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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Identification and quantification of particulate tracers of exhaust and non-exhaust vehicle emissions" *by* Aurélie Charron et al.

Anonymous Referee #2

Received and published: 30 October 2018

–GENERAL COMMENTS– Charron et al present chemical speciation of particulate matter collected from chassis dynamometer experiments as well as near-road and urban background sites. Such measurements are important for understanding both exhaust and non-exhaust emissions and their variability across changing and variable vehicle fleets as well as locations with different traffic conditions and meteorology. The wide range of measured species (which are effectively contextualized within other measurements in the literature) strengthens the dataset, but also presents challenges for clear communication. A number of editorial comments are given in order to clarify ambiguous or unclear meaning as well as to correct grammatical errors. Improving the clarity of figures would also strengthen the manuscript.

–SPECIFIC COMMENTS–

SECTION 2.1.1 and SECTION 3.2.3: (1) Please provide the sample flowrates, dilution ratios, and sampling time duration for each vehicle tested (perhaps in supplemental section I). (2) The authors note that differences in dilution ratios could affect the distribution of n-alkanes due to differences in phase partitioning among species with different vapor pressures. Differences in filter face velocity could also lead to differences in vapor adsorption (positive artifacts) and particle evaporation (negative artifacts), resulting in different degrees of over-estimation of particle-phase organic material across different sampling conditions (and thus different vehicle types). For chassis dynamometer experiments, filter face velocity differs by up to a factor of 1.67 for molecularly speciated measurements and up to a factor of 6 for EC and OC measurements. Would quartz filter sampling artifacts due to differences in sample flowrates (ie. face velocity, pressure drop) affect the authors' conclusions about distribution of n-alkanes as well as OC/EC values? (In particular, how would quartz filter artifacts affect the authors' conclusions about the impact of the particulate filter retrofit on OC and EC emissions?) REFERENCE Solomon, P. A., et al., Evaluation of PM2.5 chemical speciation samplers for use in the U.S. EPA national PM2.5 chemical speciation network, EPA Rep. EPA-454/R-01-005, Off. of Air Qual. Plann. and Stand., Research Triangle Park, N. C., 2000. REFERENCE McDow, S. R., and J. J. Huntzicker, Vapor adsorption artifact in the sampling of organic aerosol: face velocity effects, Atmos. Environ., Part A, 24, 2563 – 2571, 1990.

PAGE 5, LINE 9: Please specify which analysis method (GCMS or LCMS) is used for which organic molecules.

SECTION 2.4: Which type of MLR analysis was used in this work? (ie. Which algorithm was used to calculate the MLR relationships with HD and LD traffic?)

PAGE 9, LINE 17-18: Which constants have high p values? Those in Table 3 all have low (significant) values. In addition, the second half of the sentence implies that the

authors are comparing the urban background and remote site? Please clarify.

PAGE 10, LINE 5: What do the authors mean by “smoker vehicles”?

PAGE 11 LINES 25-29: 1) Which “unquantified compounds”? How is this rough estimate calculated if these compounds are not quantified? 2) I suggest also including the measured particulate EF’s for exhaust and non-exhaust emissions to compare both exhaust and non-exhaust EF’s with the standard.

PAGE 16 LINES 3-4: Please be more specific about which divergences were observed.

–COMMENTS ON TABLES AND FIGURES–

TABLE 1A: 1) 10th column’s title should be “R with EC”. 2) Why are parts of the table highlighted? Please include this in the caption.

TABLE 1B: Why are parts of the table highlighted? Please include this in the caption.

TABLE 3: Perhaps this would be addressed in the type-setting process, but please ensure that the units column is sufficiently wide, center the column titles, and use heavier borders to separate the HDV and LDV sections.

TABLE III-1: The title should be for a figure, not a table. Also, please indicate both data series in the legend (currently only harmonic mean speed is included).

FIGURE 2: Please indicate in the figure or caption why there are missing data points in Figure 2B (or leave these categories out of the plot). Suggestions to increase readability: Consider combining EC and OC into one plot. Instead of repeating vehicle type for each set of three conditions (UC, UH, R), consider grouping them with a bracket and labeling them together with the vehicle type.

SUPPLEMENTAL VII-1 and VII-2: Please use different colors for different species (ie. use a single color for Cu and not for any other species). Please also increase the text size of axes titles and tick mark labels.

SUPPLEMENTAL VIII: Please increase the text size of axes titles, tick mark labels, and other text.

SUPPLEMENTAL IX: Given the attention paid to ratios in this study, I would suggest adding rows for the most relevant ratios (at the least, Cu/Fn, Cu/Mn, and Cu/Sn) where data is available.

–EDITORIAL COMMENTS–

PAGE 1, LINE 13: grammar: add comma: “vehicular emissions, a large comprehensive dataset”

PAGE 1, LINE 20: ambiguous, replace “Most of the first ones” with “Light-duty traffic emission factors” and move “in absence of significant non-combustion emissions” to be beginning of the sentence (since it applies to the traffic emission factors, not the chassis dynamometer measurements)

PAGE 1, LINE 21: suggested change to correct grammar and increase clarity: “Since recent measurements in Europe including those from this study are consistent, ratios involving copper (Cu/Fe and Cu/Sn) could be used as brake-wear emissions tracers as long as brakes with Cu remain in use.”

PAGE 1, LINE 23: The sentence regarding OC/EC ratio does not seem relevant or necessary to the abstract. In addition, the language implies that the OC/EC ratio is always 0.44 in France, which is likely not the intention of the authors.

PAGE 1, LINE 26: grammar: change “markers; while, their” to “markers, since”

PAGE 1, LINE 28: grammar: “environments” (should be plural as written)

PAGE 1, LINE 30: grammar: “filters have been progressively introduced”

PAGE 1, LINE 36: soften tone: delete “It is obvious that”

PAGE 2, LINE 1: grammar: “Also, knowledge of the deleterious impacts of PM on

human health”

PAGE 2, LINE 2: grammar: “PM is responsible”

PAGE 2, LINE 11: grammar: “They also do not represent the variability”

PAGE 2, LINE 19: clarity / word choice: replace “chemistry of PM” with “chemical composition of exhaust and non-exhaust particulate emissions”

PAGE 4, LINE 26: clarity: replace “below installation of” with “below this threshold for”

PAGE 4, LINE 28: grammar: “30% and 36% respectively”

PAGE 4, LINE 33: “(see SI section IV)”

PAGE 5, LINE 12: grammar: replace “sampler” with “samples”

PAGE 5, LINE 13: ambiguity, makes it sound like there are two background and two urban sites: delete “two”

PAGE 6, LINE 11: I am not sure if perhaps the authors intended “residual” instead of “residues”?

PAGE 6, LINE 33: Is the discussion of the additional Fe and Mn source in the subsequent paragraph? I do not see it in Section 3.2 as indicated in the text.

PAGE 7, LINE 1: clarity / ambiguity: “are more scattered possibly due to differences between light-duty and heavy-duty emissions factors”

PAGE 7, LINE 12: precision / clarity: replace “relationships” with “correlations”

PAGE 7, LINES 33-34: Do the authors intend that the n-alkanes and hopanes are correlated to each (traffic, NOx, EC)? If so, change to “NOx, and EC”.

PAGE 8, LINE 25: grammar: concentrations should be singular: “one for Sr concentration”

PAGE 8, LINE 36: grammar: “All of these suggest”

PAGE 9, LINES 8-9: To increase clarity, consider moving this sentence to the end of the first paragraph of this subsection (3.1.2).

PAGE 9, LINE 18: grammar: “contributes”

PAGE 9, LINE 27: change “technics” to “techniques” for more common spelling

PAGE 9, LINE 37: grammar: “larger than could be expected”

PAGE 10, LINE 2: grammar: “as ours, and the EF for exhaust OC”

PAGE 10, LINE 5: grammar: “contribution of smoker vehicles; and rapid formation”

PAGE 10 LINE 11-12: grammar: non-retrofitted is less clear than “without” (see comment on PAGE 15 LINE 6 as well): “test diesel vehicles without particle filters”

PAGE 10 LINE 26: grammar: “third highest traffic emission rate”

PAGE 11 LINE 28: grammar: “EF”

PAGE 11, LINE 30 – PAGE 12, LINE 8: The second sentence of this paragraph (regarding Cu and Sb brake wear emissions) confused me because it suggested that the Cu/Sb ratio would be a good candidate as a tracer, which is not the case (as communicated later in the paragraph). I suggest being more direct with the conclusion earlier in the paragraph to avoid confusion.

PAGE 11 LINE 35: tone: suggestion to avoid the word “obviously.” Also, please qualify the sentence by adding “in this study.”

PAGE 11 LINE 31: Add citation for CITEPA in references

PAGE 11 LINE 35: grammar: “depend”

PAGE 12 LINE 6: replace “Then” with “Thus”

PAGE 12 LINE 23: replace “spent” with “used”

PAGE 12 LINE 31: precision / clarity: “Cu/Sn would be the strongest candidate

PAGE 14 LINE 12: grammar / clarity: “The normalized abundance of 17α,21β-norhopane (246 μg per g of OC) . . .”

PAGE 14 LINE 17: replace “data” with “emissions factors and compositions”

PAGE 15 LINE 2: grammar: “This study determined . . .identified . . .quantified” or “This study attempted to determine. . .identify...quantify”

PAGE 15 LINE 6: see comment on PAGE 10 LINES 11-12 for similar issue: “passenger diesel cars without particle filters”

PAGE 15, LINE 5: I am not sure what the authors are communicating with this first sentence (“The traffic shows the larger emission factor. . .”). Please rephrase.

PAGE 15 LINE 19: As written, implies that different sites in Europe were sampled in this study. Instead: “Cu/Fe ratios consistent with literature values from other sites suggest similar brake composition for these elements throughout Europe (as long as Cu-free brakes do not increase in use).”

PAGE 15 LINES 21-23: Ambiguous as written as to whether these ratios are good or bad tracers. Instead: “Our measurements do not support the use of Cu/Mn and Cu/Sb as tracers of brake wear emissions possibly due to additional sources of Mn as well as the introduction of Sb-free brake pads.”

PAGE 15 LINE 36: clarity: “agreement between chassis dynamometer and near-road measurements”

PAGE 15 LINE 36: delete “the change of the”

PAGE 16 LINES 5-6: I suggest a more specific concluding sentence. Perhaps replace “delivered valuable information” with “describes exhaust and non-exhaust emissions measurements”

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