"Characterization of particulate matter emissions from on-road gasoline and diesel vehicles using a soot particle aerosol mass spectrometer"

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Recommendation

I recommend that the MS entitled "Characterization of particulate matter emissions from on-road gasoline and diesel vehicles using a soot particle aerosol mass spectrometer" by Dallmann et al. be published by ACP after minor revisions.

Overview and general comments

This MS presents an array of useful data obtained during a tunnel study of the combustion emissions from a large number of heavy duty diesel vehicles (HDDVs). A soot particle aerosol mass spectrometer (SP-AMS) was used in conjunction with a multi-angle absorption photometer (MAPP), an OC/EC analyzer and gravimetric analysis of Teflon filters (and additional gas phase instruments). Data from these instruments support the authors' four main conclusions. First, they conclude that although the sensitivity of the SP-AMS is approximately a factor of four lower than that of the MAPP, the correlation between the two instruments is excellent, as is the correlation with Teflon filter PM mass. Second, the chemical composition of the emissions was surprisingly consistent for the test fleet; both the hydrocarbon ion series and the trace element analysis suggest that lubricating oil dominates contributions from diesel fuel in the HDDV combustion emissions. Third, given the cited similarity in chemical composition of oils used for gasoline and diesel vehicles, the measured similarity between the OA mass spectra for gasoline- and diesel-dominated sampling periods suggests that much of the OA from gasoline vehicles is due to lubricating oil. Fourth, this similarity between the OA mass spectra for gasoline- and diesel-dominated sampling periods is likely to confound efforts to apportion air pollution contributions from these two on-road sources.

This manuscript is clear, well-written and relatively free of faults—typographical, dictional or structural. In short, I was pleased to see that the authors took a similar degree of concern with communicating their scientific results as they did in carrying out the science itself. Therefore, my comments are mostly restricted to minor edits, although I do suggest a couple of thoughts for further consideration.

Specific comments

p. 4009, lines 1-21

The abstract could be improved by mentioning conclusions three and four (cited above in my general comments): emissions from gasoline vehicles is dominated by lubricating oil and the similarity in gas/diesel OA spectra will make apportionment difficult. In addition, it is worth mentioning the sample size (n=293 for SP-AMS and n=145 for the mass spec analysis) since it informally establishes the statistical power of the results. I would also consider mentioning the diesel truck BC "fingerprint" idea in the abstract.

p. 4011, lines 1-4
Engine age and condition are also important factors. Add reference(s).

p. 4011, line 8
Add Worton et al., 2014 reference.

p. 4011, line 14
"Kittelson," not "Kittleson"

p. 4012 lines 11-13
Why have the authors chosen to define MD and HD diesels like this? It would be more logical to use one of the standard definitions from CARB or the US EPA. Dieselnet provides the following discussion: "Heavy-duty vehicles are defined as vehicles of GVWR (gross vehicle weight rating) of above 8,500 lbs in the federal jurisdiction and above 14,000 lbs in California (model year 1995 and later)" (https://www.dieselnet.com/standards/us/hd.php). Or if they choose to retain their classification scheme, please briefly outline how it contrasts with the widely held definitions and why this choice was made. In making broad conclusions about vehicle classes (HD, MD, LD), it will be helpful if researchers from different groups are using a common language.

p. 4019, lines 7-9
Please add a one sentence explanation for why the negative artifact is expected to be low relative to adsorption.

p. 4019, lines 14-17
Why choose OA=1.25xOC? Provide references and/or quantitative explanation.

p. 4019, lines 26-28 and Fig. 2
Why include the OA from QBT? It confuses the graph and makes it harder to see that the quartz filter samples accounted for 87% of the Teflon PM mass. This figure needs a little work. First, too small and crowded; please widen it. Provide more space between the different sampling periods; right now it is difficult to see at a glance (if you don't look carefully at the x-axis labels) that the data should be viewed in three bar groups. Please add labels for weekday and weekend.

p. 4027, line 28
Please compare your emission factors with recent results from Gordon, T. D., A. A. Presto, et al. (2013). "Secondary Organic Aerosol Production from Diesel Vehicle Exhaust: Impact of Aftertreatment, Fuel Chemistry and Driving Cycle." Atmos. Chem. Phys. Discuss. 13: 24223-24262.

p. 4029, lines 6-10
Do the authors consider their estimates of the mixing ratios of trace elements to be accurate enough to provide a range for the fraction of the PM due to fuel vs. oil? Is there any way to make more

quantitative conclusions from the data in Table 1, recognizing the greater uncertainty in the Ca and Mg emission factor?

Please add a figure showing a simple cartoon of your experimental set-up. It will help readers see at a glance what was done and the instruments that were used.

Fig. 5
Enlarge figure in x and y. Enlarge legend. Split legend so that OA and BC goes with the bottom subplot and the other entries stay with the top subplot.

Fig. 6
Unclear what m/z ion signal is chosen as the point of comparison (y-value of 1). Should it be m/z 12? Identify your chosen reference m/z in caption.

Fig. 7
Enlarge figure in x and y. Put Zn and P on separate subplots, thereby changing this into a four panel plot instead of two.