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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Direct measurements of near-highway emissions in a high diesel environment" *by* H. L. DeWitt et al.

Anonymous Referee #2

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This paper presents analysis of the data from an approximately 2 week long measurement campaign at a site adjacent to a large freeway outside Grenoble, France. An extensive set of real-time instrumentation for measurement of particle- and gas-phase species was used including an SMPS for particle size/count, an HR-TOF-AMS for particle composition/size, a MAAP for BC light absorption, a TEOM for PM mass (though I did not see mention of these data), a PTR-TOF-MS for VOC measurement and a NOx analyzer. Integrated samples were collected for multiple analyses, including carbon isotope analysis to determine the modern/fossil fraction of total particulate carbon (TC). Supporting measurements included detailed traffic flow, speed and composition measurements and a meteorological station to measure wind parameters, temperature and RH. The analysis presented in the paper is similarly comprehensive and includes:

examination of diurnal trends, linkages with traffic conditions, PMF decomposition of AMS data and factors' relations with other measurements (e.g. VOCs, NOx, BC), and analysis of the fossil/modern carbon fraction of the total carbon.

This is a large and comprehensive dataset, and it appears that the data collection and analysis were well planned and executed. These data have the potential to provide valuable insights into the characteristics of emissions from a large and diverse vehicle fleet and to their contribution to urban/regional air pollution. Further, the title of the manuscript indicates that this is the goal of analysis. My major issue with this analysis is that it does not proceed in a way that can truly address this objective. Rather, as presented, the analysis treats the data set as a 'standard' ambient monitoring study, that pays little attention to the relationship between the source in question (the roadway) and transport of these emissions to the point of measurement (e.g. wind direction). If this work were presented as a routine monitoring effort at a site that happens to be near a large road, I might not take issue with this presentation. However, given that 'emission' is in the title, I expect to see more of a focus on trying to find the 'emission' signature, rather than just expecting it to be there and constant during all measurements. Therefore if the title were something like 'Comprehensive gas- and aerosol-measurements collected near a roadway in a high-diesel peri-urban environment', I would expect this kind of analysis. However, characterization of emissions should make efforts to separate or characterized the background vs roadway contributions. So, one approach could be to change the title of the paper and apply some of the changes suggested here and by the other reviewer. However, to actually address the objective implied by the title, more work needs to be done to extract the roadway 'emission' signature from the data. My sense is that not doing this additional analysis would be a missed opportunity.

There is a rather extensive literature of near-road air pollution measurements ((Karner et al. 2010) is a good place to start), which seems to be largely ignored in this analysis aside from the few studies in which an AMS was used. The important message from

these previous studies is that concentration gradients are: a) steep near roadways, b) a strong function of wind direction/speed and mixing conditions. However, the spatial and meteorological aspects of this data set are entirely absent from the analysis as presented. The air sampled at this site will be a combination of the background/regional mixture (heavily influenced by other roadways with similar diurnal various to traffic and fleet among the multitude of other sources) and emissions from vehicles on the adjacent roadway. A strength of having all of these highly sensitive, high-time-resolution instruments is that the authors should be able to pick the 'emission' signal out from the background, by sorting data based on wind speed/direction. It could be that due to the low wind speeds observed in this study (I cannot comment on wind direction, as no data were shown, nor was the relationship of the measurement site to the roadway) that is a small portion of the overall data set, but it would still get at the 'emissions' question. Based on the factor-based wind-roses in Figure S8, I would hazard a guess that the roadway was to the North/Northwest of the sampling location give the asymmetry in the HOA contribution on the wind rose (correction – just found the mention of this in Sec. 3.2). I would expect that the authors could extract portions of the data set that allow them to more directly evaluate traffic emissions, ideally by contrasting with other periods where the measurements were upwind from the roadway. Other measurements from regional air quality monitoring stations might be of use in assessing changes in regional levels to extract the roadway signal. The existing analysis can be presented along a more directional approach; there are likely valuable insights gained from each approach.

Apart from this framing/objectives concern, the main other concern I have is concerning the analysis of the fossil/modern carbon fractions. This analysis needs more attention, and I find it somewhat difficult to follow the process described, the results shown in Fig. 15 and the conclusions of this analysis. For one, a number of assumptions appear to be made in the process of apportioning the modern/fossil contribution to OA, but these are not at all clear. It appears that the modern C contribution to EC/BC is ignored, as is the potential modern C contribution to HOA, but I may not be clear on this. Given

the statement that as much as 7% of diesel may be biodiesel and that BBOA made a reasonably large contribution (Fig. 10b) to the OA loading at times, these assumptions seems to be potentially biasing and also unnecessary (given all of the other supporting data/analysis that is presented). I would like to see this process better spelled out and the uncertainties in the various assumptions required apportion the modern/fossil C better constrained and the impact of measurement uncertainties and uncertainties due to assumptions propagated through to this overall apportionment exercise. In particular, the measurement of EC/BC is a potentially large source of uncertainty that would introduce an enormous bias into this analysis. That the light-absorbing carbon is discussed as BC suggests that these quantities are based on an optical measurement, and there is huge variability/uncertainty in the absorption cross-section and thus the BC concentration measurement generated by something like a MAAP. There is mention of a thermo-optical analysis of filters, I think mainly from the other site, but no data are ever shown. Ideally, all of these comparisons would be on a mass basis (i.e. EC, not BC) and the uncertainties incorporated into the analysis. If these various uncertainties are addressed, a more quantitative apportionment of C mass will be possible, giving the discussion of SOA sources/etc. a more solid footing. In addtion, Fig. 15 is not clearly labeled – preferably segments would be labeled as either OC or EC to avoid any ambiguity.

Finally, while I am suggesting additional analysis, I agree with the other reviewer that the number of figures is excessive. Figures 2, 6, 11, 12 and 14 could easily be moved to SI without diminishing the readability of the manuscript.

Other specific points: P27376, L15-18 – Somewhat nitpicky, but the use of 'rate' here is not really appropriate, as this really depends on engine size. You could say 'emission factor' or 'ratios'. . .

P27376, L25-30 - Suggest reorganizing this paragraph as there is a transition to discussion of engine-type-specific emissions to results of several specific field measurements.

P27377, L6 – There has been no previous mention of smog chamber studies, so this way of starting the paragraph is a bit confusing.

P27377, L16-30 – Not so clear how this particular paragraph pertains to the study objectives. Seems like it could be replaced with a sentence stating that there is controversy concerning the relative contributions of gasoline and diesel engine emissions to primary and secondary OA in the US.

P27378, L17-20 – This seems like a major objective of the study, but then there is very little comparison (apart from e.g. HOA/BC ratios. This could be highlighted more and then more fully addressed.

P27379, L19 – This seems like a natural place to discuss the meteorology of the site, location of measurements relative to roadway, prevailing wind directions, etc.

P27382, L13 – It is not clear where this site is relative to the mobile lab, how these are impacted differentially by wind, and where data from the two sources are used in concert in this analysis. Were there any duplicate instruments on the two platforms on which this comparison can be made? In general, the contribution of data from the different sites could be better delineated.

P27383, L5-18 – This discussion of the fleet composition on this roadway is only really directly relevant if you are measuring emissions from these vehicles (see discussion above). Otherwise, you are better served by talking about the regional fleet composition (and other potential sources). This is a very detailed description of the source term, but is not relevant if you're not sampling from this source.

P27384, L5 – The only mention of meterology. There should be data accompanying this, including a more detailed diagram of the placement of the road, mobile lab and the regulatory monitoring site.

P27384, L21 – BC measurements need to be defined (e.g. comparison with EC, etc.)

P27385, L6-7 – The Org/BC ratio is much lower during this period as well. There

seems to be an opportunity to analyze specific periods/episodes to try to better extract the vehicle signature from other things. The BC variation is relatively consistent, but the 15th-18th has a much larger OA fraction and more of these other factors, and also much lower NO, this seems to be a different type of period than the rest of the study. Can you use differences between periods to infer the difference between roadway/regional contributors to measured levels?

P27386, L6 – this is not really 'BLH-corrected', as it's not clear that's something you can correct for. In any case, the rationale and approach could be better described. What is the motivation for this approach versus just plotting the diural variation in ratios (e.g. benzene: BC or some other primary vehicle emission tracer).

P27387, L27-28 – The evidence for this is not really shown, also need to show the mass in each size bin and the contribution of HOA/OOA to each size bin. Also, I'm not clear of showing the average values for the entire campaign. This represents a snapshot of the climatology of the local aerosol, but this analysis might be more useful if it compares times with different roadway vs. non-roadway contributions.

P27998, L26 – 'was SOA' is a very definite statement. . .

Figure S2 – It is hard to see in this presentation, but it appears that the PM data (presumably from TEOM?) has substantially more variation than the AMS data. Was the mass estimate from the MAAP benchmarked against the EC data? A scatter plot would be helpful. What about comparisons with SMPS volume data.

References: Karner, A. A., Eisinger, D. S., and Niemeier, D. A. (2010). "Near-Roadway Air Quality: Synthesizing the Findings from Real-World Data." Environmental Science & Technology, 44, 5334–5344.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 27373, 2014.