Authors Final Response to Referee 2

We are pleased that all the referees think that this work should be published after we address their concerns. We agree with many of their suggestions and have modified the manuscript as described below. We respond to the comments of each referee separately. Their comments are included in italics with numbers, followed by our responses.

In reviewing Table 1, we found an error in the VOC concentration numbers in the studies of SOA chemical composition dependence on OH exposure. The correction of the precursor VOC concentrations does not affect the results or the conclusions of those experiments. The corrected Table 1 and the VOC concentration numbers are listed in 'Minor and technical corrections by Authors' at the end.

Re-arrangement of paragraphs by Authors:

We re-arranged the paragraphs and section orders as suggested by referee 2's comment and by authors' decision.

We re-arranged the introduction section by moving the 2nd and 3rd paragraphs under Section 2 to Introduction section. Thus, we switched the number of figure 2 and 3 each other to be consistent with the manuscript revision.

We moved the "Section 3.1.2 The determination of OA mass concentration for this study" to "Section 2.4 The determination of OA mass concentration".

We deleted the section title "Section 3.1.3 Oxidation indicators f_{44} and f_{43}" and moved the paragraph under that section to Section 2.4.

We deleted the section title "3.1.1 Mass spectra of SOA".

Major comments:

1-*This paper mostly focuses on the analysis of m/z 44 and 43 fractions of the AMS mass spectra, which are important OA mass fragments to study, but for this type of laboratory based study seems somewhat restrictive. The analysis can be extended to also study changes in the overall mass spectra. How do the mass spectra evolve over the course of a single experiment and with OA mass and OH exposure? How do the spectra compare with chamber SOA and ambient PMF/PCA factors such as HOA, SVOOA and LV-OOA factors? These comparisons are discussed only qualitatively in the paper and could easily be more quantitatively correlated. Offline characterization of the chemical composition and molecular weights of species in OA would also be highly complementary to this work, as it would shed some light on the importance of oligomerization reactions in this study, which cannot be determined with AMS measurements, compared to offline analyses of chamber SOA.*

As the reviewer points out, it is possible for us to do an analysis with PMF/PCA factors, but we feel that using f_{43} and f_{44}, as Ng et al. (2010) did, demonstrates that main points that we want to make in this paper. We prefer to keep the analysis "as is", although we have made several modifications in response to the reviewer's comments.

The time-evolution is complicated by the flow through the PAM chamber, which is not a plug flow. Thus, it is not possible to assign a specific OH exposure to a specific time either after the UV lamps are turned on or the air enters the chamber. We clarify this situation in Section 2.1 with the following sentences.

Page 24058, Line 25: add these sentences "The flow in the PAM chamber is not strictly plug flow, for which residence time and distance from the inlet are linearly correlated. Instead, convection mixes the air and creates a distribution of residence times (Lambe et al., 2010).

Thus, OA sampled at any time will have experienced this distribution of OH exposures, although the distribution does have a well-defined peak value."

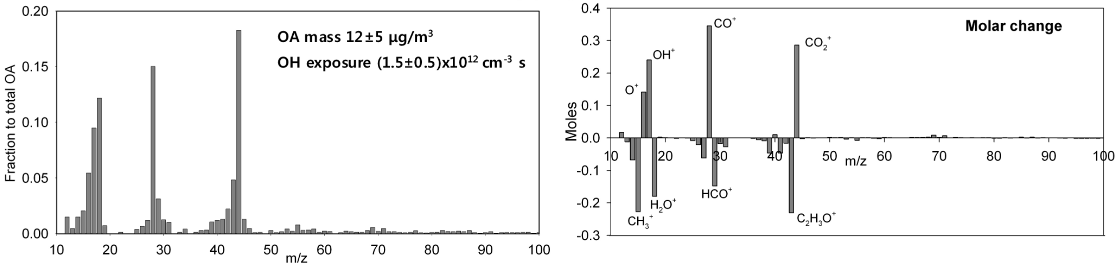
We also add a reference for work done at Boston College with a later version of the PAM chamber.
Page 24078, after Line 29: add a new reference "Lambe, A. T., Ahern, A. T., Williams, L. R., Slowik, J. G., Wong, J. P. S., Abbatt, J. P. D., Brune, W. H., Ng, N. L., Croasdale, D. R., Wright, J. P., Worsnop, D. R., Davidovits, P., Onasch, T. B.: Characterization of aerosol photooxidation flow reactors: heterogeneous oxidation, secondary organic aerosol formation and cloud condensation nuclei activity measurements, Atmos. Meas. Tech. Discuss., 3, 5211–5251, 2010."

We have added additional panel for each spectrum in Figure 3 that shows the changes in peak heights as a function of increased oxidation. These 'action spectra' illustrate the main features of the evolution of aerosols that would be revealed by an analysis of individual peaks – in essence, these spectra show how individual peaks evolve with increased oxidation. We believe that any additional analysis of other, more minor, peaks is beyond the scope of this paper.

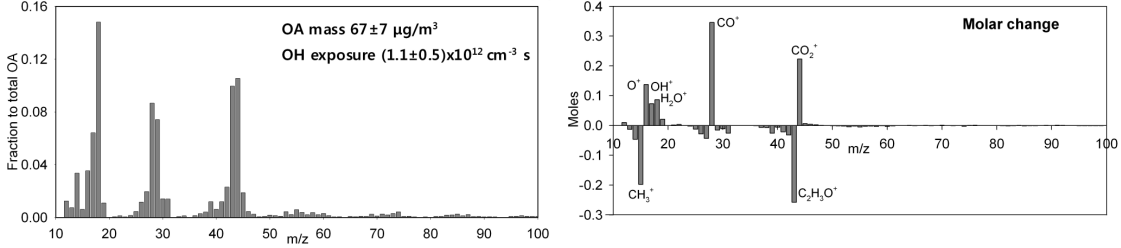
Page 24062 Line 17: We added a new paragraph "For each precursor studied, there was a series of peaks that varied primarily with m/z 43 and a different series that varied primarily with m/z 44. There were a larger number of peaks that varied with both m/z 43 and m/z 44, probably due to fragmentation of a common parent compound. The right panels in Fig. 3 show difference spectra obtained from a regression of the changes in individual m/z values as a function of changing f_{44}. A positive (or negative) value represents a gain (or loss) of a particular mass with increasing f_{44}. These difference spectra are remarkably similar and very simple: they reveal that with increased oxidation the condensed phase is enriched in species that fragment into m/z 16, 17, 28, and 44 (e.g., the expected fragments of –COOH and –C=O groups), while being depleted in species that fragment into m/z 14, 15, 27, 29, and 43 (the expected fragments of $–C_2H_3O$ and $–CH_3$ groups). For all three organic precursors, the change in the O/C ratio is -1.0 ± 0.05, strong evidence for an increase in the oxidation state of carbon from +2 to +3 in response to OH exposure (i.e., $-CH_xO-$ groups converting to –COOH), as opposed to just an increase in the number of oxidized carbon atoms in a particular compound. In the following discussions, the mass spectrum peaks at m/z 43 and m/z 44 will be used as indicators of the relative oxidation of the SOA as the VOC type and amount or the OH exposure are varied."

Figure 3: We added the right panel plots, and the figure caption is revised as follows.

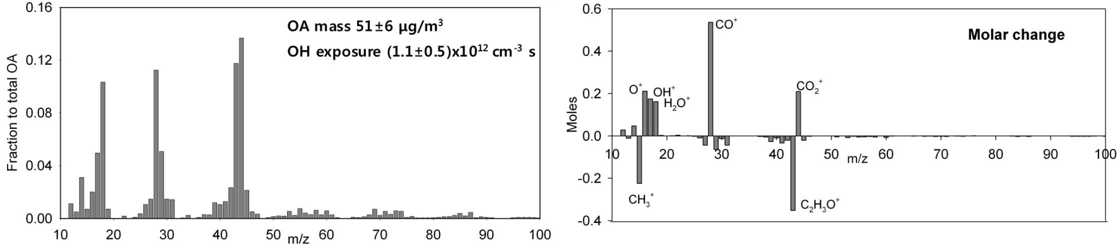
(a) α-pinene



(b) m-xylene



(c) p-xylene



"Figure 3. The left panel represents the example of mass spectra of SOA for (a) 7±1 ppbv of α-pinene, (b) 160±30 ppbv of m-xylene, and (c) 200±30 ppbv of p-xylene in the PAM chamber. OH exposure was 1.5x10^{12} cm^{-3} s for no VOC addition, and resulting OH exposure for each VOCs amount are shown in the figure. The OH exposure includes the uncertainty error bars (2σ confidence level). All spectra are the quantitative fractional contribution to the total OA mass concentration. The right panel represents the observed fractional molar change as a function of increasing f_{44} (e.g., OH exposure). Peaks are normalized to the y-intercept of the f_{44} vs. f_{43} plot (e.g., Figure 6)."

***2-** Given the possible temperature dependence of SOA yield due to partitioning of semivolatile organics, it is unclear why a correction for OA mass measured by the TEOM measurements at 30 C is not needed. This point is mentioned briefly in Kang et al., 2007, but is not explained in this work.*

Even though we discussed the means of looking for semi-volatile mass loss with the TEOM in Kang et al. (2007), we agree with the reviewer that it would be good to discuss it again in this paper. Thus, we modified the manuscript as follows.

Page 24061 Line 12: We included these sentences. "Even at 30°C, semi-volatile mass can evaporate from the TEOM filter, causing an underestimate of the mass (Wilson et al., 2006). However, as we described in Kang et al. (2007), the TEOM measures the time-rate-of-change of mass on a filter and integrates it over a time period to find the mass concentration. When the UV lamps are turned off and no more mass is being produced, the continued evaporation of semi-volatile organics from the filter appears as a negative mass. For both Kang et al (2007) and this study, the negative mass signal after the UV lights were turned off was only a few percent of the measured mass; thus we have assumed that loss of semivolatile organic mass from the TEOM can be neglected."

Page 24080 Line 11: We added a new reference "Wilson, W. E., Grover, B. D., Long, R. W., Eatough, N. L., and Eatough, D. J.: The measurement of fine particulate semivolatile material in urban aerosols. J. Air & Waste Manage. Assoc., 56, 384-397, 2006."

3-*The AMS and TEOM measurements are in good agreement in this work, where an AMS transmission efficiency of 100% is assumed. Yet recent studies such as Salcedo et al. (2006) and Virtanen et al. (2010) have shown that SOA particles are likely in amorphous solid state and therefore have an AMS transmission efficiency less than 100% due to particle bouncing. How do you reconcile your observations with these studies? Would the particle bounce effect impact TEOM results in a similar fashion to AMS measurements but still allow good agreement between the two techniques?*

Salcedo et al. (2006) excluded particulate water to calculate total aerosol mass concentration from ambient AMS mass spectra, which may cause the 20% difference of PM2.5 mass concentration between the AMS+Black Carbon+soil and the TEOM. In this study, particulate water was included in the organic mass concentration and the air sample was not dried in prior to AMS inlet. Virtenan et al. (2010) observed that atmospheric organic aerosol could be solid particles that have reduced transmission efficiency in the AMS by the particle bouncing effect. However, other studies describe that the organic aerosols absorb water even at lower relative humidity (Mikhailov et al., 2009) and the relative humidity, precursor component, and VOC concentration can affect the state of organic particles: solid, semi-solid, or liquid (Matthew et al., 2008; Virtenan et al., 2010).

The TEOM continuously collects particles as air passes through a filter mounted on the end of a glass column. No particle bouncing effect has been observed for the TEOM; instead evaporation loss or diffusion will cause the under- or over-estimation of mass (Chow et al., 2008).

Given the very different sampling conditions and methods, it would be remarkable if losses of particles in the AMS were to track nearly one-for-one those in the TEOM. In addition, if significant bounce (or, more generally, transmission losses) was occurring in the AMS, and we were to double the organic mass consistent with corrections made by others, the amounts of organic mass produced in the PAM chamber would exceed anything that has been observed in chambers or flow tubes. Thus, it is far more likely that both instruments were seeing the bulk of the organic mass produced in the PAM chamber, and we are led to conclude that the TEOM and AMS were sampling with near 100% efficiencies. The question then raise is why our AMS would behave differently than others' in similar experiments. The most likely explanation is that in our study, because the particles were not dried prior to sampling, the transmission losses are very small. Another, less likely explanation for the lack of bounce effect could be due to the lower AMS oven temperature used in this study, but this possibility needs to be explored in more detail for AMS instruments. It would be useful to repeat the PAM chamber experiments both with and without a diffusion dryer and at different oven temperatures to address these possibilities. At the time we carried out this work, the AMS community was only beginning to recognize the particle bounce issue, and because we observed excellent agreement between the TEOM and AMS total organic mass, such studies did not seem necessary.

Page 24064, Line 14-18: We removed the sentence starting with "The fact that AMS particulate...", and made a new paragraph starting withthese sentences "In this study particles were not dried prior to sampling by the AMS. The good agreement between the total OA mass measured by the AMS and TEOM suggest that transmission losses in the AMS are small under these conditions. As described in section 2.3, the evaporation losses of semivolatiles on the TEOM were also small. In addition, if the AMS results were to be corrected for particle bounce reported by others who dry the particles prior to sampling, the total organic yields would vastly exceed those observed in environmental chambers and flow tubes (Matthew et al., 2008). In light of recent studies that have discussed on the AMS sampling issues, it would be useful for future studies of the PAM chamber to investigate the role of particle phase on sampling efficiency."

Page 24064 Line 18: We replaced the uncertainty value "32%" to "±30%" with 1 significant figure.

Page 24079, after Line 3: add a new reference "Matthew, B. M., Middlebrook, A. M., Onasch, T. B.: Collection efficiencies in an Aerodyne Aerosol Mass Spectrometer as a function of particle phase for laboratory generated aerosols, Aerosol. Sci. Tech., 42(11), 884-898, 2008."

4-*Were these techniques compared with some size distribution measurements such as SMPS to check whether particle bounce was an issue?*

We do have size distributions from a UHSAS, which indicate that the AMS is not missing particles of a specific size (in fact, the UHSAS and AMS results were highly correlated). However, these measurements don't necessarily constrain the total mass, because the UHSAS measures optical diameter, which differs from vacuum aerodynamic diameter because of density and index of refraction. So all we can say is that the measurements are consistent and they appear to be no changes in systematic errors. Unfortunately, an SMPS would not have been helpful in our experiments due to the long sample times required (of order 3-5 minutes), which would not have captured the short-term changes in mass.

5-*Have the OH concentrations been directly measured in the PAM chamber with GTHOS during an oxidation experiment to confirm that the OH concentrations were consistent with modeled values for the high VOC concentrations over the course of an experiment? Volatile and semivolatile oxidation products may also react with OH radicals to reduce its overall concentration during an oxidation experiment. Have the OH reactions with these oxidation products also been taken into account in the model?*

The OH concentration was measured in the PAM chamber with GTHOS but in separate studies. The OH concentration did not change with the addition of large amounts of CO or SO_2. On the other hand, the OH measurement was within 15 cm of the PAM chamber's outlet and was not particularly sensitive to OH near the inlet, where the OH should change the most because the VOC concentrations are the greatest there. Thus, the OH exposure is most sensitive to the changes in OH that occur near the inlet, not the outlet. In the simple model used to make the corrections to the OH exposure in this study, we included a simplified form of the reaction chemistry for these three VOCs. It should be pointed out that ozone reactions with the VOCs and their products can produce OH as well as destroy it and the OH yields need to be taken into account. A more complete model, which we are in the process of developing, will be needed to check the simple model in terms of the changes to the OH exposure. As a result of the uncertainty in the simple models used, we have added estimated uncertainty error bars (2σ confidence level) on the OH exposure in Table 1 and figures. Other OH exposure value corrections were undertaken including uncertainty.

Page 24060 Line 7: We included the words from "… and smallest OH amount, VOCs were…" to "… and smallest OH amount, photochemical modeling indicates that VOCs were…".

Page 24060, Line14: add the sentence "The OH exposure calculation also includes the uncertainty error bars (2σ confidence level) to take into account the OH yields from the fact that the ozone reactions with the VOCs and their products can produce OH as well as destroy it."

Page 24074 Line 26: We added the sentences "The extreme oxidant amounts in the PAM chamber favors reactions of OH, HO_2, and O_3 with VOCs and their reaction products, so that organic peroxy radicals react only with HO_2 and not with each other as happens in the atmosphere. On the other hand, if most OA-producing and OA-aging reactions occur with OH, HO_2, O_3, and O_2 in the atmosphere, then the same oxidation reactions occur in the PAM chamber, only faster.".

Page 24067, Line 11: We replaced the OH exposure values with the ones including uncertainties." from $(1.8\pm1.0)\times10^{11}$ cm^{-3} s to $(2.4\pm0.8)\times10^{12}$ cm^{-3} s. "

Page 24068 Line 7: We replaced the OH exposure values with the ones including uncertainties to : "$(1.8\pm1.0)\times10^{11}$ cm^{-3} s to $(2.4\pm0.8)\times10^{12}$ cm^{-3} s."

Page 24072, Line 28: We replaced the OH exposure values with the ones including uncertainties. "study ($(1.8\pm1.0)\times10^{11}$ cm^{-3} s to $(2.4\pm0.8)\times10^{12}$ cm^{-3} s)."

Page 24081: We replaced the Table 1.

“Table 1. Experimental conditions of OH exposures, SOA concentrations and SOA yield for various conditions. SOA concentration was measured by a TEOM.

VOCs	ΔHC (ppbv)	OH (pptv)	OH exposure (molecules cm^{-3} s)	SOA Concentration (μg/m^3)	SOA Yield
α-pinene	7±1	260	(1.5±0.5)x10^{12}	12±5	0.31±0.14
	19±3	260	(1.4±0.5)x10^{12}	22±5	0.22±0.06
	33±5	260	(1.4±0.5)x10^{12}	62±7	0.35±0.06
	48±8	260	(1.3±0.5)x10^{12}	83±8	0.32±0.05
	57±9	260	(1.3±0.5)x10^{12}	150±13	0.49±0.08
	79±13	260	(1.2±0.4)x10^{12}	220±18	0.51±0.08
	39±6	63	(2.8±0.9)x10^{11}	110±10	0.54±0.10
	39±6	160	(7.8±2.5)x10^{11}	110±10	0.52±0.09
	39±6	260	(1.3±0.5)x10^{12}	94±9	0.45±0.08
	39±6	430	(2.3±0.8)x10^{12}	94±9	0.45±0.08
m-xylene	87±14	260	(1.2±0.4)x10^{12}	16±5	0.04±0.02
	160±30	260	(1.1±0.5)x10^{12}	67±7	0.10±0.02
	360±60	260	(8.9±5.0)x10^{11}	170±15	0.11±0.02
	430±70	260	(8.3±5.0)x10^{11}	270±22	0.15±0.03
	170±30	72	(2.4±2.0)x10^{11}	66±7	0.09±0.02
	170±30	160	(6.2±4.0)x10^{11}	63±7	0.09±0.02
	170±30	270	(1.2±0.5)x10^{12}	53±7	0.07±0.02
	170±30	390	(1.9±0.9)x10^{12}	52±7	0.07±0.02
	170±30	480	(2.4±0.8)x10^{12}	57±7	0.08±0.02
p-xylene	140±30	260	(1.2±0.4)x10^{12}	18±5	0.03±0.01
	200±30	260	(1.1±0.5)x10^{12}	51±6	0.06±0.01
	260±40	260	(9.9±6.0)x10^{11}	86±9	0.08±0.01
	370±60	260	(8.9±5.0)x10^{11}	250±21	0.16±0.03
	180±30	63	(2.1±2.0)x10^{11}	70±8	0.09±0.02
	180±30	140	(5.7±4.0)x10^{11}	61±7	0.08±0.02
	180±30	260	(1.1±0.5)x10^{12}	55±7	0.07±0.01
	180±30	330	(1.6±0.7)x10^{12}	52±7	0.07±0.01
A mixture					
(α-pinene 37±6 ppbv,		58	(1.8±1.0)x10^{11}	79±8	0.13±0.03
m-xylene 46±7 ppbv,		260	(1.1±0.5)x10^{12}	59±7	0.10±0.02
p-xylene 47±8 ppbv)		450	(2.3±0.8)x10^{12}	48±6	0.08±0.02

Figure 3, we added the error range of OH exposure as already shown above.
Figure 3, caption: We added the sentence “The OH exposure includes the uncertainty error bars (2σ confidence level).” after 2nd sentence as follows.
“Figure 3. The left panel represents the example of mass spectra of SOA for (a) 7±1 ppbv of α-pinene, (b) 160±30 ppbv of m-xylene, and (c) 200±30 ppbv of p-xylene in the PAM chamber. OH exposure was 1.5x10^{12} cm^{-3} s for no VOC addition, and resulting OH exposure for each VOCs amount are shown in the figure. The OH exposure includes the uncertainty error bars (2σ confidence level). All spectra are the quantitative fractional contribution to the total OA mass concentration. The right panel represents the observed fractional molar change as a function of increasing f_{44} (e.g., OH exposure). Peaks are normalized to the y-intercept of the f_{44} vs. f_{43} plot (e.g., Figure 6).“

Figure 4, Caption: We replace the OH exposure values with the new values including uncertainties.
“Figure 4. SOA oxidation dependent on the OA mass concentration for α-pinene, m-xylene, and p-xylene. OH exposure was ranged between (12±4.0)x10^{11} and (15±5.0)x10^{11} cm^{-3} s for

α-pinene, (8.3±5.0)x10^{11} and (12±4.0)x10^{11} cm^{-3} s for m-xylene, and (8.9±5.0)x10^{11} and (12±4.0)x10^{11} cm^{-3} s for p-xylene. OA mass concentration was measured by the TEOM. Error bars for SOA mass concentration, f_{43}, and f_{44} are at the 1σ confidence level. (a) Generated OA mass concentrations vs. precursor VOCs amount. (b) The ratio of f_{44} to f_{43} vs. OA mass concentrations. (c) The values of f_{44} and f_{43} vs. the OA mass concentrations. O:C ratio was calculated from f_{44} values based on Aiken et al. (2008)."

In Figure 5, we have not indicated the uncertainty in the OH exposure on the plots, since the precision errors are small and the absolute error is correlated for all points. Thus, the effective scale has uncertainty. We have noted this with the alterations to the figure caption as follows:

"Figure 5. SOA oxidation dependent on the OH exposure at a constant VOC amount of 39±6 ppbv of α-pinene, 170±30 ppbv of m-xylene, 180±30 ppbv of p-xylene, and a mixture of three (37±6 ppbv of α-pinene, 46±7 ppbv of m-xlene, 47±8 ppbv of p-xylene). OA mass concentration was measured by the TEOM. The error bars for OA mass concentration, f_{43}, and f_{44} are at the 1σ confidence level. Atmospheric OH aging time was obtained with a typical diurnally averaged OH concentration of 1.5x10^6 molecules cm^{-3} (Mao et al., 2009). Uncertainties in the OH exposures are correlated so that the OH exposure scale could expand or shrink by the uncertainty ranges given in Table 1. (a) OA mass concentration vs. OH exposure. (b) The ratio of f_{44} to f_{43} vs. OH exposures. Linear regressions of each lines are shown here; for α-pinene, slope=4.50x10^{-13}, intercept=0.47, r^2=0.99; for m-xylene, slope=6.29x10^{-13}, intercept=0.48, r^2=0.99; for p-xylene, slope=4.46x10^{-13}, intercept=0.64, r^2=0.99; and for the mixture, slope=1.17x10^{-12}, intercept=0.58, r^2=0.99. (c) The values of f_{43} and f_{44} vs. OH exposures. O:C ratio was calculated from f_{44} values based on Aiken et al. (2008)."

Figure 6. We replaced the legend with OH exposure with uncertainties. And figure caption is changed by adding the sentence "The OH exposure unit is cm^{-3} s with uncertainty error bars (2σ confidence level)."
"Figure 6. Change in f_{44} and f_{43} over the course of m-xylene photo-oxidation experiment for various OH exposure conditions. (a) Change in f_{44} as a function of OA mass concentrations, (b) Change in f_{44} as a function of f_{43}. Each data point is a one-minute measurement data by the Q-AMS. The OH exposure unit is cm^{-3} s with uncertainty error bars (2σ confidence level)."

6-*It would be useful to discuss the role of multigenerational oxidation chemistry and its impact on OA formed in the PAM chamber compared to other chamber and flow tube studies. In particular, how many oxidation generations do the volatile organics and SOA particles undergo with respect to OH oxidation under different OH exposures? Does the number of oxidation generations change significantly with increase in precursor VOC concentrations (i.e. experiments in Sec 3.2), and can this in part explain reduction in F44 with OA mass? Do particle-phase organics undergo enough oxidation generations to impact the chemical composition of OA through heterogeneous oxidation compared to gas phase oxidation?*

While we agree with the reviewer that this study of further oxidation steps would be useful and insightful, we feel that this rather substantial project is not within the scope of this paper. Once we have developed and tested the full gas-phase and particle-phase model, then we can undertake the excellent study that the reviewer suggests.

7-*Is it possible that the reduced OA mass with OH exposure (decrease of OA mass from transient maximum to stable concentration in Fig. 6) may not be due to fragmentation induced by heterogeneous oxidation as suggested, but other reasons, such as: a) photolysis induced fragmentation of gas-phase and/or particle-phase oxidation products, b) changes in* *transmission/collection efficiency in AMS/TEOM due to changes in particle phase/shape or c) wall loss?*

All evidence suggests that the decrease of OA mass from transient maximum to stable concentration is due to the microphysics (e.g. evaporation), not the chemical process such as oxidation. While the OA mass was decreasing, f_{44} and f_{43} values were stable. In Fig. 6 (b), the stable f_{44} and f_{43} values were reached after 3 minutes while the OA mass peaked and then decreased to a stable value over the next 10 minutes. (a) If the loss was due to photolysis induced fragmentation, the photolysis would have to fragment organics that contributed to f44 and f43 equally, which seems unlikely to us. (b) There is a excellent agreement in the masses observed by AMS,TEOM, and UHSAS at all times. It is highly improbable that all three instruments would have identical transmission/collection losses or identical response to changes in particle phase/shape. (c) The wall loss in this version of the PAM chamber is small, since the conversion of SO_2 to sulfate (as described in page 24058, line 21~page 24059, line 2) agreed with theory to within 10 %.

***8-**A reduction in particle size (size distribution measurements) would more clearly show whether the particles are in fact volatilizing. The observed OA mass loss in this work with OH exposure could more quantitatively be compared to observed mass/volume loss (particle size change) due to OH oxidation in other laboratory studies such as Kroll et al. (2009) and George and Abbatt (2010). Is the observed mass loss in this work quantitatively consistent with these studies? Can these observations be reconciled with Kang et al., 2007 (i.e. Fig. 6) where SOA yield from the photooxidation of a-pinene did not decrease with OH exposure?*

In the PAM chamber, the production of condensate and super-saturation is fast so that evaporation, condensation, and coagulation are all competing. Thus, it may not be possible to observe the particles actually decrease in size during this transient phase. This condition is different from the heterogeneous volatilization of semi-volatile products from the particle phase after the particle generation was completed, as in SOA generation and heterogeneous oxidation in other flow tubes or slow reacting chambers. Thus, in the PAM chamber we observed no size reduction in particle size with respect to different OH exposures from UHSAS data, which was not shown in this manuscript. The constant size distribution also supports that idea that the OH oxidation favorably occurs in the gas phase rather than on particle surfaces.

Our mass loss with OH exposure was about 20% at 2.5×10^{12} cm^{-3} s of OH exposure. George and Abbatt (2010) observed 10% of volume loss of SOA at similar OH exposure of ours. But their SOA was mono-dispersed (diameter 200 nm) and generated by ozonolysis and not exposed to OH until after most of the gas-phase volatile and semi-volatile organics were removed. Thus, it is difficult to make a good, quantitative comparison of the results of these studies.

The reviewer asks if we can reconcile the apparent differences between the oxidation shown in Figure 6 of Kang et al. (2007) and this paper. However, the experimental conditions of this study and our previous study (Kang et al., 2007) were different for the precursor gas mixing ratios, which was 100 ppbv for the results in Figure 7 in Kang et al. (2007) and only 39 ppbv for the results in this study. This difference alone helps explain the apparent discrepancies because 39 ppbv of α-pinene, which oxidizes fast than 100 ppbv of α-pinene. If the reviewer looks in Figure 7 in Kang et al. (2007), the SOA yield does decrease with increasing ozone, although actually OH is also increasing and is the likely main oxidant and cause of that decrease. So, the difference between Figure 7 in Kang et al. (2007) and Figure 5 in this paper can be ascribed to a combination of different conditions between the two studies and untracked experimental variability. Thus, more study is needed to determine if these results from these two papers are truly inconsistent.

***9-**The F44 and F43 values from OA produced in the PAM chamber were directly compared to the SOA particles in Jimenez et al. (2009) and George and Abbatt (2010). It should be noted that both these studies produced a-pinene SOA from ozonolysis and passed the SOA particles through activated charcoal denuders to remove volatile (and potentially semivolatile) organics before exposure to OH radicals in the flow tubes. Could these differences in SOA production – and lack of volatile organics in these studies partially explain differences in chemical composition of SOA as well as their chemical evolution with OH exposure?*

We will revise the manuscript to clearly show the experimental differences between their studies (Jimenez et al., 2009 and George and Abbatt, 2010) and ours. These differences may or may not make these comparisons invalid.

Page 24071, Line 8-9: We moved the chapter title "3.4 Comparison of SOA oxidation…" to Page 24070 before Line 21.

Page 24070, Line 29-page 24071 Line 7: We removed the 4th and 5th sentences starting with "It could be that our study…" and "Or it could be that the walls in their flow tube…", and add the following sentences instead. "Why the OA were more efficiently oxidized in this study compared to theirs is not known, but the conditions between their study and ours are quite different. In their study, OA were produced from α-pinene by ozonolysis and then passed through an activated charcoal denuder to remove volatile and probably semi-volatile organics before being exposed to OH in a reaction flow tube. Thus, the greater amount of volatile and semi-volatile organics present during OH exposure in our study could explain the greater amount of f_{44} observed."

Page 24071 Line 6-7: We changed the words "to this point in the next section" to "to this point later".

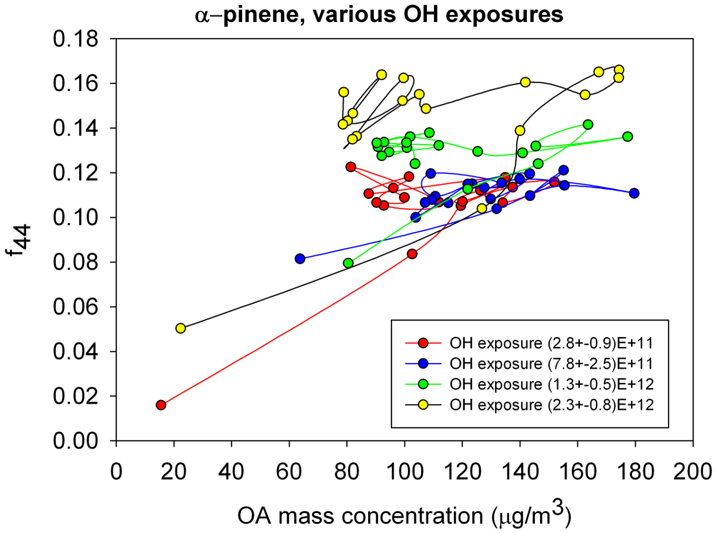
As for the comparison with Jimenez et al. (2009), we have revised our paragraph as follows.

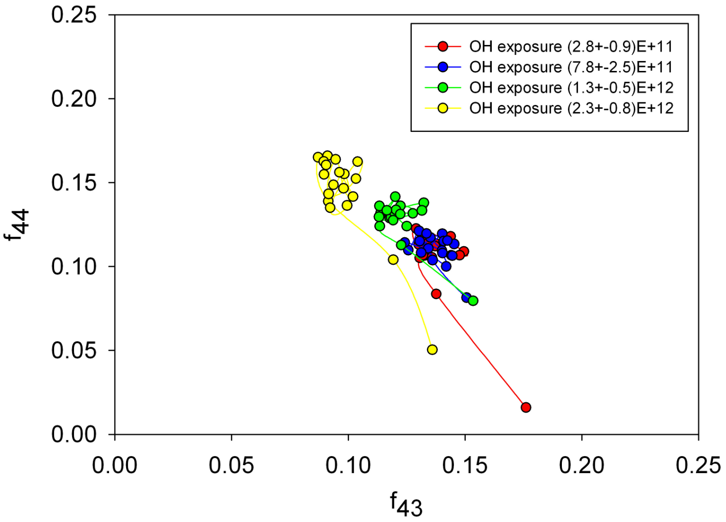
Page 24072, Line 28: We removed the last sentence starting with "While wall effects in the LBNL flow tube……". We added the sentences "However, just as in George and Abbatt (2010), Jimenez et al. (2009) produced SOA from α-pinene ozonolysis and then removed volatile organics prior to adding the OA to the reaction flow tube. It is possible that these different conditions between their study and ours result in slightly different particle composition, which then produces different f_{44} and f_{43}, although in the same proportion. However, it is also possible that the differences in the AMSs used in the two studies or in the calculations of f_{44} and f_{43} are responsible."

Page 24073 Line 4: We added the sentence "Our results in Fig. 7 can be made to overlap with the results from Jimenez et al. (2009) by shifting our f_{44} or f_{43} values up by 0.07 or by shifting a combination of both by as little as 0.04.".

***10-**In Ng et al. (2010), Figure 9 shows the evolution of F44 with organic mass concentration of SOA over the course of a chamber experiment, where photooxidation of apinene and m-xylene look quite different. Were the changes in F44 of a-pinene SOA over a single PAM oxidation experiment comparable to m-xylene shown in Fig. 6 of this work, and if so, please discuss why this is not consistent with results in Ng et al. (2010).*

Our result also showed that for alpha-pinene, the f_{44} levels off at lower values than m-xylene, which was similar to what is shown in Figure 9 of Ng et al., (2010). Please see the following figures.





We modified the manuscript as follows.

Page 24070 Line 10: We added the sentences “. The behavior for α-pinene, not shown, is qualitatively the same as for m-xylene, with similar peak values for different OH exposures, but with a slight decrease in f_{44} between the transient peaks and stable values.”

Minor Points:

1- *Pg. 24054 Line 26: "cardiovascular and respiratory systems" would be more appropriate than "cardiovascular system"*

We modified the manuscript as follows.
Page 24054 Line 26: We replaced the words from "cardiovascular system" to "cardiovascular and respiratory systems".

2- *Pg. 24055 Line 6-7: Nel (2005) and Forster et al. (2007) references should be cited on previous page when discussing aerosol climate and health effects. Jimenez et al. (2009) and Ng et a. (2010) papers do not focus on SOA particles, per se. More appropriate references here to reflect the breadth of SOA research are the recent SOA reviews, e.g. Kroll and Seinfeld (2008), Hallquist et al. (2009).*

We modified the manuscript as follows by including suggested new references. We thank for good reference suggestions.

Page 24054 Line 25: We changed the reference list to "(Foster et al., 2007; Hoyle et al., 2009; Myhre et al., 2009)".

Page 24054 Line 26: We changed the reference list to "(Nel, 2005; Jang et al., 2006; Baltensperger et al., 2008)".

Page 24055 Line 6: We changed the reference list to "(Kroll and Seinfeld, 2008; Hallquist et al., 2009)".

Page 24077 after Line 21: We added a new reference "Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoffmann, T., Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., Maenhaut, W., McFiggans, G., Mentel, Th. F., Monod, A., Prévôt, A. S. H., Seinfeld, J. H., Surratt, J. D., Szmigielski, R., and Wildt, J.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, Atmos. Chem. Phys., 9, 5155–5236, 2009"

Page 24078 after Line 23, We added a new reference "Kroll, J. H. and Seinfeld, J. H.: Chemistry of secondary organic aerosol: Formation and evolution of low-volatility organics in the atmosphere, Atmos. Environ., 42, 3593–3624, 2008"

3- *Pg 24056 Line 11: With exception of the Jimenez et al. (2009) reference, placing the other references here are somewhat misleading as they do not focus on SOA aging and hygroscopicity.*

We replaced the references with ones describing hygroscopicity and evolution of SOA. We modified the manuscript as follows.

Page 24055 Line 11: We changed the reference list to "nuclei (Jimenez et al., 2009; Massoli et al., 2010: Poulain et al., 2010).

Page 24079 before Line 1: We added a reference "Massoli, P., Lambe, A. T., Ahern, A. T., Williams, L. R., Ehn, M., Mikkilä, J., Canagaratna, M. R., Brune, W. H., Onasch, T. B., Jayne, J. T., Petäjä, T., Kulmala, M., Laaksonen, A., Kolb, C. E., Davidovits, P., and Worsnop, D. R.: Relationship between aerosol oxidation level and hygroscopic properties of laboratory generated secondary organic aerosol (SOA) particles, *Geophys. Res. Lett.*, 37, L24801, doi:10.1029/2010GL045258, 2010"

Page 24079 after Line 21: We added a reference "Poulain, L., Wu, Z., Petters, M. D., Wex, H., Hallbauer, E., Wehner, B., Massling, A., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and CCN activation for secondary organic aerosols—Part 3: Influence of the chemical composition on the hygroscopic properties and volatile fractions of aerosols, Atmos. Chem. Phys., 10, 3775–3785, doi:10.5194/acp-10-3775-2010, 2010"

4- *Pg 24056 Line 19: Shilling et al. (2009) might not be appropriate reference here as they did not study SOA aging in particular, unless by "aging" you are also including atmospheric dilution. Shilling et al. (2009) would be more appropriately cited after sentence in Line 25-26 when discussing partitioning and dilution.*

We agree with the comments because the dilution effect was also described in somewhere else, thus we moved the reference Shilling et al. (2009) from line 19 to line 26 in Page 24055. Thus, we changed the reference list as follows.

Page 24055 Line 19: "(Jimenez et al., 2009; George and Abbatt, 2010)".

Page 24055 Line 25-26: "…to oligomer formation (Donahue et al., 2006; Rudich et al., 2007; Shilling et al., 2009)."

5- *Pg. 24056 Line 16-17: Explain what is meant by the phrase "probably because the aging much less than in the atmosphere"? Do you mean aging timescale is shorter? Or the experiments do not include all aging mechanisms such as dilution and cloud processing? Also, what is specifically meant by 'this limitation'?*

We have modified the paragraph to take to clarify our comment and have replaced the middle of the paragraph with the following.

Page 24056 Line 13: We removed the 4th and 5th sentences starting with "For large environmental chambers,…" and "It has been possible to overcome…" and added these sentences "For most large environmental chambers, the chemical composition of OA is much more hydrocarbon-like than the OA observed in the atmosphere (Bahreini et al., 2005; Alfarra et al., 2006; Zhang et al., 2006; Shilling et al., 2009; Ng et al., 2010), probably because the exposure to oxidants is much less than in the atmosphere, especially in case of rural or remote regions. Even when oxidant levels are increased in large environmental chambers, it is difficult to achieve the exposure to oxidants that occurs in the atmosphere. Attempts to increase the exposure to oxidants have used highly oxidative environments in small chambers and flow tubes (Jimenez et al., 2009; Kroll et al., 2009; George and Abbatt, 2010), but the resulting chemical compositions has too much SV-OOA for the observed amounts of LV-OOA (Ng et al., 2010)."

6- *Pg. 24057-24058: Second and third paragraphs under Section 2 do not pertain to experimental methods and would be better in the introduction when discussing different OA oxidation types OOA, HOA etc. Typical values for O/C ratios would be useful to state in the second paragraph instead qualitative statements such as "SV-OOA. . . has a low ratio of oxygen to carbon (O:C)".*

We agree to move the second and third paragraphs under Section 2 to the Section 1. We modified the manuscript as follows.

Page 24057 Line 13: We removed the first sentence of this paragraph starting with "The mass spectrum of OA enables…" and added the sentence "The degree of OA oxidation is oftened studied with an aerosol mass spectrometer (AMS), which vaporizes incoming OA and then samples the mass spectrum of the vaporized, ionized molecular fragments (Zhang et a., 2005; Jimenez et al., 2009; Ng et al., 2010).” instead.

Page 24057 Line 18-19: We changed the words from “, and has more m/z 43 than m/z 44 contributions” to “, and has m/z 43 contributing more than m/z 44”.

Page 24057 Line 21: We removed the sentence starting with “While Principle Compoenent Analysis provides…”.

Page 24058 Line 4: We changed the words from “For this study, f_{43} and f_{44} are defined…” to “Typically for laboratory studies, f_{43} and f_{44} can be defined…”.

Page 24058 Line 7: We added the sentence “While Principle Component Analysis provides a more rigorous characterization of atmospheric OA, the use of f_{43} and f_{44} is sufficient to track the degree of oxidation in VOC precursor laboratory experiments (e.g., Shilling et al., 2009; Ng et al., 2010).”

And then, we moved these modified 2nd and 3rd paragraph under Section 2 to Page 24056 Line 22 prior to the last paragraph of the Section 1.

7- *Pg. 24064: Section 3.1.3 is unnecessary. The point about F44 and F43 values differing with fragmentation table modifications can be emphasized in the previous subsection.*

We agree to delete the Section title “3.1.3 Oxidation indicators f44 and f43”. Also, we moved the section “3.1.2 The determination of OA mass concentration for this study” to the Section 2 with Section title “2.4. The determination of OA mass concentration”, because we thought the OA mass concentration determination should be described as a methodology under the method section. We modified the manuscript as follows:

Page 24064 Line 19: We deleted the Section title “3.1.3 Oxidation indicators f44 and f43”.

Page 24064 Line 20: We deleted the 1st and 2nd sentences starting with “The fractions f_{44} and f_{43} were…” and “The OA mass concentration was…”, and then we moved the whole paragraph to Page 24064 Line 5 after the 4th paragraph of that section.

Page 24064 Line 7: We changed the figure number from “(Fig. 3)” to “(Fig. 2)” because we re-arranged the figures.

Page 24062 line 20: We changed section title “3.1.2 The determination of OA mass concentration for this study” to “2.4 The determination of OA mass concentration” and moved the whole section to Page 24061 Line 28 under the Section 2.

Page 24062 Line 3: We deleted the sub-section title “3.1.1 Mass spectra of SOA”.

8- *Pg 24067 Lines 16-17: “Oxidation pathways seem to be divided to fragmentation and oligomerization. . .” please also add “functionalization”. Also, conclusions cannot be made on importance of oligomerization from this work.*

Yes. The current AMS system cannot recognize the oligomerization of organics because the organic compounds are fragmented into small functional groups. We would like to replace “oligomerization” to “functionalization” and modify the manuscript as follows.

Page 24067 Line 15-16: We changed the words in the sentence from "… fragmentation can occur, resulting in more volatile organics and loss of OA mass concentration." to "… fragmentation can occur, resulting in breaking carbon bonds and loss of OA mass concentration."

Page 24067 Line 16-19: We removed the sentence starting with "Oxidation pathways…". We added the sentences "Functionalization and oligomerization can also occur (Gross et al., 2006; Jimenez et al., 2009), but in this study functionalization was not the dominant process based on the O:C ratio increase and OA mass concentration decrease with oxidation, which is similar to the conclusion of Kroll et al. (2009) and Heald et al. (2010). In this study, oligomerization is difficult to study because of the extensive fragmentation by the Q-AMS."

9- *Pg 24069 Line 3: "oxidation saturates" Please clarify this point. Do you mean that under high OH exposures OH adsorbs to particle surface and takes up all surface sites via Langmuir-Hinshelwood mechanism or that reaction kinetics of OH with surface organic species do not slow down with increasing OH exposure?*

We mean that the oxidation, as indicated by f_{44} increases linearly with OH. We revise the sentence as follows.

Page 24068 Line 29 to Page 24069 Line 4: We removed the sentences starting with "However, if the oxidation is occurring on the …" and "This observation provides additional…". And, we added the sentences "Since the OH concentration, [OH], is simply the OH exposure divided by the PAM chamber reisdence time, the oxidation indicated by the f_{44} increase and f_{43} decrease is linear with [OH]. If the oxidation is occurring on the particle surface, then the surface reaction kinetics are linear with [OH] no matter what the exact surface reaction mechanism is. If the oxidation is occuring in the gas phase, then the reaction kinetics appear to be first-order in [OH]. This linearity holds for [OH] from 10^9 cm^{-3} to 10^{10} cm^{-3}; additional studies will be needed to see if this linearity holds down to atmospheric levels of [OH] from 10^6 cm^{-3} to 10^7 cm^{-3}. Whether the oxidatin occurs in the gas phase or on the particle phase, this observation suggests that the high levels of OH in the PAM chamber can possibly be used to simulate atmospheric oxidation." instead.

10- *Table 1: Could you state the SOA yield for each experiment? Are OH exposures the corrected values?*

We added the SOA yield for each experiment in Table 1 as already shown above

OH exposures are corrected values by the photochemical modeling to include the reduction of OH exposure by the presence of the high VOC amounts as described in Page 24060 Line 11-18. In the simple model used to make the corrections to the OH exposure in this study, we included a simplified form of the reaction chemistry for these three VOCs. That is why the OH exposure value was different for various HC concentrations under the same OH concentration. It should be pointed out that ozone reactions with the VOCs and their products can produce OH as well as destroy it and the OH yields need to be taken into account. As a result of the uncertainty in the simple models used, we have added estimated uncertainty error bars (2σ confidence level) on the OH exposure in Table 1.

11- *Figure 2: Can these mass spectra be compared with reference chamber SOA spectra for SOA produced under similar conditions to those in this study, for example plot reference spectra on the respective plots and/or state correlation coefficients in the text?*

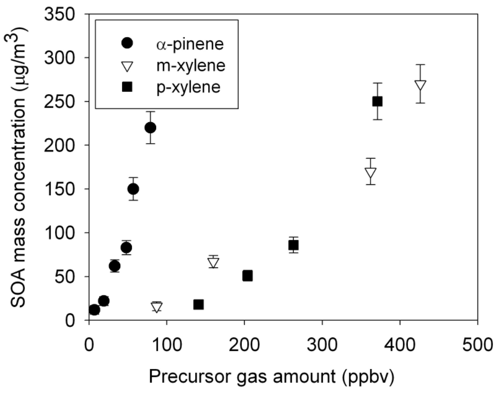
Yes, although it isn't clear to us what purpose this would serve that isn't addressed in the detailed comparisons of f_{43} and f_{44} that we show, since these are the dominant fragments that are most readily assignable to likely parent compounds. We believe that the AMS-reader can compare the representative spectra that we present in Figure 3 to those published by others. Our goal in this paper is not to assign spectra to specific compounds, which is more difficult with a Q-AMS than a high resolution ToF-AMS that many are now using. But, we have added a series of difference spectra (as described above) that help to highlight the main points of this study.

12- *Figure 4. Why do F44 values have such large errors at low OA masses?*

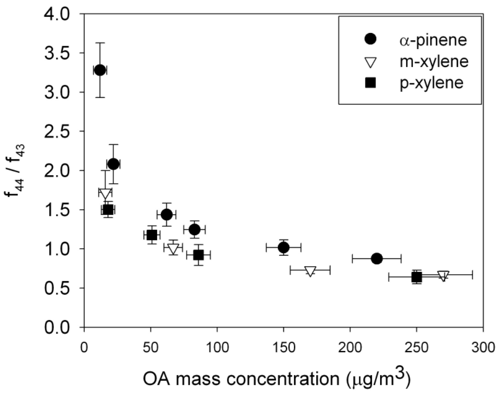
In fact, these values were computed incorrectly. We have recalculated the standard deviations for these low-organic points and the errors are much smaller and replaced the figure 4 with correctly computed error values.

Figure 4. We replaced the figure as follows:

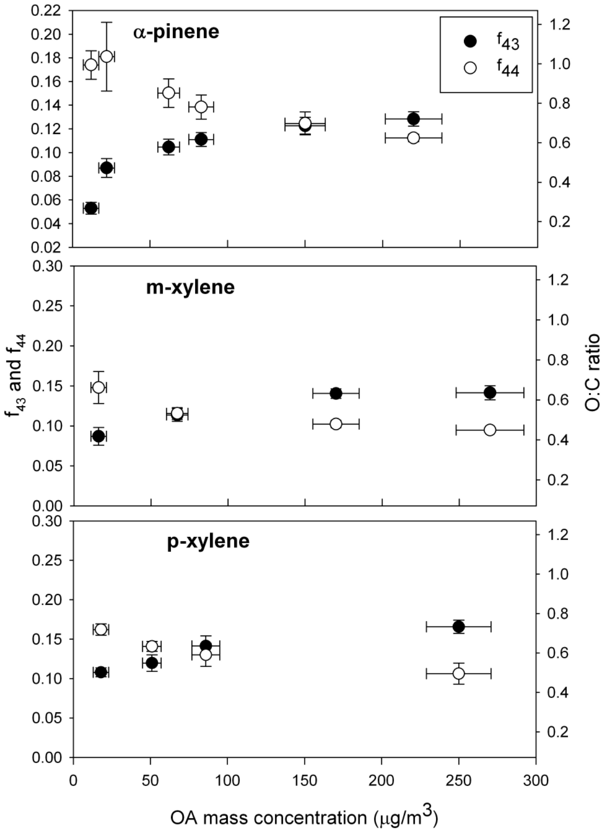
(a) OA concentration vs. Precursor amount



(b) f_{44}/f_{43} vs. OA concentration



(c) f_{43} and f_{44} vs. OA concentration

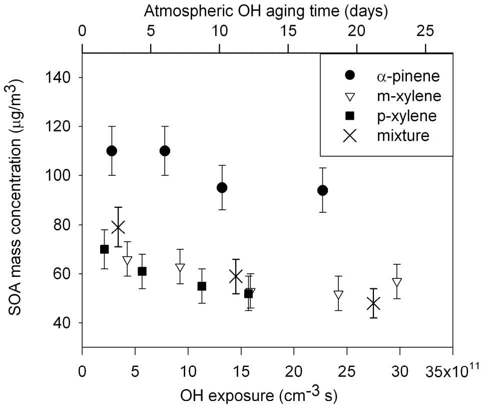


13- *Figure 5: Please show a second x axis for atmospheric OH aging time equivalent to the OH exposure assuming an atmospherically relevant average daily OH concentration.*

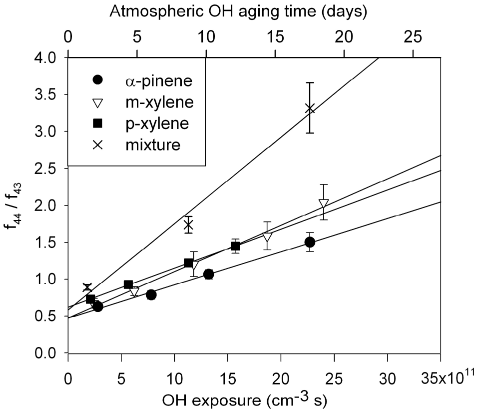
Based on the typical diurnally averaged OH concentration of 1.5×10^6 molecules cm^{-3}, atmospheric OH aging time in days was plotted in Fig. 5. We modified the manuscript as follows.

Figure 5: We replaced a modified figure with the second x axis for atmospheric OH aging time as follows:

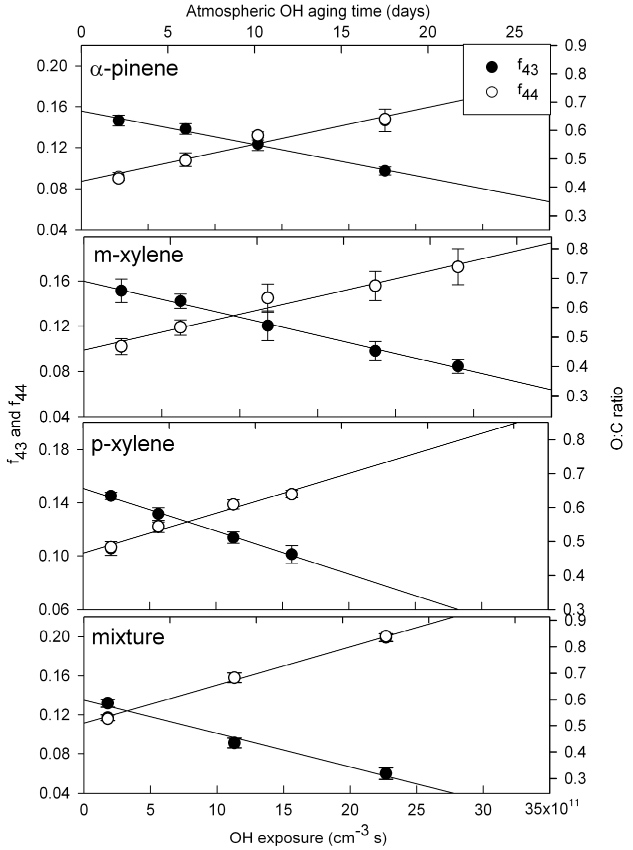
(a) OA concentration vs OH exposure



(b) f_{44}/f_{43} vs OH exposure



(c) f_{44} and f_{43} vs OH exposure



"Figure 5. SOA oxidation dependent on the OH exposure at a constant VOC amount of 39±6 ppbv of α-pinene, 170±30 ppbv of m-xylene, 180±30 ppbv of p-xylene, and a mixture of three (37±6 ppbv of α-pinene, 46±7 ppbv of m-xlene, 47±8 ppbv of p-xylene). OA mass concentration was measured by the TEOM. The error bars for OA mass concentration, f_{43}, and f_{44} are at the 1σ confidence level. Atmospheric OH aging time was obtained with a typical diurnally averaged OH concentration of 1.5×10^{6} molecules cm^{-3} (Mao et al., 2009). Uncertainties in the OH exposures are correlated so that the OH exposure scale could expand or shrink by the uncertainty ranges given in Table 1. (a) OA mass concentration vs. OH exposure. (b) The ratio of f_{44} to f_{43} vs. OH exposures. Linear regressions of each lines are shown here; for α-pinene, slope=4.50×10^{-13}, intercept=0.47, r^2=0.99; for m-xylene, slope=6.29×10^{-13}, intercept=0.48, r^2=0.99; for p-xylene, slope=4.46×10^{-13}, intercept=0.64, r^2=0.99; and for the mixture, slope=1.17×10^{-12}, intercept=0.58, r^2=0.99. (c) The values of f_{43} and f_{44} vs. OH exposures. O:C ratio was calculated from f_{44} values based on Aiken et al. (2008)."

Page 24078 after Line 33: We added a new reference, "Mao, J., Ren, X., Brune, W. H., Olson, J. R., Crawford, J. H., Fried, A., Huey, L. G., Cohen, R. C., Heikes, B., Singh, H. B., Blake, D. R., Sachse, G. W., Diskin, G. S., Hall, S. R., and Shetter, R. E.: Airborne measurement of OH reactivity during INTEX-B, Atmos. Chem. Phys., 9, 163-173, doi:10.5194/acp-9-163-2009, 2009."

References:

Chow, J. D., Doraiswamy, P., Watson, J. G., Chen, L.-W. A., Ho, S.S.H., and Sodeman, D. A.: Advances in integrated and continuous measurements for particle mass and chemical composition. J. Air & Waste Manage. Assoc. 58, 141-163, 2008

Matthew, B. M., Middlebrook, A. M., Onasch, T. B.: Collection efficiencies in an Aerodyne Aerosol Mass Spectrometer as a function of particle phase for laboratory generated aerosols, Aerosol. Sci. Tech., 42(11), 884-898, 2008.

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Salcedo, D., Onasch, T. B., Dzepina, K., Canagaratna, M. R., Zhang, Q., Huffman, J. A., DeCarlo, P. F., Jayne, J. T., Mortimer, P., Worsnop, D. R., Kolb, C. E., Johnson, K. S., Zuberi, B., Marr, L. C., Volkamer, R., Molina, L. T., Molina, M. J., Cardenas, B., Bernabe, R. M., Marquez, C., Gaffney, J. S., Marley, N. A., Laskin, A., Shutthanandan, V., Xie, Y., Brune,W., Lesher, R., Shirley, T., and Jimenez, J. L.: Characterization of ambient aerosols in Mexico City during the MCMA-2003 campaign with Aerosol Mass Spectrometry: results from the CENICA Supersite, Atmos. Chem. Phys., 6, 925–946, 2006.

Virtanen, A., Joutsensaari, J., Koop, T., Kannosto, J., Yli-Pirilä, Leskinen, J., Mäkelä, J. M., Holopainen, J. K., Pöschl, U., Kulmala, M., Worsnop, D. R., Laaksonen, A.: An amorphous solid state of biogenic secondary organic aerosol particles, Nature, 468, doi:10.1038/nature09455, 2010.

Minor and Technical corrections by Authors:

Abstract line 14: We changed the words "…, with linear f_{44} increase and f_{43} decrease." to "…, with f_{44} increasing and f_{43} decreasing."

Page 24055 Line 29: We changed the words "by the ultraviolet light (UV)" to "by the ultraviolet (UV) light".

Page 24058 Line 13: We changed the words "UV grid lamps" to "ultraviolet (UV) grid lamps".

Page 24056 Line 29: We chanted the words "During those experiments,…" to "During some additional experiments, …".

Page 24059 Line 18: We corrected the typos from "60 pptv to 500 pptv" to "60 pptv to 480 pptv".

Page 24060 Line 27: We corrected the typos from "60 pptv to 500 pptv" to "60 pptv to 480 pptv".

Page 24060 line 18: We added the reference list here "1.5×10^{6} molecules cm^{-3} (Mao et al., 2009)".

Page 24062 Line 5: We changed the figure number from "are shown in Fig. 2" to "are shown in Fig. 3" because we rearranged the figure 2 and 3.

Page 24067 Line 4-6: We corrected the VOCs concentration by replacing the sentence to "Initial VOCs concentrations were 39±6 ppbv of α-pinene, 170±30 ppbv of m-xylene, 180±30 ppbv of p-xylene, and a mixture of 37±6 ppbv of α-pinene, 46±7 ppbv of m-xylene and 47±8 ppbv of p-xylene."

Page 24065 Line 4: We corrected the number from "fixed to 259 pptv" to "fixed to 260 pptv" for 2-significant figure.

Page 24065 Line 5: We corrected the VOCs concentration numbers including error ranges from"…from 7 to 79 pptv, m-xylene from 87 to 426 pptv, and p-xylene from 141 to 371 pptv" to "from 7±1 to 79±13 ppbv, m-xylene from 87±14 to 430±70 ppbv, and p-xylene from 140±30 to 370±60 ppbv".

Page 24061 Line 10: We changed the words "… species by evaporation. Aerosol mass…" to "… species by evaporation. The aerosol mass…".

Page 24061 Line 16: We changed the words "… and only brief description …" to "… and only a brief description …".

Page 24063 Line 18: We changed the words "… sensitivity or tune of the …" to "… sensitivity or tuning of the …".

Page 24065 Line 2: We corrected the typos from "… of three VOCs, α-pinene, …" to "… of three VOCs: α-pinene, …".

Page 24065 Line 26: We changed the words "a rapid decrease in the ratio with…" to "a rapid decrease in f_{44}/f_{43} with…".

Page 24065 Line 26-27: We changed the words ", leveling off at higher OA mass concentrations" to "and a much smaller decrease at higher OA mass concentrations".

Page 24065 Line 27: We changed the numbers "mixing ratio, 7 ppbv, …" to "mixing ratio, 7±1 ppbv,…".

Page 24066 line 6-7: We change the numbers with error range and 2-significant figure values from "11.8 to 216 μg/m^{-3}. … within 16 and 271 μg/m^{-3}" to "12±5 to 220±18 μg m^{-3}., … within 16±5 and 270±22 μg m^{-3}.".

Page 24066 Line 14: We removed the sentence "The ratio of f44 to f43 shows similar behavior" and added a new sentence "The decrease in the ratio of f_{44} to f_{43} with increasing OA mass concentration is another way to show this behavior.".

Page 24066 Line 21: We changed the words from "OA mass concentration than α-pinene …" to "OA mass concentration than for α-pinene …".

Page 24067 Line 8: We changed the words from "… the relative humidity and added VOC amounts" to "… the relative humidity."

Page 24068 Line 10: We corrected the typos in the reference list from "…Bahreini et al. (2006)" to "…Bahreini et al. (2005)".

Page 24068 Line 26: We changed the words from " … to all AMS masses,…" to "…to several AMS masses,…".

Page 24071 Line 19: We added the words from "… Caltech environmental chamber are consistent …" to "…Caltech environmental chamber (orange rectangle in Fig. 7) are consistent …”.

Page 24071 Line 21: We added the full name of LBNL here to “study at Lawrence Berkeley National Laboratory (LBNL) (Jimenez et al., 2009) …”.

Page 24072 Line 24: We removed the full name of LBNL here to “The differences between the LBNL study…”.

Page 24072 Line 18: We corrected the typo by removing “… into m/z 43 and m/z44 groups (or that there…“ to “… into m/z 43 and m/z 44 groups or that there…”.

Page 24074 Line 3: For smooth expression, we changed the words from “A caveat in the comparisons of the results of this study from those of other atmospheric and laboratory studies is the possible differences …” to “The comparisons of this study to others are subject to other caveats -the possible differences…”.

We used the consistent expression “vs.” instead of “versus” for the followings.
Page 24068 Line 18: “f_{44}/f_{43} vs. OH exposure”.
Page 24068 Line 21: “f_{44} vs. OH exposure…”.
Page 24068 Line 22: “f_{43} vs. OH exposure…”
Page 24070 Line 17: “f_{44} vs. f_{43}…”
Page 24072 Line 1: “f_{44} vs. f_{43}…”

Figure 2 Line 3-4: We changed the sentence from “…are the 1σ standard deviation (precision).” To “…are at the 1σ confidence level.”