The paper proposes a mechanism of SO_2 oxidation by organic peroxides in aerosol particles, through which the formation of biogenic secondary organic aerosol is enhanced. Reactions between SO_2 and stabilized Criegee Intermediates or organic peroxides are suggested to be the dominant sinks for SO_2 under dry or humid conditions. Although the oxidation of SO_2 has been addressed by a number of studies over the past decades, rapid oxidation of SO_2, readily observed in heavily polluted area, remains unexplained by existing mechanisms. The current contribution is a welcome addition to the field.

The authors suggested that under dry conditions, particle acidity is increased due to the condensation of H_2SO_4 to the aerosols particles, thus enhancing SOA formation through acid catalyzed reactions. Under dry conditions (~ 10% RH), while H2SO4 is hygroscopic, the particles will contain less liquid water, resulting in possibly low dissociation of H_2SO_4 and hence low hydrogen activity. And in the humid condition (~ 50% RH), the authors explained that due to high liquid water content of the particles, the acidity of the aerosols were decreased thus resulting in decreased SOA yields. The aerosol water can potentially increase the dissolution of H_2SO_4, thus increasing the acidity too. However, as organics are formed, they may be hygroscopic and their presence reduces the DRH of AS so the solution will be less acidic. Overall, role of condensation of H_2SO_4 and effects of SOA on changes of phase and aerosol liquid water of the particles need to be incorporated in the discussion.

Overall:

1. OS vs. inorganic sulfate. OS and oligomers analyses are not quantitative, it is not easy to rely on their peak identifications to explain the difference in yields under wet and dry conditions.
2. Aerosol acidity has been used heavily to explain the results. Need better, at least, semi-quantitative discussions, on aerosol acidity. Dissociation of H_2SO_4 under dry condition and influence of organics/SOAs on DRH of AS need to be discussed.

There are several places in the paper are inconsistent and some explanations are a bit too obscure as detailed in the major comments below. Beyond these, I do not see any major obstacles to publication.

Major comments:

1. Line 230

For all the experiments in Figure 2, when all the limonene was consumed, the particle volume concentration seemed kept increasing, *i.e.,* Exp.#7, dark blue markers vertically stacked at 30 ppb limonene. Does that mean that particle volume concentration kept increasing after limonene was completely consumed? This is inconsistent with Figure 1 that when all the limonene was consumed, the particle volume concentration reached a plateau.

2. Line 235 & Line 245

The authors attribute the increased SOA formation to increased particle acidity.

Experiments #1 - #10 were performed at RH below 16 %, where the particles might have less water. I'm not sure particle acidity is increased as it is a reflection of hydrogen activity, where aerosol water is needed. See comments earlier.

3. Line 250

The authors stated that "increase in particle volume concentration was also observed in the presence of SO_2 under humid conditions.

In Figure 1, I cannot tell if the increase is by comparing Seed volume and Final volume between Exp #11-13 and Exp #14-15, please clarify.

4. Line 255

The authors stated that aerosol water decreased particle acidity, resulting in decreased SOA enhancement in humid condition. However, if we compare experiments without SO_2, i.e., Expt #1-3, with Expt #11-13, SOA enhancement also showed a decrease under humid condition when compared with dry condition. Acidity should not be the issue in the experiments without SO_2, does that mean it is the humidity that affects the SOA yield instead of the acid catalyzed formation?

5. Line 270

SO_2 can dissolve into the aqueous droplets, what is its contribution to the measured SO2 gas consumption?

6. Lin 320 & Figure 4

I understand the authors conducted flow tube experiments in order to collect more products for analysis. But I'm not sure the flow tube experiments are exactly comparable to the chamber ones. The key issue is that OH scavenger is not added in the flow tube experiments. It is more efficient in producing peroxides in OH system than in O_3 system due to the RO2 + HO2 reactions. Therefore, the bulk experiments using the LSOA extracts without scavenger will bias the absorption of SO_2 high compared with the real chamber SOA material where OH scavenger is present.

7. Line 300

Even H_2SO_4 can be formed, I'm not sure in terms of the fate of "Sulfur", which is formed more significantly H_2SO_4 and OS? This is related to the mechanism/yield of SOA formation under dry condition. The authors stated aerosol acidity increased SOA yield but what is the acidity of aerosols in dry condition? And the authors did observe quite some OS molecules, will this be the major reason for increased SOA yield? Please clarify.

8. Line 345

As the author said, aqueous reaction of SO_2 with O_3 in forming sulfate is proved to be quite efficient. What is the reason that SO_2 is not reacted efficiently with O_3 in these experiments? Any estimates for the rate constant of SO_2+O_3 in this experiment?

9. Line 370

What is the resolution of the mass spectrometers? I wonder if it can give four decimals for the detected m/z.

10. Line 420

"first generation of oxidation products from alpha-pinene ozonolysis may be too volatile to condense…"

There are quite some studies (*i.e.*, Ehn et al., Nature, 2014) showing that first generation of oxidation products were supposed to be HOMs, which are of extremely low vapor pressures and can condense easily. Please explain.

11. Line 455

“We present evidence to suggest that HSO_3^- can further react with organic peroxides produced from monoterpene ozonolysis”

I don’t think the authors give any explanation before line 455 about reaction between HSO_3^- with organic peroxides. The authors only show the evidence that organic peroxides decreased when bubbling SO_2 into the solution. Please explain the reaction mechanism of HSO_3^- with organic peroxides.

Reference：

Ehn, M.; Thornton, J. A.; Kleist, E.; Sipila, M.; Junninen, H.; Pullinen, I.; Springer, M.; Rubach, F.; Tillmann, R.; Lee, B.; *et al.* A Large Source of Low-Volatility Secondary Organic Aerosol. *Nature* **2014**, *506*, 476–479.