Response to Reviewer 1's comments:

We thank the reviewer for the comments. Below are our responses to the comments and corresponding modifications to the manuscript.

1. Experiments were made in presence of excess ozone to ensure the complete consumption of the terpenes. Have you performed tests experiments where this was not the case? Would expect any difference in the chemical regimes to SO_2 oxidation at lower ozone concentration? For instance, due to reduced peroxy-functions productions, concomitant presence of unsaturations, etc:

Yes, we have performed chamber experiments with limited ozone injection, as shown in Fig. R1. Ozone was injected into the chamber that was prefilled with limonene until it reached ~ 50 ppb. Additional ozone was added at 180 min. As can be observed from the figure, SO_2 consumption coincides with limonene depletion and particle growth, further confirming the interactions between SO_2 and limonene oxidation products. Comparing this result (e.g. the result of the 1st ozone injection) to the experiment conducted under ozone-rich conditions with similar SO_2 injection (Exp. #5, Table 1), lower SOA yield (41% vs. 52%) and smaller SO_2 consumption (4.0 ppb vs. 5.2 ppb) were observed, consistent with the results from other studies (Chen and Hopke, 2010; Leungsakul et al., 2005; Youssefi and Waring, 2014). This is likely due to the incomplete oxidation of the two double bonds of limonene or high volatility of second-generation reactions under ozone-limited conditions. We have not examined the detailed chemical changes of SOA products between these two scenarios (ozone-rich vs. ozone-limit). However, based on our observations regarding SOA yields and SO_2 consumption, as also mentioned by the reviewer, reduced formation of Criegee intermediates and peroxides is expected, which may weaken the effect of SO_2 on limonene SOA formation. Unsaturated compounds may also be present when limited ozone was injected (Maksymiuk et al., 2009).

The following content has been added and highlighted in the manuscript (Section 3.1):

"…Tests have also been performed by injecting ozone in two separate batches into the chamber prefilled with limonene, as shown in Fig. S1. Similar to the experiments conducted under ozone-rich conditions (e.g. Fig. 1), synergistic effects have been observed. We therefore infer from the correlation between depletion rate of SO_2 and particle formation that similar species or processes are responsible for SO_2 reaction and LSOA formation."
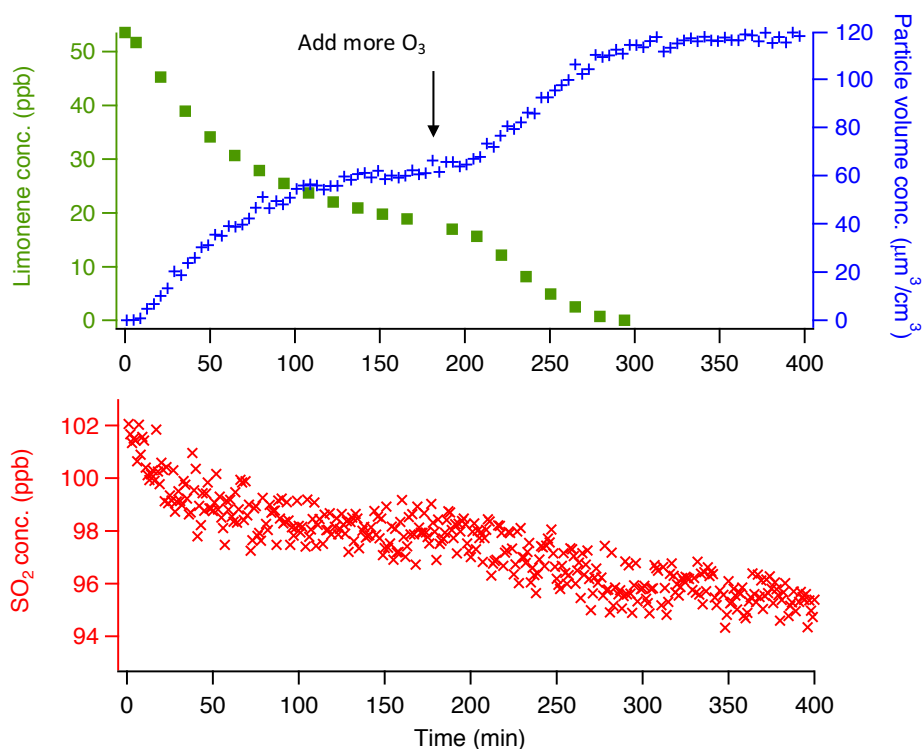
Figure R1 Particle volume concentration, limonene concentration and SO$_2$ concentration as a function of experimental time with stepwise ozone injection. Ozone was injected into the chamber that was prefilled with limonene until it reached around 50 ppb. Additional ozone was added at 180 min.

2. Maybe you could add in the experimental section 2.2, the ozone levels used in the flow tube experiments with, maybe, some indications how this compares to the chamber ones?

Thanks for the comments. The ozone concentration used in the flow tube experiments was around 3 ppm (in the absence of monoterpenes) as mentioned in Section 2.2. The inlet concentration of ozone was 6 times higher than those of monoterpenes. This concentration was chosen to achieve the greatest extent of oxidation in the flow tube. Based on literature rate constants, the lifetimes of α-pinene and limonene were calculated to be around 3 min and 2 min at these ozone concentrations, respectively. The residence time of chemical species in the flow tube reactor was 4 min.

Compared to chamber experiments, the flow residence time in the flow tube reactor is shorter, which may lead to less-oxidized SOA formation in the flow tube. However, this does not affect our conclusions regarding the effects of SO$_2$ on the change of SOA composition. The observations of organosulfate formation and increased SOA oxidation state in the presence of SO$_2$, are still valid despite the difference between the chamber and the flow tube.

The following content has been added into Section 3.4 of the manuscript:

3. You are making clear that limonene is needed to induce a SO_2 loss (line 225), but does this fully exclude that loss of SO_2 is not firstly physically driven by solubilization into the nascent aerosols? (Adding limonene would also affect such an equilibrium due to the growth rate of the particles).

Dissolution of SO_2 into the particles is governed by Henry's law. We calculated the dissolved SO_2 in two scenarios (acidic and neutral). pH = 5 is used for the calculation in acidic scenario because the pH for pure ammonium sulfate particle was estimated to be ~5 according to E-AIM Aerosol Thermodynamics Model (Model II, Clegg et al., 1998).

The effective SO_2 Henry's law constants $H^*_{S(IV)}$ under these two scenarios were taken to be 1×10^3 M atm^{-1} (pH = 5) and 2×10^5 M atm^{-1} (pH = 7), respectively (Seinfeld and Pandis, 2006). Assuming an initial SO_2 concentration of 600 ppb (the maximum SO_2 concentration used in this study), the concentrations of dissolved sulfur [S(IV)] in the aqueous phase can be calculated as 0.60 mM (pH = 5) and 120 mM (pH = 7). Assuming that the entire particle is aqueous, the aqueous mass concentration would be ~100 μg/m^3, and dissolution of SO_2 would only result in a decrease of 1.5×10^{-3} ppb (pH = 5) and 0.3 ppb (pH = 7) gaseous SO_2, which is at least one order of magnitude less than observed. Therefore, we conclude that the loss of SO_2 into particle phase solely due to dissolution is negligible.

4. The paragraph starting at line 230 is slightly confusing to me. How do you conclude/affect to the growth effect to sulfuric acid? How would that acid be produced efficiently in your system? OH reaction can be excluded due to the presence of an OH scavenger and ozone reacts quite slowly under acidic conditions. Would all this just be linked to sCI chemistry? Maybe adding a few words of explanation would be helpful.

The formation of sulfuric acid was predicted based on the chemical mass balance. By assuming that all the loss of SO_2 leads to the formation of sulfuric acid, we can estimate the upper limit of sulfuric acid that was formed in the condensed phase in our experiments.

Oxidation of SO_2 by Criegee intermediates from monoterpene ozonolysis leads to efficient production of sulfuric acid (Sipilä et al., 2014). However, it is also observed in this study that SO_2 can be oxidized by organic peroxides in the particle phase, which may also contribute to the formation of sulfuric acid especially under humid conditions. The relative contributions of each pathway under dry and humid conditions are shown in Fig. 8A and 8B.

Thanks for the comments. The role of humidity in organosulfate formation can be complicated. On one hand, increased humidity reduces acidity in the particle phase. However, particle acidity was found to promote the formation of organosulfates. Surratt et al. (2008) demonstrated that sulfate formation (including inorganic sulfate and organosulfates) in isoprene and monoterpene oxidation system increased with increasing seed particle acidity. Chan et al. (2011) observed that the abundances of organosulfates from β-caryophyllene photooxidation correlates strongly with aerosol acidity. On the other hand, increased humidity enhances the interactions between SO_2 and organic peroxides as shown in this study. Conclusion cannot be drawn without detailed mechanism studies whether organosulfate can be formed through SO_2/peroxide reactions. However, study is currently conducted in this group to examine the interactions between organic peroxide and SO_2.

With regards to the effect of ozone, we have conducted control experiment by adding SO_2, ozone, formic acid and ammonium sulfate into the chamber, as shown in Fig. S4A in the Supporting Information. No significant decrease of SO_2 was observed, indicating that dissolved ozone in the aqueous phase is not an important sink of SO_2 in this study. This is likely because that ammonium sulfate seed is slightly acidic (pH ~ 5, based on the calculation from E-AIM Aerosol Thermodynamic Model) that is less favorable for SO_2 oxidation by ozone in the aqueous phase. In addition, unlike SO_2 oxidation in a cloud droplet, the liquid water content in ammonium sulfate particles is limited. Therefore, little SO_2 depletion was observed. However, we agree with the reviewer that competition exists between SO_2/ozone reaction and SO_2/peroxide chemistry under high pH conditions and during cloud processing.

Thanks for the comments. The references were added into the manuscript.

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