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Comment on acp-2021-61

Anonymous Referee #3

Referee comment on "SO_2 and NH_3 emissions enhance organosulfur compounds and fine particle formation from the photooxidation of a typical aromatic hydrocarbon" by Zhaomin Yang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-61-RC3, 2021

Overview:

The manuscript by Yang et al. examined the effect of SO_2 and NH_3 on the formation of SOA from 1,2,4-trimethylbenzene photooxidation. After the injection of SO_2 and/or NH_3, the apparent yield of the SOA increased after wall loss correction, which demonstrated the synergistic effect of SO_2 and NH_3 in facilitating SOA formation. The authors also used ATR-FTIR, IC, and UPLC-MS to systematically analyze the particle phase composition and identified various inorganic and organo-sulfates compounds. My main comments are about the control experiment setup in this manuscript, which are discussed below. Other than the main comments, the manuscript is written clearly and comprehensive.

Major Comments:

The authors demonstrate that the addition of SO_2 and/or NH_3 during the photo-oxidation experiments can enhance the SOA formation and yield. However, part of the aerosol growth can arise from the formation of H_2SO_4 or $(NH_4)_2SO_4$particles even without the presence of any SOA, which should be deducted from the enhancement effect of SO_2 and/or NH_3.

I suggest the authors add in three control experiments (pure SO_2, pure NH_3, and SO_2+NH_3 mixtures but with similar levels of OH radicals and UV intensity using H_2O_2 or other OH generators) without any VOCs to rule out the formation of inorganic species contributing to the SOA. Figure S7 shows the mass spectra with and without SO_2 and/or NH_3 are similar, suggesting that maybe inorganic species formed with OH are the likely source of enhancement. It would be important to understand that after ruling out this part of the inorganic aerosol formation, how much enhancement the $SO_2$and/or NH_3 would add to the yield of the SOA.

Another comment I have is that the enhancement of the SOA from the addition of SO_2 and/or NH_3 can also be attributed to the increase of the surface area from the formation of inorganic species, which shifts the gas-particle equilibrium more to the particle side. Can the authors discuss more about the effect of this shift of equilibrium? It seems Figure 4 shows that the first three experiments after adding SO_2 seem to follow this rule.

Minor Comment:

L217: Can the author make an additional plot in the SI or show how the size dependent wall loss factor is generated?

Figure 1: It would be better to change the plots all in the same scale for easy comparison of different conditions.

L305: an extra space before "to"

L367: Please add the recent paper by Chen et al. 2020 also demonstrate that OS-228 could be from isoprene-derived SOA.

L389: The author should also consider adding Zhang et al. 2019 here, which discussed the effects of inorganic sulfates to organosulfates conversion in affecting aerosol growth, multiphase chemistry, and pH.

L525: As mentioned, OS-228 was observed by Chen et al. 2020 from the aging of isoprene-SOA. Even though the source might be different from the OS-228 here, the author should mention it in the conclusion instead of defining OS-228 as from unknown source.

References:

Chen, Y., et al. (2020). "Heterogeneous Hydroxyl Radical Oxidation of Isoprene-Epoxydiol-Derived Methyltetrol Sulfates: Plausible Formation Mechanisms of Previously Unexplained Organosulfates in Ambient Fine Aerosols." Environmental Science & Technology Letters **7** (7): 460-468.

Zhang, Y., et al. (2019). "Joint Impacts of Acidity and Viscosity on the Formation of Secondary Organic Aerosol from Isoprene Epoxydiols (IEPOX) in Phase Separated Particles." ACS Earth and Space Chemistry **3**(12): 2646-2658.