Referee Comments #2 (acp-2017-942-RC2-supplement) and author response.
Simone M. Pieber et al.
We thank the editor and referees for their comments. To guide the review process we have copied the referee comments in black text. Our responses are in regular blue font. We have responded to all the referee comments and made alterations to our paper (*in italic text*) and removed redundancies for clarification. Along with the revision we suggest a slightly changed title: "*Gas phase composition and secondary organic aerosol formation from gasoline direct injection vehicles with prototype particle filters investigated in a batch and flow reactor*"

General:

RC2-1: This paper describes measurements of primary emissions and secondary organic aerosol from four modern gasoline direct injection (GDI) engine equipped vehicles. The market share of GDI vehicles is rapidly increasing in the US and other countries, displacing the more traditional port fuel injection engine equipped vehicles. This paper represents probably most systematic study of SOA formation from GDI vehicles to date. The paper also investigates the impact of adding a retrofitted gasoline particulate filter (GPF) to two of the vehicles. While others have investigated the effect of this technology on primary emissions, I am not aware of previous studies that have investigated its impact on SOA formation. The paper also uses an oxidation flow reactor (OFR) and a smog chamber to investigate SOA formation, comparing the results between the two systems and with measurements made with individual compounds. The paper shows SOA formation dominates primary PM emissions (consistent with previous studies, though the SOA/POA ratio seem much larger than previous studies). The paper demonstrates that the majority of SOA formation is formed from cold-start emissions, which is not surprising but I have not seen it demonstrated before. The paper shows that somewhat more than half of the SOA formation appears due to 8 single-ring aromatics. Finally the paper shows GPF reduces primary PM emissions but does not reduce the SOA formation (or non-methane organic compound emissions). Overall the paper is well written and very comprehensive. The experiments appear to have been carefully conducted (though I agree with the other reviewer's concerns on treatment of wall loss), with results from repeated experiments shown (it would be nice to describe the precision a bit more). I think this paper makes a nice contribution and recommend that it be published in ACP after addressing the following comments.

Author Response: We thank the referee for the positive feedback and address the specific comments in the following. We provide answers to RC2 and modifications to our manuscript to the best of our abilities and have condensed and clarified the text where possible.

Specific comments

RC2-2: Abstract and a few other places "a large fraction (>0.5)" These statements refer to the mass closure of the SOA production based on measured precursors. 1 is greater than 0.5. The authors need to be more quantitative; e.g. give a range (or some other metric such as median and interquartile range).

Author Response: We agree with the referee's comment and have reformulated the statement in the abstract. As the assessment of the contribution is complex and condition-dependent, we prefer to remove the quantitative information in the abstract and rather keep the discussion of this issue in the results and discussion section 3.6 where we can address the details of this closure.

Text modifications: Abstract: "*A significant fraction of the SOA production was explained by those compounds, based on investigation of reacted NMOC mass and comparison of effective SOA yield curves with those of toluene, o-xylene and 1,2,4-trimethylbenzene determined in our OFR within this study and others from literature. Remaining discrepancies may result from diverse reasons including apart from unaccounted precursors also aging conditions, uncertainties of SOA yields for the aromatic hydrocarbons with different degrees of substitution, as well as experimental uncertainties in the assessment of particle and vapor wall losses.* "

RC2-3: Figure 6 suggests that this ratio likely varies with OA concentrations. Figure 5 suggests poor closure for 100% and 70% OFR conditions, but good closure for 50% OFR. SC has better closure at short timescales. The authors need to be more quantitative about the mass closure.

Author Response: We agree with the referee's observation. We have used two different ways to approach the closure in our manuscript: 1) with a forward mass closure as presented in initial Figure 5 as an example for 1 experiment (i.e. the reacted aromatic SOA precursor mass was weighted by a fixed SOA yield from the literature), and 2) via an indirect approach normalizing the formed SOA to the reacted ArHC mass (i.e. deriving an "effective yield" combining all reacted species), which allows to present the result as a function of OA loading, takes partitioning into account and is interpreted by comparing to the yields of classes of compounds (such as ArHC). As also Referee 1 noted that this is confusing, we have decided to remove the SOA-mass closure from initial Figure 5 (bottom panels), and instead present all our data only as a "yield closure" (method 2, Figure 6).

Text modifications: We have revised all text sections to allow this modification; instead of a mass closure we discuss the results now in terms of the yield-closure presented in section 3.6.

RC2-4: Retrofitted GPF – How was this done? How representative is it of how a true OEM designed and installed GPF-system would operate? It is hard to simply add a control system to a vehicle for which it was not designed (I have seen tests with a retrofit GPF hanging off the back of a car!), therefore I am always concerned about how representative the performance of researcher retrofitted system versus what might be done by a vehicle manufacturer. This is not to say that they are not seeing some effect of the GPF but it may be (much) less than the performance of bottom up engineered system. For example, I was surprised that the catalyzed GPF did not further reduce the NMOC emissions – what was the operating temperature of the GPF? Anyways I think the GPF results are interesting, but a bit more detail on how the retrofit was done, specifically limitations of researcher retrofitted systems should be acknowledged. Unless they can document that the retrofitted is representative of OEM designed and installed systems the conclusion section (_line 10 on page 25) is too strong.

Author Response: We agree that validity of the experiments to transfer our results to bottom up engineered systems is a crucial point. We have highlighted that our GPF system was a "retrofit" in our discussion paper, and have described the system in section 2.1.1 (page 6, line 5 onwards). It is indeed difficult to judge how such systems will be implemented by manufacturers and there will be also variability between different vehicles. For our experiments, which we designed with experts from the industry, we replaced the "muffler" which was located ca. 60 cm downstream of the three-way-catalyst (TWC) with the GPF. Pictures are provided in Figure R2-1 which has been added to the SI of our revised manuscript. The GPF was not externally heated, and its temperature dependent on the exhaust conditions, in the same way the temperature of the TWC is dependent on this.

We believe our retrofit is installed in a location that is representative for real-world retrofitting of GDI vehicles which are on the market currently, and that our experiments represent also the typical temperature conditions that can be found in such retrofits. An evaluation of this system on primary particle number emissions was published previously by a sub-set of our collaboration team (Czerwinski et al., 2017). The PCFEs of this investigation yielded ≥98% for GPF1 on either GDI1 or GDI4, and ≥86% for catGPF on GDI4, and indicate somewhat lower performance of the catGPF on primary PN reductions. This does not seem to be dependent on the location of installation in a vehicle, given that GPF1 on GDI4 showed good performance.

We don't believe that the generally somewhat lower performance of catGPF on primary PN is caused the limited effect on NMOCs and associated SOA. Instead, we believe that given that the vehicle is already equipped with a TWC, it is unlikely to see additional effects on NMOC reductions by a 2nd catalytic system during cold-starts. This is, because under cold-start conditions the NMOCs that pass the TWC will also pass the catGPF, which, at this point will likewise not have reached light-off temperatures to efficiently remove NMOCs.

We agree, that differently engineered systems (especially catGPFs that will be meant to replace the TWC), should be investigated in the future to see whether further NMOC reduction can be achieved.

Figure R2-1: *"**Pictures** of **a**) original "muffler" and GPF in comparison, **b**) retrofitted GPF, installed underfloor in replacement to "muffler"."*

Text modifications: We have modified above mentioned text sections to specify the distance between TWC and GPF (replacing muffler), which was roughly 60 cm. Along with modifications within the manuscript:

- *"GDI1 was studied i) in standard configuration, and ii) equipped with a prototype gasoline particle filter (GPF (cordierite, porosity 50%, pore size 19 μm, 2000 cells per square inch)), installed at the muffler ("underfloor"), which was located 60 cm downstream of the TWC. It's filtration quality at this configuration is equivalent to the best available technology for DPFs (personal communication by the manufacturer; particle number reductions by the application of the GPF are further assessed in Czerwinski et al., 2017, and yield PCFE ≥98%)."*
- *"GDI4 was retrofitted with i) the previously tested GPF (as above: cordierite, porosity 50%, and pore size 19 μm, 2000 cells per square inch, PCFE ≥98%), as well as ii) a Pd/Rh catalytically coated GPF (catGPF) (installed at the muffler, underfloor, while keeping the original TWC in the original position; the PCFE was ≥86%)."*

For the catGPF, additionally we had already the following statement provided in the initial version of the experimental section, which we kept in its original format:

- *"For the retrofitted catGPF, the primary purpose of the catalytically active coating is the constant self-cleaning of deposited carbonaceous material on the particle filter (personal communication with manufacturer). In future applications, such catalytic coating on a GPF might replace the existing TWC in GDI vehicles, or specifically, the TWC can be replaced with a GPF carrying the TWC coating."*

Along with this, however, we have modified the conclusions section to state more explicit, that GPFs carrying the TWC coating meant to replace the initial TWC will possibly lead to better NMOC removals than the current TWCs, and that research addressing this should be conducted:

- *"GPF application efficiently removes eBC, which is the dominant component of primary PM, and also shows small effects on the minor POA fraction. The volatile POA fraction passes through the filter in the vapor phase and later condenses when the exhaust is emitted and cooled; hence POA emission factors are not as significantly reduced as refractory PM. NMOC emissions and SOA formation are unaffected by the tested GPFs. This is particularly true when the GPF is catalytically inactive, and at cold-started driving cycles for catalytically active GPFs (i.e. when emissions pass through the TWC and the catGPF before light-off temperatures are reached.. This means that retrofitting GDI vehicles with GPFs will likely result in an important reduction of the total primary PM emitted (removal of refractory material), but will (under conditions similar to our experiments only to a small extent reduce NMHC (or NMOC) emissions including ArHC, and thereby not directly lead to a reduction of SOA. Future work on so-called "4-way catalysts", i.e. a TWC catalyst directly applied onto a GPF and installed at the location of the current TWC for simultaneous filtration of particulates and catalytic conversion of NMHC (or NMOC) should be conducted, to understand whether reductions of SOA precursors, SOA production, and semi-volatile primary PM can be achieved with further optimized systems."*

RC2-5: The dramatically higher NMOC emissions and SOA production from cold start is important. Can the authors quantify how much more important it is than hot start (e.g. using an analysis similar to that Saliba et al. EST 2017 10.1021/acs.est.6b06509 to compare hot and cold start emissions).

Author Response: We have provided a comparison of cold-started and hot-started cycles in Figure 2 (discussion paper), but had not given a comparison of the values previously, this is a highly interesting point. We now provide ratios of Ph1-SOA emission factors over Ph2-4-SOA emission factors in the manuscript, which is similar to Saliba et al., 2017-approach, and indicates that the cold-start is 20-50 times more important. However, it needs to be noted that this information should only carefully be transferred to the ambient air, additional parameters (e.g. how long/far will a vehicle be driven for during a real journey and what's the ambient temperature, i.e. "how cold" is the vehicle) need to be considered (see also Platt et al., 2017). In addition to our own ratio, we also provide a reference to a recent publication by Zhao et al., 2018, who also compare SOA from cold- and hot engine conditions.

Text modifications: *"Data are presented as a function of suspended OA for all experimental conditions of cold-started GDI1-3 (i.e. for full cW, cE; and Ph 1 (cW)), while GDI4 or hot engine conditions, i.e. Ph 2-4 (cW) are not included in our the analysis, as this data set includes only two experiments with concentrations levels close to our background measurements; a discussion of SOA yields from cold- and hot-engine emissions has recently been published by Zhao et al., 2018 for an OFR data set)."*

We have also added the following and updated the conclusions.

- *"Hot-engine emissions (Ph 2-4 sampling from cold-started WLTC, as presented in Figure 2d) also resulted in SOA formation, which was, however, 20-50 times lower in terms of EFs than SOA formed from Ph 1 sampling of a cold-started WLTC. This is in line with the trends indicated by the phase-dependent NMHC emissions (Figure 2c).*

- *"Future work should investigate the quantitative use of online OFR data in further detail for additional quantification of cold- and hot-start contribution of SOA to the total SOA burden; a discussion of the associated technical issues (i.e. changes in OH-exposure and condensational sink as well as the equilibration time inside the OFR reactor) has been recently published by Zhao et al., 2018."*

RC2-6: The conclusion section largely repeats conclusion from earlier in the paper. The paper would be improved if they put the results in context with the growing literature in this area. In particular I was interested if the results are consistent with the existing body of knowledge on SOA formation for PFI vehicle exhaust. My sense is that it is. You tested the vehicles using two different cycles? Were there any cycle dependencies or was cold start just dominant?

Author Response: We have modified and shortened the manuscript and provided additional comparison with literature. Figure 2a/b includes indeed data from WLTC and EDC cycle, comparing the cold/hot cycles. We don't observe any significant cycle-dependent differences which are larger than vehicle-by-vehicle or test-by-test variability, especially during cold-started cycles. As the EDC cycle was tested only as the full cycle and not split into separate phases, no explicit analysis of the SOA contribution to the total cycle can be made in comparison to the WLTC. This information has already been provided in the discussion paper and is now also stated more clearly in the conclusions section. Further points are discussed along the referee comments below specifically to Figure 2.

Text modifications:

- *"While no drastic cycle-dependencies (WLTC vs. EDC) were observable from our tests (especially during cold-started cycles), EFs of primary NMHC and THC were reduced by a factor of 90 under hot-started conditions."*

- *"Emissions of all cold-started vehicles, technologies and driving tests showed significant SOA formation upon photochemical oxidation (Figure 2b), in line with other studies on GDI as well as PFI systems (Platt et al., 2017; Gordon et al., 2014; Saliba et al., 2017; Zhao et al., 2018).*

- *"Overall, the SOA potential (in terms of an emission factor) of the tested vehicles agreed with recent literature reports from both, GDI and port fuel injection systems (PFI)."*

RC2-7: Figure 2 – The y-axes are five orders of magnitude. This illustrates large changes, but changes of a factor of 2 or 3 can also be interesting. For most tests it does appear that the GPF is reducing the POA emissions, but not as dramatically as the EC. I did not get that impression reading the text but it does appear in the figure. More attention needs to be paid to these trends.

Author Response: We agree that Figure 2 is packed with details, and we have revised it (see below). Further, we refer to our related publication (Munoz et al., 2018), which discusses the difference between cold- and hot-started cycle emissions in detail for GDIs in standard configuration, regarding CO, NOx, particle number and genotoxic PAHs. We agree on the observation of POA removal with the GPF and have adjusted our statements:

Text modifications: *"Retrofitted GPFs (including catGPF behind the standard TWC) appeared also to reduce the POA fraction.",* is added in the results and have also updated the abstract ("*GPF retrofitting was found to greatly decrease primary particulate matter (PM) through removal of eBC, showed partial removal of the minor POA fraction, ...*") and conclusions ("*GPF application efficiently removes eBC, which is the dominant component of primary PM, and also shows small effects on the minor POA fraction.*").
We also added the following: *"A detailed discussion on emissions of CO, NOx, particle number and genotoxic PAHs from cold- vs. hot-started cycle driven GDI vehicles in standard configuration can be found in our related publication by Munoz et al., 2018.*

RC2-8: Figure 2 – The SOA production seems surprisingly high. For GDI1 the total NMHC (most of which are not SOA precursors) is around 1000 mg/kg (Figure 2a). The SOA production is between 200 and 600 mg/kg (Figure 2a) – the SOA production from the GPF equipped experiments with GDI1 seem incredibly high. This suggests an effective of SOA of 20-60% of the total NMHC emissions of which less than half is aromatics (Figure 4b is misleading because the NMOC measured by PTR is only 65% of NMHC measured by FID). The SOA production seems higher than previous studies of modern vehicles (they are more similar to 25 year old vehicles). I guess Figure 6 suggest SOA yields are "reasonable", but I was confused looking at Figure 2 (maybe it is just the log scale with 5 orders of magnitude). Are there background issues?

Author Response: We agree with the very high SOA production of especially GDI1-3, but specifically GDI1. For the corresponding SOA emission factors in relation to previous publications, there are two additional things to consider. 1) the OA mass at which these SOA-emission factors are determined, and 2) that most previous literature is using exclusively data from SC and not OFRs (data points at the upper end of the SOA emission factors in our experiments are derived from OFR experiments). Additionally, previous SC experiments which the referee refers to were conducted typically at lower OH exposures than our experiments, and at different ratio of NO/NO$_2$ or total NO$_x$/VOC, points which are discussed later in the manuscript in the section "SOA yield analysis" and in Zhao et al. 2017, which we have added to our reference list
   Regarding background issues, we have provided additional information in our answer to RC1 as well as within other answers herein. Experiments were conducted with high purity air after extensive cleaning (described in the main text). Background was insignificant compared to our vehicle testing data, except for Ph2-4 experiments which were close to background levels in terms of the NMOCs (noted in Figure caption to Figure 4).

Text modifications: Figure 2 was modified for clarity (see below).

RC2-9: Figure 2c – There is a lot of vehicle to vehicle variability (2+ orders of magnitude). Are the reductions between cold and hot start consistent across vehicles? Plotting ratios may be more informative. What is up with the experiments with an NMHC emission rate of 0.1 mg/kg? Are those valid data?

Author Response: Vehicle by vehicle differences between cold- and hot-started cycles can be seen from Figure 2a and we have already provided ratios in our discussion in section "3.1 Pollutants as function of vehicle technology and driving cycle"; We provide median and interquartile ranges for data presented in Figure 2c now; 0.1 mg/kg is the detection limit of our NMHC measurements, and data were below this limit in some cases.

Text modifications: Figure 2 was modified for clarity (see below).

RC2-10: Figure 2 is very busy (especially panel d). It is basically impossible to sort out the trends. Pick the key points you want to make and plot just that data. The SOA production appears surprisingly close to the NMHC emissions (it even exceeds it for some vehicles).

Author Response: Indeed, (OFR-from-SC)-SOA is very close to the FID-based NMHC emission factors determined in the SC. SOA emission factors never exceed the emission factors of aromatic hydrocarbons (which is the relevant information for SOA in our case). Hence, the information provided is fully consistent. We have revised the figure for clarity.

Text modifications: The figure was revised, see below, and have clarified section 3.1.
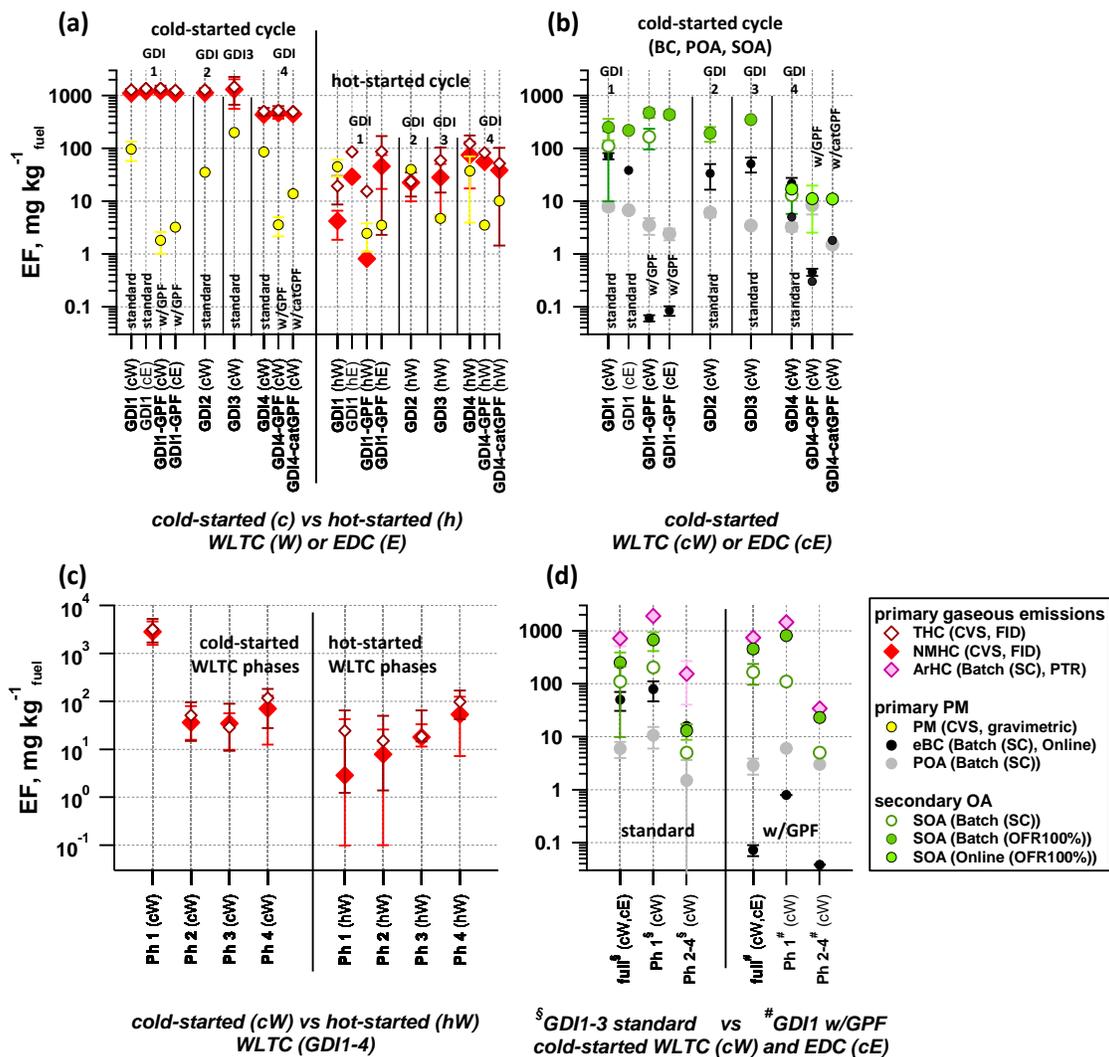
Figure RC2-1: new version of Figure 2; new figure caption reads as follows:
"Emission factors (EF) of pollutants from cold-started ("c") and hot-started ("h") test cycles (WLTC ("W") and EDC ("E")). Individual cW and hW phases are indicated as "Ph" 1-4. (a) Total and non-methane hydrocarbons (THC, NMHC) and primary gravimetric particulate matter (PM) from CVS measurements over entire test cycles for different vehicle configuration and test conditions (average±1SD), (b) primary PM (equivalent black carbon (eBC) and primary organic aerosol (POA)), and secondary organic aerosol (SOA) from SC and OFR-from-SC experiments, and from online OFR operationat 100% UV per vehicle configuration for cold-started test cycles (average±1SD), (c) THC/NMHC of cW and hW experiments from (a) separated into individual cycle phases (median, and P25-P75 range are shown). (d) POA, eBC, aromatic hydrocarbons (ArHC) and SOA over the full cW and cE, compared to individual phases of cW from SC batch experiments and OFR-from-SC (average±1SD). (a-d) EF calculation is detailed in the SI. The time-resolved SOA

profile from online OFR measurements conducted on GDI4 in 2015 (standard and catGPF) is provided in Figure S14."

RC2-11: Additional ArHC (page 23). The analysis in Zhao et al. (EST 50(8): 4554-4563 2016) suggests some of the IVOCs are alkylated single ring aromatics larger than those included in the analysis here. How does including IVOC component measured by Zhao et al. change the analysis? His analysis suggests that IVOCs contribute somewhat less than half of the SOA in gasoline vehicle exhaust.

Author Response: We agree with the referee that those compounds could give additional SOA mass in our experiments. However, due to the different analytical techniques applied (we use a PTR-ToF-MS vs. the TD-GC-MS technique by Zhao et al., 2016), the data cannot be simply combined, as at the current moment, we are uncertain of which fraction of the IVOC (which makes up ca. 50% of the SOA in Zhao et al., 2016) overlaps with a fraction accounted for in our experiments, as we might be able to see fragments of e.g. alkyl-substituted aromatics that would fall into the IVOC category in Zhao et al., 2016 in our understanding. Ignoring a potential double count here, would likely allow us to conclude that those compounds make up a big fraction of the 50% of the missing mass seen by the FID but not PTR-ToF-MS, and would also bring our yield analysis in closer agreement with the yields determined for the vehicle exhaust (taking these additional compounds into account) would agree with that of single aromatic compounds.

Text modifications: We have added small adjustments to make this point more explicit in our manuscript.

RC2-12: Page 3 "diesel PM emissions have been greatly reduced." – This is true for new diesel particulate filter (DPF) equipped vehicles but there are a lot of old diesels on the road, especially in Europe so human exposure to diesel particles has probably not yet been greatly reduced. Eventually it will be when the fleet is completely turned over. May want to refine this statement.

Author Response: We agree with the comment and have refined our statement to specify we are referring to test bench measurements and emission factors of the recent vehicle fleet, which isn't fully compliant with the fleet on the road.

Text modifications: *"Due to the regulatory attention and the improved after-treatment systems, diesel PM emissions from new generation vehicles have been greatly reduced, and fleet modernization will help to reduce their burden in the ambient air."*

RC2-13: Page 4 "modern diesel vehicles" Modern is too generic. You should be more precise catalyzed-DPF equipped diesel vehicles. I don't necessarily think modern = DPF.

Author Response: We agree with the comment and have specified our terms.

Text modifications: *"modern"* was replaced with *"catalyzed-DPF equipped"*

RC2-14: Page 8 "experiment. Control experiments were conducted regularly in the SC to estimate the contribution of the SC background to SOA formation." Please provide another sentence or two here that describes results from control experiments. How much SOA was formed in controls and how does it compare to what is measured in an experiment with vehicle exhaust. Did you run control experiments with the OFR – what were the background levels in that system?

Author Response: Thanks for the comment. In brief: control experiments were SOA experiments conducted with the SC and OFR after the standard cleaning procedures, and in both cases, SOA formed during control experiments was insignificant compared to SOA formed during vehicle testing, except for Ph2-4 which are close to background levels in the SC as stated in cation to Figure 4, and and eventually GDI4 experiments which formed less SOA. SOA-control experiments with ammonium sulfate as seed in the SC yielded a SOA background < 1 µg m^{-3} after 2 hours of aging (i.e. comparable to the typical vehicle SC SOA experiments). Control experiments with UV on where also conducted in the OFR. When sampling test bench room air through our 1:8 dilution system (prior to online tests during cWLTC and hWLTC), background levels where <10 µg m^{-3} (which is far below the online vehicle SOA measurements of 100-2000 µg m^{-3}). When sampling pure air from the cleaned SC before OFR-from-SC experiments, background levels where < 2 µg m^{-3}.

Text modifications: We have added the following:

- *"Background measurements of the clean chamber were conducted prior to each experiment, and was insignificant compared to our measurements except for Ph2-4 or GDI4 experiments as stated in the results. Photochemistry control experiments were conducted regularly to estimate the contribution of the SC background to SOA formation; these experiments were conducted after the standard cleaning procedure. Instead of vehicle exhaust, pure air was used as a sample and ammonium sulfate (50 ug m^{-3}) injected as seed. Other experimental procedures were in line with the typical vehicle experiments. We found a SOA background of < 1 µg m^{-3}, which is below the SOA concentrations formed during vehicle exhaust aging Concentration-levels of our SC experiments were representative for urban ambient conditions, as reported in Table S4-S7."*
- *"Background levels were <2 µg m^{-3} SOA before OFR-from-SC experiments (when sampling from cleaned SC) and <10 µg m^{-3} when sampling diluted (1:8) test bench room air prior online-experiments."*

RC2-15: Section 2.2.6 – I am pretty sure that you are calculated yields using the reacted aromatic mass in the denominator however this statement is confusing "the ratio of the SOA mass to the reacted SOA-forming mass, delta_NMOCreacted" My understanding is that delta_NMOCreacted is not the same as the reacted aromatic mass. This needs to be cleaned up to avoid confusion. May also want to state this in the caption for Figure 6 to reminder reader of how yields are calculated.
Author Response: We calculated yields by normalizing the formed SOA mass to the reacted delta of the 8 selected aromatic hydrocarbons which dominated the identified NMOC fraction. We have revised this statement and clarified this also in the discussion to Figure 6.
Text modifications: According above description, the new text reads: *"SOA yields analysis is based on SC and OFR-from-SC experiments with GDI1-3. An effective SOA yield (Ye), was calculated as the ratio of the SOA mass to the reacted SOA-forming species i (in Δµg m^{-3}, Eq. (2)). We take into account all our identified SOA precursors (which refers to the 8 dominant aromatic hydrocarbons presented in Figure 4d), neglecting non-reactive and non-SOA forming precursors and assuming that all relevant SOA precursors are measured.*

$$Ye = \frac{\Delta SOA}{\sum_i \Delta SOA\_precursor_{i,reacted}} \qquad (2)"$$

RC2-16: Using "NMOC" to describe the sum of the PTR measurements is confusing as it is measuring less than 2/3rds of the organic gas emissions as measured with FID. This limitation needs to be stated more clearly (it is in the intro but the reader will likely forget – e.g. adding to caption of Figure 4 would be good and in other places in the main text when you discuss NMOC.
Author Response: We agree and have added the information throughout the manuscript and in the caption of Figure 4; further we have shortened section 3.4 and moved detailed discussion of O_2^+ charge processes and fragmentation of alkyl-substituted aromatics to the SI to make this section more concise on the point of SOA-precursor identification.
Text modifications: As described in our response. The revised figure caption reads as follows:
"(b) Relative composition of the PTR-ToF-MS derived NMOC fraction (which makes up 65%±15 of the FID-based NMHC signal on a carbon-basis for cW/cE/Ph 1(cW)), (c) total ArHC EFs (which make up 49±8% of the FID-based NMHC signal on a carbon-basis for cW/cE/Ph 1(cW), and (d) relative contribution of the 8 dominant ArHC (correspond to 96.7±3.3% of the total ArHC signal for cW/cE/Ph 1(cW)).".

RC2-17:
Emissions data from tests in mg/kg-fuel needs to be provided in tables in supplemental.
Author Response: Emissions data presented in Figure 2 (in mg/kg fuel) can be made available to others upon request. Median values are stated in section 3.1.
Text modifications: No modifications were made to the text.

References to Author's Response:

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