Interactive comment on “Gas phase composition and secondary organic aerosol formation from gasoline direct injection vehicles investigated in a batch and flow reactor: effects of prototype gasoline particle filters.” By Simone M. Pieber et al

This paper evaluates the gas phase composition and secondary organic formation from gasoline vehicles in a batch and flow reactor. Excusive results about primary emission factors, gas vapor composition and SOA formation are shown. This paper is well written and organized. The paper describes a large amount of data, but critical evaluation and analysis is missing, that is needed to have confidence on the quantification of results. Therefore I recommend that the paper may be published in ACP after addressing the major revisions below.

Major revisions:

1) The analysis of vapor losses to walls is inadequate. Enormous progress has been made on this area recently, and there is no excuse to ignore those corrections in current studies. Comparability with past studies that were performed when vapor wall losses were not understood is not an excuse to ignore this major issue. Comparability with future studies, for which all the good ones will include analysis of this effect, should be the relevant criterion. It will benefit the citation of this paper for showing both results.

This is an essential correction which could introduce large bias (a factor of 3-4 in Zhang et al. 2014) and is related to multiple key calculation including SOA yield, NMHC composition etc. Taken vapor loss corrections into account, comparison results can be more accurately assessed, especially crucial for some main focuses of this paper: SOA yield between SC and OFR, SOA yield of vehicle exhaust vs single precursors. As the authors stated in page 21 line 27-29 “we expect both SC and OFR yields to be underestimated, by factors of approximately 1.5-2 (SC) and 1.25 (OFR) ((Platt et al., 2017);(Zhang et al., 2014);(Palm et al., 2016)) due to vapor wall losses. Corrections would reduce the discrepancy between the two systems.” Thus a gas vapor wall loss correction is needed for this study. The model of Krechmer et al. (2016) can be used for the SC, and has been recently shown ((Ye et al., 2016); also at AAAR 2017) to be consistent among all Teflon chambers. The model of Palm et al. (2016) should be applied to the OFR.

2) The comparison on SOA formation between OFR and SC should also consider gas vapor loss in the tubing (Pagonis et al., 2017). The different tubing lengths and materials can also result in a SOA yield difference between SC and OFR. A vapor loss calculator can be found in Pagonis et al. (2017). Please clarify the tubing material and length as well.

Page 16 line 17-18: In addition to the residence time of OFR, this SOA delay was also possibly caused by the delayed gas vapor in the tube, as suggested in Pagonis et al. (2017).

3) The particle losses due to heating in the sampling line and the hotter temperature in SC when UV light is on should be addressed. The aerosol loss due to tubing (under no heating condition) needs to be estimated as well. The model for aerosol loss calculation in the tubing can be found in (von der Weiden et al., 2009). The particle loss due to heating can be experimentally determined.

4) Page 10 line 20-30: The SOA photolysis in OFR should be considered as well. For example in OFR with full UV light setting (100%), half of the SOA from toluene SOA (OH chemistry) or naphthalene SOA (OH chemistry) can be photolyzed under 254 UV light with a low quantum yield of 0.1, as shown in Fig. 8b in (Peng et al., 2016). This photolysis effect on SOA formation and SOA yield calculation under 50% and 100% UV light setting should be considered.

5) Page 14 line 1-2. How did the mass quantification between AMS and SMPS compare? It is essential to document this comparison in the form of scatterplots and regressions. Collection efficiency (CE) vary with chemical composition and aerosol phase (Middlebrook et al., 2012). CE for AMS quantification should vary in this study since relative OA and NO3 fraction in total aerosol changed a lot. Why do the authors choose a CE ~1 here. I would expect a slight variation on RIE since there were POA dominated periods during the studies, for which RIE may be higher (Jimenez et al., 2016; Murphy, 2016). Attention needs to be paid for the size cut differences between AMS and SMPS as well, when nucleation was happened.

6) Schematic of the sampling strategy is confusing. I did not get the timing for the sampling strategy. Was the UV light setting is constant during vehicle testing cycle? Then the background of aerosol and gases under UV light is off in SC and OFR was obtained by repeating the testing cycle of the cars? Sufficient detail needs to be provided, that would enable someone else to repeat the experiments, as it is standard in scientific publications.

Other revisions:

Page 5 Line 8: EDC was defined as “older” low-road European Driving cycle (EDC), which is inconsistent with the definition of “New” EDC in the abbreviation/definition list.

Page 7 line 26-27: Please specify the dilution factor for smog chamber (SC) and oxidation flow reactor (OFR).

Page 8 line 14-15: What is the aerosol background level in SC and OFR experiments with clean air when the UV light is on? If it is high, this background needs to be subtracted in the calculation of formed SOA mass concentration.

Page 8 line 26-29: Figure S1 shows the aerosol and gas-phase species are sampled through the same tube in the center of OFR. If it is true, there will be large loss either on VOCs species (if stainless or copper tube is used) or on aerosols (if Teflon tube is used)

Page 9 line 26-27: The particle loss can be determined by measuring the aerosol concentration before and after OFR when the UV-light is off. The aerosol concentration before the OFR can also be roughly determined with aerosols in the SC chamber if the dilution factors and volatility of OA are known in OFR and SC. There have also been some reports from FIREX (Jesse Kroll’s group) that particles containing BC can be charged by the UV lights and be lost much faster, also see (Federer et al., 1983). Was this effect evaluated?

Page 10 line 16: “1000-5000 nm2 cm-3” Is this unit true? These values indicate the particle surface areas in this study are very small, which is inconsistent with mass values reported in Fig. 3. It is ~10$^{\wedge 6}$ times less than those in the typical chamber studies, e.g. (Zhang et al., 2014).

Page 11 line 19-24: Does the OH exposure estimated from BuOH-D9 agree with the OH exposure calculated based on Peng et al. (2015)? A plot showing the comparison of these two methods will beneficial for readers to understand how accurate of the OH exposure used here.

Page 12 line 2: Please specify the dilution factor.

Page 12 line 5-7: Why heat the sample before CPC? Why 300 °C.

Page 14 line 19-24: These descriptions cannot be found in the Fig. 1 e.g.. No graph compares “Ph 1 of cW and hW vs. Ph 2-4 of cW and hW” in line 20.

Page 15 line 2-3: It is hard to draw such a conclusion based on Fig. 2b. A scatter plot between POA+BC vs PM is needed or at least please give the value POA+BC.

Page 15 line 9: Please give the value ranges for “previous finding”.

Page 15 line 16-19: A smaller vapor loss in the OFR is also a possible explanation.

Page 18 line 5: publishing year is required for “Jordan et al”

Page 20 line 1: How to define the “high NO condition”.

Page 20 line 21: No OH exposure is shown in Fig. 6.

Page 21 line 5: Please clarify “Limited experimental statistics”

Page 21 line 30-32: The fragmentation effect on aerosol phase under high OH exposure in OFR should also be considered.

Page 22 line 12: “This generally indicates that we are able to identify the most relevant SOA precursors in the vehicle exhaust.” This statement is not true. The SOA yield in SC and OFR was calculated based on a larger group of ArHC than merely OXYL/TOL. The author should calculated the SOA formation based on OXYL/TOL consumption in the SC (and OFR) and yield from OXYL/TOL experiment. Then compare the calculated SOA to the SOA formed in the SC.

Page 24 line 13-14: To conclude this, it is better to plot a graph showing O:C comparison as a function of OH exposure between SC and OFR. The SC shows similar O:C ratios with OH exposure of $1.2*10^{11}$ molec cm-3 s to the those in OFR under 4.5 molec cm-3 s, which seems not agreeable. The different vapor losses between the SC and OFR might also a reason.

Fig. 2(b) POA point is missing for GDI4-catGPF (CW)

Fig. 5 Better to show the OH exposure range as well.

References:

Federer, B., Burtscher, H., Schimidt-Ott, A., and Siegmann, H. C. (1983). Photoelectric charging and detection of ultrafine particles, Atmospheric Environment (1967). 17, 655-657, https://doi.org/10.1016/0004-6981(83)90140-3

Jimenez, J. L., Canagaratna, M. R., Drewnick, F., Allan, J. D., Alfarra, M. R., Middlebrook, A. M., Slowik, J. G., Zhang, Q., Coe, H., Jayne, J. T., and Worsnop, D. R. (2016). Comment on “The effects of molecular weight and thermal decomposition on the sensitivity of a thermal desorption aerosol mass spectrometer”, Aerosol Sci Tech. 50, i-xv, 10.1080/02786826.2016.1205728

Krechmer, J. E., Pagonis, D., Ziemann, P. J., and Jimenez, J. L. (2016). Quantification of Gas-Wall Partitioning in Teflon Environmental Chambers Using Rapid Bursts of Low-Volatility Oxidized Species Generated in Situ, Environ Sci Technol. 50, 5757-5765, 10.1021/acs.est.6b00606

Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R. (2012). Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, Aerosol Sci Tech. 46, 258-271, 10.1080/02786826.2011.620041

Murphy, D. M. (2016). The effects of molecular weight and thermal decomposition on the sensitivity of a thermal desorption aerosol mass spectrometer, Aerosol Sci Tech. 50, 118-125, 10.1080/02786826.2015.1136403

Pagonis, D., Krechmer, J. E., de Gouw, J., Jimenez, J. L., and Ziemann, P. J. (2017). Effects of gas–wall partitioning in Teflon tubing and instrumentation on time-resolved measurements of gas-phase organic compounds, Atmos. Meas. Tech. 10, 4687-4696, 10.5194/amt-10-4687-2017

Palm, B. B., Campuzano-Jost, P., Ortega, A. M., Day, D. A., Kaser, L., Jud, W., Karl, T., Hansel, A., Hunter, J. F., Cross, E. S., Kroll, J. H., Peng, Z., Brune, W. H., and Jimenez, J. L. (2016). In situ secondary organic aerosol formation from ambient pine forest air using an oxidation flow reactor, Atmos. Chem. Phys. 16, 2943-2970, 10.5194/acp-16-2943-2016

Peng, Z., Day, D. A., Stark, H., Li, R., Lee-Taylor, J., Palm, B. B., Brune, W. H., and Jimenez, J. L. (2015). HOx radical chemistry in oxidation flow reactors with low-pressure mercury lamps systematically examined by modeling, Atmos. Meas. Tech. 8, 4863-4890, 10.5194/amt-8-4863-2015

Peng, Z., Day, D. A., Ortega, A. M., Palm, B. B., Hu, W., Stark, H., Li, R., Tsigaridis, K., Brune, W. H., and Jimenez, J. L. (2016). Non-OH chemistry in oxidation flow reactors for the study of atmospheric chemistry systematically examined by modeling, Atmos. Chem. Phys. 16, 4283-4305, 10.5194/acp-16-4283-2016

von der Weiden, S. L., Drewnick, F., and Borrmann, S. (2009). Particle Loss Calculator – a new software tool for the assessment of the performance of aerosol inlet systems, Atmos. Meas. Tech. 2, 479-494, 10.5194/amt-2-479-2009

Ye, P., Ding, X., Hakala, J., Hofbauer, V., Robinson, E. S., and Donahue, N. M. (2016). Vapor wall loss of semi-volatile organic compounds in a Teflon chamber, Aerosol Sci Tech. 50, 822-834, 10.1080/02786826.2016.1195905

Zhang, X., Cappa, C. D., Jathar, S. H., McVay, R. C., Ensberg, J. J., Kleeman, M. J., and Seinfeld, J. H. (2014). Influence of vapor wall loss in laboratory chambers on yields of secondary organic aerosol, Proceedings of the National Academy of Sciences, 10.1073/pnas.1404727111