Response to Reviewer #1

General Remarks:

This paper presents an experimental study of SOA formation from dilute exhaust from two gasoline powered vehicle (Euro I and Euro 4) operated on Euro 3 gasoline. The vehicles were tested at idle. A total of five experiments were performed. The dilute exhaust was photo-oxidized in a smog chamber and substantial SOA was formed (exceeding the primary organic aerosol emissions). The paper calculated effective yields (~3 to 17%) and evaluated SOA mass closure (50-90% of SOA could be explained with measured precursors).

The basic experiments are quite similar to other recent papers on SOA formation from gasoline vehicle exhaust. The results also fall within the range of results from hose previous paper. It does expand the dataset on gasoline vehicle exhaust. The paper emphasizes the Chinese context as novel (“first” experiments performed in China). Not clear how important it is to perform these experiments in China, since China follows European emissions and fuel standards. The very small number of vehicles (2) and limited tests (5) all performed at idle (a very limited operating mode) makes it hard to draw much insight into SOA formation from in use vehicle operations. Therefore, the paper contributes little new knowledge.

The paper is well organized and the experiments were performed with high quality instrumentation.

Reply: As shown in the “Introduction” section in the manuscript, though China follows European fuel standards, gasoline fuel in China has relatively higher mass content of alkenes and aromatic hydrocarbons than that in US (Schauer et al., 2002; Zhang et al., 2013). The emission factors of $PM_{2.5}$, organic carbon (OC), element carbon (EC), NO_x, SO_2, NH_3 and non-methane hydrocarbons (NMHCs) for on-road vehicles in China were quite different from those in other countries (Liu et al., 2014; Y. L. Zhang et al., 2015). Given that the possession of LDGVs in China was 98.8 million in 2012 and increased at a rate of approximately 20% per year since 2005 (NBSC, 2013), and that gasoline cars are considered “cleaner” than diesel ones in

China, it is urgent to investigate the SOA formation from LDGVs in China in order that we can assess both primary and secondary contribution from gasoline vehicles and help formulate future vehicle emission control strategies taking the secondary formation into consideration. As diesel vehicles are considered dirtier, in many China's large cities they are not allowed to enter the core urban areas as a measure to lower urban pollution. Our study suggests gasoline cars are not so innocent and clean if considering secondary contributions. This is particularly important since SOA contributed substantially to $PM_{2.5}$ and ozone pollution is becoming more and more serious in China's megacities. We know that it would be much better to conduct more chamber simulations with more cars. However, it is not so easy to do this in a large chamber with regard to the costs and managements. We would find resources to do more based on this study. We have added some words to mention this quite limited number of experiments in the revised manuscript (lines 359-362):

"It is important to note that the reported data are only based on five chamber experiments with two LDGVs under idling conditions. More tests are needed to assess SOA formation from gasoline vehicle exhausts in China."

Specific comments:

Q1- The exhaust was transferred through Teflon transfer line and a GAST rotary vane pump (not clear if the system was heated). This set up likely caused substantial losses of both PM and VOCs before the chamber (we never use Teflon lines for PM sampling). Were these losses characterized? If not, it makes the results largely qualitative and every presentation of effective yield or mass closure needs to be qualified with this substantial uncertainty. Presumably there is less issues with the composition information (Figure 8).

Reply: A flow rate of as high as 20 L min^{-1} and a transfer line of as short as 5 m were used to provide residence time within seconds, and thus reduce the losses of particles and VOCs in the transfer lines. Furthermore, before being introduced into the reactor, gasoline vehicle exhausts were generally pumped through the transfer lines for half an hour to saturate the transfer lines with particles and VOCs while warming the catalytic converter. Losses of particles and VOCs in the introduction lines were determined by comparing the concentrations of total particle number and VOCs in the directly emitted exhausts and the ones after passing through the transfer lines. As shown in Figure 1, the distributions of particle number in the directly emitted exhausts and the ones after passing through the transfer lines were similar. The loss of total particle number was estimated to be less than 3%. The penetration efficiency of particles in the transfer line was also estimated by a laminar diffusional deposition model (Gormley and Kennedy, 1949). For particles with diameters larger than 10 nm, the penetration efficiency was higher than 95%, indicating minor losses of particles in the transfer line. The losses of VOCs in the transfer line were estimated to be less than 5%, which might lead to a small underestimation of SOA production.

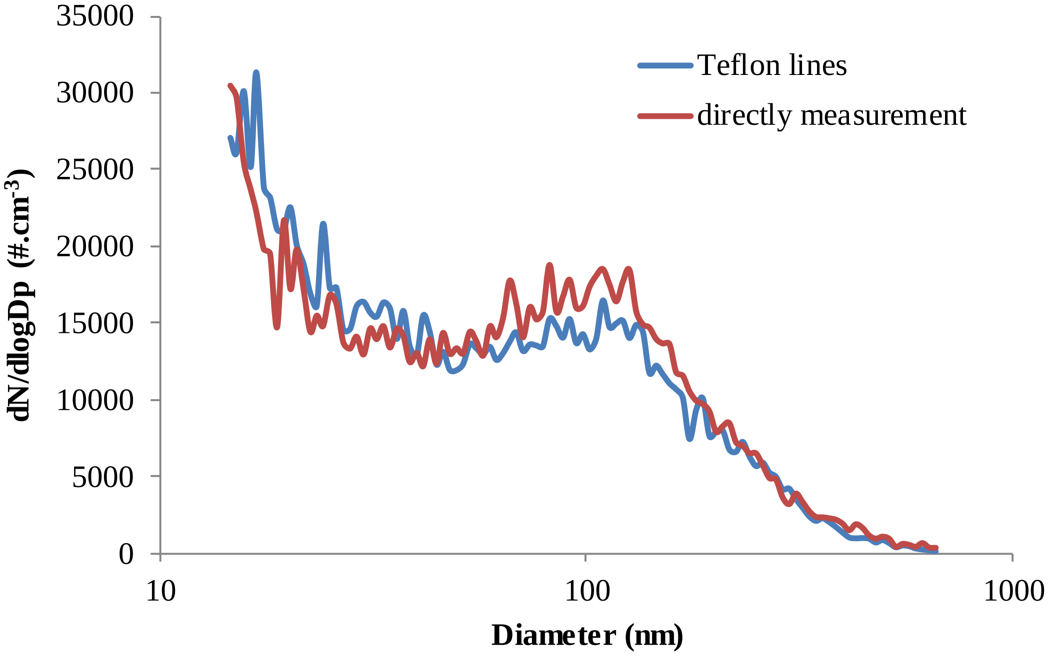


Figure 1. Particle number distributions of the directly emitted exhausts and the ones after passing through the transfer lines.

The following text has been added to the revised manuscript.

"During the introduction of exhausts, particles and VOCs might deposit to the surface of the transfer lines. Therefore, a flow rate of as high as 20 L min^{-1} and a transfer line of as short as 5 m were used to provide residence time within seconds, and thus reduce the losses of particles and VOCs in the transfer lines. Furthermore, before being introduced into the reactor, exhausts were generally pumped through the transfer lines for half an hour to saturate the transfer lines with particles and VOCs while warming the catalytic converter. Losses of particles and VOCs in the introduction lines were determined by comparing the concentrations of total particle number and VOCs in the directly emitted exhausts with the ones after passing through the transfer lines. The loss of total particle number was estimated to be less than 3%. The penetration efficiency of particles due to diffusion in a cylindrical tube, η(dp), can be also estimated by a laminar diffusional deposition model (Gormley and Kennedy, 1949). For particles with diameters larger than 10 nm, the penetration efficiency was higher than 95%, indicating minor losses of particles in the transfer line. The losses of VOCs in the transfer line were estimated to be less than 5%, which might lead to a small underestimation of SOA production."

Q2- The high, end of experiment SOA/POA ratios (Table 4) could largely be an artifact of substantial POA losses in the sampling system.

Reply: Conductive silicon tubes were used as sampling lines to reduce electrostatic losses of particles. In addition, the residence time of within seconds would also reduce the losses of particles. High SOA/POA ratios, mainly caused by the low concentrations of POA, might be related to the idling condition. Nordin et al. (2013) also observed high SOA/POA ratios and low concentrations of POA during the aging of emissions from idling Euro 2-4 light-duty gasoline vehicles.

The following text has been added to the revised manuscript.

"Conductive silicon tubes were used as sampling lines for HR-TOF-MS and SMPS to reduce electrostatic losses of particles."

Q3- This paper focuses on organic aerosol. What about total PM (that is what is regulated not organic aerosol)? Were there substantial refractory (e.g. BC) emissions?

Reply: Inorganic nitrate and ammonium were also formed synchronizing with SOA in some experiments. Formed nitrate mass could even reach 4–5 times as high as SOA. However, this paper mainly focused on organic aerosols, the formed inorganic aerosols will be discussed in detail in a subsequent paper. Theoretically, the difference of PM mass measured by AMS and SMPS should be attributed to black carbon. As shown in Fig. S3 (Fig. 3 in the manuscript) in the SI, the initial mass of PM measured by SMPS was comparable with that measured by HR-TOF-AMS, thus we assumed that the mass of black carbon (BC) in the reactor was negligible.

The following text has been added to the revised manuscript.

"As shown in Fig. S3, the mass of primary particles measured by SMPS was comparable with that measured by HR-TOF-AMS, thus we assumed that the mass of black carbon (BC) in the reactor was negligible."

Q4- It does not appear that seed particles were used in the experiment. Therefore there is likely substantial loss of condensable vapors to the chamber walls (Zhang et al. PNAS 2014). This will reduce the SOA production.

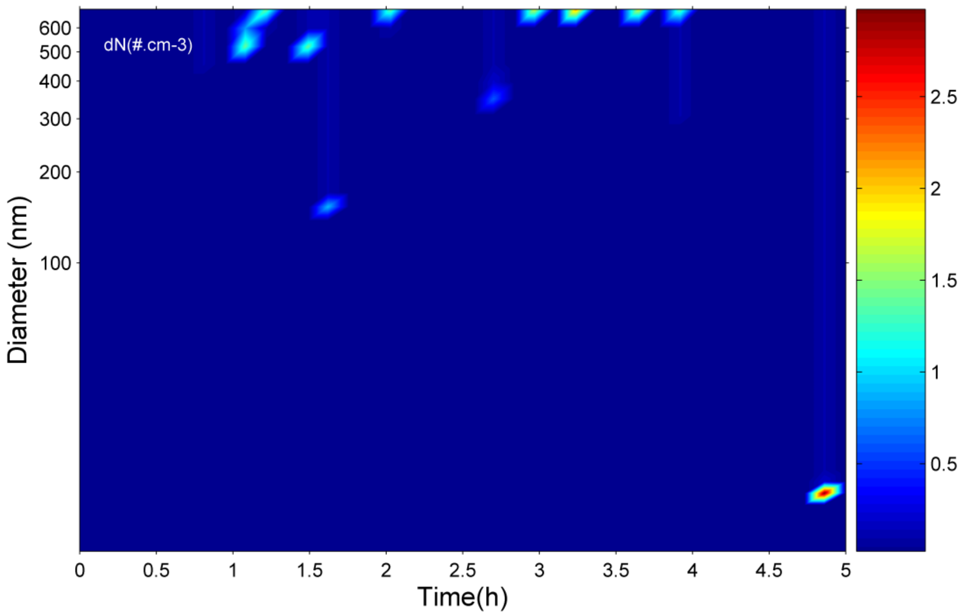
Reply: The wall loss rate coefficient of vapors is related with the numbers of carbon and oxygen in the molecule (X. Zhang et al., 2015). Here, we take $C_7H_8O_4$, a product of the photo-oxidation of toluene as an example. The loss of $C_7H_8O_4$ to walls would be 7% in an hour before SOA formation when a wall deposition rate of 2×10^{-5} s^{-1} was used (X. Zhang et al., 2015). After SOA formation, the surface concentrations of particles increased fast to as high as 2000 μm^2 cm^{-3} in an hour, which would reduce the vapor wall losses. Therefore, the underestimation of SOA production due to vapor wall losses seems minor.

The following text has been added to the revised manuscript:

"Deposition of SOA-forming vapors to the walls might lead to the underestimation of SOA production. The wall loss rate coefficient of vapors is related with the numbers of carbon and oxygen in the molecule (X. Zhang et al., 2015). Here, we take $C_7H_8O_4$, a product of the photo-oxidation of toluene as an example. The loss of $C_7H_8O_4$ to walls would be 7% in an hour before SOA formation when a wall deposition rate of 2×10^{-5} s^{-1} was used (X. Zhang et al., 2015). After SOA formation, the surface concentrations of particles increased fast to as high as 2000 μm^2 cm^{-3} in an hour, which would reduce the vapor wall losses."

Q5- Blanks – Did they run any blank experiments (with no vehicle exhaust) to quantify contamination?

Reply: Blank experiments with no vehicle exhaust introduced were performed to quantify the reactivity of the matrix gas. After 5 h of irradiation, the number and mass of formed particles were <5 cm^{-3} and 0.1 μg m^{-3}, respectively. Particle number distribution in a blank experiment was shown.



The following text has been added to the revised manuscript:

"Blank experiments with no vehicle exhaust introduced were performed to quantify the reactivity of the matrix gas. After 5 h of irradiation, the number and mass of formed particles were <5 cm^{-3} and 0.1 μg m^{-3}, respectively."

Q6- "alkanes shared about 42.9 and 66.2% of the total speciated NMHCs measured with the GC-FID/MSD by mass, respectively, dominating the NMHCs emissions in gasoline vehicle" not sure what "shared" means? Contributed might be a better word. It is not clear that alkanes dominate NMHC emissions, only NMHCs that were speciated in this study. This needs to be rephrased.

Reply: "shared" was changed to "*contributed*" as suggested in the revised manuscript. "dominating the NMHCs emissions" was changed to "*dominating the speciated VOCs emissions*" in the revised manuscript. NMHCs were changed to "*VOCs*" for purpose of consistency as suggested by referee 2.

Q7- "emission factors of NMHCs and aromatic hydrocarbons for vehicle I were 2.1 and 0.8 g kg^{-1}, approximately 1.3 and 0.5 times lower than those for vehicle II, respectively" Not clear what 0.5 times lower means. Does this mean the aromatic emissions were actually higher for the newer vehicle. Also these seem like pretty modest emission reductions between a Euro 1 and a Euro 4 vehicle. I am not familiar with these specific standards but suspect that the expected reduction in NMHC emissions would be much greater than a factor of 2. This may reflect the fact that the experiments were performed at idle. Idle exhaust temperature are relatively low and therefore may not be sufficient to fully activate the catalytic converter. This further complicates interpreting the results (e.g. at idle, Euro 1 and Euro 4 vehicles may have similar emissions).

Reply: Emission factor of aromatics for Euro 4 vehicle was actually 43.5% of that for Euro 1 vehicle. This sentence has been changed in the revised manuscript and listed as follows: "The averaged emission factors of VOCs and aromatic hydrocarbons for Euro 4 vehicle were 2.1 and 0.8 g kg^{-1}, approximately 26.0% and 43.5% of those for Euro 1 vehicle, respectively." The VOCs emission factors based on g km^{-1} for Euro 1 and 4 vehicles were comparable with the previous reported values for Euro 1 and 4 gasoline vehicles in China (Huo et al., 2012; Huang et al., 2015). In this study, emission factors and compositions of VOCs for Euro 1 and 4 vehicles were significantly different.

The following text has been added to the revised manuscript.

"Using 7.87 L/100 km as the average fuel efficiency (Wagner et al., 2009), we obtained the VOCs emission factors based on g km^{-1} for Euro 4 and 1 vehicle to be 0.12 and 0.46 g km^{-1}, respectively, comparable with the previous reported values for Euro 1 and 4 gasoline vehicles in China (Huo et al., 2012; Huang et al., 2015). According to previous studies, there is a clear reduction of VOCs emissions from gasoline vehicles with stricter emission standards (Huo et al., 2012; Huang et al., 2015)."

Q8- "approximately 10 min, indicating dramatic new particle formation. After nucleation occured, the mean diameter increased from 20 to 60nm in about 1.5 h. Because particles with diameters lager than 50nm can act as cloud condensation nuclei (CCN) (McFiggans et al., 2006) and influence the radiative forcing, SOA from vehicle" The nucleation underscores the weakness of not having seed aerosol. The reference to McFiggans and CCN is misleading since in the atmosphere there is substantial existing particle mass which will likely suppress nucleation of gasoline vehicle exhaust.

Reply: This issue was also mentioned by referee 2 (Q2). The starting surface concentrations of particles were all below a critical value (100–2000 μm^2 cm^{-3}) (Wehner et al., 2004), which benefited the formation of new particles. The sentence "*As shown in Fig. 5c, the total particle number concentration increased fast from 82 to 116143 cm^{-3} in approximately 10 min, indicating dramatic new particle formation. After nucleation occurred, the mean diameter increased from 20 to 60 nm). (Section 3.2, Page 10565, Line 11 – 17)*"has been revised and now reads:

"As shown in Fig. 3c, the total particle number concentration increased fast from 82 to 116143 cm^{-3}, indicating dramatic new particle formation, which might be due to that the starting surface concentrations of particles were all below a critical value (100–2000 μm^2 cm^{-3}, Table S1) (Wehner et al., 2004). As shown in Table S1, primary particle numbers in the reactor in this study ranged from 82 to 18948 cm^{-3}, 1-2 orders of magnitude higher than that of a Euro 2 car operated at idling with a similar dilution ratio (Nordin et al., 2013), indicating that the small starting particle number concentrations might mainly due to the idling condition of tested cars rather than the losses in the introduction lines. In addition, upon entering into the chamber, emitted particles would partition due to dilution similar as in the atmosphere, regardless of the temperature and concentration in the sampling system, which might lead to the decrease of starting number concentrations. A certain extent of primary particles under the detection limit of 14 nm of SMPS also contributed to the measured small starting number concentration of particles."

Q9- "aromatics and naphthalene accounted for 51–90% of the measured SOA, comparable to the estimation that classical C6–C9 light aromatics were" Claiming this level of mass closure is very problematic since they do not account for loss of condensable vapors to the walls. This seems more like the maximum amount of SOA they can explain.

Reply: We agree that this will be the maximum amount of SOA can be explained if wall losses of condensable vapors are not accounted for. If we take $C_7H_8O_4$ as an example, a maximum underestimation of SOA production due to wall losses of organic vapors throughout the experiments would be 30%, single-ring aromatics and naphthalene would thus account for 43%-80% of the measured SOA. This will not change the conclusion that aromatics and naphthalene are main SOA precursors in LDGV exhausts. The following text has been added to the revised manuscript.

"Wall losses of organic vapors were not considered in this study, which would lead to the underestimation of SOA production. Therefore, the mass closure analysis estimated the maximum amount of SOA that could be explained by aromatics."

Q10- "The wall-loss rate constant was determined separately for each experiment by fitting the SMPS and AMS data when no new particles were formed." I do not understand this. No new particles formed suggests to me no nucleation. I suspect they mean before there is any photo-oxidation – needs to be clarified in the text.

Reply: SMPS and AMS data were fit with first-order kinetics after UV lamps were turned off. This sentence has been changed to "The wall-loss rate constant was determined separately for each experiment by fitting the SMPS and AMS data with first-order kinetics when UV lamps were turned off." in the revised manuscript.

Q11- "The w = 1 wall-loss correction is not suitable for the experiments here in which nucleation occurred and no seed particles were added" I do not understand why this is the case. The ~1 hr induction period before SOA formation could simply reflect loss of condensable vapors to the walls. I can understand why the w=1 case may not be representative of the vapor losses, but it seems incorrect (based on the results of Zhang) to ignore the wall losses of vapors. Also, wall losses with vapors will mean that you likely underestimate the amount of SOA production and therefore that your mass closure analysis estimates the maximum amount of SOA that can be explained with single ring aromatics.

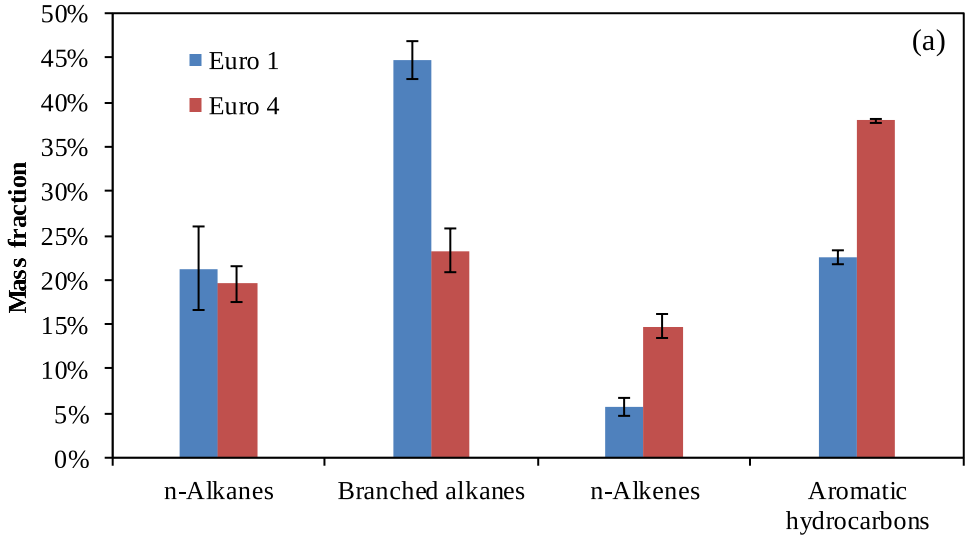
Reply: For the case ω=1, the following equation is used to constrain the vapor wall loss (Hildebrandt et al., 2009).

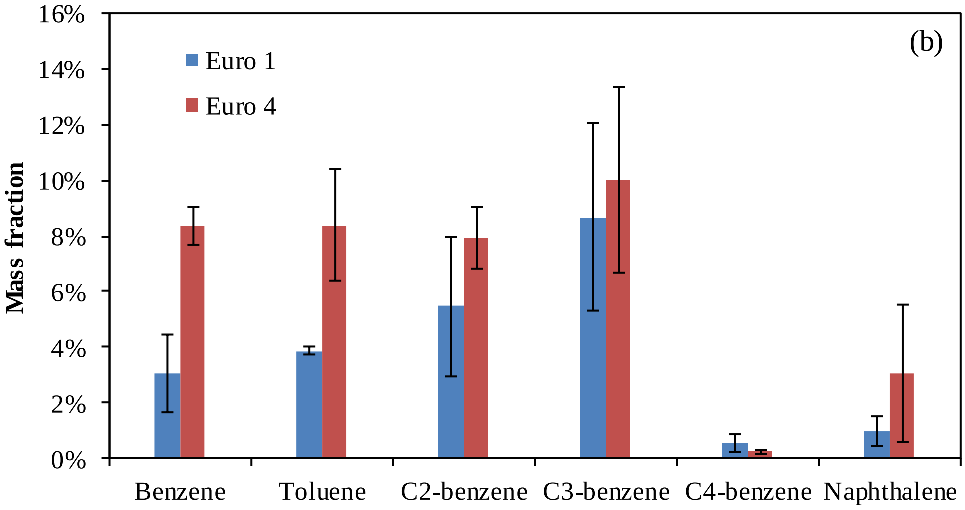
$$C_{OA}(t) = \frac{C_{OA}^{sus}(t)}{C_{seed}^{sus}(t)} C_{seed}^{sus}(t=0)$$

Suspended seed particles are needed to correct the vapor wall loss. Thus the ω= 1 wall-loss correction is not suitable for the experiments in which no seed particles are added. 1 h introduction period would lead to around 7% loss of organic vapor to walls if we took $C_7H_8O_4$ as an example. The subsequently formed SOA, with surface concentrations as high as 2000 $\mu m^2\ cm^{-3}$, provided condense sinks to reduce the vapor wall losses.

Q12- "Table 5. Chemical compositions of the aromatic hydrocarbons in the exhaust of different vehicles, listed as weight percentages." Weight percentages of what? Speciated NMHCs?

Reply: Additional information in Table 5 was added to Figure 4, now Fig. 2 in the revised manuscript, as suggested by referee 2. Chemical compositions were presented as weight percentages of speciated NMHCs, this has been clarified in the caption of Fig. 2.





Q13- Figure 2. Particle number (left) and mass (right) distributions for a typical smog chamber experiment (experiment). The right hand panel suggests that there is substantial mass in the nucleation mode (much more than the 3% suggested in the text). Wall loss of these very small particles is more rapid than the larger particles. How was this corrected for?

Reply: Nucleation mode only includes particles with diameters smaller than 20 nm. Less than 3% of the particle mass is in the nucleation mode ten minutes after nucleation for all the experiments. The SOA formation is sufficiently fast (~ 1 h) and the particle volume distribution shifted not much after SOA reached the maximum value in about 1 h. Therefore, the impact of the nucleation event on wall-loss estimate is considered to be negligible as other nucleation studies did (Cocker et al. 2001; Pathak et al., 2007). In addition, Keywood et al. (2004) concluded that particle wall loss rates could not be accurately quantified for those particles generated in a nucleation event.

Q14- Figure 4 – Mass fraction of what?

Reply: Mass fraction of speciated NMHCs. This has been clarified in the caption.

Q15- Figure 6. They attribute lower yields measured here compared to Nordin et al. to not using seed particles, which leads to higher losses of vapors to the smog chamber walls. That seems like a reasonable explanation. If that is the case then the amount of unexplained SOA is twice what they report. This caveat needs to be included in the text. Also, did Nordin have the same or higher OH exposures. This might also impact the yield.

Reply: The existence of seed particles in the study of Nordin et al. (2013) might reduce the wall loss of semi-volatile organic vapors and thus increase the effective SOA yield (Kroll et al., 2007; Zhang et al., 2014; X. Zhang et al., 2015). However, Cocker et al. (2001) found that SOA formation from m-xylene and 1,3,5-trimethylbenzene photo-oxidation was unaffected by the presence of ammonium sulfate seed aerosols. The influence of seed particles on SOA yields still needs further investigations. Higher OH concentrations in the study of Nordin et al. (2013) would also result in higher SOA yields (Ng et al., 2007). The discussion in the manuscript has been rewritten and now reads:

"The effective SOA yields in the study of Nordin et al. (2013) were 60%-360% higher than those in this study at same concentrations of M_0. In their calculation of the reacted SOA precursors, C4-benzene and naphthalene were excluded. The effective SOA yields would increase 7%-34% when C4-benzene and naphthalene were excluded in this study, which could explain a small portion of the discrepancy. According to the estimation above, the loss of VOCs in the transfer lines was less than 5%. A little higher than VOCs, if assumed to be 20%, losses of IVOCs and SVOCs in the transfer lines would increase the SOA effective yields by a factor of 2%-10% when the unexplained SOA discussed later was all attributed to the contribution from IVOCs and SVOCs. The existence of seed particles in the study of Nordin et al. (2013) might reduce the wall loss of semi-volatile organic vapors and thus increase the effective SOA yield (Kroll et al., 2007; Zhang et al., 2014; X. Zhang et al., 2015). However, Cocker et al. (2001) found that SOA formation from m-xylene and 1,3,5-trimethylbenzene photo-oxidation was unaffected by the presence of ammonium sulfate seed aerosols. The influence of seed particles on SOA yields still needs further investigations. Faster oxidation rates caused by higher OH concentrations in the study of Nordin et al. (2013) would also result in higher SOA yields (Ng et al., 2007)."

Q16- The effective yields are defined based on a relatively small number of single ring aromatics. Did Nordin use the same set of compounds to define their effective yield? If not, is it fair to compare the new results with Nordin et al. results based on yields. The fact that the effective yield was defined relative to this small subset of compounds needs to more clearly stated in paper, including the presentation of the effective yield results and in the caption of Figure 6.

Reply: In the study of Nordin et al. (2013), C4-benzene and naphthalene were excluded when calculating the mass of reacted SOA precursors. The effective SOA yields would increase 7%-34% when C4-benzene and naphthalene were excluded in this study, which could explain a small portion of the discrepancy. The calculation of effective yield has been stated in section “2.4.3 Effective SOA yields” and in the caption of Fig. 4 in the revised manuscript.

Q17- Figure 7 – is this just plotting numbers in Table 3? If so, Table 3 could go into the supplemental.

Reply: Table 3 has been moved to SI.

Q18- Given that they only have two vehicles; I would refer to them in the text by their certification standard, not Vehicle 1 and Vehicle 2. I had to keep looking up which one is which.

Reply: As suggested the tested vehicles were presented as Euro 1 and Euro 4 throughout the revised manuscript.

Q19- “In this study, the SOA yield of benzene and other single-ring aromatics were estimated using the two-product model curves taken from Borrás et al. (2012) and Odum et al. (1997), respectively. While the SOA yield of naphthalene was taken from Shakya et al. (2010).” How representative are these yields of the rest of the literature. E.g. I think that the Odum yields are on the low end of the literature yields for single ring aromatic (in part because they did not use seed aerosols). Do conclusions about the mass closure change if the analysis is done with different yields. Were the yields taken from the literature measured under similar experimental conditions as these experiments (e.g. VOC/NO_x, OA concentration). Given the uncertainty in the yields, the analysis should be repeated with different published yield data to generate error bars in the mass closure estimates (51 to 90%).

Reply: In the study of Odum et al. (1997), around 5000 to 10000 cm^{-3}, 5–10 μm^3 cm^{-3} of ammonium sulfate were introduced as seed aerosols. The adoption of SOA yield curves of toluene and m-xylene from Ng et al. (2007) would increase the mass closure estimation to 55%-114%. But the experimental conditions of Ng et al. (2007) were quite different from this study. In the study of Ng et al. (2007), about 10000 cm^{-3} and 15 $\mu\mathrm{m}^3\ \mathrm{cm}^{-3}$ of ammonium sulfate served as seed aerosols. In addition, nitrous acid was used as OH precursor to provide higher oxidation rates, resulting in higher SOA yields. Therefore, the SOA yield curves of Ng et al. (2007) were not suitable for this study. Considering that the study of Odum et al. (1997) provided a systematic estimation of SOA yields from toluene, C2-benzene, C3-benzene and C4-benzene, we mainly used the two-product curves from Odum et al. (1997) to estimate the SOA production. The following text has been added to the revised manuscript.

"SOA yield curves of toluene and m-xylene from Ng et al. (2007) were also widely used to estimate SOA production (Platt et al., 2013). However, the introduction of seed aerosols and OH precursor made the SOA yield curves in the study of Ng et al. (2007) not suitable for this study. Considering that the study of Odum et al. (1997) provided a systematic estimation of SOA yields from toluene, C2-benzene, C3-benzene and C4-benzene, we mainly used the two-product curves from Odum et al. (1997) to estimate the SOA production."

Q20- Page 10555 line 15 – "form" should be "from"

Reply: Revised as suggested.

Q21- Table 2 – Is this supposed to be ppbv carbon for NMHCs?

Reply: It is just ppbv instead of ppbv carbon.

References

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