Atmospheric Chemistry and Physics Discussions

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# Comment on acp-2021-61

Anonymous Referee #1

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-RC1, 2021

This smog chamber study investigated the effects of $NH_3$, $SO_2$, and $NO_x$ on SOA formation under UV irradiation from 1,2,4-trimethylbenzene (TMB) over a period of 5-6 hours. The smog chamber was monitored in real time using a scanning mobility particle sizer; gas analyzers for $SO_2$, $NO_x$, $NH_3$, and $O_3$; and gas chromatography-flame ionization for TMB. Offline analysis used infrared spectrophotometry, ion chromatography, and liquid chromatography-mass spectrometry. The study found that the presence of $NH_3$ and $SO_2$ both individually and synergetically increase SOA yield, and that $SO_2$ speeds nucleation, possibly through uptake onto $H_2SO_4$ surfaces. New organosulfates were identified and reaction schemes and structures were proposed for some; some organosulfates had molecular weights consistent with TMB-derived aerosol components found in the atmosphere. $NH_3$ was found to react to form organic nitrogen compounds in the aerosol phase, but only in the presence of $SO_2$, attributed to formation of ammonium sulfate. Aerosol components had a wide range of volatility almost nine orders of magnitude, as predicted by elemental composition from mass spectrometry.

This study is a good match for the scope and aims of ACP, and it does an excellent job conveying its own novelty even if the reader is not well versed in aromatic aerosol chemistry. It represents a necessary contribution to the aerosol research community's understanding of $SO_2$ and $NH_3$ dynamics, and has implications for reactions of a large number of aromatic VOCs as well as human health in the findings regarding enhanced UFP fraction.

## Major Comments

This article is solid, if not concise, and all sections contribute to the main point as a cohesive whole. The introduction, methods section, and discussion of limitations are excellent, and while there is far more description of results than there is analysis of results I believe that both the analysis and discussion of atmospheric implications are sufficient. All of my suggestions for improvement are minor, relating to clarity and readability.

## Minor Comments

line 13: Consider a brief explanation of the atmospheric relevance of TMB in the abstract.

line 69-70: "Equivocally not originated from biogenic VOCs" is a little unclear. Possibly change to "unidentified OSs with C2-C25 skeletons that may not have originated from biogenic VOCs" or similar.

line 254-55: Make it clear whether ammonium sulfate particles were introduced during experiments or as an independent wall loss experiment. Is this dependent on humidity or hygroscopicity of the particles? Not VERY important, but may be a limitation worth discussing.

line 284: Were these from two single experiments? If so, include experiment numbers. Otherwise I want to know the standard deviations for these values.

line 291-95: Before noting that UFP are more harmful to human health, you may want to make it explicit in the text analysis of figure 1 that increased $SO_2$ concentration increases the fraction of UFP in the aerosol size distribution.

line 323: I would be interested in more detail about the mechanics of $H_2SO_4$ as a "condensed surface for key compounds." I take this to mean reactive uptake or heterogeneous reactions of VOCs with $H_2SO_4$. However, you don't justify this assertion; I might recommend citing Wang et al. 2010 (doi.org/10.1021/es9036868) and/or Zhang et al. 2019 (doi.org/10.1021/acsearthspacechem.9b00209) for VOC heterogeneous reaction with sulfuric acid surfaces.

line 329-30: As neither this work nor that of Julin et al. attempts to explain the "nonlinear dynamics of aerosol populations," rather than attributing your results to these dynamics it might be more effective to say something like "The nonlinear response of the mean particle diameter to $SO_2$ initial concentration is similar to results found by Julin et al. (2018) in a modelling study."

line 336: Is this the mean particle diameter after 30 minutes? Be explicit. You defined initial growth rate as average for 0-30 minutes, but did not define a timeframe for mean diameter.

line 383-84: If you believe you have not sufficiently justified the necessity of chemical composition studies, I recommend you do so in the introduction instead.

line 589: Figure 9 may belong in the supplement, because it has only a brief mention in the text that is limited to the observation that positive mode MS spectra skew toward lower m/z's.

line 594, 600: The contents of Figure 10 are also not discussed in the text, so it could be moved to the supplement as well.

line 509-10: use comparable measures to compare the $SO_2$-involved and $SO_2$ free conditions. I would recommend including the multiplicative factor of the change for both.

line 524-26: The sentence "Initial growth rate... as shown in Fig. S6" is redundant, and to the next sentence "In $SO_2$-free experiments... initial growth rate of aerosol particles" I would add something like "compared to the $SO_2$-involved condition."