

## Responses to Referee #1's comments

**General comment:** Writing with an easy and fluent style. No obvious grammar problems. Easy to read and understand. However, use some conjunctions and sentence patterns again and again. For example, meanwhile, in addition, this phenomena indicated that... Conclusion is too weak. In other word, do not just use references to explain the normal results. Show the highlight of your own research.

**Response:** Many thanks for your constructive comments and valuable suggestions. The manuscript has been carefully revised according to your suggestion both in conjunctions and conclusions. We also added lots of discussion about our experimental results. And the conclusion has been rewritten more concrete in the revised manuscript.

### **Revision in the manuscript:**

**Line 28, 89, 123, 154, 260, 344, 367, 391, Delete** “Meanwhile”

**Line 79, Change** “Meanwhile” to “**In previous studies**”

**Line 205, Change** “Meanwhile” to “**Additionally**”

**Line 373, Change** “Meanwhile” to “**Moreover**”

**Line 336, Change** “This phenomenon further indicated that  $\text{NH}_4\text{NO}_3$  became a dominant nitrate species in the presence of  $\text{NH}_3$ .” **to:** “**Therefore,  $\text{NH}_4\text{NO}_3$  was the dominant nitrate species in the presence of  $\text{NH}_3$ .**”

**Line 341, Change** “These phenomena indicated” **to:** “**, which revealed**”

**Lines 435-436, Change** “This phenomenon indicated that the formation of more-oxygenated organic aerosol and nitrogen-containing organics was enhanced with the increase of  $\text{NH}_3$  concentration.” **to:** “**Thence, the formation of more-oxygenated**

organic aerosol and nitrogen-containing organics will be enhanced with the increase of NH<sub>3</sub> concentration.”

**The conclusion in the revised manuscript has been rewritten more concrete as follows:** In this study, SA formation from the photo-oxidation of gasoline/NO<sub>x</sub> in the presence of SO<sub>2</sub> or NH<sub>3</sub> was investigated. Our experimental results demonstrated that SA was enhanced by a factor of 1.6–2.6 or 2.0–2.5, respectively, with the increase of SO<sub>2</sub> or NH<sub>3</sub> concentration (0–151 ppb and 0–200 ppb, respectively). Meanwhile, both secondary organic aerosol (SOA) and secondary inorganic aerosol (SIA) were increased by varying degrees. In the presence of SO<sub>2</sub>, SO<sub>4</sub><sup>2-</sup> was the most sensitive linear increase with the increase of SO<sub>2</sub> concentration, and SOA was also greatly enhanced due to the acid catalytic effect and the formation of sulfur-containing organics. In the presence of NH<sub>3</sub>, NH<sub>4</sub>NO<sub>3</sub> was most enhanced, following by organic aerosol. The formation of nitrogen-containing organics was also promoted by the presence of NH<sub>3</sub>. Meanwhile, conspicuous new particle formation (NPF) and particle size growth were enhanced in the presence of SO<sub>2</sub> or NH<sub>3</sub>.

In this study, a linear relationship between the SA yield and SO<sub>2</sub> or NH<sub>3</sub> concentration was also obtained (Fig. S13). Considering the typical concentrations of SO<sub>2</sub> and NH<sub>3</sub> of 40 ppb and 23 ppb in haze pollution in the north China plain (Cheng et al., 2016), and the lower aromatics content (~ 10%) in vehicular evaporative emissions (Zhang et al., 2013), the SA yield is roughly estimated to be about 0.20. Recently, an updated emission inventory of vehicular evaporative emissions was reported to be 1.65 Tg yr<sup>-1</sup> (Liu et al., 2017a). Then, the SA formed from the photo-

oxidation of VOCs emitted by vehicular evaporation in the presence of SO<sub>2</sub> and NH<sub>3</sub> is roughly estimated to be 0.33 Tg yr<sup>-1</sup>, which is about 1.5 times as much as the primary PM<sub>2.5</sub> emissions from transportation (0.21 Tg yr<sup>-1</sup>) in China (Jing et al., 2015; Zhang et al., 2007) and accounting for about 21 % of the SOA production (1.6 Tg yr<sup>-1</sup>) from anthropogenic precursors estimated by global chemical transport model (Farina et al., 2010). In addition, the photo-oxidation of long-chain alkanes (> C<sub>6</sub>, IVOCs) contained in evaporative emissions would also contribute to SOA formation (Pye and Pouliot, 2012; Tkacik et al., 2012; Presto et al., 2009; Lim and Ziemann, 2005; Zhao et al., 2016). This estimate suggests that vehicular evaporative emissions will be a significant source of SA in the presence of SO<sub>2</sub> and NH<sub>3</sub>, although the estimate might have a high uncertainty due to the fact that SA yield might vary considerably under different atmospheric conditions. Meanwhile, in the presence of NO<sub>x</sub>, SO<sub>2</sub> and NH<sub>3</sub>, vehicular evaporative emissions may be a potential source of sulfur- and nitrogen-containing organics, according to the results obtained from our study. Sulfur- and nitrogen-containing organics will have an adverse influence on the climate by light absorption and/or by affecting aerosol hygroscopicity (Staudt et al., 2014; Nguyen et al., 2012), and they also have a significant contribution to SOA and nitrogen or sulfur budgets (Lee et al., 2016; Shang et al., 2016).

Therefore, under the compound pollution conditions of SO<sub>2</sub> and NH<sub>3</sub>, synergistic emission reduction of vehicular evaporative emissions, SO<sub>2</sub> (e.g., coal-fired flue gas) and NH<sub>3</sub> (e.g., emitted from agricultural non-point source and traffic emissions) should be taken into consideration by policy makers for future management, which will

contribute to reducing the burden of PM<sub>2.5</sub>, and then cut the environmental, economic and health costs caused by PM pollution. Our work will provide a scientific basis for taking corresponding control measures to relieve haze events in China. Additionally, there might be some differences between the VOCs composition of gasoline vapors directly injected to the smog chamber and vehicular evaporative emissions. Thus, further work should be focused on SA formation directly from vehicular evaporative emissions under coexisting SO<sub>2</sub> and NH<sub>3</sub> conditions to shed light on the formation mechanism of SA under more atmospherically relevant conditions.

**Comment 1:** Line 21-22: It is good to use specific number to show the results.

**Response:** Thank you very much for your suggestions. The concentration ranges of SO<sub>2</sub> and NH<sub>3</sub> have been supplemented in the revised manuscript.

**Revision in the manuscript:**

**Lines 21-23, Change** “It was found that increase in SO<sub>2</sub> and NH<sub>3</sub> concentrations could promote linearly the formation of SA, which could be enhanced by a factor of 1.6–2.6 and 2.0–2.5, respectively.” **to:** “It was found that increase in SO<sub>2</sub> and NH<sub>3</sub> concentrations (0–151 ppb and 0–200 ppb, respectively) could promote linearly the formation of SA, which could be enhanced by a factor of 1.6–2.6 and 2.0–2.5, respectively.”

**Comment 2:** Line 116-119: “For gaseous NO<sub>x</sub>, O<sub>3</sub> and SO<sub>2</sub>, a chemiluminescence...”

Each gaseous species has been described in detail later, so it is repeated to show NO<sub>x</sub>,

O<sub>3</sub> and SO<sub>2</sub> here.

**Response:** Thank you very much. According to your valuable comment, the repeated gaseous species have been deleted in the revised manuscript.

**Revision in the manuscript:**

**Lines 126-128, Delete** the repeated “NO<sub>x</sub>, O<sub>3</sub> and SO<sub>2</sub>”

**Comment 3:** Line 128: Why use this equation  $\rho = d_{va}/d_m$  to calculate mass concentration?

**Response:** Thanks for your comment. The equation  $\rho = d_{va}/d_m$  mentioned by DeCarlo et al. (2004) was used to calculate the density of PM, and then a relatively reliable PM mass concentration could be calculated based on the measured mobility diameter of particles with the SMPS. Many previous studies have demonstrated that PM effective density could be estimated by comparing mobility diameter from the DMA (i.e.,  $d_m$ ) and vacuum aerodynamic diameter from an Aerodyne AMS (i.e.,  $d_{va}$ ) in parallel (Jimenez et al., 2003a, b; Bahreini et al., 2005; Alfarra et al., 2006; Ng et al., 2007; Sato et al., 2010). According to this equation, the density of PM was calculated to be 1.5–1.6 g cm<sup>-3</sup>, which was in the range of density of SOA derived from aromatic hydrocarbons (1.24–1.48 g cm<sup>-3</sup>) (Sato et al., 2010) and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>, 1.72 g cm<sup>-3</sup>) (Bahreini et al., 2005) and could be comparable with the previous studies (Li et al., 2018).

**Comment 4:** Line 133-134: Better to describe RH control system here.

**Response:** Thanks for your comment. The RH control system is achieved by vaporizing Milli-Q ultrapure water contained in a 5.0 L high pressure resistant container and the water vapor is flushed with purified dry zero air into the smog chamber. And the humidification process does not introduce detectable hydrocarbons or particles into the chamber. RH inside the smog chamber could be adjusted from < 5 to 80 %. In this study, ~50 % RH was adjusted prior to each experiment.

**Revision in the manuscript:**

**Lines 153-155, Add:** “As for the RH control system, it is achieved by vaporizing Milli-Q ultrapure water contained in a 5.0 L high pressure resistant container and the water vapor is flushed with purified dry zero air into the chamber.”

**Comment 5:** Line 164: Pls explain why inject NO<sub>x</sub>.

**Response:** Thank you very much. In the experiments, NO was injected into the chamber through the standard gas cylinder (1020 ppm in N<sub>2</sub>, Beijing Huayuan). Considering there was a small fraction of NO<sub>2</sub> (< 1 %) in the standard gas cylinder of NO, so NO<sub>x</sub> was used as the proxy for injected NO. In addition, high concentrations of NO<sub>x</sub> have been observed in China haze pollution episodes (He et al., 2014; Zou et al., 2015). In order to avoid this misunderstanding, NO<sub>x</sub> in the revised manuscript has been replaced by NO.

**Revision in the manuscript:**

**Line 185, Change “NO<sub>x</sub>” to: “NO”**

**Line 188, Change “NO<sub>x</sub>” to: “NO”**

**Comment 6:** Line 165: How to combine experimental gasoline vapor with real case?

**Response:** Thanks for your valuable comment. In this study, gasoline vapors were used as a substitute for evaporative emissions. In recent years, vehicle ownership in China has increased rapidly, and vehicle-related pollutants (e.g. VOCs) have exacerbated the severity of compound atmospheric pollution, and caused frequent regional haze events in China. Meanwhile, with the implementation of tailpipe exhaust emission control measures, the proportionate share of vehicular evaporative emissions to this pollution has grown, so that it has become a non-negligible contributor (39.20 %) to ambient VOCs from anthropogenic sources (Liu et al., 2017). In addition, the long-chain alkanes (> C<sub>6</sub>, IVOCs) contained in evaporative emissions would also contribute to SOA formation (Pye and Pouliot, 2012; Tkacik et al., 2012; Presto et al., 2009; Lim and Ziemann, 2005; Zhao et al., 2016). Therefore, the study of the contribution of VOCs emitted by gasoline evaporation to secondary aerosol formation has an important environmental significance for understanding the causes of haze events.

We agree that there might be some differences between the VOCs composition of gasoline vapors directly injected to the smog chamber and evaporative emissions. Thus, further work should be focused on SA formation directly from vehicular evaporative emissions to shed light on the formation mechanism of SA under more atmospherically relevant conditions. Corresponding discussions have been supplemented in the revised manuscript.

**Revision in the manuscript:**

**Lines 476-480, Add:** “Additionally, there might be some differences between the VOCs composition of gasoline vapors directly injected to the smog chamber and vehicular evaporative emissions. Thus, further work should be focused on SA formation directly from vehicular evaporative emissions under coexisting SO<sub>2</sub> and NH<sub>3</sub> conditions to shed light on the formation mechanism of SA under more atmospherically relevant conditions.”

**Comment 7:** Line 181: Can not find Fig. S2 and S3.

**Response:** Thanks for your careful check. Fig. S2 and Fig. S3 could find in the Supplement and corresponding annotation has been added in the revised manuscript.

**Revision in the manuscript:**

**Line 202, Add:** “in the Supplement”

**Comment 8:** Line 186: Can not find Fig. S4.

**Response:** Thanks for your comment. Fig. S4 is shown in the Supplement and corresponding annotation has been added in the revised manuscript.

**Revision in the manuscript:**

**Line 208, Add:** “, in the Supplement”

**Comment 9:** Line 205: Can not find Fig. S6.

**Response:** Thanks for your careful check. Fig. S6 is given in the Supplement and corresponding annotation has been added in the revised manuscript. According to your



Comment 10, Fig. S6 has been changed to Fig. S8 in the revised Supplement.

**Revision in the manuscript:**

**Line 321, Add:** “, in the Supplement”

**Comment 10:** Line 204-210: ammonium aerosols should be discussed in 3.3.

**Response:** Thanks for your suggestion. The corresponding discussion has been moved to Section 3.3 in the revised manuscript.

**Revision in the manuscript:**

**Lines 320-328, Add:** “It is worth noting that ammonium aerosols were formed without the addition of gaseous  $\text{NH}_3$  (Fig. S8, in the Supplement), which signified that some  $\text{NH}_3$  was present in the background air in the chamber or introduced during the humidification process of the chamber (Liu et al., 2015c). Unfortunately, appropriate instruments are unavailable to measure the exact concentration of background  $\text{NH}_3$  in the chamber. According to the concentration of generated ammonium aerosols, the concentration of background  $\text{NH}_3$  was estimated to be ~15 ppb using the E-AIM model (Clegg and Brimblecombe, 2005; Wexler and Clegg, 2002; Clegg et al., 1998). Therefore, for the experiments with the presence of  $\text{NH}_3$ , the concentration of injected  $\text{NH}_3$  (150–200 ppb) was much higher than this value to identify the effect of  $\text{NH}_3$  on SA formation.”

**Comment 11:** Line 213: Delete #.

**Response:** Thank you. # has been deleted in the revised manuscript.

**Revision in the manuscript:**

**Line 237, Delete:** “#”

**Comment 12:** Line 212-213: There were many studies indicated the relationship between SO<sub>2</sub> and secondary aerosol. What is the highlight of your experiment?

**Response:** Thanks for your comment. You are right. Indeed, there were many previous studies have demonstrated that secondary aerosol (SA) formed from typical biogenic (e.g., isoprene and  $\alpha$ -pinene) (Lin et al., 2013; Jaoui et al., 2008; Kleindienst et al., 2006; Edney et al., 2005) and anthropogenic (e.g., toluene, o-xylene, and 1,3,5-trimethylbenzene) precursors (Chu et al., 2016; Liu et al., 2016; Santiago et al., 2012) could be greatly enhanced by the presence of SO<sub>2</sub>. While, at the present time, the effects of SO<sub>2</sub> on SA formation have rarely been studied under highly complex precursors, such as VOCs from vehicular evaporative emissions, especially in China. Previous studies have reported that vehicular evaporative emissions have become non-negligible contributors (39.20 %) to ambient VOCs from anthropogenic sources compared with vehicular tailpipe emissions (Liu et al., 2017). In addition, China has the highest concentration of SO<sub>2</sub> in the world due to a large proportion of energy supply from coal combustion (Bauduin et al., 2016). Therefore, in this study, the influence of SO<sub>2</sub> on SA formation from evaporative emissions was investigated to simulate the case in the ambient air, which has practical significance for shedding light on the formation mechanism of SA under more atmospherically relevant conditions, especially in China. Meanwhile, the different roles of SO<sub>2</sub> and NH<sub>3</sub> in secondary organic aerosol (SOA)

chemical properties were further compared by applying positive matrix factorization (PMF) analysis to the HR-ToF-AMS data. Our study indicates that the photo-oxidation of VOCs from vehicular evaporative emissions will be a significant source of SA in the presence of high concentrations of SO<sub>2</sub> and NH<sub>3</sub>. Moreover, these emissions might also be a potential source of sulfur- and nitrogen-containing organics. Our work provides a scientific basis for the synergistic emission reduction of secondary aerosol precursors, including NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub> and particularly VOCs, to mitigate PM pollution in China.

**Comment 13:** Line 215: Pls revise “were enhanced by one order of magnitude” to “enhanced by XXX to one order of magnitude”.

**Response:** Thanks for your suggestion. This sentence has been revised in the revised manuscript.

**Revision in the manuscript:**

**Lines 239-241, Change** “were enhanced by one order of magnitude in the presence of SO<sub>2</sub> (~130 ppb) in the photo-oxidation of high concentration toluene/NO<sub>x</sub>” **to:** “were enhanced by the presence of SO<sub>2</sub> (~130 ppb) to one order of magnitude in the photo-oxidation of high concentration toluene/NO<sub>x</sub>”

**Comment 14:** Line 240: “This phenomenon” means “the H<sup>+</sup> concentration was increased” and.

**Response:** Thanks for your comment. “This phenomenon” means that “the higher SOA concentration and SOA yield could be well explained by the enhancement of the particle

acidities”. In order to avoid this misunderstanding, “This phenomenon” was specifically pointed out in the revised manuscript.

**Revision in the manuscript:**

**Line 285, Change** “This phenomenon” **to:** “The higher SOA concentration and SOA yield”

**Comment 15:** Line 247-251: Why use large gap between  $\text{SO}_4^{2-}$  and  $\text{SO}_2$  rather than S-bearing organic fragments ( $\text{C}_x\text{H}_y\text{O}_z\text{S}$ ) to show the reason for existing of organic sulfur?

**Response:** Thanks very much for your comment. First, we found that there was a large gap between the concentration of formed  $\text{SO}_4^{2-}$  and the amount of consumed  $\text{SO}_2$  (after wall loss correction for  $\text{SO}_2$ , sulfuric acid gas and sulfate). As for this large gap, there are might be many possible reasons including the underestimation of deposition and heterogeneous reaction of sulfur species on the wall and the formation of organic sulfur-containing products and so on. In order to confirm the existing of sulfur-containing organics, we further utilized the characteristic ions  $\text{CSO}^+$ ,  $\text{CH}_3\text{SO}_2^+$  and  $\text{CH}_3\text{SO}_3^+$  detected by HR-ToF-AMS to quantify the amount of sulfur-containing organics. According to the estimation method for sulfur-containing organics mentioned in Huang et al. (2015), we found that the signal of these characteristic ions and the concentrations of sulfur-containing organics indeed increased with the  $\text{SO}_2$  initial concentration. Evidences of both two aspects were used to prove the photo-oxidation of gasoline vapor in the presence of  $\text{SO}_2$  was a noteworthy source of sulfur-containing organics.

In order to more directly explain the presence of sulfur-containing organics, we refer to

your comment, the S-bearing organic fragments ( $C_xH_yO_zS$ ) detected by HR-ToF-AMS were used to prove the presence of sulfur-containing organics.

**Revision in the manuscript:**

**Line 276, Delete:** “According to the linear fitting between the concentration of formed  $SO_4^{2-}$  and the amount of consumed  $SO_2$  (after wall loss correction for  $SO_2$ , sulfuric acid gas and sulfate), there was a large gap between the slope of the line and the ratio of  $M(SO_4^{2-})$  and  $M(SO_2)$ , as shown in Fig. S8. There are some possible reasons for this, including the underestimation of deposition and heterogeneous reaction of sulfur species on the wall, the formation of organic sulfur-containing products, and small leaks of pollutants from the smog chamber.”

**Comment 16:** Line 282: Too many “This phenomenon indicated”.

**Response:** Thanks very much for your comment. The sentences involving “This phenomenon indicated” have been modified in the revised manuscript.

**Revision in the manuscript:**

**Line 336, Change** “This phenomenon further indicated that  $NH_4NO_3$  became a dominant nitrate species in the presence of  $NH_3$ .” **to:** “Therefore,  $NH_4NO_3$  was the dominant nitrate species in the presence of  $NH_3$ .”

**Line 341, Change** “These phenomena indicated” **to:** “, which revealed”

**Lines 435-436, Change** “This phenomenon indicated that the formation of more-oxygenated organic aerosol and nitrogen-containing organics was enhanced with the increase of  $NH_3$  concentration.” **to:** “Thence, the formation of more-oxygenated

organic aerosol and nitrogen-containing organics will be enhanced with the increase of NH<sub>3</sub> concentration.”

**Comment 17:** Line 286: Too many “This result indicated that”.

**Response:** Thanks very much. The sentences involving “This result indicated that” have been deleted and modified in the revised manuscript.

**Revision in the manuscript:**

**Line 339, Delete** “This result indicated that NH<sub>3</sub> was incorporated in the photo-oxidation of gasoline vapor.”. Similar description has been expressed in the above sentence.

**Line 467, Delete** “Previous studies have indicated that”.

**Comment 18:** Line 402-403: Repeated conclusion.

**Response:** Thanks for your comment. This repeated sentence has been deleted in the revised manuscript.

**Revision in the manuscript:**

**Line 451, Delete:** “Our results indicate that the photo-oxidation of gasoline/NO<sub>x</sub> in the presence of SO<sub>2</sub> and NH<sub>3</sub> is a significant source of SA. Therefore, in order to mitigate PM<sub>2.5</sub> pollution in China, emission control strategies should not only pay attention to primary particulate emissions, but also focus on synergistic reduction of the emission of SA precursors including NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub> and, particularly, VOCs.”

**Comment 19:** Line 422-423: Repeated conclusion.

**Response:** Thank you very much for your valuable comment. This repeated sentence has been deleted and modified in the revised manuscript.

**Revision in the manuscript:**

**Line 471, Delete:** “Therefore, more attention should be paid to collaborative control reductions in vehicular evaporative emissions and gaseous pollutants, including NO<sub>x</sub>, SO<sub>2</sub>, and NH<sub>3</sub>.”

**Lines 471-475, Change** “This will contribute to reducing the burden of PM<sub>2.5</sub>, and then cut the environmental, economic and health costs caused by PM pollution. Corresponding emission controls should be taken into consideration by policy makers for future management.” **to:** “Therefore, under the compound pollution conditions of SO<sub>2</sub> and NH<sub>3</sub>, synergistic emission reduction of vehicular evaporative emissions, SO<sub>2</sub> (e.g., coal-fired flue gas) and NH<sub>3</sub> (e.g., emitted from agricultural non-point source and traffic emissions) should be taken into consideration by policy makers for future management, which will contribute to reducing the burden of PM<sub>2.5</sub>, and then cut the environmental, economic and health costs caused by PM pollution.”

**Comment 20:** Line 410-412: SA formation still be a small part from vehicular evaporation emissions. What is the meaning of your study?

**Response:** Thanks for your comment. First, an updated emission inventory of vehicular evaporative emissions have demonstrated that they have become a non-negligible contributor (39.20 %) to ambient VOCs from anthropogenic sources compared with

vehicular tailpipe emissions (Liu et al., 2017). In the past two decades, policy makers and researchers have mainly focused on tailpipe emissions but paid little attention to evaporative emissions from vehicles in China. For example, the evaporative emission limits in the China V standards, which was implemented nationally in 2018, are not much different from the limits in the China I standards implemented in 2000. Our results indicates that the photo-oxidation of gasoline vapor especially in the presence of SO<sub>2</sub> and NH<sub>3</sub> have a huge secondary aerosol formation potential. When the lower aromatics content (~ 10%) in vehicular evaporative emissions was considered (Zhang et al., 2013), the SA yield is roughly estimated to be about 0.20 and the SA production is roughly estimated to be 0.33 Tg yr<sup>-1</sup>, which is about 1.5 times as much as the primary PM<sub>2.5</sub> emissions from transportation (0.21 Tg yr<sup>-1</sup>) in China (Jing et al., 2015; Zhang et al., 2007) and accounting for about 21 % of the SOA production (1.6 Tg yr<sup>-1</sup>) from anthropogenic precursors estimated by global chemical transport model (Farina et al., 2010). Thence, under the compound pollution conditions of SO<sub>2</sub> (e.g., coal-fired flue gas) and NH<sub>3</sub> (e.g., emitted from agricultural non-point source and traffic emissions), the contribution of vehicle evaporative emissions to SOA formation cannot be ignored. Secondary, our results have revealed that vehicular evaporative emissions is a potential source of sulfur- and nitrogen-containing organics in the presence of NO<sub>x</sub>, SO<sub>2</sub> and NH<sub>3</sub>. Sulfur- and nitrogen-containing organics will have an adverse influence on the climate by light absorption and/or by affecting aerosol hygroscopicity (Staudt et al., 2014; Nguyen et al., 2012), and they also have a significant contribution to SOA and nitrogen or sulfur budgets (Lee et al., 2016; Shang et al., 2016).



Therefore, more attention should be paid to this primary emissions source (i.e., vehicular evaporative emissions), and especially its associated secondary aerosol formation, which is also the significance of our study. According to our study, synergistic emission reduction of vehicular evaporative emissions, SO<sub>2</sub> (e.g., coal-fired flue gas) and NH<sub>3</sub> (e.g., emitted from agricultural non-point source and traffic emissions) should be taken into consideration by policy makers for future management, which will contribute to reducing the burden of PM<sub>2.5</sub>, and then cut the environmental, economic and health costs caused by PM pollution.

Additionally, we agree that there might be some differences between the VOCs composition of gasoline vapors directly injected to the smog chamber and vehicular evaporative emissions. Thus, further work should be focused on SA formation directly from vehicular evaporative emissions to shed light on the formation mechanism of SA under more atmospherically relevant conditions.

**Revision in the manuscript:**

**Lines 455-463, Change** “Considering the typical concentrations of SO<sub>2</sub> and NH<sub>3</sub> of 40 ppb and 23 ppb in haze pollution in the north China plain (Cheng et al., 2016), the SA yield is roughly estimated to be about 0.3. Then, the SA formed from the photo-oxidation of VOCs emitted by vehicular evaporation in the presence of SO<sub>2</sub> and NH<sub>3</sub> is roughly estimated to be 0.49 Tg yr<sup>-1</sup>, which is about twice as much as the primary PM<sub>2.5</sub> emissions from transportation (0.21 Tg in 2007) in China (Jing et al., 2015; Zhang et al., 2007).” **To** “Considering the typical concentrations of SO<sub>2</sub> and NH<sub>3</sub> of 40 ppb and 23 ppb in haze pollution in the north China plain (Cheng et al., 2016), and the lower

aromatics content (~ 10%) in vehicular evaporative emissions (Zhang et al., 2013), the SA yield is roughly estimated to be about 0.20. Recently, an updated emission inventory of vehicular evaporative emissions was reported to be 1.65 Tg yr<sup>-1</sup> (Liu et al., 2017a). Then, the SA formed from the photo-oxidation of VOCs emitted by vehicular evaporation in the presence of SO<sub>2</sub> and NH<sub>3</sub> is roughly estimated to be 0.33 Tg yr<sup>-1</sup>, which is about 1.5 times as much as the primary PM<sub>2.5</sub> emissions from transportation (0.21 Tg yr<sup>-1</sup>) in China (Jing et al., 2015; Zhang et al., 2007) and accounting for about 21 % of the SOA production (1.6 Tg yr<sup>-1</sup>) from anthropogenic precursors estimated by global chemical transport model (Farina et al., 2010).”

**Lines 479-483, Add:** “Additionally, there might be some differences between the VOCs composition of gasoline vapors directly injected to the smog chamber and vehicular evaporative emissions. Thus, further work should be focused on SA formation directly from vehicular evaporative emissions under coexisting SO<sub>2</sub> and NH<sub>3</sub> conditions to shed light on the formation mechanism of SA under more atmospherically relevant conditions.”

#### References:

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