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Interactive Comment

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

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We thank the referee for valuable feedback. Referee #3 had some significant and valid concerns regarding the paper – the chief concerns included a lack of a comparison to validate time-series based approaches to estimate background, whether sufficient data was utilized and shown in the paper, and the inclusion of some analyses that the reviewer suggested to be of lower interest. We've undertaken some major revisions that we believe will address Reviewer #3's concerns and we feel have significantly improved the paper. To start off with, we realized in retrospect that we did not clearly present the amount of mobile monitoring data that were utilized in the paper – it is an extensive data set, with measurements collected over 12 routes repeatedly with over

140,000 observations of multiple pollutants recorded. We've added a map showing all of the routes.

In response to comments by all of the referees, we have undertaken significant additional analyses to strengthen the background analysis section of the paper and also removed the time lag/alignment section that multiple referees commented as of lower importance. In the course of this additional analysis and considering referee comments, we decided to remove the overly complicated time series algorithm we developed ("flexible window") and have simplified the comparison to be between the location-based background and the time-series based background (spline of minimums).

The location-based approach of estimating background concentrations, i.e., using the mean or median concentration measured in designated background areas (characterized by low traffic away from known sources) was applied to the eight mobile monitoring routes which included background areas. The time-series approach (spline of minimums) was also applied to these routes and the results were compared. In response to a comment by Reviewer #1, we further examined whether the spline of minimums method could be used when the sampling vehicle did not pass through designated background areas by artificially removing measurements made in the background areas and re-calculating the background using the spline of minimums.

The original questions and comments are shown below, followed by our point-by-point responses. The revised manuscript has been attached as a supplement.

General Comments: 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

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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 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.

Author response: We thank the referee for highlighting the lack of clarity about the purpose of the manuscript. Mobile monitoring has been applied to answer a variety of research questions (Table 1), not just to discern emissions factors, although that is one important application. As titled, the manuscript is primarily focused on understanding data processing implications on spatial air pollution trends derived from mobile monitoring. By including a section exploring time lag effects on estimated emission ratios (and the reviewer is correct that it was better defined as an emission ratio, not a factor), we may have caused confusion. In the improved manuscript, from the outset we introduce the topic of mobile monitoring from the broadest perspective possible and describe the multiple applications in research studies. We then focus the analysis and discussion on the utility of mobile monitoring to derive spatial patterns of air quality in urban-suburban settings.

As the reviewer commented, the paper needed improvement in providing an independent validation of the background estimation. The suggested approach – utilizing a second monitoring data set positioned in a background location – is logistically difficult and has drawbacks for the vast majority of mobile monitoring studies. One of the largest issues is that mobile monitoring vehicles, including the one in this study, utilize very expensive and sensitive instruments in order to accurately measure air pollutant concentrations in real-time while in motion. Replicating the identical instruments (e.g., an engine exhaust particle sizer for ultrafine particles or quantum cascade laser for carbon monoxide) is impossible for most research groups. Using alternative instruments for comparison – such as a typical trace-level carbon monoxide NDIR analyzer – has issues in using a slower and less-sensitive instrument as the benchmark.

An alternate approach we propose and one that aligns well with published mobile monitoring studies is to define an area of the route as background and use the average of the measurements collected on that section of the route (e.g., Hagler, 2010; Van Poppel, 2013). In the revised manuscript we selected a subset of the routes (8 of the 12) measured in this study, which had clear low traffic sections we could designate as representative of urban background. We then compared the median concentration measured during each pass of the background area with the concentration determined using a purely time series based approach (spline of minimums). The logical next question might be – well, how well could you estimate background using a time series algorithm if the driving route did not include low traffic background roads. In order to address that question, for the 8 routes we artificially removed the sections of the time series that had be labeled as "background areas" and ran the algorithm again. In the new figure and section added to the manuscript, we illustrate how the - spline of minimums time series approach (with and without designated background areas included) closely aligns but is generally slightly lower than the mean of the background estimated by location.

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

Author response: The abstract was updated in response to the referee comments and new additions added to the paper.

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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.

Author response: We added new content to the methods as well as a figure showing a map of all the routes. New text includes: "The study being reported here utilized 40 hours (> 140,000 observations) of mobile monitoring data collected on a roadway network in central North Carolina to explore common data processing strategies including local emission plume detection, background estimation, and averaging techniques for spatial trend analyses. One-second time resolution measurements of ultrafine particles (UFPs), black carbon (BC), particulate matter (PM), carbon monoxide (CO), and nitrogen dioxide NO2 were collected on twelve unique driving routes that were each sampled repeatedly. The route with the highest number of repetitions was used to compare local exhaust plume detection and averaging methods."

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?

Author response: Changed to: "Analyses demonstrate that the multiple local exhaust plume detection strategies reported produce generally similar results and that utilizing a median of measurements taken within a specified route segment (as opposed to a mean) may be sufficient to avoid bias in near-source spatial trends."

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

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here.

Author response: The reviewer is correct in that it is “emission ratios” presented in the original version of the manuscript. The improved version of the paper seeks to clarify that the focus of the analyses presented is on spatial trends analysis, where the focus on emissions is primarily to discern whether a vehicle emitted exhaust in very close proximity to the sampling inlet and potentially caused a bias in the spatial information derived from the data set. The same strategy of local exhaust plume identification, however, could be applied for studies where emission factor estimation is a priority.

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

Author response: As noted above, the primary focus on the analyses presented is on spatial variability of pollutant concentrations. We have clarified the text to note: “In near-source environments and general air quality surveys, pollutant concentrations attributable to local sources can vary on the scale of tens of meters or smaller.”

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.

Author response: The Referee is correct that “background” is an ambiguous and often contested term, and that we need to take care in explicitly defining how the term is used

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for the manuscript. We have added text to clearly define that the analyses presented are seeking to identify urban-suburban background component, calling it for simplicity “background” in the paper. To clearly explain the components of interest, we’ve added in the following text: “In this study we considered three components of mobile monitoring data: 1) local exhaust plumes (i.e., tail pipe exhaust near the sampling inlet); 2) local air pollution (e.g., traffic-related air pollution); 3) urban-suburban background (i.e., ambient air quality in the area sampled). Gas and aerosol concentrations change in a continuum of spatial and temporal scales, from the point of emissions to ultimate fate in the environment. Our definitions of local exhaust plumes, local air pollution, and background were derived from the various investigations that have utilized mobile monitoring (Table 1). Local exhaust plumes are defined as short-term events characterized by extremely high pollutant concentrations that can be attributed to directly sampling exhaust from a nearby vehicle. Local air pollution is defined here as well-mixed air that is affected by one or more known sources and modulated by local wind, such as air flow from a major highway to local residential areas. Finally, urban-suburban background, henceforth called “background” for simplicity, is defined on the scale of the route (∼5-20 km) as representative of ambient air quality conditions without detectable impact of a nearby source.”

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?

Author response: The referee identified what may be an important source of confusion to the reader. In the context of the analyses shown, the primary focus is understanding analysis strategies employed to understand spatial trends. Often, local exhaust plumes that occur on otherwise low traffic roadways (e.g., an incidental truck that emit-

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ted nearby the monitoring vehicle on one lap) are screened out to minimize their effect. We've updated the text to improve clarity.

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.

Author response: This section has been updated to improve clarity. New text includes: "Estimating background is a second key feature of interest to isolate in mobile air monitoring time series. Background air quality often varies diurnally and daily due to meteorology and long-distance transport of pollution. Accounting for the variable background may be conducted through optimal sampling design where an area representative of background is frequently sampled (e.g, Van Poppel et al., 2013). However, when a route completion exceeds the time frame within which the regional background changes or comparisons are being made between multiple routes measured on different days, additional strategies are needed. An alternative approach is to assume the baseline of the time series – represented simply as a low percentile of the data range or a more sophisticated time-varying baseline – is representative of background."

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.

Author response: Changed to "As a final data processing step, temporal or spatial smoothing is often applied either to reduce variation due to atmospheric variability or more effectively display trends. "

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).

Author response: Changed to “Applying a rolling median or mean can be used to maintain the temporal resolution while reducing the amount of instrument noise and influence of extreme outliers.”

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.

Author response: Added “The dataset consists of 40 hours of mobile monitoring data collected during weekday morning rush hour on 24 days and spanning 12 routes that covered areas of traffic delay, high traffic volume, transit routes, and urban background.”

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?

Author response: Added: “Calibration checks were routinely performed before and after each run. All instruments utilized minimal tubing length (<2 m) and pulled from manifolds connected to two co-located inlets mounted through a side passenger window location. Particle instruments utilized antistatic tubing with minimal bends to avoid particle loss. Further information on the general sampling vehicle set-up is available in Hagler et al. (2010) Wind speed and direction were measured with a highly sensitive 3-dimensional ultrasonic anemometer (Model 81000, RM Young Company, Traverse City, Michigan) placed at a stationary sampling site on each route.”

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".

Author response: Changed to "Four methods of removing the influence of local exhaust plumes were compared: the running coefficient of variation (COV) method used by Hagler et al. (2012) the standard deviation of the background (SD) method used by Drewnick et al. (2012), the rolling 25th percentile method used by Choi et al. (2012), and aggregating the data by route segment using outlier resistant statistics such as the median. The first two methods: the COV method (Hagler, 2012) and the SD method (Drewnick, 2012) are both methods of detecting and flagging local exhaust plumes."

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.

Author response: The Referee brings up a good point, that the emission event detection methods are optimized for specific conditions. We've added some text to clarify the best use and where the methods would likely fail. Added: "These methods are most effective for conditions where an individual vehicle's emissions causes a significant deviation in an otherwise low emissions environment, such as a truck passing the mobile monitoring vehicle on a low traffic residential road. In recent history, these approaches have been developed specifically for understanding local-scale air pollution effects from a nearby source, such as a major roadway, with the mobile sampling vehicle being driven along low-traffic side roads. Applying these approaches in environments with higher traffic, such as while driving on highways, likely only detects major outliers as the within-source pollutant levels are likely consistently high and dynamic."

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 measurements, I found this aspect to be severely lacking.

Author response: In this context the 25th percentile method proposed by Choi et al. (2012) is a rolling function of time, while the median is a method of aggregating the measurements by route segment. The revised manuscript has been updated to clarify this distinction.

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.

Author response: As noted in the initial summary comments, we felt this was a critical comment made by the Referee and conducted some significant new analyses to address this concern. While replicate instruments positioned at a site representative of background would be ideal, it is hardly ever feasible at the level of advanced instrumentation usually applied for mobile air monitoring and even then has instrument comparison steps that need to be applied to ensure bias between replicate instruments does not cause error. However, we wanted to provide an alternate approach to evaluate time series based background estimates. New content has been added demonstrating how time series-based background estimates (spline of minimums) compare against observations for “background” labeled sections of eight unique routes. We went beyond that to also show how the time series approach estimate would perform in driving route situation where the route did not include any background sections, by artificially removing the “background” identified data from the time series and re-running the algorithm.

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 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.

Author response: We thank the reviewer for the comment, this section was removed and instead we provided some more content on the background analyses, as noted above.

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

Author response: This part of the sentence was removed.

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

Author response: Changed to: “Before background standardization, the regional background variation obscured the variation in PM2.5 due to the highway (I-40).”

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

Author response: We have standardized the terminology in the revised manuscript: Changed to: “To illustrate the possibility of comparing different routes sampled on different days, we standardized the background of the concentrations measured on 4 routes on 8 different days by subtracting the estimates produced by the spline of minimums method from the measured concentrations. We then compared the PM2.5 concentrations with and without background standardization. “

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 2014. 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.

Author response: We thank the reviewer for highlighting the need for background standardization. This is the point that we were trying to make, albeit poorly. The low value on route B is due to differences in background concentrations. Even though the all the routes were measured during weekday morning rush hour, changes in the background concentrations from one day to the next and with different routes performed on different days, obscured the influence of the highway. We’ve updated the text to improve clarity.

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Page 10457, Line 19: there is no Sect. 4.2.

Author response: Corrected to 3.2

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

Author response: Based on the Referee comments on the low value for this portion of the manuscript, Figure 3 was removed.

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.

Author response: With the Referees helpful comments, we feel we have provided a more clear presentation of an extensive mobile monitoring data set and added some important new insight into the field.

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 emis- sion source characteristics. Environ. Sci. Technol. 38, 5694–5703 (2004). C4779

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/6/C4854/2014/amtd-6-C4854-2014-supplement.pdf

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