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Interactive comment

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

Anonymous Referee #1

Received and published: 3 December 2018

The study by Charron et al. documents measurements of PM10 samples collected at a roadside in France. Overall, the results appear to be of high quality and are of potential use to further source apportionment work. The authors contextualize the study with the increasing importance of non-exhaust PM to total vehicular emissions, which is an interesting and intuitive concept. Justification for sampling PM10 should be included in the paper. What is the typical size distribution for vehicular emissions? Exhaust emissions, especially those treated with a particulate filter onboard the vehicle are dominated by ultrafine particle emissions, which have vanishingly small mass, especially in comparison to the remainder of PM10. Therefore, does PM10 over-sample the resuspended and non-exhaust emissions? Are aerosol size distributions of non-





exhaust emissions known? The size-segregated composition of the PM in this study probably varies strongly, and will have an influence on the findings. Please discuss this aspect of the study in greater detail. Coarse particles also have fast deposition rates, limiting their influence on respiratory exposure compared to PM2.5 (or size classes with smaller upper size limits).

Inclusion of more data visualizations throughout the manuscript, especially related the early sections of results (temporal profiles) is strongly advised. The reviewer's comprehension of the text was heavily improved by opening the SI, which should not be a requirement of reading a paper. This recommendation will likely improve the manuscript by an important margin. Combining metals onto fewer panels may help so that the entirety of SI Section VII does not need to be included as-is (for instance). Please consider this idea for all sections of the manuscript.

The presentation of the manuscript overall could use some English grammar editing and overall proofreading. A number of small grammatical or usage errors exist throughout the document. A few salient ones are pointed out in the minor comments below, but this should not be considered an exhaustive list of corrections.

Overall opinion: This study should be considered for publication after addressing the comments of this review in a major revision.

Introduction: The study is contextualized well, focused on the importance of nonexhaust emissions from vehicles, which may now be the most important aspect of vehicular emissions. Findings were, however, associated with both exhaust and nonexhaust emissions, which is clear.

Methods: Methods were clearly described, with the exception of a lack of definition for TEOM-FDMS (a minor comment).

Results: The use of the term 'incremental' is confusing (occurs throughout 'Results'). Referring to the "increment" with a clearer name would greatly clarify the presentation

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this important quantity.

Section 3.1.1: It would be helpful to see a graphical representation of the contribution of each component of PM10 to the total mass.

Page 6, Line 32: It is becoming clear that the influence of particle size may be evident in the data (also see broader comments above). Please include a description or reference to what is known about the size-resolved composition of vehicle emissions from both exhaust and non-exhaust sources. [Such a discussion is not necessarily relevant to insert at this point in the manuscript, however, its importance began to become clear at this point.]

Page 7, line 38 – Page 7, line 2: Can a consideration for super-emitting vehicles be included in this part of the discussion?

Page 8, lines 20-22: The measured 'dominant regional contribution' to OC may be masking the vehicular primary OC due to differences in the size distribution. Regional, secondary OC may be significantly aged, and therefore larger in size. While this will manifest as an overwhelming signal in a PM10 sample, it may not be so in a PM1 sample, or even a PM0.1 sample. Please discuss the significance of what is known about size-dependent composition of particles associated with vehicle emissions.

Page 8, lines 33-38: If Co does not show any correlation with traffic emissions, how can it be reasonably concluded from the data collected in the present study that Co is a contributor to brake wear? The authors clearly attempt to make a case using the literature. If the contribution of other elements to one another is going to used to explain their similar vehicular sources (Fe, Cu, etc), then the same standard must be held to Co. At best, perhaps there is some other, more consistent, low level source of Co that is flattening the temporal profile. A conclusion as written in this passage, however, is dubious.

Page 9, lines 11-15: Can the impact of variability within the parts of the vehicle fleet

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be incorporated? In such highly controlled emissions from vehicles, a small number of super-emitters may be quite impactful. Page 9, lines 34-35: This is a salient point for air quality management and regulation.

Page 10, line 5 (and other instances): Please give a more succinct definition to the term "smoker vehicles". Perhaps the authors could call these super-emitters? (see also comment about Page 9, lines 11-15)

Page 11, line 22: The authors use and cite a finding that a dominant fraction of brake wear emissions come from the disc and not the brake pad. This seems hard to believe considering the fact that the brake pad is the primarily consumable part, and that brake discs (rotors) do not need to be replaced as often.

Page 11, line 35: Please clarify and/or define "PM fraction". This term is not used routinely in this manuscript. A change in wording may help in this instance.

Page 11, line 34 - Page 12, line 8: It may help to define Cu/Sb in the brake materials themselves. How might consistency in the brake materials themselves drive atmospheric Cu/Sb? Could the act of particle formation (temperature, breaking force, etc) influence the ratio?

Page 15, line 17-19: Has Cu/Fe been reported in any other proportion in the atmosphere? Please illustrate (perhaps in the results section on this topic) that the \sim 4% value is unique to vehicles.

Page 15, line 24-28: This is a strange placement for an overview paragraph about the significance of redox-active metals, which have only been mentioned as such in the introduction. This paragraph is probably better off at the beginning of the conclusions section, mirroring the structure of the paper itself.

Page 15, line 29: While studies such as this one may be scarce in the literature, the authors have reported agreement with these studies throughout the manuscript – suggesting that the science is highly convergent. Please provide a clear, summary as-

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sessment of the novel findings of this study.

Page 16, lines 5-6: This concluding statement seems to highlight the fact that this is a characterization study with little in the way of entirely new findings. (see previous comment) Do the authors believe feel that this is the case? If not, a revised summary statement or declaration of a way forward in light of the present study is in order.

Minor comments: Line 20-21: "Most of the first ones" – please be specific, most of the first 'what'? What do you mean by first? I honestly do not know to which prior items this sentence refers.

Line 24-26: "On the contrary,..." to what??

Page 8, line 9: should refer to "SI Section VII"

Page 9, line 27: change "technics" to "techniques"

Page 10, line 2: define DPF, first usage

Page 10, line 26: change to "third highest"

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