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Title: Plume-Based Analysis of Vehicle Fleet Air Pollutant Emissions and the Contribution from High Emitters
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Overview:
The authors present results from a near-road motor vehicle emissions measurement campaign in Toronto, Canada. Concentrations of a variety of pollutants were measured 15 m from a busy roadway, and an algorithm was developed to (1) automatically identify vehicle emission plumes using CO_2 data and (2) calculate fuel-specific pollutant emission factors for individual vehicle plumes. The authors identify a total of 103,000 individual plumes in their data set, though the number of plumes for which pollutant emission factors can be quantified is significantly lower due to instrument sensitivities and plume dilution. Results are used to investigate fleet-average emission factors, impacts of high-emitting vehicles, and the efficacy of local vehicle emission control regulations.

In general, I thought the authors present an interesting new technique for collection and analysis of near-road, high time-resolution air pollution data. The sample size of individual vehicle plumes considered here is certainly impressive and the authors use results for some interesting investigations. However, given the aim and scope of AMT, I would like to see a more thorough explanation of the algorithm used in this study for automated data analysis. This seems to be the novel contribution of the study in regards to measurement techniques, and the current treatment of the algorithm in the text left me somewhat confused and limited my confidence in presented results. I believe this issue should be addressed prior to consideration for publication in AMT. Specific comments are included below.

Specific Comments:
Lines 51-52: I found this paragraph to be slightly confusing, partially due to the use of “top-down” and “bottom-up” when referring to emissions modeling and emission factor characterization, respectively. This terminology indicates that emission models and emission factor characterization are two complimentary approaches for evaluating vehicle emissions; however this is not the case. I suggest deleting the terms “top-down” and “bottom-up”. On a related note, it was not clear to me how real-world emission measurements can be used to “bridge the gap between laboratory and ambient observations”. The authors should clarify what differences between laboratory and ambient measurements they seek to reconcile with their measurements.

Lines 67: Remote sensing typically refers to a specific method for measuring pollutant concentrations in vehicle exhaust plumes employing cross-road spectroscopy. The authors incorrectly refer to all near and on-road emission measurement techniques (e.g. tunnel studies, plume capture, remote sensing) as remote sensing here. This indicates unfamiliarity with distinctions between methods used to evaluate vehicle emission factors in near-road settings (despite the large number of cited studies in this paragraph). I suggest the authors revise this paragraph to more clearly explain similarities and differences between real-world emission measurement techniques.

Lines 102-103: From a statistical perspective, is it possible to estimate the number of individual vehicle emission measurements needed to accurately estimate the fleet-average EF?

Lines 211-213: Please include the actual number of stable ambient periods considered in the calculation of instrument sensitivities. Did sensitivities remain constant in each of the 4 seasonal sampling periods? Also, please explain how EF_{DL} values for each species were calculated and include EF_{DL} values in Table 1.

Section 2.3: Given the aim and scope of this journal, I expected to see a more complete description of the automatic plume identification and integration method included in the main text. It seems to me that the novel aspect of this work in regards to atmospheric measurement techniques is the algorithm used for the analysis of high time-resolution data collected at a near-road sampling site. Unfortunately, the description of the algorithm is limited to six lines in the main text, and I remain somewhat confused about how (1) vehicle plumes were identified, (2) how integration start and end points were identified, and (3) how values for the two criteria for a "successful" plume capture were determined (plume width greater than 9 seconds and average delta [CO_2] greater than or equal to 5 ppm).

Section 2.3: An important question related to my comment above is the how data streams from each of the instruments were time-aligned in order to assure that plume start and end times derived from CO_2 data were appropriate to use for other species. For example, were any offsets made to the data to account for different instrument response times?

In the Supporting Information, the authors state that a 20 second buffer was added on each side of the plume length for CO integrations due to the lower time response of the CO instrument. How does this impact CO EF calculations for successive plumes separated by less than 20 seconds (as shown in the right panel of Figure S1)? It seems as if this would result in integration of the same selection of CO data for two separate plume events.

Lines 207-208, SI Individual EF analysis section: In lines 207-208, the authors state that background concentrations are set to the minimum concentration level at the beginning or end of a plume. In the supporting info, the authors describe a slightly different method for determining background concentrations for BC, which involves averaging lower value points to determine a minimum value. This is a relatively minor difference, but in relation to my comment above on Section 2.3, these inconsistencies make it difficult to understand how the algorithm developed by the authors actually evaluated EFs for exhaust plumes.

Section 3.1: For the 103,000 successful plume captures, please include descriptive statistics (range, mean, median, etc.) for $\Delta[CO_2]$, dilution ratio, and plume width.

Lines 238-251: The authors compare particle number EFs calculated from measurements made at 3 m and 15 m from roadside for 287 vehicle plumes as a method validation step. This is a useful comparison; and I suggest that, in addition to comparing the mean PN EF calculated at 3 m and 15 m, the authors include a scatter plot showing the comparison of PN EFs calculated at 3 m and at 15 m for the 287 individual plumes. This would provide an indication of how effective the authors' methods are in quantifying EFs for individual plumes. This plot can be included in the supplementary information.

Lines 264-278: The authors select a subset of 152 plumes that had "elevated pollutant concentrations" to examine the effect of individual vehicle types (e.g. truck vs. passenger car). The selection criteria used is vague and seems to be somewhat arbitrary. For example, was an individual plume selected only if concentrations of all species were elevated? Given that this analysis only involves ~0.1% of the authors data set of captured plumes, I feel that the selection criteria should be more objectively defined and explained more clearly in the manuscript.

Line 306: It's not clear why the CO emission factor reported here is more representative of LDVs than CO EFs reported in cited studies. This wording implies other studies exclude low-emitting vehicles from fleet-average EF calculations, which is not the case for tunnel and remote sensing studies.

Minor Comments:

Lines 24-25: Suggest changing BC and PN emission factor units from mg kg^{-1} and kg^{-1} to mg (kg $fuel)^{-1}$ and (kg $fuel)^{-1}$, respectively.

Line 36: This sentence seems to imply there were only 3.7 million deaths globally in 2012. I suspect this refers to mortality attributable to air pollution. I suggest rephrasing the sentence to make this connection clear for the reader.

Figure 1d: Please include units for wind speed and the radial axis.

Lines 143-147: How were diurnal profiles for LD and HD vehicle activity at the site obtained? Suggest mentioning methods in the text.

Lines 184-185: Please include units for BC conversion factor.