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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Gasoline direct injection vehicles exceed port fuel injection ones in both primary aerosol emission and secondary aerosol formation" *by* Zhuofei Du et al.

Anonymous Referee #2

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Du et al. report on emissions and chamber experiments performed on port fuel injection (PFI) and gasoline direct injection (GDI) gasoline vehicles and their potential to form secondary organic aerosol (SOA). They find that the PFI vehicle emits more VOCs compared to the GDI vehicle but the GDI vehicle emits both primary particles and forms more SOA than the PFI vehicle. They claim that the higher SOA from GDI vehicles could be attributed to higher emissions of intermediate volatility organic compounds (IVOCs).

The study builds on previous work done with PFI and GDI vehicles to answer an important question currently on the minds of researchers and regulators: are GDI vehicles

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a cause for more primary and secondary pollution in the future? Thus, the work is well-motivated and very topical. The manuscript could benefit from a copyedit from someone with fluency in English since there are some styling and phrasing issues. The methods are appropriate and the experimental results are worth publishing although there are some minor issues that need to be resolved (see comments below). My major concern is the study design that only used one vehicle of each technology type and that the conclusions are generalized for all vehicles in that technology type. I recommend publication after the authors have had an opportunity to respond to my comments.

Major comment: Vehicle-to-vehicle variability - Vehicles certified to the same emissions standard can vary significantly in their tailpipe emissions and their potential to form SOA (order of magnitude or more). For example, see any of the large studies done over the past decade and a half (Kishan et al., 2008; May et al., 2014). In the United States where successive emissions standards for any given pollutant do not change by an order of magnitude, it follows then that it is likely that a randomly picked vehicle certified to a newer standard emits more pollutants than a randomly picked vehicle certified to an older standard. In the context of this work then, one needs to be careful in comparing absolute emissions/production from one PFI vehicle against another GDI vehicle and using those comparisons to make broader conclusions about PFI versus GDI vehicles. For example, it is imprecise to imply that all GDI vehicles had higher SOA production factors than PFI vehicles (as mentioned in the title) or that the GDI versus PFI SOA difference could be attributed to higher S/IVOC emissions (as mentioned in the discussion section). If this study would have included many more vehicles or somehow performed on the same vehicle but with interchangeable injection methods, this comment would not apply but since only one vehicle was chosen for each technology type this becomes a concern. Can the authors comment on why the vehicles they picked are representative of their technology type and why the differences from those vehicles (e.g., Table 3 and 4) can be extrapolated to a whole class of vehicles?

Other comments: 1. Line 34-44: There is a lot more modeling and measurement work done around understanding the motor vehicle contribution to OA in urban areas. Perhaps work on a better literature review to motivate the work?

2. Line 40: Does Huang et al. (2014) only refer to Chinese cities?

3. Line 42: 'exhaust' not 'exhausts'.

4. Line 47-49: In comparison to what type of vehicle? My understanding is that one of the major advantages of the GDI is that it eliminates pumping losses.

5. Line 51-52: Are these percentages for sales of new vehicles only?

6. Line 67: Gentner et al. (2017) is a review paper. Please state the primary study.

7. Line 72: 'formation' not 'formations'.

8. Line 89: How many experiments were performed? Figure 4 suggests two for the GDI and four for the PFI. Mention this in the methods section.

9. Line 97-99: How was the setup designed to reduce losses of gases and particles from the CVS to the chamber? Were the transfer lines heated? Were they coated to reduce losses of vapors and particles? How long were the transfer lines? Were the gas and particle losses in the transfer line characterized before performing the vehicle experiments?

10. Line 110-111: How were the concentration data corrected for additional dilution?

11. Does a DMS500 thermally denude the particles before measurement? This should be mentioned. Also, what is the denuding temperature?

12. Line 147-148: Are the BJC versus NEDC differences shown in this work?

13. Line 150: What was the benzene to toluene ratio in the fuel and does this align with the emissions measurements?

14. Line 166: What was the EC+POA mass compared to? Teflon filters? Were artifacts

on the Quartz filters considered in the comparison? What OM to OC ratio was used to get POA mass?

15. Line 172: POA was not higher in Saliba et al. (2017) only EC was.

16. Line 181-184: The tailpipe aerosol size distribution can vary substantially with atmospheric processes (e.g., coagulation, evaporation) on very short timescales so care needs to be exercised in comparing source measurements with ambient measurements for particle size. Also, is it possible that the bimodal ambient measurements are influenced by nucleation rather than exhibiting a vehicle signature?

17. Line 212-214: It would be better to see a description of the correction for dilution, particle, and vapor wall-losses in the methods section.

18. Line 226-229: Zhao et al. (2017) argued that some of the differences in SOA formation between the vehicles they tested could be explained by differences in the NOx levels. Is it possible that differences could be explained by different NOx levels?

19. Line 239: Explain what underestimated here means?

20. Line 252: Yang et al. (2017) recently suggested that ozonolysis of alkenes in gasoline exhaust could form SOA through aldol condensation reactions. Perhaps cite that study here?

21. Line 255: Units for VOC/NOx ratio? How does this ratio compare to those in megacities?

22. Line 258: Figure 7 shows SOA measurements that are not corrected for vapor wall losses but presumably the SOA mass yields for the aromatic precursors include the effects of vapor wall losses. What is the implication of this? Discuss here.

23. Line 264-271: Did the authors consider the detailed speciation data of Zhao et al. (2016) to model the SOA formation from S/IVOCs? Can S/IVOCs explain the unexplained SOA?

24. Line 272-276: My reading of Saliba et al. (2017) (and even May et al. (2014)) is that they do not find any differences in speciation between GDI and PFI vehicles (aromatics versus S/IVOCs) where this work does see differences in SOA production, which it then attributes to differences emissions of aromatics versus S/IVOCs. Elaborate on this discrepancy.

25. Line 294: GDI vehicles are 25% of the on-road vehicle stock or new vehicle sales?

26. Figure 2: Can you add more detail to the caption? What instrument are these data from? Are these from the CVS or chamber? Are these time-averaged? Are the particles denuded?

27. Figure 4: There are definitely more PAH studies that can be compared here, e.g., Hays et al. (2017).

28. Figure 5: This figure is misleading and needs to be redone accounting for the right OH exposures for each study since clearly some of the studies had different OH exposures for different vehicles.

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