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Atmospheric Measurement Techniques
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Discussions

Interactive comment on "Sensitivity of a Q-ACSM to chamber generated SOA with different oxidation states" *by* Xiaoxiao Li et al.

Anonymous Referee #2

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General Comments:

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.

Specific Comments:

Writing and Presentation of Key Concepts

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.

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.

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.

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?

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.

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.

Volatility Effects

One potential issue that should be addressed is the possibility of the SOA being partially volatile. Particles with low f44 (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 f44?

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 "f44" or "average oxygen content" depending on the context.

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

Page 1 Line 21 (Abstract): revise "SOA samples were generated . . ." to "SOA was generated . . ."

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.

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 ... "

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."

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.

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.

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 f44, 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.

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.

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

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 f44, 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.

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 f44 are consistent with the changes in the response factor without a varying RIE. Specifically, if CE=1 instead of 0.5 for the lowest f44 values, the response factor would decrease from 2.15 to 1.1 and if CE=0.3 instead of 0.5 for the highest f44, 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.

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).

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”

Page 9 Lines 207-208: f44 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.

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+/-3%), making the 0.28 value outside the uncertainty band.

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.

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.

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.

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

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.

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).

Page 14 Lines 336-337: While this statement may be true, the section on CE as a function of f44 seems to indicate that the large changes in RF are mostly due to changes in CE. Once the CE is properly taken into account for the RF, 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.

Table 1: Add errors, mainly to PM, density, f44, and ACSM/SMPS columns.

Table 2: Digits listed for ammonium sulfate, Org, f44, 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.

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.

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.

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).

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

Figure 5: The conclusions of this paper about the response factor (and effective density) varying with f44 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 f44=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.

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.

Figure 7: This plot is potentially the key to understanding the reason for changes in the response factors as a function of varying f44. However, it does not contain sufficient conclusive evidence on how f44 and coating thickness independently relate to CE. Are the changes in CE at f44 of about 0.2 as a function of increasing Org/SO4 ratios significant? Why are the range of values for f44 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.

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.

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