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***Interactive comment on* “Characterization of particulate matter emissions from on-road gasoline and diesel vehicles using a soot particle aerosol mass spectrometer” by T. R. Dallmann et al.**

Anonymous Referee #2

Received and published: 11 April 2014

General comments

This paper presents measurements of vehicle exhaust from a traffic tunnel near San Francisco. The primary purpose of the paper is to present SP-AMS measurements at this location and to use these measurements to characterize vehicle emissions and derive emission factors for individual vehicles. One major finding in the paper is that lubricating oil is a major source of organic aerosol in the exhaust of diesel trucks, a finding that supports previous studies. Overall, the paper is well-written and presents

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a new SP-AMS dataset that can be used in the future to understand and give context to future SP-AMS measurements. Therefore, I recommend publication after my comments below are addressed.

Specific comments

pg 4009 - line 19 : Why does the presence of trace elements in particular raise a concern about ash deposits? This specific link is not made in the text.

4014 - 19 : What is the actual duty cycle of the SP-AMS? That is, how much of that 1 s is spent with the beam open and collecting ambient data?

4015 - 1 : What was the inner diameter of the copper tubing? The inner diameter is more relevant than the outer diameter.

4015 - 7 : What “experimental measurements and theoretical calculations” were done to quantify particle transmission efficiency in the sampling line? Because you quote absolute numbers later in the paper and are comparing to a gas-phase instrument (a CO₂ instrument), sampling artifacts should be more carefully considered. If this is answered in the previous paper, it should be cited here.

4015 - 18 : It might be helpful to talk more about potential evaporation or condensation in the sampling line that could affect chemical composition of the particles. You assume that these processes are not happening, but why is that a good assumption? How does the temperature of the sampling line change from 50 m inside the tunnel to outside the tunnel to inside the mobile lab? Is the ventilation duct drawing air into or out of the tunnel? Are there any references for what the volatilities are for typical engine exhaust?

4016 - 19 : Tied in with the comment below about page 4019 - line 24, I am a little confused about exactly what is at play in determining the calibration of the SP-AMS. This is probably all discussed in the previous SP-AMS literature, but I think there is room for a few more sentences of explanation in this paper so that readers aren't required to read the previous literature in order to understand this work. Considering

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just the species vaporized by the laser, i.e. black carbon and some of the lubricant-derived trace species, black carbon is the only species with an RIE_s not equal to 1. Despite this large correction for the ionization efficiency, there is still another large correction for collection efficiency, which I assume is really vaporization efficiency of black carbon. Does this collection efficiency factor back into the calibration of RIE_{BC} in any way? Basically, my cause for concern is that you apply two large correction factors to the black carbon mass measured by the SP-AMS in order make the measurements align with a MAAP instrument, and yet other species that are assumed to be vaporized in the same manner as BC in the SP-AMS are not corrected in the same way (because data and calibrations specific to these species do not exist). Then, ratios of these species to each other are used to conclude that the source of aerosol is dominated by lubricating oil rather than diesel fuel. Because the treatment of the individual species signals in the SP-AMS plays such a critical role in the final conclusions of the paper, I would like a little more detail throughout this section of the paper, particularly with regards to the assumptions made regarding the calibrations.

4017 - 5 : Everything after “assuming” is confusing and I’m not sure what you are trying to convey. What you have before that might be enough – you are saying that calcium and magnesium ions may be generated inside the SP-AMS and therefore may have higher than expected ion signals which means an overestimation of the mass of those species.

4018 - 25 : Does the fact that Massoli et al., 2012 reports a factor of 9 and this study reports a factor of 4 difference in SP-AMS BC and MAAP BC indicate that alignment of the particle beam and laser were better in this study than the previous study?

4019 - 6 : Was the SP-AMS ever run in laser-only mode during this campaign? Is there any way of knowing if there were significant coatings on the BC particles?

4019 - 24 : It seems like a bit of a stretch to assume that all lubricant-derived trace species have a CE of 1. I can understand this being the case for non-refractory species

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(mainly because of previous literature), but for the refractory species which are vaporized in the same way that black carbon is, why wouldn't the CE be much lower than 1 and closer to the CE of black carbon? Also, you are assuming that calcium and magnesium are heated by the BC particles vaporizing, which means you are assuming that calcium and magnesium are attached to the black carbon. So if only 27% of the black carbon particle mass is being vaporized, why can you assume that 100% of the BC coating mass is vaporized? Even if the calcium and magnesium were separate particles from the BC particles but were primarily vaporized by the laser, they would still suffer from the same beam alignment issue you discuss on pg 4018 - line 25. I think justification for this assumed CE needs to be stronger in this section. Also, what is the uncertainty of the black carbon CE?

4020 - 15 : Is "equivalent OA mass" the same as OM/OC ratio discussed by Hallar et al., 2013? It would be worthwhile to discuss your correction factor in context with others present in the literature.

Hallar, A. G., D. H. Lowenthal, S. L. Clegg, V. Samburova, N. F. Taylor, L. R. Mazzoleni, B. K. Zielinska, T. B. Kristensen, G. Chirokova, I. B. McCubbin, C. Dodson, and D. R. Collins (2013) Chemical and hygroscopic properties of aerosol organics at Storm Peak Laboratory, . *Geophys. Res. Atmos.*, 118(10), 4767–4779, doi:10.1002/jgrd.50373.

4020 - 17 : What specifically was evaluated using the SP-AMS? How might this evaluation be affected by the higher OA concentrations measured with the SP-AMS versus the filters?

4021 - 24 : Is this a heavy-duty truck?

4022 - 12 : Is the tunnel background mass spectra subtracted out from the exhaust plume mass spectra?

4023 - 8 : What does the average tunnel background BC mass spectrum look like?

4023 - 16 : The discussion here now switches back to Figure 5 - consider reorganizing

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this section slightly so that discussion of Figure 6 comes after all of the discussion of Figure 5.

4024 - 19 : What does the word “slightly” mean here? Are these quantifiably unsaturated compounds?

4025 - 6 : Aren't these literature results opposite your findings? These results suggest ratios less than 1 when you report ratios greater than 1 on line 3. This also calls into question your conclusion on lines 13-14.

4026 - 5 : Can you define “ash” and how it relates to your measurements?

4030 - 5 : What is the uncertainty on the 2.6 value?

4030 - 21 : Assuming that the background tunnel mass spectra are not subtracted from these mass spectra in Figure 8, what effect might that have on this correlation? It is probably impossible to do this subtraction for the IOPs, but it should be able to be done for the single gasoline and diesel vehicle events to ensure that the correlation is still strong. My concern is that this correlation is dominated by the background tunnel signals and any actual differences are masked by that background signal.

4031 - 1 : Did the gas vehicle event show BC signals above background levels?

4031 - 4 : To be clear, the conclusion is that the carbon fragments in the SP-AMS the same way on the weekdays as it does on Sunday, which may or may not say something about how the carbon was emitted in the first place.

4031 - 8-9 : This finding just says that all OA fragments in the same way in the SP-AMS regardless of the number of diesel trucks. If the OA is vaporized primarily by the heater and not the laser, and the presence of diesel trucks is more pronounced in the BC signal - which is vaporized by the laser - then this finding isn't necessarily surprising.

4031 - 28 : For the single gas vehicle event shown in Figure 8, were the lubricant-derived ions above background like they are in the diesel vehicle events?

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page 4043 : What does it mean that absorbed vapors is as much as the real OA for some of these sample periods?

Technical corrections

pg 4013 - line 18 : “operational” should be “operating”

4014 - 4 : “impact on...” should be “will impact on...”

4014 - 28 : It would be more clear if “prior to” is replaced with “from” or “inside”.

4017 - 25 : It might be helpful if the notation were “ EF_S ” rather than “ EF_P ” because “S” is clearly defined as species.

4022 - 24 : The sentence beginning with “In the average diesel PM mass spectrum...” is found in the figure caption and doesn’t need to appear in the text.

4025 - 4 : “mixtures” should be “ratios”

4025 - 5 : “and” should be “to”

4026 - 16 : Technically, BC has the highest correlation with OA as presented in the figure.

4026 - 26 : “in” should be “between”

4028 - 1 : “Note that ...” sounds better than just “Note ...”

4028 - 6-7 : This sentence is a repeat of page 4027 - line 21.

4028 - 7 : “Note that ...” sounds better than just “Note ...”

4030 22 : Remove the comma in this sentence, assuming that you mean that all BC mass spectra are highly correlated with that single spectrum from a diesel truck.

4031 - 21 : Consider making a Conclusions section starting here.

4031 - 27 : “omitted” should be “emitted”

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4033 - 4 : Should “abd” be “and”?

page 4043 : In the legend, isn't “OA (QBT)” really absorbed vapors and not organic aerosol?

page 4044 : The legend for the red circle and blue triangle is incomplete. Also, the “IOP2212” notation is confusing in the context of this figure - this text box could just be stated in the figure caption.

page 4046 : The sentence beginning with “Only trucks with ...” is found in the text of the paper and doesn't need to be repeated in both location. Either remove from the text or remove from this caption. Also, in the figure, it is hard to see a difference between the bars and error bars. Might it be better to have a third panel with the error bars plotted on that? Or come up with some other way that more clearly shows where the relative ion signal is such that it isn't masked by the error bars.

page 4047 : See the comment about the bars and errors bars for page 4046.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 4007, 2014.

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