We sincerely thank the Referee for the valuable comments. Our manuscript has been revised according to the comments from the Referee and our responses to the comments are as follows. For clarity, the comments are reproduced in blue, authors' responses are in black and changes in the manuscript are in red.

Responses to Anonymous Referee #1

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.

Author Reply:

Aromatic hydrocarbons can dominate the VOC budget in certain urban areas with TMB being one of the most significant species (Ran et al., 2009). TMB is mostly emitted from anthropogenic sources such as solvent use (Mo et al., 2021). Recent studies have recognized TMB as an important species in SOA formation in the atmosphere (Wang et al., 2020; Zaytsev et al., 2019; Mehra et al., 2020). Hence, the following text has been added in the abstract to explain the relevance of TMB in the atmosphere.

Aromatic hydrocarbons can dominate the volatile organic compounds budget in the urban atmosphere. Among them, 1,2,4-trimethylbenzene (TMB), mainly emitted from solvent use, is one of the most important secondary organic aerosols (SOA) precursors.

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.

Author Reply:

We have modified the sentence in the revised manuscript as: Recent field studies reported that some unidentified OSs with C_2–C_{25} skeletons may not be originated from biogenic VOCs.

line 254-55: Make it clear whether ammonium sulfate particles were introduced during experiments or as an independent wall loss experiment.

Author Reply:

Ammonium sulfate (AS) seed particles were not introduced into the chamber over the course of particle formation experiments. We used inert AS particles only in independent wall loss experiments in order to characterize the wall loss rate of AS particles, which was adopted to correct aerosol particle concentrations. This particle wall-loss correction method has been commonly used in previous studies (Chen et al., 2019; Charan et al., 2020). For clarity, we have included the following sentences in the revised manuscript.

Page 5, lines 168–169

Seed particles were not introduced into the chamber over the course of particle formation experiments.

Page 10, line 275

In order to determine the particles wall loss rates, we carried out independent wall loss experiments.

Is this dependent on humidity or hygroscopicity of the particles? Not VERY important, but may be a limitation worth discussing.

Author Reply:

We agree with the Referee that particle wall depositions are affected by the physicochemical properties of particles to some extent. It should be noted that the description of an important step of the wall loss experiments was missing in our original manuscript. Indeed, the AS solution was added to a TSI atomizer (Model 3076) to produce droplets, which passed simultaneously through a silica gel diffusion dryer to produce dry AS particles. Although the hygroscopicity of AS particles is not totally similar to aerosol particles generated, the wall-losses of particles are commonly evaluated by independent seed experiments where inert AS particles are used (Chen et al., 2019; Charan et al., 2020).

Aerosol particles can deposit onto the chamber walls due to gravitational settling, Brownian diffusion, convection, and electrostatic effects (Crump and Seinfeld, 1981). The wall-loss rate of particles depends mainly on the smog chamber design parameters

(Charan et al., 2019). Previous studies showed that elevating RH inside the chamber did not remarkably influence the particle wall loss rates (Yu et al., 2011; Ge et al., 2017). Ge et al. (2017) used a same wall loss constant for the particle wall-loss corrections in experiments with RH ranged from 11% to 90%. Although the changes in RH can influence the hygroscopicity, density, size, and chemical composition of particles (Hinks et al., 2018; Yu et al., 2011), almost identical humidity ((25 ± 1) %) condition was achieved among each experiment in the current work. Therefore, we believe that the particle wall depositions are not basically influenced by humidity or hygroscopicity of the particles. The description about the wall loss experiments has been included in the revised manuscript as follow.

Page 10, lines 276-279

An aqueous solution of ammonium sulfate was fed to a constant output atomizer (Model 3706, TSI, USA) to produce droplets, which passed simultaneously through a silica gel diffusion dryer to introduce dry particles into the chamber. The size distributions of ammonium sulfate particles were measured by SMPS for 480 min. The wall losses of particles are size-dependent and, thus, we used a size-dependent particle wall-loss correction approach, which is described in detail in the supplement

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

Author Reply:

Yes, the reported particle number concentrations were obtained in independent SO_2-free experiments with different initial NO_x levels. We have highlighted the experiment numbers in the revised manuscript as:

After 300 min UV irradiation without SO_2 introduction, the total maximum number concentration of aerosol particles was only 2.7×10^4 cm^{-3} and 2.9×10^4 cm^{-3} under low-NO_x (Exp. 1) and high-NO_x (Exp. 5) experiments, respectively.

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.

Author Reply:

The SMPS instrument could measure particles larger than 4.50 nm in this work. In order to more clearly analyze the effects of SO_2 on the ultrafine particle formation, the number concentrations of ultrafine particles in the range of 4.50–100 nm were summed up and they are shown in Fig. R1, where it can be seen that the total number concentration of ultrafine particles is significantly enhanced with increasing SO_2 levels. Therefore, the following text was added in the revised manuscript and Fig. R1 was added as Fig. S2 in the revised supplement.

Page 10, lines 309–315

Interestingly, the particle maximum number concentration considerably increased with increasing SO_2 levels, regardless of low- or high-NO_x conditions (Table 1). As shown in Fig. 1, although the unimodal particle size distribution in the presence of SO_2 was similar to that in its absence, particles with mobility diameter ranging from 10 to 80 nm, especially, dominated particle number concentrations with SO_2 addition. The time series of total ultrafine particles (diameter < 100 nm) number concentration are shown in Fig. S2, where it is seen that SO_2 considerably enhanced ultrafine particle formation.

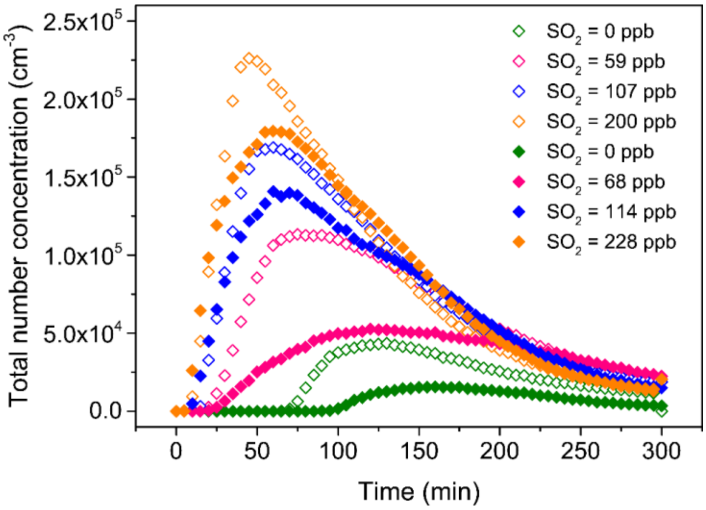

Figure R1. The total number concentrations of ultrafine particles (< 100 nm) as a function of reaction time (Exps. 1–8). The open symbols and solid symbols represent low- and high-NO*x* experiments, respectively.

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.

Author Reply:

The oxidation of SO_2 can contribute to the formation of H_2SO_4 in SO_2-involved experiments. The formed H_2SO_4 plays a key role in nucleation and can contribute to the increase in nucleation rate (Sipila et al., 2010; Lehtipalo et al., 2018; Yao et al., 2018), which are responsible for the short nucleation time observed in the TMB/NO_x/SO_2 regime (Wyche et al., 2009). Several H_2SO_4 nucleation mechanisms have been suggested, including binary nucleation, ternary nucleation, ion-induced nucleation, and so on (Sipila et al., 2010; Lehtipalo et al., 2018; Yao et al., 2018). SOA-forming compounds can be produced in the gas-phase via photooxidation of TMB, and the chamber walls and particle surfaces are competitive condensation sinks for these compounds (Charan et al., 2020). Seed particles were not introduced into the chamber during particle production experiments in this work. In the original sentence, we meant that the H_2SO_4-induced new particles are an attractive condensation sinks for SOA-forming compounds and can subsequently provide surfaces like seed particles onto which the compound can condense.

We have carefully read the two recommended papers. Wang et al. (2010) found that H_2SO_4 can effectively uptake gas-phase alkylamines, hence contributing to the growth of aerosol particles. Zhang et al. (2019) highlighted the significance of uptake of isoprene-derived epoxydiols onto sulfate aerosol particles. This process can influence the phase state, morphology, and acidity of aerosol particles. Reactive uptake or heterogeneous reactions of organic vapors with H_2SO_4 are important, which can promote the growth of particle and the increase in particle mass (Deng et al., 2017; Wang et al., 2010). However, it should be noted that we focused on analyzing SO_2 effects on the particle nucleation time in line 323 of the original text. The heterogenous reactions promote mainly the formation of the accumulation mode particles instead of the nucleation mode particles (Lu et al., 2019). The original sentence has been revised as: The formed H_2SO_4 could induce nucleation and increase the nucleation rate (Zhao et al., 2018; Blair et al., 2017), and these processes are responsible for the short nucleation time observed in the TMB/NO_x/SO_2 regime (Wyche et al., 2009).

We fully agree with the Referee that acidic surfaces on aerosol particles can promote the reactive uptake of organic vapors (Zhang et al., 2019). The acid-catalyzed heterogenous reactions were already discussed in Sect. 3.1.2 in the original manuscript as follows.

"The increase in aerosol acidity could be largely responsible for the observed enhancements in SOA formation in SO_2-invloved experiments. The OH oxidation of TMB can result in the formation of multifunctional carbonyl compounds (Liu et al., 2012; Zaytsev et al., 2019), which could promote SOA formation via acid-catalyzed heterogeneous reactions."

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."

Author Reply:

We have carefully read the study of Julin et al. (2018) again, who suggested nonlinear response of particle size distributions to ammonia emissions. In the revised manuscript, the sentence has been revised as follows:

The nonlinear response of the particle mean diameter to SO_2 initial level is similar to the findings of Julin et al. (2018), who found the response of particle size distribution to NH_3 emissions to be also nonlinear.

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.

Author reply:

Figure R2 shows the mean diameter of aerosol particles at different reaction times, which was measured with SMPS. The particle mean diameter can stabilize after about 300 min photooxidation. Therefore, the particle mean diameter measured at 300 min in each experiment are compared in Fig. 2 in the main manuscript. The related text and Fig. 2 have been updated in the revised manuscript as follows.

Page 11, lines 331–335

The particle size rapidly increased within 30 min after nucleation, and gradually reached a stable level within 300 min photooxidation (Fig. 1). Consequently, the initial growth rate (GR_{inital}), calculated based on the method of Kulmala et al. (2012), is defined as the particle growth rate within 30 min after nucleation (Li et al., 2018). The mean diameter reported in this work represents the particle mean diameter measured at 300 min in each experiment.

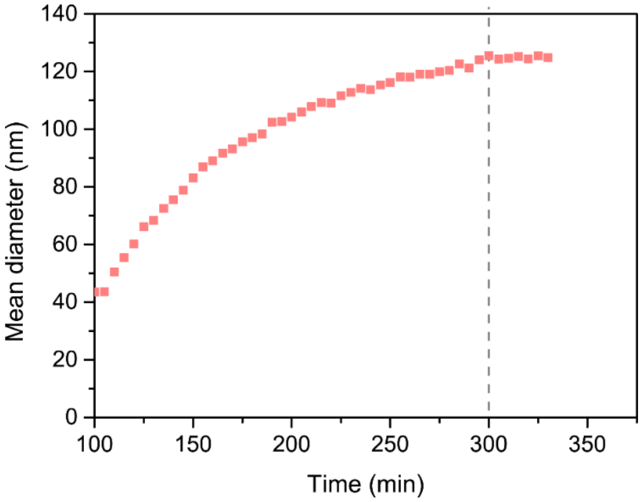

Figure R2. The mean diameter of aerosol particles generated as a function of reaction time.

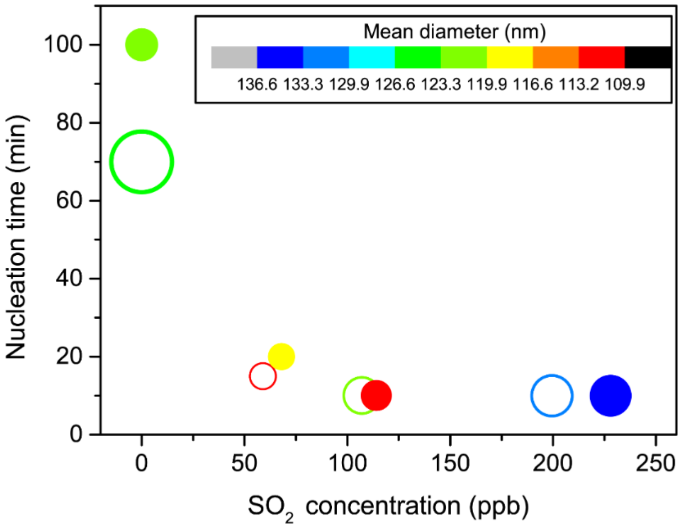

Figure 1. Particle nucleation time as a function of initial SO_2 concentration under low-NO_x (open circles) and high-NO_x (solid circles) conditions (Exps. 1–8). The symbol color indicates the particle mean diameter and symbol size represents the particle initial growth rate. Values of the particle parameters are listed in Table S1.

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.

Author Reply:

In the Introduction, we initially highlighted the necessity to analyze the chemical composition of particles as follows:

"To better understand TMB-SOA formation and growth, detailed laboratory characterization of TMB-SOA composition and TMB oxidation mechanisms with inorganic perturbation are required."

"The mechanisms leading to secondary aerosol formation in the urban environment remain highly elusive and controversial, particularly for the processes related to changes in secondary aerosol mass and chemical composition."

However, based on the Referee's suggestion, we moved the original sentence to the Introduction to further supplement the necessity of studying particle chemical composition.

Page 2, lines 55–62

High levels of SO_2, NH_3, and VOCs have been reported in certain regions such as

Guangzhou (Zou et al., 2015), Beijing (Meng et al., 2020), Handan (Li et al., 2017) in China. During haze pollution episodes, Li et al. (2017) observed that SO_2 levels can be up to 200 ppb in Handan, China. A recent study also showed significant increasing NH_3 levels in the atmosphere over the United States and the European Union (Warner et al., 2017). However, less focus has been placed on the SO_2 and NH_3 perturbations on SOA formation and chemical composition. Aerosol particles contain a multitude of compounds with different physicochemical properties.

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.

Author Reply:

Figure 9 has been moved to the supplement.

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.

Author Reply:

Figure 10 presents the predicted saturation mass concentrations of detected products, which is an important part of our discussion. Therefore, we keep Figure 10 in the revised manuscript.

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.

Author Reply:

We have modified the original sentence following the Referee's suggestion as: Increasing the NH_3 level to 200 ppb enhanced the particle maximum number concentration by factors of 2.0 and 1.7 under SO_2-free and SO_2-involved conditions, respectively.

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."

Author Reply:

We have deleted the sentence "Initial growth rate... as shown in Fig. S6" in the revised manuscript. In addition, we already compared the initial growth rate of particles in SO_2-free and SO_2-involved experiments in the original manuscript as: "In SO_2-free experiments, the increase in NH_3 initial concentrations led to remarkable increase of the initial growth rate of aerosol particles. High initial growth rates were also found in the photooxidation of other aromatics such as toluene and o/m/p-xylene with NH_3 addition (Li et al., 2018). This result may be explained by the fact that under SO_2-free condition, NH_3 mainly reacts with acids to produce ammonium salts. Previous studies have reported that ammonium salts could partition into the initial growth process of new secondary aerosol particles and thus increase the particle initial growth rate (Li et al., 2018; Zhu et al., 2014). More interestingly, NH_3 level did not substantially affect the initial growth rate of particles in the presence of SO_2 (Fig. S9)."

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