SUMMARY This paper investigated the SOA formation from the ozonolysis of ethyl methacrylate (EM) in the presence of acidic sulfate seed, which was nebulized using aqueous sulfuric acid solution or/and SO_2 oxidation, in the indoor chamber. The resulting SOA and acidic sulfate were characterized using HR-ToF-AMS fragmentation patterns and the Van Krevelen diagram under varying the amount of acidic sulfate. Overall, it is difficult to follow the flow of the manuscript. The interpretation for the impact of aerosol acidity on SOA yields and the trend in product distributions was somewhat weak. In addition, the implication of the resulting study to the ambient air was not well described.

Answer: Thank you very much for your review and giving us valuable suggestions. The manuscript has been revised substantially according to your comments (See below).

Q1. Gas product aging. For the chamber study, the authors included CO to remove OH radicals that were yielded from the decomposition of ozonolysis products of EM. The experimental condition in the absence of UV light and OH radicals suggests that further oxidation of ozonolysis products in the gas phase can be very slow or insignificant. It is unclear how the ozonoysis products of this study can be further oxidized under such experimental conditions.

A1. Before 200 min, it could be found that both Factor 1 and 2 were almost produced simultaneously. This indicated that the ozonolysis products in Factor 1 and 2 were produced via different reaction pathway rather than the further oxidized. The reaction between sCI and SO_2 was the major formation pathway of carbonyls; Proposed H-shift process in Scheme 1 or the acidic-catalyzed oligomerization may result in the formation of high oxidized species. After 200 min, less oxidized species in Factor 2 gradually converted to more oxidized species in Factor 1 due to the acid-catalyzed role.

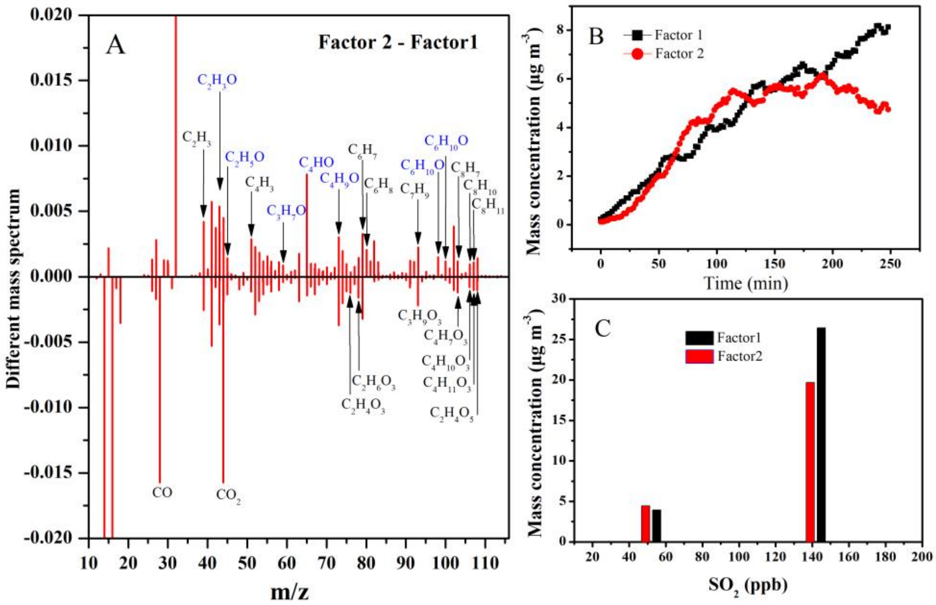

Figure 3. Two-factor solutions for PMF analyses of SOA under different SO_2 concentrations: (A) Different mass spectra between two factors (Factor 2-Factor 1) at 135 ppb SO_2; (B) Time series of factor concentrations; (C) Maximum concentration of two factors at 55 ppb and 135 ppb SO_2.

Moreover, the evolution of both m/z 44 (the characterized fragment of high oxidized species) and m/z 43 (the characterized fragment of carbonyl species) concentration along with their ratio (f44 and f43) to total signal in the component mass spectrum were also investigated. From Figure S16, the mass concentration of m/z 44 (the characterized fragment of high oxidized species) and the ratio of m/z 44 to total signal in the component mass spectrum (f44) were observed to continuedly increased with reaction time. And m/z 43 mass and its ratio (f43) first increased before 200 min and then decreased after 200 min. The consistent result in Figure 3B and Figure S16 further supported above conclusion.

This has been added in the revised manuscript (Line 301-306 and Line 309-314).

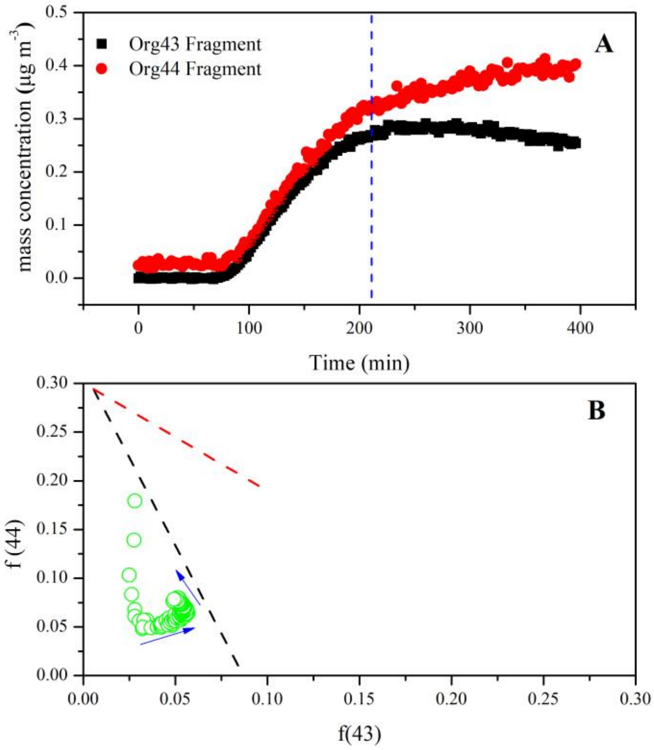

Figure S16, Time-dependent growth curves of m/z Org43 and m/z Org44 fragment (A); the triangle plot of f(43) vs f(44) of SOA (B) at 55 ppb SO_2 in absence of seed particles. f44 (ratio of m/z 44 to total signal in the component mass spectrum) and f43 (defined similarly).

Q2. 2nd paragraph in page 5, Cleaning chamber. (1) The paper described that the smog chamber was cleaned for at least 24 h until certain condition was reached. Authors need to explain the cleaning procedure with more details. In addition to the concentrations of background gas and particles, the cleaning procedure can influence the wall conditions. (2) Off-gassing of some preexisting organic gases from chamber takes time. No chamber can be perfectly sealed. The organic compounds deposited on the chamber wall can influence atmospheric chemistry and SOA formation. Ammonia can slowly intrude to the chamber wall and stick to chamber wall. This ammonia can be off-gassing from the wall and neutralize acidic seed in the absence of SO_2.

A2. (1) During the chamber cleaning, the chamber was first inflated using purified and dry zero air with a flow rate of 120 L min^{-1} for 10 min, subsequently air pump began to run for 5 min. The stainless-teel fan installed at the bottom of chamber was kept to run during the whole cleaning process. Prior to each experiment, Teflon chamber was repeatedly and circularly cleaned by purified and dry zero air using above method for about 24 h until almost no NOx could be detected or the particle number concentration was < 30 cm^{-3}. The cleaning procedure of chamber was consistent with that described in our previous studies.(Liu et al., 2019; Chen et al., 2019a)

This has been added in the revised manuscript (Line 126-133).

(2) Yes, we admitted that no chamber can be perfectly sealed. From revised Figure 1C, it could be found that the desorption or off-gassing of some organic gaseous products from chamber wall could be absorbed by seed particles to some extent during introducing seed particles (~25 min before reaction). However, the influence of these preabsorbed OA to newly produced SOA could be ignorable based on the comparison of their concentrations.

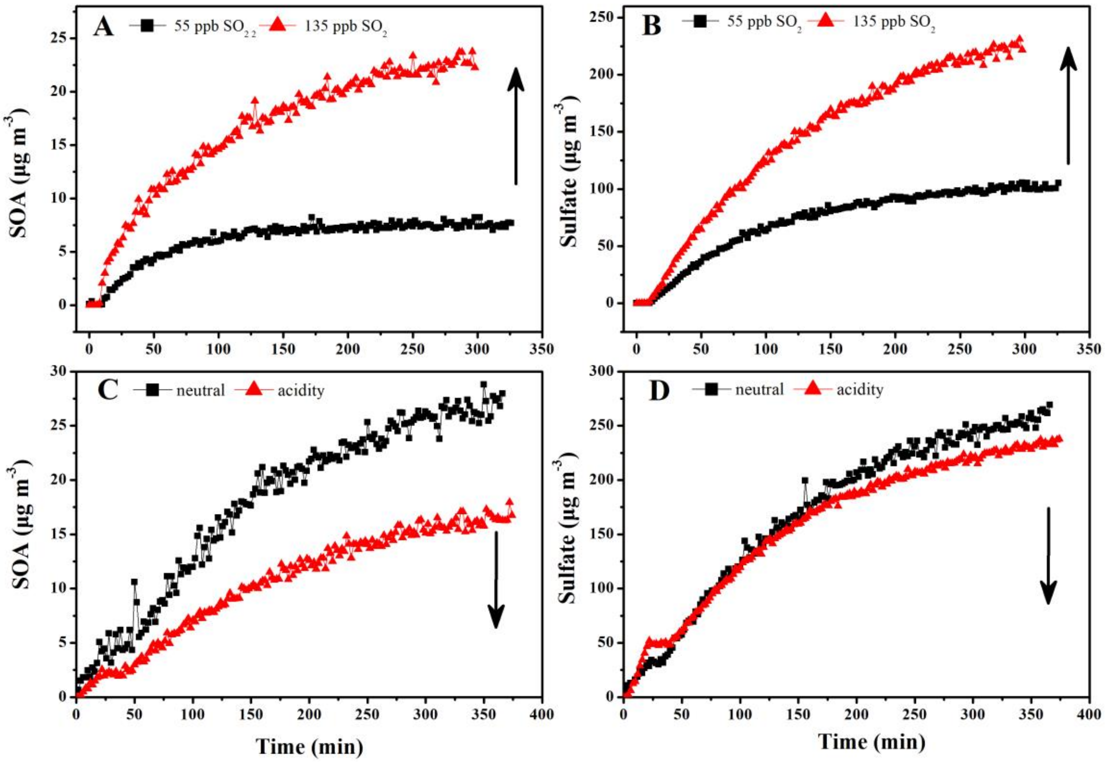

Figure 1. Time-dependent growth curves of SOA (A) and sulfate (B) under different initial concentrations of SO_2 in absence of seed particles; SOA (C) and sulfate (D) after subtracting seeds in presence of neutral and acidic seed particles.

Similarly, the concentration of ammonium was significantly lower than sulfate (Figure S2), thus the influence of NH_3 to particle acidity should also be ignorable.

This has been added in the revised manuscript (Line 153-157).

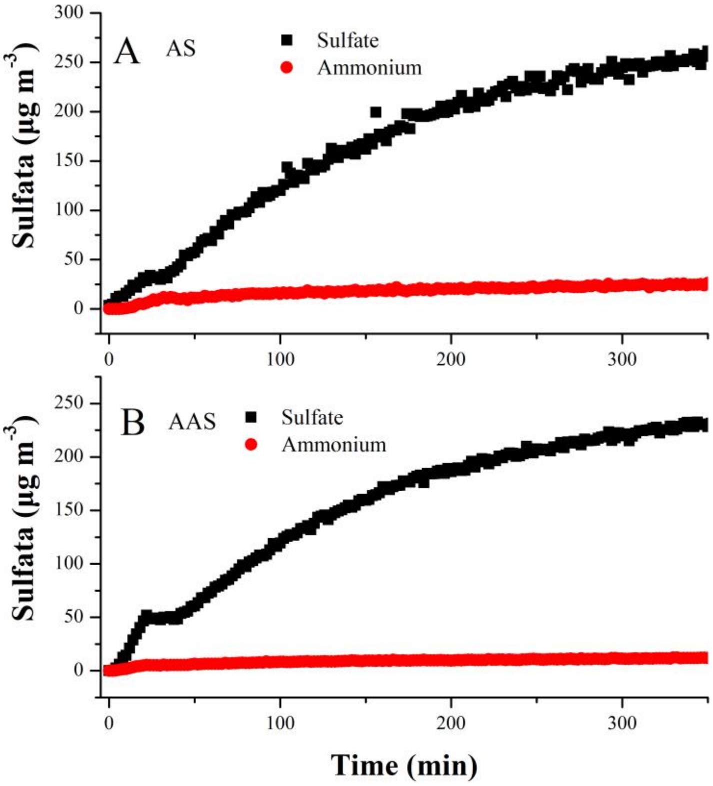

Figure S2. Time-dependent growth curves of ammonium and sulfate in the presence of AS (A) and AAS (B).

Q3. Missing of EM profiles: Authors showed the time profiles of ozone and SO_2 (Figure S2) but did not show EM time profiles. Please include the EM time profiles. No SOA yield was reported.

A3. EM time profiles and consumption along with its uptake on acidity seed in the chamber could be supplied in this work due to the lack of proton transfer reaction time-of-flight mass spectrometry (PTR-TOFMS) in our laboratory. Thus, SOA yield could not be estimated either. We hope that these quantitative problems could be solved after purchasing PTR-TOFMS in the future.

This has been added in the revised manuscript (Line 483-487).

Q4. Impact of acidic aerosol on SOA formation (Section 3.1). The explanation for the influence of acidic inorganic seed on SOA needs to be more clarified by characterizing the impact of the wall on SOA formation. Most explanations in this paper are qualitative. SOA growth need to be explained with mathematical parameters. SOA growth mechanisms should be explained based on thermodynamic partitioning and the kinetic factor in heterogeneous chemistry.

(1) SOA formation is not necessarily dependent of the quantity of acidic aerosol or the quantity of wet-inorganic aerosol because SOA growth via aqueous reactions of organic species can be limited by the quantity of reactive chemical species in gas phase. The significance of the aqueous reaction of organic species are related to the characteristic time of heterogeneous chemistry. The lifetime of EM could be several hours (longer than experimental duration). The gaseous organic products formed in the experimental condition of this study may be relatively stable in the absence of OH radicals and UV light. In the case when the characteristic time of the aqueous reaction or organic products is relatively short, the aqueous phase reaction does not need the high concentration of wet-seed. The authors did not show the profile of EM as well as the consumption of EM over the cause of the experiment. Based on Figure 1, sulfate concentrations were much higher than SOA. In the presence of the high concentration of acidic sulfate, the sensitivity of reactive chemical species to aerosol acidity can be small. On the other hand, if aqueous reaction is relatively slower than ozonolysis, heterogeneous reactions can be sensitive to the quantity of acidic wet-seed. In this case, the impact of the wall can be significant because both the characteristic time of particle loss to the wall and characteristic time for organic deposition can be relatively important. Therefore the experimental data reported in this study need in-depth analyses. Overall, experimental date is not strong to support the author's explanation on the impact of acidic sulfate on SOA growth.

A4-1: Yes, I agree with that SOA formation is not necessarily dependent of the quantity of acidic aerosol or the quantity of wet-inorganic aerosol and is related to reactive chemical species produced in gas phase. However, it should be noted that SOA formation may be also impacted if internal adsorbed precursor would not be oxidated, especially in the absence of OH radicals, once reaction products was formed on outermost surface. Moreover, all experiments were carried out at relative condition (at 10% and 45% RH) in this work and a diffusion dryer was placed after the atomizer to remove liquid water from the seed particles. Thus, the aqueous reaction of organic species on wet-seed almost could not occur in our study since high RH (RH > 80%) was very necessary if AS seed was air-slaked in the chamber.(Matthew et al., 2008)

(2) Figure 1: Authors need to explain how Figure 1 was obtained (what instrument and what procedure?). The subtraction of the large quantity of sulfate aerosol from the total aerosol is not appropriate to accurately quantify the SOA mass produced. The acidic aerosol deposition to the chamber wall can acidify the chamber wall and actively absorb the reactive gas species. The characteristic time of the wall loss of organic compounds can be influenced by the dimension of the chamber. In general, the surface area of wall can be larger than the surface area of aerosol. If the oxidized products from the ozonolysis of precursor hydrocarbons are not highly reactive for aerosol phase reactions, the impact of the wall can be high as discussed above. To demonstrate the actual impact of acidic aerosol, the characterization of the chamber wall should be performed.

A4-2: After completing injection, seed particles were allowed to stabilize for approximately 30 min in the chamber. Figure 1 in the previous manuscript was obtained using the maximum concentration of secondary particles at the end of experiment subtract the initial seed concentration. To accurately describe and compare the evolution of secondary particle, the evolution of secondary particles with the initial seed concentration has been added in Figure 1C and 1D.

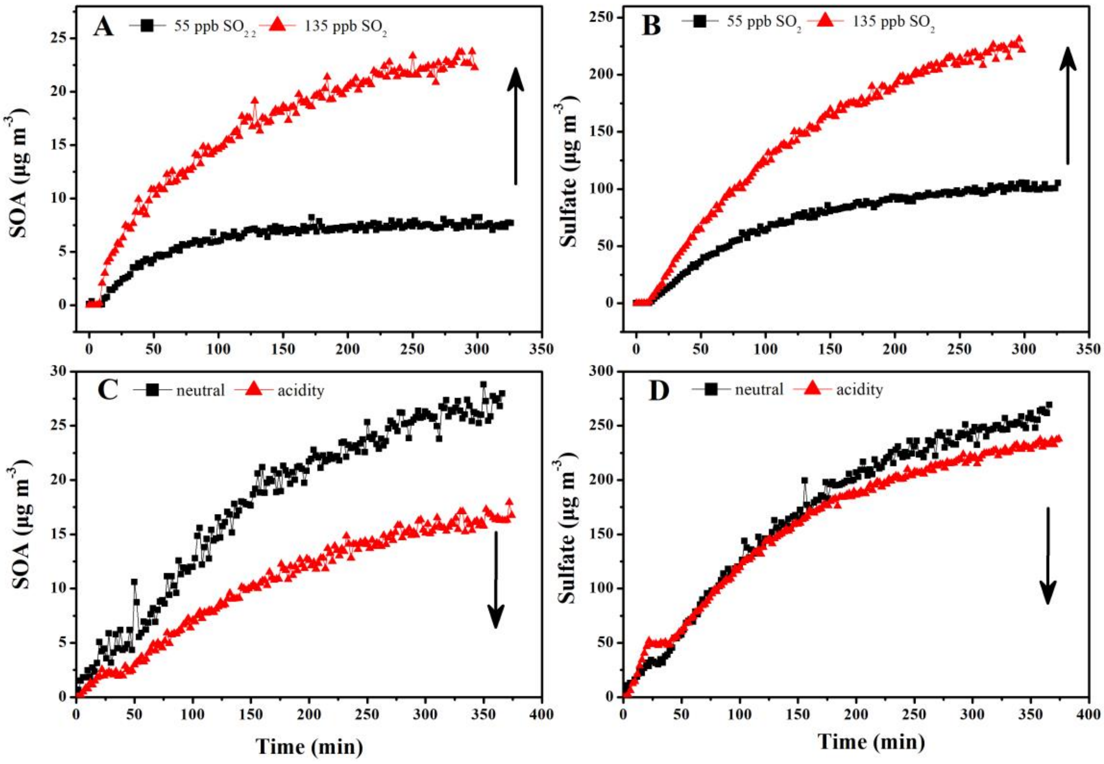

Figure 1. Time-dependent growth curves of SOA (A) and sulfate (B) under different

initial concentrations of SO_2 in absence of seed particles; SOA (C) and sulfate (D) after subtracting seeds in presence of neutral and acidic seed particles.

We admitted that the acidic aerosol deposited to the chamber wall can acidify the chamber wall and actively absorb the reactive gas species. However, from Figure S6, it could be found that the wall loss of both AS and acidic AS seeds could be almost ignorable after the correction of wall loss rate of $(NH_4)_2SO_4$ particles (Figure S1). This indicated that effect of acidic aerosol deposited on the chamber to SOA formation should also be negligible. New Figure 1 has been added in the revised manuscript (Line 221).

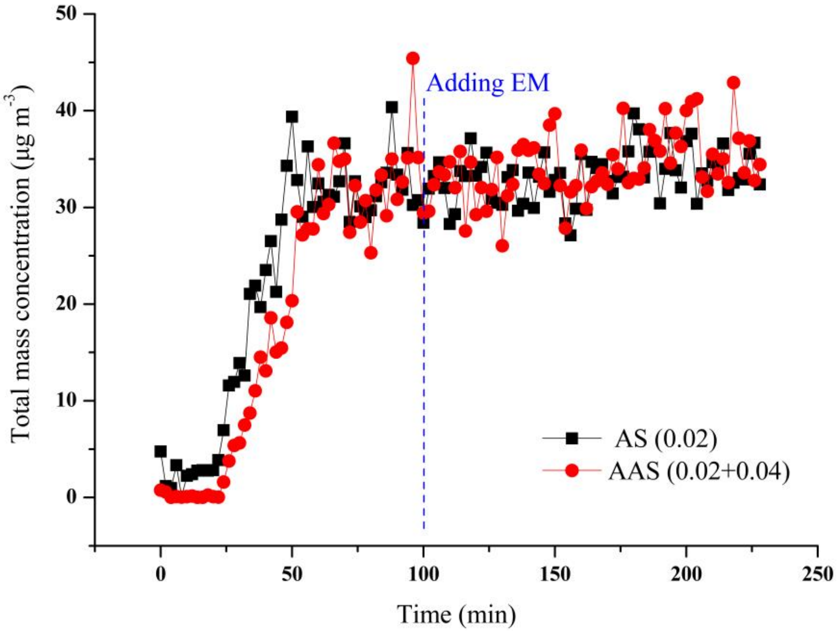

Figure S6. Evolution of secondary particles mass concentration with AS (black) and AAS (red) in the absence of SO_2 (SMPS data).

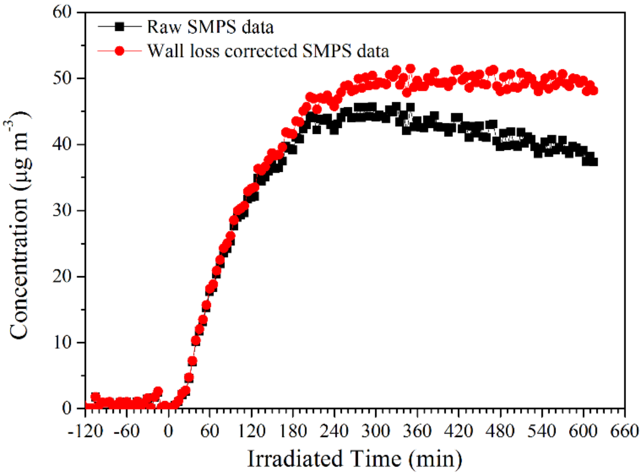

Figure S1. The correction of wall loss rate of $(NH_4)_2SO_4$ particles.

(3) SO_2 oxidation in the absence of preexisting wet aerosol produces many small particles increasing the surface area of aerosols (Figure 7). In the presence of wet-ammonium sulfate, SO_2 can partition to aqueous phase and SO_2 can be heterogeneously oxidized with ozone. Thus, wet-seed can suppress the formation of new sulfuric acid particles that can increase surface areas of particles. In the presence of wet-AS aerosol, it will take time to build up aerosol acidity as increasing sulfate fraction in the internal mix of ammonium sulfate and sulfuric acid. I do not convince that SOA formation is suppressed in the presence of more acidic sulfate because the large amount of acidic seed is not necessarily linked to the large SOA mass except a slow organic heterogeneous reaction. The quantity of reactive organic species may be relatively small compared to the large quantify of acidic sulfate.

A4-3: In this study, all experiments were carried out at relative dry condition (at 10%) and a diffusion dryer was placed after the atomizer to remove liquid water from the seed particles. Thus, the heterogeneous reaction between SO_2 and O_3 on wet-seed almost could not occur in our study since high RH (RH > 80%) was very necessary if AS seed was air-slaked in the chamber.(Matthew et al., 2008) This was also supported by one recent work from Ye et al., in which the effect of 6 h-reactions between SO_2 (289 ppb) and O_3 (485 ppb) either in the gas or particle phase on SO_2 consumption was negligible under 50% RH.(Ye et al., 2018)

Q5. Line 233. What is Fourier spectra? Please also clarify what is mass spectrum instrument.

A5. Fourier spectra represents the in situ attenuated total internal reflection infrared (ATR-IR) spectra, which was recorded using a NEXUS 6700 (Thermo Nicolet Instrument Corp.) Fourier transform infrared (FT-IR) spectrometer equipped with a high-sensitivity mercury-cadmium-telluride (MCT) detector cooled by liquid N_2. All spectra reported here were recorded at a resolution of 4 cm^{-1} for 100 scans in the spectral range of 4000−650 cm^{-1}.(Han et al., 2013; Ma et al., 2019)

This has been added in the Supporting Information (Line 100-109).

Q6. Line 241. Please check "Sci".

A6. "Sci" has been revised to "sCI" (Line 266).

Q7. Line 244-247. The connection of alpha–pinene data with EM ozonolysis data is unclear. The appearance of alpha-pinene SOA data is somewhat awkward in the flow of the paper. The SOA from EM ozonolysis will be different from the SOA from alphapinene ozonolysis in hydrocarbon lifetime, volatility of products, reactivity in aerosol phase, organic vapor deposition to the wall, and solubility of products in salted aqueous phase. It is hard to understand why alpha-pinene SOA behaves in the same with EM SOA for the impact of sulfate on SOA.

A7. The α-pinene SOA data shown in this work was to check whether the reducing effect of increasing particle acidity to secondary particle in the presence of SO_2 also happened for other systems. Moreover, previous studies on α-pinene SOA proved that the increase in particle acidity promotes or have a negligible effect on SOA formation, but these studies were conducted in the absence of SO_2 rather than in the presence of SO_2. However, the description of α-pinene SOA has been deleted from the revised manuscript to make this work be better understanded.

Q8. Lines 257-260. For the last two decades, there have been numerous studies to investigate the impact of sulfuric acid on SOA formation. The authors' citations mainly pointed the publication from own lab. There were landmark publications to launch the investigation of acid-catalyzed reactions of organics in the aerosol phase. The authors need to give a credit to early scientific efforts by citing land breaking publications in this area.

A8. The related references on the impact of sulfuric acid on SOA formation have been cited in the revised manuscript (Line 282-283).

"Recent studies have suggested that a higher proportion of H_2SO_4 in aerosol can result in greater formation of oligomers and high-oxygenated organic aerosol via acceleration of the acid-catalysis process (Zhang et al., 2019; Liu et al., 2019; Shu et al., 2018; Kristensen et al., 2014; Iinuma et al., 2004; Rodigast et al., 2017)."

Q9. PMF analysis (page 10-13) (1) In page 10, authors mentioned that Factor 1 is for less-oxygenated organic aerosol (carbonyls) and factor 2 is for more oxygenated organic aerosol (carboxylic acid). Authors mentioned that after 200 min, Factor 1 continuously increased but Factor 2 decreased suggesting chemical conversion of part of less-oxygenated species in Factor 2 to more-oxygenated products in Factor 1 in the latter period of reaction. Authors need to rationale how this conversion occurs. In addition, the description in page 10 is contradicted between several sentences.

A9-1: Factor 2 and Factor 1 represented the less and more-oxygenated organic aerosols. These contradicted sentences have been corrected in the revised manuscript (Line 295-299 and 308-309). In the presence of SO_2, carbonyl species (such as aldehyde) was the major species from the reaction between SO_2 and sCI during EM ozonolysis. And some oxygenated products with oxygen-containing group (such as OH and C=O) may be also produced as shown in Scheme 1. When the particle acidity reached a certain concentration with the increase of H_2SO_4, the enhanced acidity-catalyzed process could promote the formation of oligomers with multi-oxygen atoms via aldol condensation and hemiacetal reaction. This has been added in the revised manuscript (Line 301-306 and 309-314).

(2) Lines 288-293. Authors mentioned condensation of both less oxidized species and more oxidized species. However, this description cannot be connected to acid-catalyzed heterogenous reaction. What does the condensation means in this paper? The authors need to clarify aerosol growth mechanisms that mainly influence EM SOA.

A9-2: From Figure 1A and 1B, it could be found the quantity of SOA was significantly lower compared to the large quantify of sulfate. This implied that the reaction between sCI and SO_2 remarkably promoted the homogeneous nucleation of sulfate relative to SOA formation. (Figure 2B). SOA growth should be mainly attributed to the adsorption and condensation of both less oxidized species and more oxidized species on particle before 200 min. This has been added in the revised manuscript (Line 301-306 and 309-314).

(3) In general, if aerosol growth is dominated by acid-catalyzed heterogeneous chemistry, the concentration of carbonyl species can significantly decrease due to the reaction of carbonyl species. Authors need to clarify the mechanistic role of sulfuric acid on SOA growth on the trend of mass peaks.

A9-3. From Figure S16, the mass concentration of m/z 44 (the characterized fragment of high oxidized species) and the ratio of m/z 44 to total signal in the component mass spectrum (f44) were observed to continuedly increased with the decrease of m/z 43 (the characterized fragment of carbonyl species) and the ratio of m/z 43 to total signal (f43) after 200 min, which is consistent with that observed in Figure 3B. This result also supported our proposed conclusion in which the chemical conversion of less oxidized species to more oxidized species could occur when the H_2SO_4 proportion (acidity) in the particle-phase reached a certain concentration

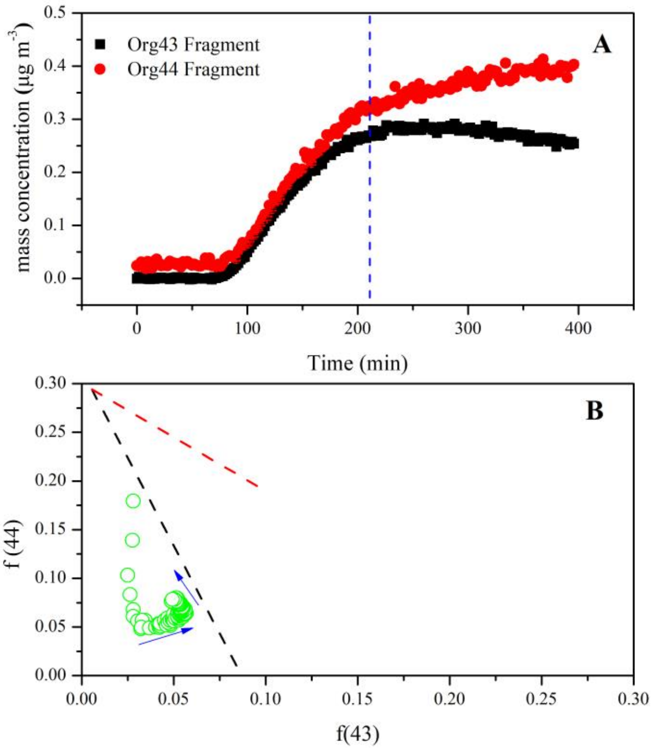

Figure S16. Time-dependent growth curves of m/z Org43 and m/z Org44 fragment (A); the triangle plot of f(43) vs f(44) of SOA (B) at 55 ppb SO_2 in absence of seed particles. f44 (ratio of m/z 44 to total signal in the component mass spectrum) and f43 (defined similarly).

This has been added in the revised manuscript (Line 301-306 and 309-314) and Supporting Information (Line 125-138).

Q10. Lines 346-347. "SOA increased with decreasing particulate pH" This sentence may be true for a specific situation when aerosol composition changes (neutralization with ammonia and change in humidity). The concern is what is the definition of aerosol acidity in the authors' point. The pH is calculated from the proton concentration (mol/L) in aqueous phase. The increasing of the quantity of acidic sulfate cannot necessarily increase pH. pH mainly changes with inorganic composition and humidity. Authors should clarify how pH changes in the paper and how to define aerosol acidity in the discussion in the numerous places. The change in pH is different from the change in the quantity of sulfate without changing aerosol composition.

A10. According to the methods reported by Peng et al (2009),(Peng et al., 2019) we estimated the pH in the particles. For the E-AIM model, pH was estimated as follows:

$$pH = -log_{10}(f_{(H^+)} \times X_{(H^+)})$$

Where $f_{(H^+)}$ is the activity coefficient of H^+, $X_{(H^+)}$ is the molar fraction of H^+.

The acidity for nucleated H_2SO_4 particles (pH) under different SO_2 concentration have been estimated to be 3.27 and 3.46, respectively (Figure 5). The acidities for AS and AAS (pH) have been estimated to 7.3 and 4.1, respectively. As expectedly, it could be found that the formation of more H_2SO_4 particles and high acidity of seed reduced the pH of particles. This has been added in the revised manuscript (Line 370-373) and Supporting Information (Line 139-146).

Q11. Figure 7. The curved arrow points in clockwise direction with different reasons in both the left side and the right side boxes. It is difficult to understand the information in the right side.

A11. Figure 7 has been redrawn in the revised manuscript (Line 412).

Q12. Kinetic mechanisms on page 18. Reaction III may represent the H-shift followed by autoxidation. In general, H-shift occurs 5-membered geometry or 6-membered geometry. The H-shift that is depicted in III is based on a 8-membered ring. The H-shift with a higher than 6-member ring geometry may unlikely occurs. The authors need to clarify this.

A12. The kinetic mechanism of EM ozonolysis was proposed based on previous studies.(Newland et al., 2018; Jokinen et al., 2014; Bianchi et al., 2019) Moreover, during the ozonolysis of α-phellandrene and α-pinene, some highly oxidized multifunctional compounds could be produced via the 1,7- and 1,8-H shift process.(Kurtén et al., 2015; Mackenzie-Rae et al., 2018) Thus, we concluded that the H-shift followed by autoxidation could be proposed to be a formation pathway of highly oxidized multifunctional compounds.

This has been added in the revised manuscript (Line 429-434).

Q13. (1) Lack of the description of atmospheric implication of this study. In general, the concentration of EM in ambient air will be much lower than the chamber condition andtheir oxidation will be processed in the presence of OH radicals and UV light (daytime).

(2) Is ozonolysis is the main mechanisms of EM oxidation in ambient air? If the OH radical is not scavenged, what are the impacts of acidic sulfate on EM SOA formation?

(3) In polluted ambient areas, there is preexisting particulate matter. Is the EM ozonolysis + SO_2 (Craigie intermediate + SO_2) mechanism significant in such ambient air?

A13. (1) Thanks for giving us such valuable suggestions. In fact, the studies on the reaction between OH radicals and two unsaturated esters (including cis-3-hexenyl acetate and EM) have also been investigated in our chamber to further understand secondary particle formation of these unsaturated esters. Experimental results would be published in our next work.

(2) One of our recent works indicated that higher CO levels (90 ppm) were found to significantly change the chemical composition of SOA (prompting monomer while reducing oligomer formation) relative to low CO level (Zhang et al., 2020). Thus, we concluded that acidic sulfate would also have a significant impact on SOA formation via changing the composition of EM SOA. However, estimating contribution of O_3 and OH radical oxidation to the formation of EM SOA, PTR-TOFMS would be very

necessary.

(3) In China, O_3 pollution is gradually becoming serious environmental problem with the decrease in PM2.5 concentration recent years. Especially, sCI from the alkene ozonolysis has been frequently reported to exhibit high oxidation capability in the conversion of SO_2 and NO_2 to secondary particles.(Newland et al., 2018) Thus, we concluded that the ozonolysis of EM in the presence of SO_2 may not only be a pathway of EM degradation, but contribute to the formation of fine particles.

This has been added in the revised manuscript (Line 171-174, 428-433, and 487-490).

Q14. The author needs to discuss about organosulfate (OS) formation in different acidic aerosols and the potential impact of OS on SOA growth.

A14. Chen et al. suggested that the difference in sulfate fragments between AS and organosulfate could be used to verify the organosulfate formation.(Chen et al., 2019b) In order to make clear whether organosulfates were produced during EM ozonolysis, thus the sulfate fragments between AS and AAS experiments were also compared. As shown in Figure S17, it could be found that the distributions of sulfate in both of seed experiments were almost the same. Moreover, the S/C ratio were also the same in both of seed experiments. Taken together, it could be concluded that the contribution of organosulfates to SOA formation and OSc could be ignored. The increased oxidation state (OSc) of the aerosol may be attribute to the formation of more highly functionalized products. These discussions have been added in the revised manuscript (Line 390-394) and Supporting Information (147-160).

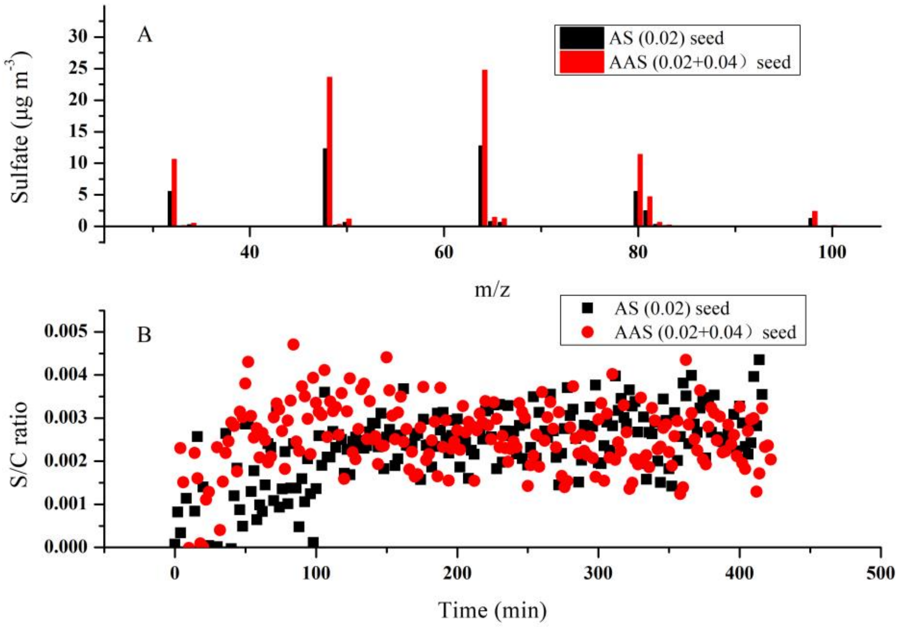

Figure S17. Mass spectrum (A) and S/C ratio (B) of sulfate in the presence of AS and AAS seed particles.

Reference

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