) and volume concentrations.”

14. *“Page 2 Line 29 (Abstract): The sentence starting here needs to be revised. Several parts of it are not clear and not valid. It indicates that “considerable errors” in the SMPS calibration occurred from variations in the effective density, but the 23% difference between the reported effective densities do not correspond to the factor of 3 difference in the measured response factors (from about 0.75 to about 2.15), which appeared to be corrected for varying effective density.”*

Response: The sentence has been revised as: “Relative response factor (RRF) of the Q-ACSM to organics was determined by dividing Q-ACSM measured mass (the relative ionization efficiency (RIE_{org}) and organic collection efficiency (CE_{org}) were set to default values of 1.4 and 0.5, respectively) by SMPS-APM deduced mass (amended by aerodynamic lens transmission efficiency (E_L) and particle loss due to non-spherical shape (E_s)).”

15. *“Page 2 Line 31 (Abstract): The term “can change substantially” should be replaced with the actual values with uncertainties. The measurements indicate that effective density changes from 1.1 to 1.35, but the accuracy of the method was not demonstrated to interpret the significance of the change.”*

Response: The sentence has been revised as: “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.”

16. *“Page 2 Lines 31-33 (Abstract): Should replace “sensitivity” here and elsewhere with “response factor.” This statement and the title of the manuscript are similarly imprecise, because the paper reports changes in the RF as a function of f_{44} , which is a measure of the average oxygen content in the total organic aerosol mass and not the “oxidation state.” A problem with this sentence and following statements is that the values for the response factors and the RIE/CE combination used to obtain them were not stated specifically in the abstract.”*

Response: The terms “sensitivity” and “oxidation state” have been replaced with “response factor” and “ f_{44} or oxygen content” throughout the manuscript.

17. *“Page 2 Line 33-34 (Abstract): This statement should be revised because it implies that RIE and/or CE should be decreased relative to the values used (1.4 and 0.5, respectively) as the oxygen content increases.”*

Response: It has been revised as: “The anti-correlation between RRF and f_{44} might be due to the decreased RIE_{org} or CE_{org} for more oxidized SOA.”

18. *“Page 2 Lines 34-37 (Abstract): Sentence has grammatical and technical errors and needs to be rewritten. There is no context for changes in sulfate signals.”*

Response: The sentence has been revised as: “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.”

19. *“Page 2 Lines 37-39 (Abstract): More grammatical and technical errors in this sentence. There is no evidence in this paper of transforming SOA “gradually” from a liquid state to a solid (or glassy) state. This work does show that the CE values vary depending on the average oxygen content as indicated by f_{44} , and the statement should be revised accordingly. However, the CE values for the solid (or glassy) state are reported here as “0.2-0.5,” whereas the measured values in the paper show that CE is above 0.3. Knowing the CE values precisely is an important part of reducing the uncertainties in the response factors, since CEs of 0.2, 0.3, and 0.5 represent changes in RF by factors of 5, 3, and 2, respectively.”*

Response: The sentence has been revised as: “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.”

20. *“Page 2 Lines 39-41 (Abstract): Again, there is no evidence in these observations of SOA transformations occurring as stated (here from hydrocarbon-like OA to more oxygenated OA). Furthermore, the changes in CE as a function of f_{44} are consistent with the changes in the response factor without a varying RIE. Specifically, if $CE=1$ instead of 0.5 for the lowest f_{44} values, the response factor would decrease from 2.15 to 1.1 and if $CE=0.3$ instead of 0.5 for the highest f_{44} , the response factor would increase from 0.75 to 1.25. Both revised values for the response factors (1.1 and 1.25) are well within the uncertainties of ACSM measurements without changing the RIE significantly from 1.4.”*

Response: The sentence has been revised as: “The large uncertainties of CE_{org} was principally caused by the uncertainty in SMPS measurement. 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.”

21. *“Page 2 Lines 41-44 (Abstract): More grammatical and technical errors in this sentence. It appears that the term “RIE” was used here where the term “response factor” would be more accurate. The claims of “underestimation” and “overestimation” need to be quantified to be meaningful. The variations in the response factors appear to be consistent with the previously reported changes in CE as a function of average oxygen content for chamber-generated SOA (Docherty et al., 2013) and the 38% uncertainty in AMS organic mass concentration (Bahreini et al., 2009).”*

Response: The statements containing OOA or HOA have been removed. The sentence has been revised as: “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 chamber studies and the necessity to calibrate RF in laboratory experiments.”

22. *“Other Corrections:*

There are other numerous grammatical errors in the rest of the manuscript, not specified here. Instead,

noticeable technical errors are highlighted below:

Page 3 Line 57: add the word “air” or “aerosols”

Page 4 Line 92: The citation for Zhou et al., 2016 does not seem appropriate for the “widely used . . . around the world” context.

Page 5 Lines 119-121: There are many examples of ambient datasets showing the AMS mass concentrations agree well with other observations (Jimenez et al., 2016). This should be mentioned here.

Page 6 Line 130: This is a good place to cite the recent ACSM characterization paper (Xu et al., 2017).

Page 7 Line 169: mention the inner diameter instead of the “OD”

Response: The manuscript has been revised by a native English speaker for grammatical errors. All errors above have been revised accordingly.

23. *“Page 9 Lines 207-208: f_{44} could also be artificially low when particles initially form if the ion signals from large m/z are not properly included in the total organic mass (and dividing the high S/N peak of m/z 44 with an artificially lower total organic signal). This issue could be checked for the first three data points in Figure 2 by comparing relatively high m/z ion peak intensities in their spectra with spectra from higher aerosol mass concentrations.”*

Response: The sentence has been revised as: “The initial high values of f_{44} could partially due to f_{44} calculation bias in low concentration ($0\text{-}5\ \mu\text{g m}^{-3}$) or the fact that initially formed SOA particles were highly oxidized because of gas to particle partitioning (Shilling et al., 2009).” (Lines 201-203)

24. *“Page 12 Lines 284-285: Does “0.28 in this work” refer to the Matthew et al. study (Matthew et al., 2008) or the work for the manuscript? The Matthew et al. paper states a lower CE with uncertainties assigned to it ($24\pm 3\%$), making the 0.28 value outside the uncertainty band.”*

Response: The value of 0.285 ± 0.067 was measured during the AS seed experiments by comparing the AS signals with SMPS measurements without organic coating. The CE was slightly higher than that reported by Matthew et al. (2008) (0.24 ± 0.03), which may be caused by the uncertainties of SMPS-APM measurement or the bias of RIE_{SO_4} from the default RIE_{SO_4} (1.15).

25. *“Page 12 Lines 286-290: The Matthew et al. work showed that the number of sulfate ions per particle did not change with additional water or organic coating, so it should be cited here that the RIE for sulfate does not change during the coating experiment. This point is important because any changes in the sulfate mass loadings during the course of the experiment are not because the RIE for sulfate changed.”*

Response: This sentence has been revised as: “A default RIE_{SO_4} (1.15) was used here as suggested by previous studies (Canagaratna et al., 2007; Ng et al., 2011; Petit et al., 2015). As RIE_{SO_4} would not change during the organic coating experiments, any changes in sulfate signal was due to variations in CE_{AS} (Matthew et al., 2008).” (Lines 239-242)

26. *“Page 12 Line 292 and following: the “coating thickness” should be calculated using the size distribution data and included in Table 2. The size distribution data would also indicate other changes, like*

newly formed SOA particles for the “extreme condition” experiment.”

Response: The coating thickness has been calculated and included in Table 2. The size distributions were also included in the supplementary information (Figs. S4 and S5): “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 contributed 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.” (Lines 271-277)

27. *“Page 13 Lines 302-305: Both sentences appear to say that the light blue points are AS cores with an organic shell, but the connecting word is “however”. Need to revise to clarify.”*

Response: Both sentences have been removed from the manuscript. The color represented the coating thickness in the current manuscript, instead of Org/SO₄ ratio in the original manuscript. SOA layer of 18-25 nm can fully cover the AS seed particles.

28. *“Page 13 Lines 314-315: “SOA may transit from liquid phase . . .” Need to revise/check grammar.”*

Response: This statement has been moved to lines 340-342 and revised as: “Decreasing CE_{org} with f_{44} might be explained by the phase transition from liquid-state into more solid-like/glassy state of SOA particles consisting of more oxidized organic compounds that normally assume a higher ρ_{eff} .”

29. *“Page 13 Lines 315-316: Higher oxidation state and higher effective density are not necessarily linked to changing phase states if the composition is also changing. Need to revise.”*

Response: The statement has been revised as: “Decreasing CE_{org} with f_{44} might be explained by the phase transition from liquid-state into more solid-like/glassy state of SOA particles consisting of more oxidized organic compounds that normally assume a higher ρ_{eff} .”

30. *“Page 14 Line 323: To complete the section on CE, studies on CE of HOA and aged ambient OA could be added/mentioned, such as (Slowik et al., 2004; Zhang et al., 2005; Middlebrook et al., 2012).”*

Response: Former studies on CE_{org} was discussed briefly in the introduction as: “Although organics quantified using a CE_{org} of 0.5 has been found correlating well with independent organic carbon (OC) or VOC measurements in most field works (Allan et al., 2004; de Gouw et al., 2005; Takegawa et al., 2005; Venkatachari et al., 2006; Middlebrook et al., 2012), studies have shown that CE_{org} can vary substantially for various chamber generated organic particles (Bahreini et al., 2005; Docherty et al., 2013).” (Lines 96-100).

Detailed inter-comparison between our results and former CE_{org} calibrations has also been discussed in detail in lines 303-317 as “In previous studies conducted by AMS, 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 (Alfarra, 2004; Matthew et al., 2008; Docherty et al., 2013; Robinson et al., 2015). However, during field studies AMS measurement results based on default $RIE = 1.14$ and $CE = 0.5$ appeared to be in reasonable agreement with other aerosol measurement techniques (Slowik et

al., 2004; Zhang et al., 2005; Middlebrook et al., 2012). 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 (Chen et al., 2009; Allan et al., 2014). 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 (Vaden et al., 2010; Virtanen et al., 2010; Shiraiwa et al., 2011), which might lead to lower CE_{org} . For example, CE_{org} of liquid squalane ($CE = 1$) decreased after SOA condensed on the particle (Robinson et al., 2015). Most recent CE_{org} calibration with light-scattering single-particle (LSSP) module also suggested that CE_{org} (0.25-0.4) 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)."

31. *"Page 14 Lines 336-337: While this statement may be true, the section on CE as a function of f_{44} seems to indicate that the large changes in RRF are mostly due to changes in CE. Once the CE is properly taken into account for the RRF, RIE for the organics studied here does not seem to be varying by as much as the studies cited in the previous sentence. Consider adding more statements at the end of this section."*

Response: After conducting additional calibrations, the uncertainty of measured CE_{org} was better constrained. Accordingly, we revised the statement as: "With our experiments, we still cannot rule out the contribution of RIE_{org} to the overall RRF trend even though the variation of CE_{org} with f_{44} could largely explain the observed RRF changes." (Lines 297-298)

32. *"Table 1: Add errors, mainly to PM, density, f_{44} , and ACSM/SMPS columns."*

Response: Errors have been inserted into Table 1.

33. *"Table 2: Digits listed for ammonium sulfate, Org, f_{44} , and CE are not significant. Revise and add errors to these columns. Add one more column (with errors) for the calculated thickness of organic coating, based on size distribution data and derived effective density."*

Response: Error bars and coating thickness have been inserted into Table 2.

34. *"Figure 1: The air in the chamber (how generated and introduced) is not shown. Is the chamber filled like a balloon or is it a flowing reactor? How is air pumped out of the chamber? Is air added to the chamber while the instruments are sampling? It is unclear how the ammonium sulfate seeds are dried and added to the chamber. The figure caption needs a few more details explaining a typical experiment."*

Response: Air inside the chamber was generated with a zero-air generator (Aadco 737, USA). The chamber was basically a flexible Teflon bag. When inflated the chamber was assuming a cubic shape with a total volume of 4.5 m^3 and the inside pressure was equal to the ambient pressure. During the experiment, the chamber zero-air inlet was sealed and air was sucked out by the instruments. Total of $\sim 0.7 \text{ m}^3$ air was consumed for each experiment. At the end, the chamber collapsed a little but would still assume a cubic shape and hold the same pressure as the room pressure. The description of the chamber operation was given in lines 134-139.

35. *"Figure 2: This is the only figure with number distributions. They should be converted into mass*

distributions, the effect of the ACSM lens transmission should be shown (calculated transmission into the ACSM), and additional mass distributions should be added to supplementary information. Should add the “start time” for the experiment, along with an arrow indicating when the system was considered to be in “relative steady-state.” Add that the AMS mass concentrations were calculated using the default RIE=1.4 and CE=0.5. Also include in supplementary information how the system was calibrated for the ionization efficiency of ammonium nitrate and how accurately the ammonium nitrate effective density was measured using this method.”

Response: Average number size distributions during “relative stable state” in all the experiments and the measured lens transmission efficiency have been given in the supplementary information (Figs. S4 and S6). Arrows were added to indicate the start time and the “relative stable state”. The default values of RIE = 1.4 and CE = 0.5 were added on Fig. 2. Details of Q-ACSM calibration with ammonium nitrate have been given in the supplementary information (section 1-2).

36. *“Figure 3: Should state in the caption what value of effective density was applied to the SMPS mass concentrations. Also add the default RIE=1.4 and CE=0.5 was used for the ACSM mass concentrations. Note that a bias of 22% (slope here) is within the +/-38% uncertainties of the AMS measurements (Bahreini et al., 2009).”*

Response: The original Fig. 3 has been removed. New Fig. 3 includes both the correlation between RRF vs f_{44} and effective density vs f_{44} . The error bars in new Fig. 3 were based on all data points obtained during all experiments listed in Table 1. The $\pm 38\%$ measurement uncertainty reported by Bahreini et al. (2009) has been indicated by the shaded area in new Fig. 3a.

37. *“Figure 4: This could be moved to the supplementary information, since it is not directly relevant to the discussion.”*

Response: Figure 4 has been moved into the supplementary information (Fig. S7).

38. *“Figure 5: The conclusions of this paper about the response factor (and effective density) varying with f_{44} rely mainly on a few points affecting the trends (linear relationships). Errors need to be propagated to provide strength to this observation. There is a cluster of data points near $f_{44}=0.14$, that have average effective densities of 1.25 ± 0.05 and response factors of 1.25 ± 0.3 . These values are within the combined uncertainties. Toluene is listed twice in the effective density (bottom plot) legend – green triangles should be isoprene. Add to the caption the default RIE=1.4 and CE=0.5 were used for the calculations of the ACSM mass concentrations.”*

Response: Figure 3 is the original Fig. 5. Figure 3a shows that RRF for the chamber-generated SOA decreased from 2.16 to 0.81 with f_{44} increasing from 0.057 to 0.191. Fig. 3b shows that ρ_{eff} increased from 1.09 to 1.34 g cm⁻³ with increasing f_{44} . After comprehensively evaluating the uncertainties, the error bars in both Fig. 3a and Fig. 3b increased a lot. However, RRF for the two isoprene derived SOA points were still outside of the range of Q-ACSM measurement uncertainty (the gray area). The anti-correlation between RRF and f_{44} was still very clear. Moreover, the overall positive deviations of RRF could possibly be explained by a higher than default RIE_{org} value (1.6 ± 0.5), as suggested by Xu et al. (2018). (Lines 327-330)

39. *“Figure 6: The experiment shown here is described in the text but does not appear to be listed in Table 2. As a second panel, the mass distributions should be shown. Why are the sulfate concentrations not “zero”*

before the seeds were added? What VOC precursor was added? When were the lights turned on? What are the various organic coating thicknesses estimated as the organic mass increases for a relatively constant sulfate mass? This experiment possibly indicates that the CE changed slightly when the SOA began to be detected, then did not change (within some uncertainty band) as the sulfate particles become coated with more organic material. Unfortunately, the sulfate mass concentrations do not appear to be stable enough to state this conclusively. Were there other experiments where the sulfate concentrations were more stable before adding the VOC and turning on the lights? See other comments on the discussion of this figure on the lack of stability in the sulfate concentration during the course of the experiment and changes to the mass distributions potentially affecting lens transmission.”

Response: All the experiments were numbered (#1 to #22) in the revised paper, the two typical experiments shown in details were #1 and #19. Figure 4 is the original Fig. 6. Another panel of aerosol 3-D size distribution time series plot was inserted into Fig. 4a. The details about the experiment conditions and the default CE and RIE values have been inserted into Fig. 4b. Arrows were added to show the relative stable states that were used for average. The experiment started when AS was first started to injected into the chamber. Arrow 1 indicated the AS particle concentration became stabled after stopping injection. Arrow 2 indicates when SOA started to form. The period between Arrows 3 and 4 was when the SOA coated AS particles reached “relative stable state” and was used for data averaging. In all the experiments, Sulfate concentrations had more or less variations, possibly due to the use of a relatively low sampling interval (~8 min). As inspired by the reviewer, we found that 18-25 nm organic shell can fully cover the 80 nm AS seed particles. As the situation in this experiment (Exp. 19), “Two hours after the experiment started, the Q-ACSM measured organic concentration was still increasing but the measured sulfate stopped increasing and kept constant during the rest of the experiment. By the time when sulfate signal stopped increasing, d_g increased to 115 nm, the average coating thickness (growth in particle radius) was about 18 nm assuming uniform coating. This indicated that 18 nm SOA shell was already enough to cover the 80 nm seed particles in this experiment.” (Lines 253-257)

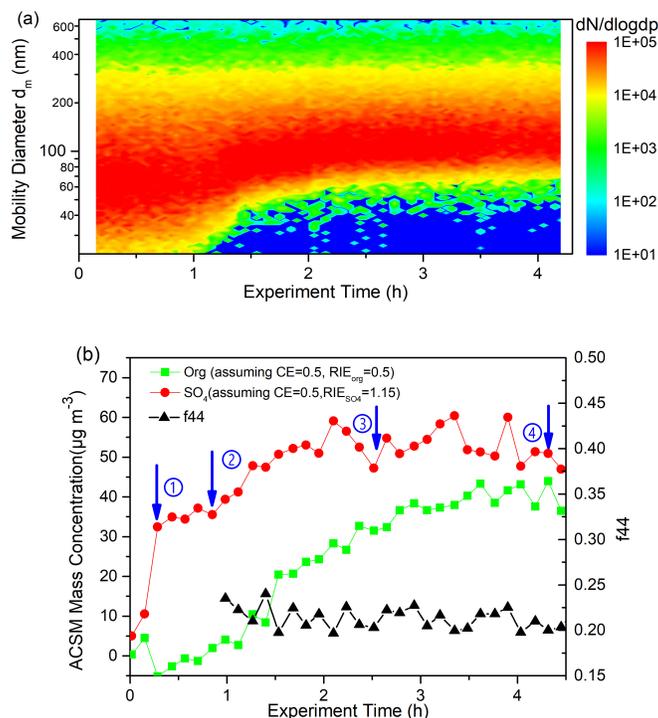


Figure 4. Time series of a typical experiment with SOA coating on AS (Exp. 19): (a) aerosol size distribution measured by SMPS; (b) Organic, sulfate mass concentrations and f_{44} measured by Q-ACSM (assuming $CE=0.5$, $RIE_{org}=1.4$, $RIE_{SO_4}=1.15$). Arrow 1 indicates when the injected AS particles reached stable, Arrow 2 indicates when SOA started to form. The period between Arrows 3 and 4 was when the SOA coated AS particles reached “relative stable state” and was used for data averaging.”

40. “Figure 7: This plot is potentially the key to understanding the reason for changes in the response factors as a function of varying f_{44} . However, it does not contain sufficient conclusive evidence on how f_{44} and coating thickness independently relate to CE. Are the changes in CE at f_{44} of about 0.2 as a function of increasing Org/SO4 ratios significant? Why are the range of values for f_{44} in these experiments forming SOA from alpha-pinene or toluene inconsistent with those shown in Figures 4 and 5? Which points are from alpha-pinene and which are from toluene? All of the mass distributions for these experiments (and the effect of ACSM lens transmission) should be shown in the supplementary information. Do the mass distributions indicate that the ammonium sulfate particles were “uniformly” coated, rather than new organic particles being formed? If so, the color scale should indicate coating fraction. Need error bars applied to CE.”

Response: Figure 5 is the original Figure 7. As suggested by the reviewer, the coating thickness were calculated and color coded in Figure 5. As stated in the revised manuscript, “For most of the experiments, the SOA coating layer was thick enough (radius growth >18~25nm) to cover AS seed particles. This was confirmed with the fact that after coating with 18~25 nm SOA, ACSM measured sulfate concentration stopped increasing with further organic increase. While CE_{org} from Exp. 16, 20, 21 (coating layer < 18-25 nm) might be the result of a combination of AS and organic particles because ACSM measured sulfate increased all the way until organics stopped to increase.” Line 278-282. As stated before, only one of the experiment had experienced negligible amount of new particle formation. Based on the additional calibrations, error bars have been added into both CE and f_{44} values. The data can also be found in Table 2. The f_{44} ranges from the two sets of experiments were shown in Fig. S7 and S8, it was still not clear why f_{44} in AS coating experiments was overall higher than pure SOA experiments, which made it impossible to directly calculate RIE.

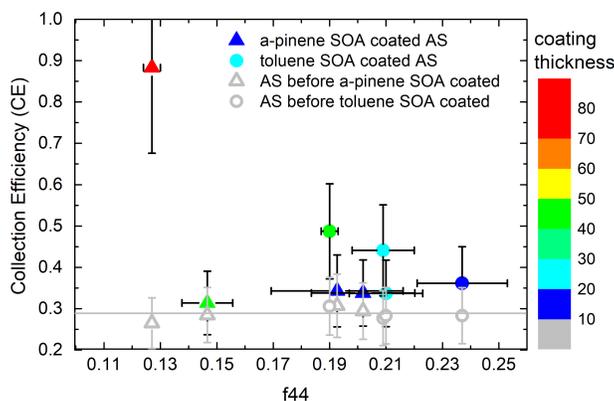


Figure 5. CE of SOA coated ammonium sulfate (AS) particles. Gray points represent CE_{AS} before SOA were coated onto AS in experiments with different VOCs. Colored triangles were CEs of AS coated with α -pinene oxidized SOA and colored circles were CEs of AS coated with toluene oxidized SOA. The color scale represents SOA coating thickness; for 80 nm AS particles generated, an SOA coating layer with a thickness of 18-25 nm should be able to fully cover the AS core. All the data can be found in Table 2.

41. *“Figure 8: This figure could be deleted because it does not show quantitatively the responses to organic aerosols for the ACSM measurements shown in this paper.”*

Response: Figure 8 has been removed from the manuscript.

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