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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 appreciated the in-depth review provided by Referee #2, whose comments have helped us revise the manuscript to be more accessible to a broader audience. One of the main concerns of the reviewer was the terminology used and lack of detail provided to make Figures clearly understood – we have made an effort throughout the paper to improve the readability.

In addition, the Referee had some concerns regarding the background estimation section of the paper. In response to comments on this subject 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

Full Screen / Esc

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Interactive Discussion

Discussion Paper



[Interactive
Comment](#)

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.

Anonymous Referee #2

Overall, the manuscript is useful. Its apparent purpose seems to fall somewhere between a review paper and original research. As an original research contribution, however, it has only a few new insights; as a review paper, it has modest scope and depth. The manuscript is a struggle to read, due to several major issues in the writing style, please see also below. There are instances of jargon, undefined terms and poor paragraph structure and organization, and many sentences are unclear. I had to read them again and again in multiple places to get their meaning, and even then I needed to make some assumptions and guesses to arrive at a meaning. Because figures are very minimally captioned they help little. Insights and conclusions are mostly absent.

Some points of confusion are outlined below but I beg the authors to go through the

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manuscript with a fine-toothed comb or otherwise find a way to improve the clarity throughout. Perhaps get a colleague who is a good writer but not so close to the work to read it and provide feedback.

The manuscript could be strengthened by including some discussion of the applicability of various approaches to various types of data. I also couldn't really understand where/what the "framework" the authors refer to was, unless it is embodied in Figure 1, but the framework in Fig. 1 is not new; it is implicit in many earlier papers—or possibly even explicit somewhere. Referencing: the authors have a tendency to reference previous techniques and analyses sparingly, even though they may come up in discussion repeatedly. This is not the usual practice (or standard) of giving proper credit. Further, referencing more would make the manuscript clearer and easier to follow; sometime it is easier to remember a person's name than a term for an analysis approach—please say the name of the method and the reference every time you refer to them. Further It would further be helpful to the reader to use terms like "(modified Drewnick (2012) method)" (or whichever method is being improved) etc. where appropriate. In summary, a solid if not exceptionally novel or exciting paper, should be published with major revisions as indicated above and below.

Author response: We thank the reviewer for highlighting the use of jargon and lack of clarity. We have standardized and more clearly defined the terminology in the revised manuscript and have added additional citations to the references to previous methods.

Specific comments:

10448 We really need to know the instrument response time. Sampling rate and response time are often not the same, and if they are not it makes an enormous difference to the data interpretation.

Author response: We added the following text to the methods section, to clarify the sampling versus response times: "For the current instrument setup, the time between a concentration change (high efficiency particulate air filter for particle instruments, gas

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Interactive
Comment

standard for gas instruments) at the inlet and visual inspection of instrument response ranged from 0 s to 5 s for both real-time gas and particle instruments. The response time of the QCL (CO) and APS (particle count in fine to coarse range) was within 1 s, the CAPS (NO₂) and aethalometer (BC) were 4 s, and the Li-COR (CO₂) and EEPS (UFP) were 5 s.”

10451 lines 8: what happens as a consequence of data being flagged?

Author response: We added the following text to provide some explanation on the utility of flagging those time periods. “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. For studies characterizing near-source air pollution spatial gradients, one approach may be to remove these flagged periods to avoid confounding influence from side road traffic. Studies focused on personal or localized exposure, however, may not want to remove the influence of the local exhaust plumes. For studies emphasizing emissions characterization, the time periods where local exhaust is detected may be of most interest to isolate and further analyze.”

10451 Lines 14 – 16, instead of “distance” I think you mean “measurement”, if not, then distance of what?

Author response: We thank the reviewer for highlighting the lack of clarity in the wording of this sentence. The intended word was distance, in reference to the distance of the sampling vehicle from the highway. The sentence was changed to: “Choi et al. (2012) used a 53 s time window (26 s before and after the center data point) when the sampling vehicle was more than 1 km away from a freeway, 31 s (15 s before and after) for distances between 300 m and 1 km, and 3 s (1 s before and after) within 300 m of a freeway.”

10452 lines 1 – 2 this sentence is very confusing. Assuming I figured out what you mean, perhaps you should give your “new method” (not clear to me what it is) a name so that the reader can make that connection if it comes up again later?

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

[Interactive
Comment](#)

Author response: The section was updated to improve clarity. The flexible window algorithm was removed and the new time-series based method proposed was named the “Spline of minimums method”

10452 line 3 what is “This”?

Author response: The sentence was removed.

10452 paragraph 2, please make a connection between the speed of the sampling vehicle and the smoothing intervals, if there is one.

Author response: The text was changed to: “The average speed of the monitoring vehicle on the route used to compare temporal and spatial smoothing was approximately 10 m/s. The smoothing intervals chosen for comparison were 10 m, 50 m, and 100 m segments and the time intervals necessary to traverse each distance at the average speed which equate to 1 s (raw data), 5 s, and 10 s, respectively.”

10452 line 14 window should be windows.

Author response: Changed to intervals.

10453 The “engineering method” (please rename for clarity) and the time correlation method both need references throughout, since they are from the literature.

Author response: This section was removed based on a comment from Reviewer #3, who advised that this section was of lower importance compared to other mobile monitoring data processing analyses. With the removal of this section, we were able to provide more in-depth analysis of how background is estimated and further information on the mobile monitoring data set employed in this paper.

10453 What happens if you just do time lag correlation and skip the “engineering method”; does that work for your data?

Author response: This section was removed based on a comment from Reviewer #3.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

10453 line 20 etc. Please clarify how you are deriving emissions factors, and from what. Why are you calculating a 10s average, and of what? The 10 s average seems too long for the plumes of individual vehicles.

Author response: This section was removed based on a comment from Reviewer #3.

10454 The inquiry you carried out begs the question what sort of emission factor (again, not sure how you are using that term) difference you would get if you had sub-second data.

Author response: This section was removed based on a comment from Reviewer #3.

10455 Please explain what you mean by “or can be added to a model as a random effect”.

Author response: This phrase was removed.

10455 My understanding of a “rolling minimum” is something that selects minima of a moving window of some (usually constant) size. The stair step pattern shown seems to be what would result from moving a fixed window step by step in a way that each data point contributes only once to the min. Need to clarify/reword/etc.

Author response: Referee #2 has a correct understanding of a rolling minimum, however that calculation counterintuitively does produce the stair step pattern shown. A rolling mean would produce a smooth curve, because it would change slightly as the window progressed through each data point. Because the minimum is based on the lowest value, it produces the stair step pattern. As the window rolls through the data the minimum remains the same until a value less than the current minimum is encountered (step down) or the window no longer includes the value that was the previous minimum (step up).

10455/Fig. 7 This data is very spiky and populated by wide peaks, not clear why, instrument response time? Background? Need to discuss the efficacy of various methods when you have different types of data/contribution of the background. For example,

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

treatment of BC with 6% contribution from the background will be very different from treatment of CO₂ with an 85% contribution from the background.

Author response: The Referee is correct in the observation that real-time air pollution readings are highly variable. This data was collected highly sensitive, real-time monitoring instruments in an environment where emissions are in close proximity. In studies where instruments have slower response time (e.g., 1 minute) or data are logged at a slower rate, the pollution trends can seem less dynamic. Further analysis has been provided to deepen the discussion on how background is estimated. A new figure was added to the revised manuscript that compares the concentrations in driving route sections designated as background with the background determined algorithmically for BC, CO, NO₂, PM_{2.5} and UFP.

10456 Para. 3. Do the meteorological conditions change significantly over the course of 2 hours? Can you back up your statement with data? I suspect there are other factors at play such as WD and upwind sources, etc.

Author response: The Referee brings up an important topic that affects how mobile monitoring sessions are implemented and how data is interpreted. Added: “During this run the wind speed increased from a mean of 0.3 m/s during the first half hour to a mean of 0.7 m/s for the last half hour and the wind direction was fairly consistently from the southwest: mean wind direction was 217 deg and 249 deg during the first and last half hour, respectively. The decrease in background concentrations over the two hour time span is likely related to an increase in the atmospheric mixing height during the morning period, however further analysis would be required to fully explore the causes of background variation.”

10456 Para. 3 Why do CO and NO₂ behave differently than the other pollutants?

Author response: We do not have a definitive answer for why the between run variation of these pollutants was greater than the hourly variation, unlike the other pollutants. It may be due to differences in long range transport.

10456 line 18 within run SD of what? W/ or w/o the “standardization”?

Author response: Changed to “To compare the variation in background concentrations estimated using the spline of minimums method, the mean background value for each run was calculated and the between-run standard deviation (SD) was determined from the resulting 24 mean background values. Additionally, the within-run SDs of the estimated background concentrations were calculated by first calculating the SD of the background concentrations for each run and then taking the range of those values.

10456 starting with “Kimbrough” -> end of paragraph: this discussion should be with your similar results and separated from this paragraph.

Author response: We thank the reviewer for the suggestion and have moved this section about the discussion of the variation in background.

10457 line 1 “downwind measurements” in which study?

Author response: Changed to: “The background contributions measured by Kimbrough et al. (2013) are higher than those calculated for the current study, likely because the downwind measurements made by Kimbrough et al (2013) were collected 20 m from the road, while many of the measurements in the current study were collected on the highway or on roads with high traffic volume causing the total concentrations to be higher and the fraction attributable to regional background to be lower.”

10458 Please explain why spatial smoothing increases correlations more than temporal smoothing and what the implications of this result are. “ADTT” is never actually defined that I could find.

Author response: The routes presented in this paper were driven repeatedly within a total driving period of several hours. Spatially averaging a repeated route leads to combining more points into the averaging-length segments (e.g., 10 m segments) in comparison to temporally averaging over the equal time window it takes for the vehicle to drive the same distance (e.g., it takes 1 second to drive 10 m). Table 4 shows there is

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

[Interactive
Comment](#)

a lower “N” for the spatial averaging versus temporal averaging. While a lower N does not necessarily always lead to higher correlation, this study demonstrates that spatial versus temporal averaging of mobile air monitoring observations does appear to provide higher correlation values. The rationale for this may be that spatial averaging may more clearly isolate trends of higher versus lower air pollution environments (highways versus background). Added the text, “Spatial smoothing results in a smaller sample size used to determine the correlations compared with temporal smoothing due to the repeated laps. While a smaller sample size does not necessarily lead to higher correlation, this study demonstrates that spatial versus temporal averaging of mobile air monitoring observations does appear to provide higher correlation values. The results indicate that spatial averaging may more clearly isolate trends of higher versus lower air pollution environments (highways versus background).”

We have added the definition of AADT at first use.

Table 5: add the instrument response times. Why do you expect/what is the insight from pollutants being correlated on different length scales, and more so than on time scales? The flexible window method developed by the authors is a complex approach, potentially unnecessarily so. This approach involves picking the minimum values, and if there is no appropriate value, a process of picking a value related to the standard deviation of the lowest 10 percent value. These choices at least need to be accompanied by some justification. Further, there is no explanation of why two or three points in the sample trace, lower than all neighboring points, were not chosen as minimum points. Overall, the flexible window approach seems to produce results that are nearly identical to the two simpler methods to which it is compared, and thus does not offer an obvious advantage. Picking a baseline is an issue that is problem specific to the question at hand, and, at best, tricky. A clear statement of the intent of the baseline choice should be added.

Author response: The insightful comments provided by the Referee are appreciated. The Referee is correct that estimation of background is a major issue for mobile air

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monitoring analyses and one where the rules are still being written. While this paper does not seek to recommend one single method, it looks to explore various background estimation methods. In response to this Referee as well as Referee #3 comments, we provided additional analyses of the background estimation methodology. In the process, we simplified the paper by removing the overly complicated formula to provide a more simple comparison of background estimated by location selection (e.g., a section of the route far from sources and with low local traffic) with background estimated through a purely time series approach (e.g., using a spline to fit the minimum concentrations for a given time window).

Word choice: 1. Background “standardization”; a better word might be “normalization” 2. The “engineering method” doesn’t tell the reader anything. Words to actually describe what you are referring to would be much more effective.

Author response: In the revised manuscript background standardization is defined and discussion of the engineering method was removed.

Figures: Figures should have captions that make them understandable on their own without referring to the text. Please use their captions and legends to make clear what is being plotted.

Author response: We have made an effort to ensure figures are well-captioned and provided with clear legends to be easily understood.

Figure 2. a. What data is this? A candle test of your instruments? Roadway data? b. It is standard to indicate the meaning of each panel in the caption, although in this case it is fairly obvious. c. The legend is a mystery. The same lines are indicated to mean different things -?? Both emissions events and roadway concentrations? d. What do you mean by emission event? c. Purple is barely differentiable from black.

Author comment: As noted previously, the section describing in detail how time lag analyses were estimated were removed based upon review comments by Referee #3

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

and further information on background estimation strategies was added. Therefore, Figure 2 was removed.

Figure 3. Not very interesting, but if you keep it, include a more complete description. Author comment: As noted previously, the section describing in detail how time lag analyses were estimated were removed based upon review comments by Referees #2 and #3. Therefore, Figure 3 was also removed.

Figure 5. That's not a transect. It's a route. "Green lines represent . . ." Is referring to what?

Author response: Figure 5 was updated to highlight the various components of the route and to help the reader visualize the local exhaust plumes.

Figure 6. This figure needs the original data. Also the methods used need their literature/this work citations in the legend, preferably (e.g., "Mean (this work)" etc.)

Author response: Added the citations, including the original data would obscure the rest of the figure, the original data for NO₂ is shown in Figure 8.

Figure 7. Indicate what the data are and what the gaps are. I am unclear how you can get the step function behavior in b) from a running average. That would be expected from windows one after another—?

Author response: Caption was updated. See previous explanation of rolling minimum.

Figure 8. The legend says points are percentiles and the caption says medians. Please clarify/correct.

Author response: As noted previously, we've made effort in response to the Referee guidance to improve Figure labeling and captions. For this particular figure, the caption is noting that each point shown is a median of all the measurements made within that 50m route segment and that the points are colored by the percentile of PM_{2.5}.

Figure 9. What is this data? What are the different panels trying to show?

Author response: The caption was expanded to provide more detail of the figure description. For this figure, the panels are showing the effect of temporal and spatial smoothing on NO₂ measurements collected on the 900 m transect of the route shown in Fig. 3 by distance from the highway: a) raw data; b) data after spatial smoothing by calculating mean concentrations by 10 m, 50 m, and 100 m route segments; c) data after temporal smoothing by calculating discrete 5 s, 10 s, and 20 s averages; d) data after combination of temporal and spatial smoothing. The model $\text{NO}_2 = m * \log(\text{distance}) + b$ was fit for each smoothing case. The regression lines are plotted with the respective data and the color corresponds with the points used

Figure 10. Needs more description in the caption.

Author response: Figure 10 illustrated the flexible window algorithm that was removed as mentioned previously.

Please also note the supplement to this comment:

<http://www.atmos-meas-tech-discuss.net/6/C4842/2014/amtd-6-C4842-2014-supplement.pdf>

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