Response to Reviewer 1

We greatly appreciate the time and effort that reviewer 1 spent in reviewing our manuscript. The comments are really thoughtful and helpful to improve the quality of our paper. Reviewer 1 has provided both main comments and other specific comments. Below we make a point-by-point response to these comments. According to editor's requirement, the response to the referee 1 is structured in the following sequence: (1) comments from the referee in black color, (2) our response in blue color, and (3) our changes in the revised manuscript in red color.

Zhang et al. presented a chamber study that examined the effect of RH on SOA mass yields and composition. This paper is potentially useful to the SOA community. However, there are portions of the manuscripts that need to be addressed before the manuscript can be considered for publication.

1. Page 4 line 3: Clarify how H2O2 and m-xylene were introduced into the chamber. Via an injection into a glass bulb using a syringe? Using a bubbler? How did the authors determine when the chamber contained 20 ppm of H2O2? Was the concentration of gas-phase H2O2 in the chamber measured in real-time? If yes, what instrument was used?

H_2O_2 and m-xylene were introduced into the reactor along with the zero air flow via an injection into a three-way tube using a syringe. The concentration of gas-phase H_2O_2 in the reactor was not measured but calculated. To obtain a certain concentration of H_2O_2, the density and mass concentration of injected H_2O_2 solution, and the volume of the reactor were used to calculate the volume of H_2O_2 solution that needed to be injected.

2. Page 4 line 5: Explain the rationale behind not using any seed aerosols in this study. Seed aerosols are typically used in chamber studies to promote the condensation of SOA-forming vapors onto seed aerosol instead of the chamber walls. The mass yields reported by the authors are likely under-estimated since most of the vapors are likely lost the chamber walls in these experiments (See examples provided in Zhang et al., PNAS 2014, Nah et al., ACP 2016, 2017). Vapor wall loss is also going to affect the

types of products formed in these SOA experiments since highly oxygenated and least volatile compounds are lost to the chamber walls are faster rates (See Zhang et al., ACP 2015). The authors should comment on how vapor wall loss affects their results. Can they also provide an estimation on how much their SOA mass yields are underestimated by?

Nah, T., McVay, R. C., Zhang, X., Boyd, C. M., Seinfeld, J. H., and Ng, N. L.: Influence of seed aerosol surface area and oxidation rate on vapor wall deposition and SOA mass yields: a case study with α-pinene ozonolysis, Atmos. Chem. Phys., 16, 9361-9379, https://doi.org/10.5194/acp-16-9361-2016, 2016.

Nah, T., McVay, R. C., Pierce, J. R., Seinfeld, J. H., and Ng, N. L.: Constraining uncertainties in particle-wall deposition correction during SOA formation in chamber experiments, Atmos. Chem. Phys., 17, 2297-2310, https://doi.org/10.5194/acp-17-2297-2017, 2017.

Zhang, X., Schwantes, R. H., McVay, R. C., Lignell, H., Coggon, M. M., Flagan, R. C., and Seinfeld, J. H.: Vapor wall deposition in Teflon chambers, Atmos. Chem. Phys., 15, 4197-4214, https://doi.org/10.5194/acp-15-4197-2015, 2015.

We agree with the reviewer that seed aerosols can promote the condensation of SOA-forming vapors onto seed aerosol instead of the chamber walls. However, inorganic salt can both participate into the SOA formation and change the reaction environment such as providing acidic surface and aqueous environment. These properties of seed aerosols probably interfere with the RH effect on SOA formation, as the RH combined with seed aerosols complicate the m-xylene-OH system. Losses of organic vapors to the chamber wall can be substantial. The fact that seed aerosols were not artificially introduced can probably lead to the underestimation of SOA. Thus, we have added a paragraph to comment on how vapor wall loss affects our results at the end of Sec. 3.1 in the revised manuscript, but we cannot provide a factor of underestimation of SOA yields.

It should be noted that seed aerosols were not artificially introduced throughout all the experiments, which could lead to the underestimation of SOA, as SOA-forming vapors partly condense to the chamber walls instead of particles (Matsunaga and Ziemann, 2010; Zhang et al., 2014). The extent to which vapor wall deposition affects SOA mass yields depends on the specific parent hydrocarbon system (Zhang et al., 2014; Zhang et al., 2015; Nah et al., 2016; Nah et al., 2017). Zhang et al (2014) have estimated two

m-xylene systems under low NOx conditions and concluded that SOA mass yields were underestimated by factors of 1.8 (Ng et al., 2007) and 1.6 (Loza et al., 2012) under low RH conditions. In addition, the excess use of H_2O_2 can lead to an excess OH radicals, leading to a less underestimation of SOA formation as the losses of SOA-forming vapors can be mitigated via the use of excess oxidant concentrations (Nah et al., 2016). Thus, the underestimation of SOA formation can be limited. In fact, the wall loss of m-xylene was not taken into consideration of calculation of mass yields, which generally overestimates the mass yields.

Loza, C. L., Chhabra, P. S., Yee, L. D., Craven, J. S., Flagan, R. C., and Seinfeld, J. H.: Chemical aging of m-xylene secondary organic aerosol: laboratory chamber study, Atmos. Chem. Phys., 12, 151-167, 10.5194/acp-12-151-2012, 2012.

Matsunaga, A., and Ziemann, P. J.: Gas-wall partitioning of organic compounds in a Teflon film chamber and potential effects on reaction product and aerosol yield measurements, Aerosol Sci. Technol., 44, 881-892, 10.1080/02786826.2010.501044, 2010.

Nah, T., McVay, R. C., Zhang, X., Boyd, C. M., Seinfeld, J. H., and Ng, N. L.: Influence of seed aerosol surface area and oxidation rate on vapor wall deposition and SOA mass yields: a case study with α-pinene ozonolysis, Atmos. Chem. Phys., 16, 9361-9379, 10.5194/acp-16-9361-2016, 2016.

Nah, T., McVay, R. C., Pierce, J. R., Seinfeld, J. H., and Ng, N. L.: Constraining uncertainties in particle-wall deposition correction during SOA formation in chamber experiments, Atmos. Chem. Phys., 17, 2297-2310, 10.5194/acp-17-2297-2017, 2017.

Ng, N. L., Kroll, J. H., Chan, A. W. H., Chhabra, P. S., Flagan, R. C., and Seinfeld, J. H.: Secondary organic aerosol formation from m-xylene, toluene, and benzene, Atmos. Chem. Phys., 7, 3909-3922, 10.5194/acp-7-3909-2007, 2007.

Zhang, X., Cappa, C. D., Jathar, S. H., McVay, R. C., Ensberg, J. J., Kleeman, M. J., and Seinfeld, J. H.: Influence of vapor wall loss in laboratory chambers on yields of secondary organic aerosol, Proc. Natl. Acad. Sci. U. S. A., 111, 5802-5807, 10.1073/pnas.1404727111, 2014.

3. Page 4 line 11: How were the particle wall loss rates determined? In seed aerosols only experiments? Were these particle wall loss rates measured by tracking the decay of the aerosol mass or volume? How often were particle wall loss experiments

conducted? Were the reported particle wall loss rates consistent with previously measured rates? Was the particle wall loss rate always faster in high RH experiments or is this measurement within experimental uncertainty?

Particle wall loss rates were generally measured in seed aerosol experiments by tracking the decay of the aerosol volume. For the same volume of new reactor, the wall loss rates were evaluated. We also checked the wall loss rate for the old reactor. Particle wall loss rate constant varies from 3×10^{-5} s^{-1} to 6×10^{-5} s^{-1} at the RH range of 5% to 90% with a trend of increase with RH, but their relationship is not statistically significant. The average particle wall loss rate constant is $(3.8 \pm 0.8) \times 10^{-5}$ s^{-1} at (13 ± 10)% RH and $(4.2 \pm 1.8) \times 10^{-5}$ s^{-1} at (79 ± 10)% RH, respectively. The relatively large wall loss rate at high RH and small wall loss rate at low RH are used in our correction of particle wall loss to look at RH effects in this study.

4. Page 4 line 14: It is not clear how the aerosol LWC was calculated. More details should be provided.

Taking the reviewer's advice, we have added some sentences about the details of LWC measurement at the end of the first paragraph of Sec. 2.2.

Thus, here a brief introduction is only given. After the lights were turned off in high RH experiments, the SMPS was modified to the dry mode through adding a Nafion dryer (Perma Pure MD-700-12F-3) to the sampling flow and a Nafion dryer (Perma Pure PD-200T-24MPS) to the sheath flow, leading to the reduction of RH in the sample air to 10 % and that in the sheath to 7 %. After modifying to the dry mode, the humid air in SMPS was quickly replaced by dry air through venting the sheath air at 5 L min^{-1}, and then the dry aerosol was measured by SMPS. The LWC was determined by the difference of the particle mass concentrations before and after the modification of the dry mode.

5. Page 4 line 20: The PILS only samples water-soluble species in the SOA, not the total SOA composition. Hence, the compositional results reported by the authors in this study are really the water-soluble species, and the authors should specify this in their

manuscript. On a related note, why did the authors decided to collect aerosol samples with a PILS instead of on filters. Filter collection and analysis would have allowed them to analyze both the water-soluble and water-insoluble species. Do the authors know what fraction of the SOA formed is composed of water-soluble vs. water-insoluble species?

We agree with the reviewer that the PILS samples water-soluble species in the SOA. Nevertheless, after the FTIR measurement of SOA samples collected on ZnSe windows, the ZnSe window was washed with ultrapure water and was measured by FTIR again, no absorbance was observed on FTIR spectra. It can be believed that the SOA compositions are almost all water-soluble species and the PILS samples almost all SOA components. In addition, we agree with the reviewer that filter collection samples both water-soluble and water-insoluble species in the SOA. However, the filter-based analysis has its limitation, including adsorption of organic vapors and evaporation of semi-volatile organic compounds from the filter surface, leading to some uncertainties in the identification of SOA components. Moreover, Bateman et al. (2010) compared the off-line mass spectra of SOA samples from limonene ozonolysis collected by PILS with those collected on filters and found that the peak abundance, organic mass to organic carbon ratios, and the average O:C ratio are essentially identical. Water-soluble species account for the vast majority of SOA.

Bateman, A. P., Nizkorodov, S. A., Laskin, J., and Laskin, A.: High-resolution electrospray ionization mass spectrometry analysis of water-soluble organic aerosols collected with a particle into liquid sampler, Anal. Chem., 82, 8010-8016, 10.1021/ac1014386, 2010.

6. Page 5 line 8: Show the corresponding reaction time profile of m-xylene measured by the GC-MS that accompanied the observed SOA growth for the four experiments. This can be placed in the supplementary information. It is currently unclear how quickly the reactions took place. Perhaps the time profiles can be used to explain the differences in SOA formation in dry vs. humid conditions? For example, did m-xylene react faster in the dry experiments thus resulting in higher SOA mass yields? Ng et al., ACP 2007 previously showed that SOA formation in the m-xylene system will be faster at faster oxidation rates. From Fig. 1, it looks like peak SOA growth was not achieved at the end

of the dry experiments (SOA mass looks like it may still increase). Why the authors decide to stop these dry experiments early? Won't that affect their calculated SOA mass yields?

Ng, N. L., Kroll, J. H., Chan, A. W. H., Chhabra, P. S., Flagan, R. C., and Seinfeld, J. H.: Secondary organic aerosol formation from m-xylene, toluene, and benzene, Atmos. Chem. Phys., 7, 3909-3922, https://doi.org/10.5194/acp-7-3909-2007, 2007.

Taking the reviewer's advice, we have added the reaction time profile of m-xylene measured by the GC-MS that accompanied the observed SOA growth for the four experiments in the supplementary information (see Fig. S1).

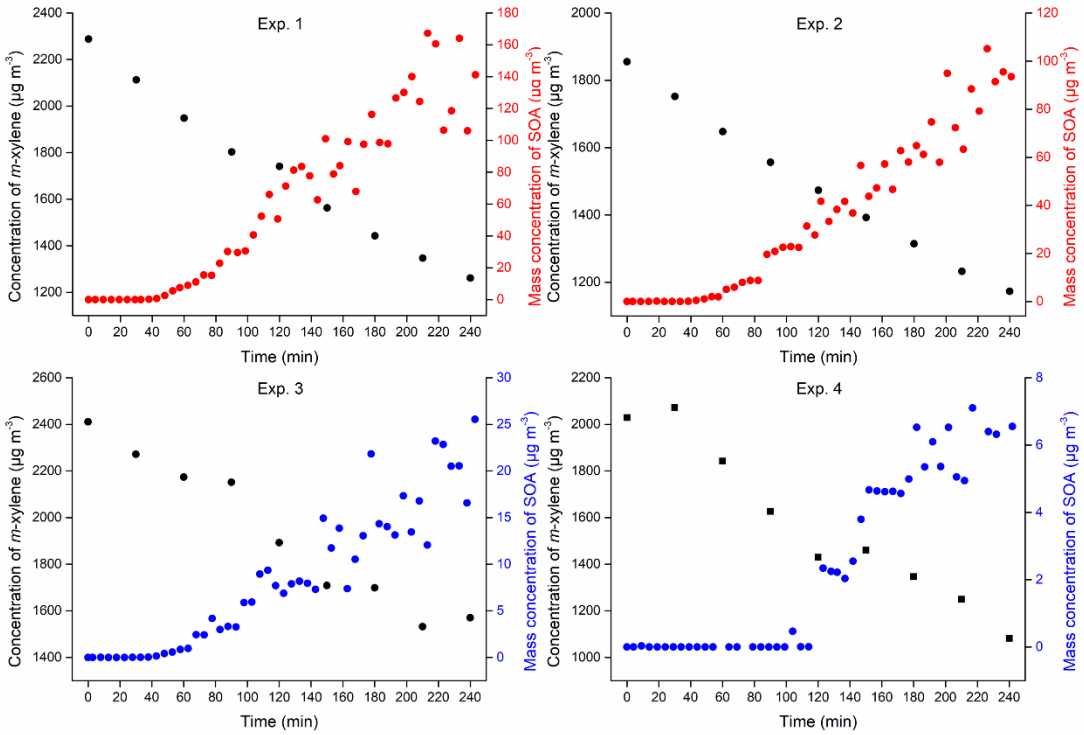


Fig. S1. Reaction time profiles of *m*-xylene measured by the GC-MS that accompanied the observed SOA growth for the four experiments

As shown in the time profile in Fig. S1, the reacted *m*-xylene account for around 40% of the initial *m*-xylene in both high and low RH experiments. *m*-Xylene did not react faster in the dry experiments which may lead to the higher SOA formation.

As the reviewer pointed out, peak SOA growth was not achieved at the end of the dry experiments from Fig. 1 and SOA mass still increase. As the experiments were

conducted under low NO_x condition, the SOA mass will increase unless *m*-xylene has all reacted. The SOA mass formation and reacted *m*-xylene are both nearly linear. The SOA yields will be basically constant no matter when we stop the reaction. In addition, the SOA-forming vapor could loss less if the reaction time was relatively short 4 h. Thus, we decided to stop the experiments early.

7. Page 5 line 21: Regarding the authors' definition of SOA yield, did they calculate the SOA yield by dividing the SOA mass obtained at the end of the experiment by the total reacted m-xylene at the end of the experiment? If yes, why did they decide to use this calculation? Previous chamber studies calculated the SOA mass yield by taking the ratio of the SOA mass at peak SOA mass divided by the mass of VOC reacted. Was peak SOA mass only reached at the end of each experiment (reaction time profiles of SOA mass growth with the corresponding reacted m-xylene for the four experiments will be useful; see comment 6)? Related to this point, are the authors confident that peak SOA mass have already occurred before they ended their experiment. Given that the authors are comparing their measured SOA mass yields with previous studies, they should make sure that their calculation of SOA mass yields are consistent with those of previous studies before they compare mass yields.

As the reviewer pointed out, the SOA yield in this study is defined by the ratio of the SOA mass obtained at the end of the experiment to the total reacted m-xylene at the end of the experiment. As the experiments were conducted under low NOx condition, the SOA mass would increase unless all m-xylene reacted. The experiment for 4-6 h is a ubiquitous reaction time used in many previous studies. Indeed, the SOA yield generally increases with time. If the relationship between the yield and time is extrapolated to 6 h, the yield is increased by 45% relative to that at 4 h, which can be compared with many previous studies (Cao and Jang, 2010; Hinks et al., 2018). Most importantly, as the purpose of our study is to investigate the RH effect on SOA formation, the reaction time of 4 h is sufficient to compare the SOA formation and to sample for SOA component analysis. Furthermore, a relatively short reaction time can minimize the wall loss of oxidized species and limit the further SOA mass uncertainty.

Cao, G., and Jang, M.: An SOA model for toluene oxidation in the presence of inorganic aerosols, Environ. Sci. Technol., 44, 727-733, 10.1021/es901682r, 2010.

Hinks, M. L., Montoya-Aguilera, J., Ellison, L., Lin, P., Laskin, A., Laskin, J., Shiraiwa, M., Dabdub, D., and Nizkorodov, S. A.: Effect of relative humidity on the composition of secondary organic aerosol from the oxidation of toluene, Atmos. Chem. Phys., 18, 1643-1652, 10.5194/acp-18-1643-2018, 2018.

8. Page 5 line 23: How was LWC subtracted from the SOA measurement? How did the authors determine the amount of LWC in the aerosols? The authors should briefly describe this process even if this was previously mentioned in one of their previous paper. The sentence "It should be pointed out that...would evaporate back into the gas phase when aerosol water is removed" is confusing. The experimental section did not mention that authors removed aerosol water prior to SMPS measurement. If aerosol water was not removed prior to SMPS measurement, then this sentence seems out of place. Unless the authors are proposing a hypothetical situation?

Taking the reviewer's advice, we have added some sentences about the details of LWC measurement at the end of the first paragraph of Sec. 2.2 (same with the reply of Comment 4).

Thus, here a brief introduction is only given. After the lights were turned off in high RH experiments, the SMPS was modified to the dry mode through adding a Nafion dryer (Perma Pure MD-700-12F-3) to the sampling flow and a Nafion dryer (Perma Pure PD-200T-24MPS) to the sheath flow, leading to the reduction of RH in the sample air to 10 % and that in the sheath to 7 %. After modifying to the dry mode, the humid air in SMPS was quickly replaced by dry air through venting the sheath air at 5 L min^{-1}, and then the dry aerosol was measured by SMPS. The LWC was determined by the difference of the particle mass concentrations before and after the modification of the dry mode.

When we measured the LWC, the aerosol water should be removed after the SMPS was modified. For clarification, we have rephrased the sentence pointed out by the reviewer, "The removal of aerosol water during the LWC measurement may cause the dissolved species that are probably volatile/semi-volatile compounds to evaporate back into the gas phase. Thus, SOA concentrations for high RH conditions were slightly

underestimated, but the underestimation is extremely low and can be negligible."

9. Page 5 line 27: Table 1 should also state the m-xylene concentration in ug/m^3 so that readers can more easily compare this study's reaction conditions with those of previous studies.

Taking the reviewer's advice, we have modified the m-xylene concentration in ug/m^3 in Table 1.

Table 1. Experimental conditions, SOA concentrations and yields at the end of the experiments in *m*-xylene-OH oxidation system.

Exp. No.	$[m\text{-xylene}]_0$ ($\mu g\ m^{-3}$)	$[m\text{-xylene}]_{reacted}$ ($\mu g\ m^{-3}$)	RH (%)	T (°C)	$[SOA]_e$ ($\mu g\ m^{-3}$)	SOA yield (%)
1	2287.9	1026.3	13.6	25.9	150.3 ± 15.0	14.6 ± 1.5
2	1855.5	682.0	13.7	25.3	95.5 ± 9.5	14.0 ± 1.4
3	2410.8	941.4	73.6	27.5	21.0 ± 2.1	2.2 ± 0.2
4	2029.1	946.9	79.1	27.4	7.5 ± 0.7	0.8 ± 0.1

$[SOA]_e$ indicates the mass concentration of SOA at the end of each experiment with particle wall loss corrected.

10. Page 5 line 28: Why were the temperatures in the high RH experiments higher than those in the low RH experiments?

The accuracy of temperature controller led to this fluctuation that the temperatures in the high RH experiments were higher than those in the low RH experiments. The highest difference between low and high RH experiment was 2 °C. The temperature effect on SOA formation has been investigated in some previous studies about the m-xylene oxidation. According to previous studies about the temperature effect of SOA formation from m-xylene oxidation (Takekawa et al., 2003; Qi et al., 2010), an increase of 2 °C can lead to a mean SOA mass decrease by 4.6%. It can be concluded that the 2 °C higher temperature in high RH experiments cannot significantly affect the results of RH effect on SOA formation in this study.

Qi, L., Nakao, S., Tang, P. and Cocker, D. R., III: Temperature effect on physical and chemical properties of secondary organic aerosol from m-xylene photooxidation, Atmos. Chem. Phys., 10, 3847-3854, 10.5194/acp-10-3847-2010, 2010.
Takekawa, H., Minoura, H. and Yamazaki, S.: Temperature dependence of secondary organic aerosol formation by photo-oxidation of hydrocarbons, Atmos. Environ., 37, 3413-3424, 10.1016/s1352-2310(03)00359-5, 2003.

11. page 7 line 25: A magnified view of the mass spectra shown in Fig. 3 would be more useful for comparison purposes.

Taking the reviewer's suggestion, we have magnified the view of the mass spectra shown in Fig. 3.

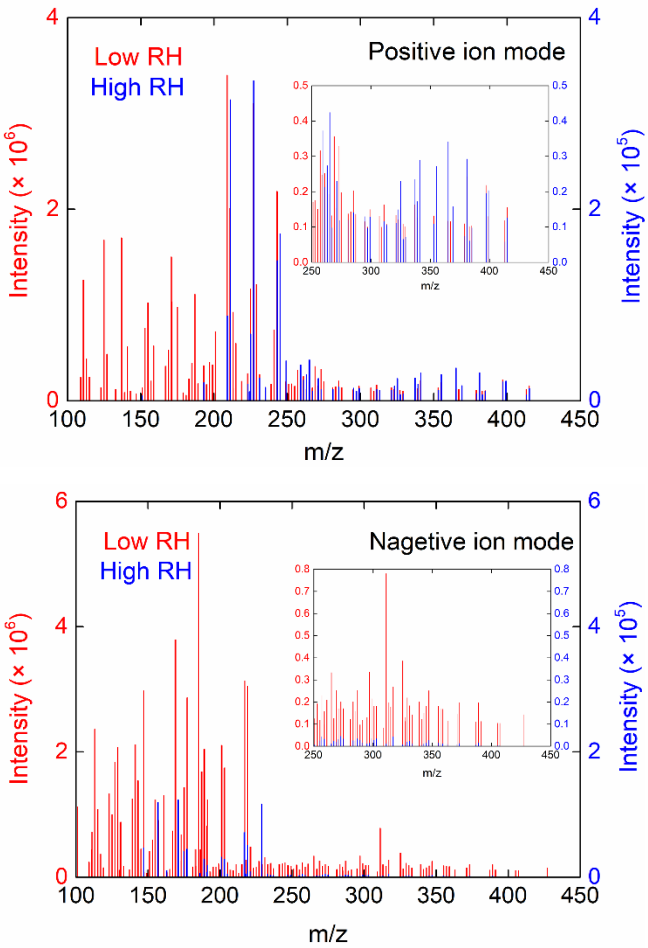


Figure 3. Selected background-subtraction HESI-Q Exactive-Orbitrap MS results of SOA in both positive and negative ion modes from the photooxidation of *m*-xylene-OH under both low and high RH conditions (Note that the Y-axis scales for low and high

RH are largely different, 10^6 at low RH and 10^5 at high RH).

12. Page 7 line 27: The sentence "It should be pointed out that the signal intensities…" is confusing. Were the mass spectra for the different experiments obtained using different MS operation conditions (e.g., ESI spray conditions, MS collision gas)?

The sentence pointed out by the reviewer is indeed confusing and thus we have deleted this sentence in the text. The mass spectra for the different experiments were obtained using exactly same MS operation conditions. Thus, the mass spectra for different experiments were comparable.

13. Page 7 line 25 to page 8 line 11: The mass peaks discussed here do not seem to be the major peaks shown in Fig. 3. Why did the authors choose to focus their discussion only on these selected peaks? The major peaks seem to be m/z > 200. How were these products formed? The authors should include a list of all the product ions identified. Do these identified products match their proposed reaction mechanism show in Scheme 1?

The mass peaks discussed between Page 7 line 25 to page 8 line 11 are the most abundant peaks in Fig 3, so we gave the proposed structures and discussed here. The m/z > 200 peaks are not discussed in this paragraph, but we discussed the m/z > 200 peaks and explained how these products formed in the Sec 3.4. In addition, taking the reviewer's advice, we have added a list of all the product ions identified in Table S1 in the supplementary information. These identified products that match their proposed reaction mechanism show in Scheme 1 are marked in Scheme 1.

Table S1(a). List of all the SOA product ions identified from ESI-HRMS in positive mode.

Low RH		High RH		Formula
m/z	intensity	m/z	intensity	
415.12032	1.55E+05	415.11937	1.26E+04	C18 H23 O11
413.10459	1.18E+05	413.10393	5.79E+03	C18 H21 O11
399.12542	1.30E+05	399.12470	2.03E+04	C18 H23 O10
397.10976	2.18E+05	397.10926	1.95E+04	C18 H21 O10

381.11482	2.42E+05	381.11434	2.92E+04	C18 H21 O9
379.09902	1.09E+05	379.09866	7.43E+03	C18 H19 O9
365.11992	1.17E+05	365.11944	3.41E+04	C18 H21 O8
385.10957	1.03E+05	385.10901	9.72E+03	C17 H21 O10
383.09399	1.01E+05	383.09330	6.04E+03	C17 H19 O10
369.11481	1.06E+05	369.11446	1.57E+04	C17 H21 O9
367.09911	1.15E+05	367.09892	-	C17 H19 O9
355.09919	1.88E+05	355.09870	2.72E+04	C16 H19 O9
353.08352	1.31E+05	353.08319	1.08E+04	C16 H17 O9
339.10481	1.27E+05	339.10408	1.72E+04	C16 H19 O8
337.08857	1.62E+05	337.08800	2.34E+04	C16 H17 O8
321.09386	1.33E+05	321.09328	1.11E+04	C16 H17 O7
341.08370	2.11E+05	341.08322	2.89E+04	C15 H17 O9
325.08882	1.54E+05	325.08817	2.30E+04	C15 H17 O8
323.07306	1.22E+05	323.07263	1.49E+04	C15 H15 O8
309.09403	9.96E+04	309.09236	-	C15 H17 O7
307.07817	1.31E+05	307.08028	-	C15 H15 O7
329.08383	1.01E+05	329.08320	7.12E+03	C14 H17 O9
327.06797	1.08E+05	327.06751	6.51E+03	C14 H15 O9
313.08917	1.08E+05	313.08830	1.05E+04	C14 H17 O8
311.07309	1.62E+05	311.07260	1.15E+04	C14 H15 O8
297.09553	9.65E+04	297.09353	9.68E+03	C14 H17 O7
295.07852	1.11E+05	295.07782	1.29E+04	C14 H15 O7
281.10079	8.07E+04	281.09867	-	C14 H17 O6
299.07316	1.49E+05	299.07245	1.29E+04	C13 H15 O8
297.05750	9.95E+04	297.05697	7.39E+03	C13 H13 O8
283.07818	1.43E+05	283.07558	-	C13 H15 O7
281.06267	1.37E+05	281.06220	1.19E+04	C13 H13 O7
267.08471	1.06E+05	267.08299	-	C13 H15 O6
265.06795	9.57E+04	265.06732	1.06E+04	C13 H13 O6
287.07312	1.36E+05	287.07266	-	C12 H15 O8
285.05754	2.03E+05	285.05710	1.41E+04	C12 H13 O8
271.07849	1.33E+05	271.07786	2.30E+04	C12 H15 O7

269.06262	3.57E+05	269.06157	-	C12 H13 O7
267.04693	1.32E+05	267.04613	9.73E+03	C12 H11 O7
265.02894	1.92E+05	265.02852	4.24E+04	C12 H9 O7
263.01347	1.17E+05	263.01304	2.73E+04	C12 H7 O7
255.08462	8.73E+04	255.08340	-	C12 H15 O6
253.06787	1.75E+05	253.06731	-	C12 H13 O6
251.05217	1.70E+05	251.05172	-	C12 H11 O6
249.03409	1.97E+05	249.03366	4.15E+04	C12 H9 O6
231.02374	1.62E+05	231.02326	2.38E+04	C12 H7 O5
275.07311	1.97E+05	275.07260	-	C11 H15 O8
273.05759	3.28E+05	273.05718	1.18E+04	C11 H13 O8
271.04182	1.54E+05	271.04129	6.63E+03	C11 H11 O8
259.07844	2.48E+05	259.07786	3.72E+04	C11 H15 O7
257.06264	3.15E+05	257.06224	-	C11 H13 O7
255.04708	1.51E+05	255.04659	-	C11 H11 O7
243.08360	2.76E+05	243.08304	5.15E+04	C11 H15 O6
239.05231	1.71E+05	239.05191	-	C11 H11 O6
235.01848	9.71E+04	235.01853	1.38E+04	C11 H7 O6
225.07285	1.87E+05	225.07253	-	C11 H13 O5
223.05754	8.06E+04	223.05702	-	C11 H11 O5
261.05771	2.51E+05	261.05727	2.13E+04	C10 H13 O8
259.04210	1.99E+05	259.04157	8.26E+03	C10 H11 O8
245.06288	1.36E+06	245.06243	1.74E+05	C10 H13 O7
243.04720	2.19E+06	243.04677	1.46E+05	C10 H11 O7
241.03138	7.38E+05	241.02850	-	C10 H9 O7
229.06802	1.21E+06	229.06751	-	C10 H13 O6
227.05224	3.10E+06	227.05182	3.34E+05	C10 H11 O6
225.03667	1.16E+06	225.03624	6.95E+04	C10 H9 O6
223.01851	2.82E+05	223.01812	1.69E+04	C10 H7 O6
213.07305	1.30E+05	213.07248	-	C10 H13 O5
211.05741	2.01E+06	211.05702	3.14E+05	C10 H11 O5
209.04177	3.40E+06	209.04141	8.84E+04	C10 H9 O5
193.04693	3.66E+05	193.04652	-	C10 H9 O4

181.08606	5.51E+04	181.08581	-	C10 H13 O3
179.07026	6.43E+04	179.06998	-	C10 H11 O3
231.04717	2.73E+05	231.04688	1.32E+04	C9 H11 O7
229.03161	1.01E+05	229.03122	-	C9 H9 O7
215.05216	5.96E+05	215.05182	-	C9 H11 O6
213.03667	9.22E+05	213.03617	-	C9 H9 O6
199.05730	3.73E+05	199.05692	-	C9 H11 O5
197.04179	4.02E+05	197.04137	-	C9 H9 O5
195.02624	1.68E+05	195.02591	-	C9 H7 O5
193.00815	3.16E+05	193.00767	1.88E+04	C9 H5 O5
185.08084	1.38E+05	185.07750	-	C9 H13 O4
183.06534	1.05E+05	183.06212	-	C9 H11 O4
179.03120	8.18E+04	179.03112	-	C9 H7 O4
167.07030	1.20E+05	167.06986	-	C9 H11 O3
147.05008	7.10E+04	147.04975	-	C9 H7 O2
201.03665	7.21E+05	201.03628	-	C8 H9 O6
189.07567	1.76E+05	189.07240	-	C8 H13 O5
187.06003	1.11E+06	187.05678	-	C8 H11 O5
185.04165	3.88E+05	185.04139	-	C8 H9 O5
183.02618	2.25E+05	183.02609	-	C8 H7 O5
171.06509	1.03E+06	171.06488	-	C8 H11 O4
169.04959	5.29E+05	169.04638	-	C8 H9 O4
167.03115	3.59E+05	167.03077	-	C8 H7 O4
155.07013	1.02E+06	155.06985	-	C8 H11 O3
153.05453	7.56E+05	153.05425	-	C8 H9 O3
151.03891	1.33E+05	151.03874	4.89E+03	C8 H7 O3
137.05962	1.70E+06	137.05931	-	C8 H9 O2
219.01631	2.01E+05	219.01588	-	C7 H7 O8
189.03670	1.66E+05	189.03673	-	C7 H9 O6
171.03328	1.50E+06	171.03295	-	C7 H7 O5
171.02640	2.77E+05	171.02571	-	C7 H7 O5
157.04949	2.03E+05	157.04906	-	C7 H9 O4
155.03117	8.61E+04	155.03096	-	C7 H7 O4

141.05445	5.63E+05	141.05420	-	C7 H9 O3
139.03886	8.46E+04	139.03859	-	C7 H7 O3
125.05974	1.68E+06	125.05950	-	C7 H9 O2
123.04411	1.34E+05	123.04389	-	C7 H7 O2
109.06503	2.41E+05	109.06480	-	C7 H9 O
209.02861	6.79E+04	209.02812	-	C6 H9 O8
175.03264	1.17E+05	175.03264	-	C6 H7 O6
175.02542	9.74E+05	175.02510	-	C6 H7 O6
157.01758	8.63E+04	157.01737	-	C6 H5 O5
143.03380	9.78E+04	143.03354	-	C6 H7 O4
127.03897	4.85E+05	127.03868	-	C6 H7 O3
113.05988	4.34E+05	113.05967	-	C6 H9 O2
111.04425	1.26E+06	111.04405	-	C6 H7 O2
215.03908	9.91E+04	215.03895	-	C5 H11 O9
199.04426	7.18E+04	199.04416	-	C5 H11 O8
115.03911	2.47E+05	115.03889	-	C5 H7 O3

Table S1(b). List of all the SOA product ions identified from ESI-HRMS in negative mode.

Low RH		High RH		Formula
m/z	Intensity	m/z	Intensity	
309.17388	8.22E+04	309.17346	-	C17 H25 O5
427.02033	1.43E+05	427.01930	3.35E+02	C16 H11 O14
407.11955	1.04E+05	407.11842	2.19E+02	C16 H23 O12
405.10387	1.04E+05	405.10320	2.63E+02	C16 H21 O12
391.12466	1.12E+05	391.12383	2.93E+02	C16 H23 O11
389.10906	1.96E+05	389.10828	1.48E+03	C16 H21 O11
387.09325	1.09E+05	387.09280	5.09E+02	C16 H19 O11
373.11399	1.97E+05	373.11329	1.26E+03	C16 H21 O10
371.09821	1.44E+05	371.09766	6.70E+02	C16 H19 O10
357.11903	1.69E+05	357.11856	9.95E+02	C16 H21 O9
355.10338	1.80E+05	355.10274	1.62E+03	C16 H19 O9
359.09835	1.67E+05	359.09778	1.15E+03	C15 H19 O10

343.10330	1.50E+05	343.10278	1.01E+03	C15 H19 O9
341.08758	1.21E+05	341.08664	5.45E+02	C15 H17 O9
339.20000	2.00E+05	339.19922	-	C15 H31 O8
327.10844	1.12E+05	327.10799	5.27E+02	C15 H19 O8
325.09285	1.08E+05	325.09217	6.23E+02	C15 H17 O8
265.14792	3.33E+05	265.14776	-	C15 H21 O4
218.03824	2.71E+05	218.03779	5.27E+03	C15 H6 O2
363.09335	1.13E+05	363.09077	9.01E+02	C14 H19 O11
347.09836	2.51E+05	347.09758	2.67E+03	C14 H19 O10
345.08263	1.80E+05	345.08202	1.35E+03	C14 H17 O10
331.10347	1.83E+05	331.10283	2.34E+03	C14 H19 O9
329.08781	2.19E+05	329.08696	1.87E+03	C14 H17 O9
327.07190	1.29E+05	327.07119	9.89E+02	C14 H15 O9
325.18438	3.85E+05	325.18366	-	C14 H29 O8
313.09287	1.96E+05	313.09204	1.88E+03	C14 H17 O8
311.07715	1.52E+05	311.07670	1.01E+03	C14 H15 O8
297.09786	1.32E+05	297.09724	1.05E+03	C14 H17 O7
295.08212	1.29E+05	295.08163	9.81E+02	C14 H15 O7
333.08273	1.42E+05	333.08206	1.23E+03	C13 H17 O10
331.06692	9.75E+04	331.06613	7.28E+02	C13 H15 O10
317.08774	2.67E+05	317.08714	4.21E+03	C13 H17 O9
315.07210	1.77E+05	315.07401	-	C13 H15 O9
311.16878	7.80E+05	311.16806	-	C13 H27 O8
301.09273	1.82E+05	301.09215	2.77E+03	C13 H17 O8
299.07727	2.15E+05	299.07641	3.44E+03	C13 H15 O8
297.06154	1.09E+05	297.06002	1.42E+03	C13 H13 O8
285.09789	1.00E+05	285.09726	1.77E+03	C13 H17 O7
283.08221	2.00E+05	283.08162	2.54E+03	C13 H15 O7
281.06697	1.22E+05	281.06609	-	C13 H13 O7
267.08726	1.26E+05	267.08657	2.19E+03	C13 H15 O6
265.07197	1.61E+05	265.07106	1.19E+03	C13 H13 O6
247.06280	1.81E+05	247.06092	3.33E+02	C13 H11 O5
231.06771	3.10E+05	231.06592	-	C13 H11 O4

303.07184	1.82E+05	303.07152	3.35E+03	C12 H15 O9
301.05629	9.34E+04	301.05576	1.42E+03	C12 H13 O9
297.15292	3.35E+05	297.15230	-	C12 H25 O8
287.07698	2.52E+05	287.07691	3.35E+03	C12 H15 O8
285.06133	1.57E+05	285.06109	2.27E+03	C12 H13 O8
271.08217	1.69E+05	271.08173	3.03E+03	C12 H15 O7
269.06606	2.53E+05	269.06611	2.03E+03	C12 H13 O7
267.05024	9.81E+04	267.05010	7.33E+02	C12 H11 O7
255.08719	1.17E+05	255.08664	-	C12 H15 O6
253.07111	1.91E+05	253.07089	-	C12 H13 O6
251.05515	1.24E+05	251.05505	5.59E+02	C12 H11 O6
237.07648	1.36E+05	237.07600	-	C12 H13 O5
221.08136	8.88E+04	221.08108	-	C12 H13 O4
291.07203	1.18E+05	291.07139	1.57E+03	C11 H15 O9
289.05636	9.61E+04	289.05566	2.67E+03	C11 H13 O9
275.07729	1.70E+05	275.07673	3.31E+03	C11 H15 O8
273.06161	2.00E+05	273.06105	4.47E+03	C11 H13 O8
259.08219	1.57E+05	259.08171	3.21E+03	C11 H15 O7
257.06651	2.28E+05	257.06598	4.31E+03	C11 H13 O7
255.05088	1.07E+05	255.05025	2.48E+03	C11 H11 O7
243.08714	8.57E+04	243.08664	-	C11 H15 O6
241.07147	2.04E+05	241.07093	-	C11 H13 O6
225.07641	1.52E+05	225.07596	-	C11 H13 O5
223.06071	1.40E+05	223.06018	-	C11 H11 O5
209.08133	1.02E+05	209.08102	-	C11 H13 O4
207.06573	1.11E+05	207.06531	-	C11 H11 O4
193.08630	7.02E+04	193.08600	-	C11 H13 O3
261.06155	2.09E+05	261.05977	-	C10 H13 O8
259.04588	9.74E+04	259.04511	3.26E+03	C10 H11 O8
245.06644	2.22E+05	245.06586	-	C10 H13 O7
243.05084	2.08E+05	243.05037	-	C10 H11 O7
229.07132	2.03E+05	229.07087	-	C10 H13 O6
227.05570	2.51E+05	227.05525	-	C10 H11 O6

211.06064	1.96E+05	211.06027	-	C10 H11 O5
209.04512	9.50E+04	209.04452	-	C10 H9 O5
195.06563	1.61E+05	195.06536	-	C10 H11 O4
193.05002	8.88E+04	193.04978	-	C10 H9 O4
181.08628	9.12E+04	181.08592	-	C10 H13 O3
163.07558	5.18E+04	163.07520	-	C10 H11 O2
249.06096	1.71E+05	249.06092	2.65E+03	C9 H13 O8
247.04556	1.43E+05	247.04500	3.44E+03	C9 H11 O8
233.06614	1.99E+05	233.06580	4.43E+03	C9 H13 O7
231.05066	1.62E+05	231.05023	-	C9 H11 O7
229.03504	7.84E+04	229.03443	-	C9 H9 O7
217.07129	1.69E+05	217.07080	-	C9 H13 O6
215.05569	2.04E+05	215.05511	-	C9 H11 O6
201.07618	2.38E+05	201.07580	-	C9 H13 O5
199.06058	1.95E+05	199.06031	-	C9 H11 O5
197.04500	1.59E+05	197.04464	-	C9 H9 O5
183.06553	1.70E+05	183.06531	-	C9 H11 O4
181.04989	1.64E+05	181.04952	-	C9 H9 O4
165.05484	1.21E+05	165.05454	-	C9 H9 O3
149.05981	6.31E+04	149.05947	-	C9 H9 O2
237.06126	9.92E+04	237.06075	2.61E+03	C8 H13 O8
235.04566	2.21E+05	235.04511	2.88E+03	C8 H11 O8
233.03005	1.23E+05	233.02891	-	C8 H9 O8
221.06623	4.82E+05	221.06579	8.46E+03	C8 H13 O7
219.05057	3.05E+06	219.05011	5.02E+04	C8 H11 O7
217.03488	3.13E+06	217.03444	7.13E+04	C8 H9 O7
205.07117	2.33E+05	205.07077	4.95E+03	C8 H13 O6
203.05550	1.74E+06	203.05513	2.87E+04	C8 H11 O6
201.03983	2.10E+06	201.03967	3.18E+04	C8 H9 O6
199.02428	2.08E+05	199.02409	-	C8 H7 O6
187.06049	1.68E+06	187.06013	-	C8 H11 O5
185.04483	5.49E+06	185.04449	-	C8 H9 O5
183.02923	4.41E+05	183.02901	-	C8 H7 O5

171.06543	5.43E+05	171.06511	1.23E+05	C8 H11 O4
169.04976	3.79E+06	169.04954	-	C8 H9 O4
167.03412	7.36E+05	167.03368	-	C8 H7 O4
153.05472	5.91E+05	153.05442	-	C8 H9 O3
151.03908	4.09E+05	151.03878	-	C8 H7 O3
137.05970	1.18E+05	137.05943	-	C8 H9 O2
205.03496	2.28E+05	205.03286	-	C7 H9 O7
191.05540	1.23E+06	191.05504	1.88E+04	C7 H11 O6
189.03975	2.04E+06	189.03942	2.91E+04	C7 H9 O6
187.02422	1.80E+05	187.02389	-	C7 H7 O6
175.06041	1.80E+05	175.06006	-	C7 H11 O5
173.04471	6.71E+05	173.04440	-	C7 H9 O5
171.02908	6.75E+05	171.02879	-	C7 H7 O5
169.01357	1.20E+05	169.01317	-	C7 H5 O5
157.04965	9.08E+05	157.04939	-	C7 H9 O4
155.03403	1.23E+06	155.03374	-	C7 H7 O4
153.01828	2.32E+05	153.01805	-	C7 H5 O4
141.05463	2.11E+06	141.05439	-	C7 H9 O3
139.03897	1.25E+06	139.03869	-	C7 H7 O3
125.05961	6.55E+05	125.05940	-	C7 H9 O2
123.04397	1.33E+06	123.04376	-	C7 H7 O2
229.05210	1.79E+05	229.04953	-	C6 H13 O9
213.05676	6.20E+04	213.05473	-	C6 H13 O8
191.01907	1.26E+05	191.01873	7.54E+03	C6 H7 O7
177.03967	2.86E+06	177.03930	4.45E+04	C6 H9 O6
175.02402	1.43E+06	175.02363	4.04E+04	C6 H7 O6
173.00836	2.54E+05	173.00529	-	C6 H5 O6
161.04464	1.30E+06	161.04430	-	C6 H9 O5
157.01330	3.67E+05	157.01334	-	C6 H5 O5
147.06522	8.02E+04	147.06492	-	C6 H11 O4
145.04957	3.71E+05	145.04929	-	C6 H9 O4
143.03391	1.54E+06	143.03365	-	C6 H7 O4
141.01827	7.79E+05	141.01797	-	C6 H5 O4

139.00264	6.12E+04	139.00224	-	C6 H3 O4
127.03890	1.83E+06	127.03866	-	C6 H7 O3
125.02325	9.93E+05	125.02297	-	C6 H5 O3
113.05952	7.21E+05	113.05925	-	C6 H9 O2
111.04388	7.15E+05	111.04361	-	C6 H7 O2
109.02824	2.35E+05	109.02805	-	C6 H5 O2
201.05694	1.09E+05	201.05479	-	C5 H13 O8
163.02399	7.06E+04	163.02364	1.05E+04	C5 H7 O6
147.02887	2.97E+06	147.02878	4.63E+04	C5 H7 O5
145.01320	4.46E+05	145.01289	-	C5 H5 O5
131.03383	8.78E+05	131.03360	-	C5 H7 O4
129.01819	2.07E+06	129.01793	-	C5 H5 O4
127.00253	1.97E+05	127.00232	-	C5 H3 O4
115.03882	1.08E+06	115.03858	-	C5 H7 O3
113.02311	2.36E+06	113.02278	-	C5 H5 O3
111.00750	4.41E+05	111.00726	-	C5 H3 O3
133.01316	1.69E+05	133.01282	-	C4 H5 O5
119.03381	1.48E+05	119.03355	-	C4 H7 O4
117.01807	3.64E+05	117.01775	-	C4 H5 O4
115.00245	3.19E+05	115.00215	-	C4 H3 O4
101.02308	1.11E+06	101.02289	-	C4 H5 O3

14: General comment: What compounds are the -ve MS mode sensitive to? Were these compounds identified in their collected mass spectra?

In positive mode analysis, ions are produced by protonation. Thus, groups that more readily accept a positive charge, such as carbonyls, are often observed in this mode. As listed in Table 3, the proposed compounds obtained by HRMS in positive ion mode are all with the carbanyl group. Negative mode analysis leads to formation of deprotonated ions. Thus, molecules containing functional groups that readily lose a proton, such as carboxylic acids, are frequently observed in this mode. Also, the esters compounds can be obtained in the negative ion mode (Hamilton et al., 2008; Camredon et al., 2010; Ge et al., 2017). In the MCM prediction about m-xylene-OH oxidation, many carbonyls are included. It can be deduced that many carboxylic acids can be formed via OH

oxidation of these carbonyls and these carboxylic acids can be measured in the negative ion mode.

Camredon, M., Hamilton, J. F., Alam, M. S., Wyche, K. P., Carr, T., White, I. R., Monks, P. S., Rickard, A. R., and Bloss, W. J.: Distribution of gaseous and particulate organic composition during dark α-pinene ozonolysis, Atmos. Chem. Phys., 10, 2893-2917, 10.5194/acp-10-2893-2010, 2010.
Ge, S., Xu, Y. and Jia, L.: Secondary organic aerosol formation from propylene irradiations in a chamber study, Atmos. Environ., 157, 146-155, 10.1016/j.atmosenv.2017.03.019, 2017.
Hamilton, J. F., Lewis, A. C., Carey, T. J., and Wenger, J. C.: Characterization of polar compounds and oligomers in secondary organic aerosol using liquid chromatography coupled to mass spectrometry, Anal. Chem., 80, 474-480, 10.1021/ac701852t, 2008.

15. General comment: The authors mentioned in the experimental system that they used a HPLC-MS system in their study. It is not clear from their presented results whether this was the case. Was HPLC not used to separate the products via their volatilities prior to MS analysis?

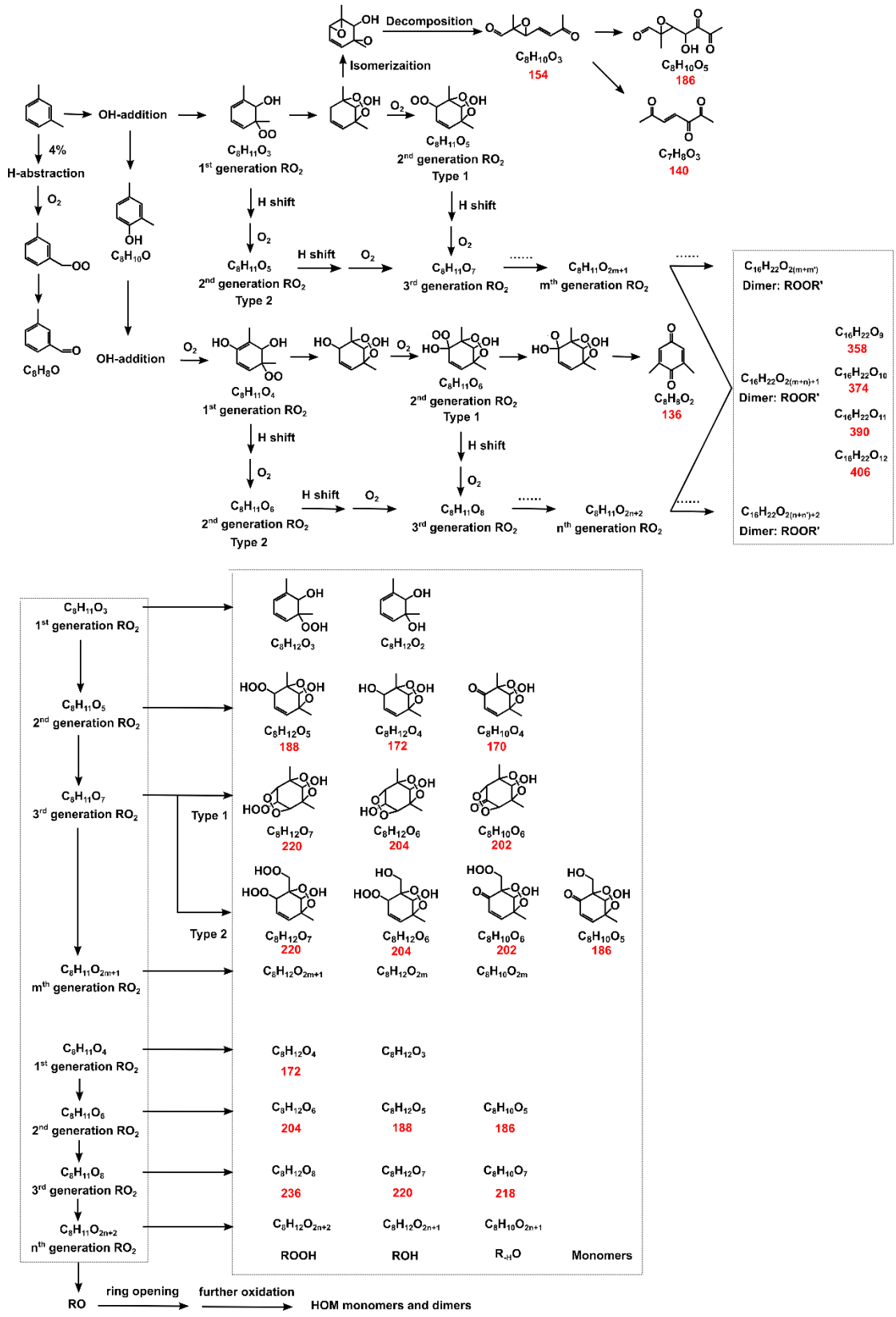
HPLC was used in our experiments as the injection system before HRMS analysis. We used the high resolution of mass analyzer for the separation of major SOA components instead of HPLC.

16. Page 9 line 30: The authors claimed that they used the distribution of relative intensity of SOA products with the same carbon number to investigate the potential RH effect on HOMs. The rationale behind this course of action seems to contradict their previous statement in Page 7 line 27 that signal intensities can be biased by ionization properties.

The statement was incorrect and confusing in Page 7 Line 27 and we have deleted it from the text. The mass spectra for the different experiments were obtained using same MS operation conditions. Thus, the mass spectra for different experiments were comparable (see the reply of Comment 12).

17. Scheme 1: The authors should indicate explicitly in Scheme 1 which are the products that they have identified.

Taking the reviewer's advice, we have modified Scheme 1 in which the products identified are marked with a molecular weight number below the molecular formula.



Scheme 1. The route of OH-initiated *m*-xylene oxidation. The red number below the molecular formula is its molecular weight, which is determined by HRMS to exist in the particle phase.

18. Page 10 line 27: The sentence “Together with the previous study on toluene SOA, it is conceivable that the effect of RH on SOA yield is a common feature of SOA formation from oxidation of all OH-initiated aromatics” is too generalized and needs to be rephrased. As discussed by the authors in their introduction, an increase RH does not necessarily cause a decrease in SOA mass yields in aromatics SOA systems. Other factors such as NOx can also alter the effect that RH has on SOA mass yields in these systems.

Taking the reviewer’s advice, we have rephrased the sentence in Page 10 line 27.

Together with the previous study on toluene SOA, it is conceivable that the effect of RH on SOA yield is a common feature of SOA formation from aromatics oxidation under low NO_x conditions and using H_2O_2 as the OH radical source.