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Atmospheric Measurement Techniques Discussions

Interactive comment on "Mobile air monitoring data processing strategies and effects on spatial air pollution trends" *by* H. L. Brantley et al.

Anonymous Referee #3

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This manuscript compares different methodologies to discern emission factors from mobile laboratories, particularly methods to deduce "background" from local emission spikes (on various spatial scales). Mobile laboratories have shown great potential for capturing spatial variability of trace gas concentrations near emission sources. The goal of the study – to quantify robustly the diverse and appropriate methods to discern emission factors in mobile laboratories – is greatly needed. However, the manuscript doesn't show enough data to convince me that there is a path forward on using mobile laboratory data more robustly. In fact, I expect that the "best use practices" for using mobile laboratory data would be highly dependent upon location, types of gases, meteorology, etc. I was hoping this manuscript would show examples of this and develop a set of general recommendations for moving forward (even if they were specific to

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their conditions). Instead, I don't see any validation of the methodologies for determining a "background" and instead just see a series of statistical comparisons/analyses that don't provide significant conclusions, even in the authors' specific datasets. The only way one can determine which methodology accurately defines a "background" is to have an independent, validated measurement to compare to the mobile laboratory data. Without such data, I cannot recommend publication. Major revisions are needed – the datasets are unique and high-quality and there may be more data available for analyses, but what is in the manuscript is not sufficient.

The abstract needs to be more quantitative/descriptive, more details needed in the experimental section, too. Specific comments:

Page 10444, line 10: instead of saying "a large mobile monitoring dataset", the authors should quantify the temporal/spatial coverage of their dataset, and which part of the dataset would be actually used in the following analyses.

Page 10444, line 17: clarify the sentence about mean and median. Is it mean/median of a time window, spatial window, or the entire road trip?

Page 10445, line 12-17: emission estimation is a very broad concept. According to the methods and literature listed in this sentence, and the following section of this manuscript, the discussion here only implies estimation of on-road vehicle "emissions" using emission ratios/emission factors. The authors should be specific about what aspects of "emissions" they can measure. Actually, the mobile measurements presented in this manuscript can only get emission ratios, and it is impossible to quantify emission factors without knowing the vehicle fuel consumption or total carbon emissions. There is still value in measuring emission ratios/factors but the wording needs to be tighter here.

Page 10445, lines 18-19: Why are emissions only at scales of 10s of meters? Seems more to be instrument/platform resolution than any specific aspect to 10s of m being unique. Emissions occur at scales of tailpipes coupled with the distribution of atmo-

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spheric eddies – so there will be structure well below this size.

Page 10446, line 3, 24, and page 10447, line 8: the authors should clarify the concept of “local background”, “local-scale influences from regional background”, and “regional background”. Generally this manuscript is not very clear about the concept of “background”. Trace gas/aerosol concentrations change in a continuum of spatial/temporal scales, from directly at the tailpipe, to on-road environment, to roadside urban environment, to urban-suburban scale. Synoptic meteorological patterns change concentration at even larger scales. Anyone of these can be “background” when comparing to variation at a smaller scale. Hence the authors are suggested to define clearly where the gap is when contrasting local and background.

Page 10446, line 16-19: please clarify the difference between “general spatial variation attributed to local sources” and “short-term concentration spikes caused by nearby exhaust events”. The reader may think these two are the same, but it does not make sense in this sentence. Do the authors mean that the “short-term concentration spikes” from surrounding vehicles are more local than “local sources”? How is the concept of “local” defined?

Page 10446, line 20: again, the term “background standardization” is confusing, like noted above. This sentence refers to Table 2, but it is called “background estimation” instead in Table 2. I realize the authors are trying to summarize multiple literature reports, but setting up the problem better before this point may be helpful to the reader.

Page 10447, line 12: “noise” used here and elsewhere – please clarify that you aren’t talking about noise due to measurements but rather atmospheric variability near sources.

Page 10447, line 14: after applying the rolling mean/median, the data has been downsampled at some level and therefore is not as high as resolution (note that outliers and extreme events are reduced, so therefore it is of lower resolution).

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Page 10447, line 24: this is a good place to describe the total spatial/temporal coverage of the dataset. Hours of driving? Distance of driving? Details on the types of road conditions (freeway, side roads, stop-and-go traffic, etc.), time of day (afternoon with turbulent mixing maximized or early morning when conditions are more stable?), etc. are fine as noted in section 2.1, but some overview of what the reader may expect is helpful.

Page 10448, section 2.1: are all the sensors sharing the same inlet? This is critical to get useful statistics of emission ratios. Note the precisions of the measurements at the relevant sampling frequencies and overall accuracies. How often were the instruments calibrated – before, during, or after each drive? Start/end of campaign? Were any meteorological sensors included, and what are the accuracies/resolutions of wind speed/direction?

Page 10450, line 19: the authors discussed how they calculate emissions factors using emissions events above. Hence it is more appropriate to use something like “emission events labeling”. The vehicle emission events are still useful, so they are not for “removal” or “filtering”.

Page 10451, line 1-20: the efficiency and limitation of these emission event detection methods should be discussed and quantified. In Fig. 5, clearly there are many vehicle emission spikes that are not detected by the COV method. In high traffic volume, the measurements would be overwhelmed by emissions from surrounding vehicles, and the efficiency of these methods is questionable. The authors should utilize their data at higher traffic density regions to evaluate these methods.

Page 10451, line 18: the median method is basically the same thing comparing to the rolling 25th percentile method, as median is just the 50th percentile, and 25th percentile is somewhat arbitrary. More justification is necessary about which percentile to use. Also, what are the sensitivities with respect to the averaging windows for a given percentile? For a manuscript with a goal on trying to quantify such mobile-based

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measurements, I found this aspect to be severely lacking.

Page 10452, line 5: it is a very important question whether mobile measurements can capture the background. Stationary monitoring is a crucial part to serve as a ground truth. The most effective method would be to compare the on-road measurements with a series of stationary sites upwind and right beside the road. I realize the authors do not have such measurements, and this is more of a fault of the mobile laboratory methodologies in general, but just using a fixed algorithm/detection window/percentile is insufficient without validation. In fact, I would guess that this changes dramatically based upon stability and wind speed – e.g. some days a 10% threshold within a certain window may be fine, while other days it may be a lower threshold and narrower window.

Page 10453, section 3.1: the methods shown in this section have already been widely adapted in mobile measurements. Cross-correlation method has been demonstrated for mobile data synchronization for over a decade (e.g. Figure 4.13 of McManus et al. (2002)1). Therefore the necessity of showing Fig. 4 and Table 3 is questionable. It also seems unnecessary to discuss the emission factors calculated at longer averaging time. On-road concentration changes at a time scale of $\sim$10 s (e.g. Figure 4.1.2 of McManus et al. (2002)1, Figure 3 of Kolb et al. (2004)2, Figure 7 of Jimenez et al. (2001)3), so it is needless to say that averaging to 10 s would result in a loss most of the emission information. The authors may justify this section if they can show more innovative analyses, like how sensor response times influence the emission factor, or if there is a trade-off of averaging time, between losing emission information and gaining signal-to-noise ratio.

Page 10455, line 7: “can be added into a model as a random effect”. Which model? How is the “random effect” generated?

Page 10457, line 7: how can “regional background variation” obscure the “spatial variation”? The spatial variation should be just what was measured.

Page 10457, line 8: “background removal” is confusing. Does it related to “emission

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event removal", "background standardization", "background estimation" mentioned in the previous sections? The authors should consider standardize the terminology when referring to mobile data processing throughout the manuscript.

Page 10457, line 7-16, and Fig. 8: In Fig 8(a), it is hard to believe in reality the on-road concentration would change so much between Route A and Route B. Route A and B are contiguous on I-40, so most vehicles will cover both in a very short period of time. The vehicles are the overwhelmingly dominant sources of PM2.5 on the freeway, and in addition, there are no other significant aerosol sources in the area shown in Fig. 8 (I-40 between Route 54 and Research Triangle Park) according to NEI 20114. Therefore, it is unrealistic to explain the low value on Route B by spatial variation. A more likely explanation is that Route B was covered many times at lighter traffic (as indicated by the authors in Page 10451, line 22), which made the statistics on Route B lower. In this case, different regions have different representativeness, and this is not a fair comparison. Page 10457, Line 19: there is no Sect. 4.2.

Figure 3 has only one time label on the x-axis - so I can't tell the duration of the measurement.

Final thought: the manuscript/data have potential, the subject matter is rapidly-developing and useful methodology (mobile vehicle measurements) but the end result is not terribly quantitative or robust enough beyond what has already been done in the literature.

References:

1. McManus, J. B. et al. Measurements and Analyses of Urban Metabolism and Trace Gas Respiration. (NASA, 2002). at <http://hdl.handle.net/1721.1/33460>

2. Kolb, C. E. et al. Mobile laboratory with rapid response instruments for real-time measurements of urban and regional trace gas and particulate distributions and emission source characteristics. Environ. Sci. Technol. 38, 5694–5703 (2004).

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3. Jimenez, J. L. et al. Cross road and mobile tunable infrared laser measurements of nitrous oxide emissions from motor vehicles. Chemosphere-Global Chang. Sci. 2, 397–412 (2000).

4. NEI. The National Emission Inventory 2011. (2013). at <http://www.epa.gov/ttnchie1/net/2011inventory.html>

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 10443, 2013.