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Interactive comment on "Morphology and size of the particles emitted from a GDI-engine vehicle and their ageing in an environmental chamber" by Jiaoping Xing et al.

Anonymous Referee #2

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The paper discusses the composition, size and mixing of single particles emitted by a gasoline direct injection (GDI) vehicle as determined using single particle electron microscopy and spectroscopy. I found the topic very interesting and important. I think the overall data and analysis is of interest and sufficient to warrant publication; however, I believe the paper could improve substantially with some changes and maybe a few relatively minor additional analyses that could be performed with the data available. Before the paper is published, the authors need to address and discuss several items as mentioned in the following general and specific comments.

General comments: - If I understood correctly the conclusions of this study are based

C1

on only one vehicle. While I totally understand that single particle analyses are very time consuming and analyzing emissions from several engines would be prohibitive, it is well known that there is tremendous vehicle to vehicle emission variability, even for the same model and engine (several works published in the literature are available on the topic). So my request here is not that to add data, which would be well beyond the scope of the study, but to clearly mention this caveat and limitation and discuss briefly its potential implications. Related to that, is the vehicle representative of the average GDI vehicles in Beijing? If so how? If not, how can the conclusions of the paper be generalized as the authors attempt to claim? (E.g., In the method section, they mention that the vehicle represents a "leading-edge design", does that means that most of the other GDI engine would perform worse than that in terms of emissions?)

- The paper is quite biased and limited in terms of citations of existing literature (some example will be discussed in the specific comments but more pervade the paper) and the paper would be much more impactful if put in perspective of a large body of existing literature for example on PM vehicle emission (not only in China but also in other countries), single particle analysis, particle optical properties measurements, and effects of single particle mixing geometries on calculated or measured optical properties and radiative effects, etc.
- Overall the paper is quite clearly written, but some additional English grammar checks would improve readability; this should include reducing typos and checks for tense consistency.
- The atmospheric implications section is too generic and not always substantiated by the results or the provided citations (more on this in the specific comments below). This section should be made more concrete and provide a deeper and more significant discussion.

Specific comments: - Title: It is a matter of personal taste, but I typically prefer not to use acronyms in the title, so the authors could consider spelling out GDI to target a

wider audience.

- Highlights: "Particles from a GDI-engine vehicle and their ageing were studied." To me this is not a highlight, it is just the topic of the paper.
- Line 38: "... must be paid enough attention." Something seems missing here and the sentence reads awkward. Consider rephrasing it.
- Line 45: "grain size" does not seem to be a very common term in the community... maybe "particle size" or just "size"?
- Lines 65-66: circular sentence, as it is now it reads as if particulate matter PM is a source of airborne particles. The authors instead mean vehicles are a source of PM, I think. Consider rephrasing.
- Line 81: change "concerned" with "concerning"
- Line 82: "compressed" maybe should be "reduced"?
- Line 130: "could provide" should be "provide" or "provided"
- Line 153: This is mostly a curiosity for me, but it seems like 6% of sulfur in the fuel is quite high. Is that the norm? Are those fractions by weight or by mass?
- Line 159: A max speed of 50 kmh-1 seems a bit low, even if that might be the speed limit in the city, do real vehicle actually respect that limit? Can the author discuss this point?
- Line 178: Please provide the manufacturer and model of the impactor.
- Line 215: Please provide more information on the image analysis procedure, including, if available, citations to existing literature, software or methods.
- Figure 2 reports the number size distribution of the particles emitted. I believe that's from the electron microscopy, that should be made clear. Additionally, what engine state is the distribution representing, or is it a composite of all the particles collected,

С3

maybe this was mentioned somewhere, but I could not find that information. Related to it, on line 230 the authors mentioned a range of 60 to 2500 nm, but earlier on, when describing the impactor, they mention a lower 50% size-cut of 250 nm, so the 250 to 60 nm contribution is probably severely underestimated, therefore the detailed of the first mode (140-240 nm) is probably severely biased by the sampling. Related to this, I think there would be a lot of benefit to either add a plot in figure 2 or present a separate plot with the size distribution that should be available from the DMS500 instrument mentioned in line 172.

- Line 260: It is interesting that the particle concentration was higher for hot conditions that cold conditions. This sparks the question though if the size distribution of the particles also changed and with it the mass emission... again using the DMS500 data should easily answer that question at least for the ensemble particle size distributions.
- Line 272: "of" missing after between "type" and "particles"
- The sentence from line 288 to 290. This is an example (there are other instances throughout the paper) of verb tense inconsistency.
- Line 299: It would be nice to know if there is a semi-quantitative assessment of what the coating material was for the most part. Sulfates? Organics? Others...
- Line 300: "...organic particles changed..." what kind of changes, please elaborate?
- Line 314: "...the majority..." in number, but is that the case in mass as well, please discuss.
- Lines 350-351: It would be useful to provide information on what is the most common coating material and, if possible, provide an estimate of the core to shell ratio. That ratio can be key to optical calculations to understand the impact of absorption increase due to coating and mentioned later in the paper. A quantitative determination of the core to shell ratio for soot, that might be possible to be determined from the data available to the authors, could be very useful to the community and make the paper substantially

more impactful.

- Line 356: How high were the gas concentrations? Were they representative of realistic atmospheric conditions? If not, could one extrapolate on what would be an equivalent aging time in more realistic atmospheric concentrations? Some comments on this issue are needed.
- Line 360: the sentence sparks the question, was there any contribution from break and tire tear emissions? Were those captured? Probably not because, if I recall correctly, these samples were collected on an engine dynamometer, but it might be good to comment (one can use several results from previous studies available in the literature) on potential contributions of tailpipe emissions vs. break and tires emissions.
- Line 375: The acid-catalyzed mechanism is mentioned here and even mentioned as one of the highlights of the paper, but the discussion is minimal, or inexistent here. This can be an interesting and important point, so please provide some elaboration of this topic and provide some unbiased citations of relevant literature on the topic.
- Line 380: The large contribution of GDI emissions here is argued, but there is no data collected or model performed (even if just conceptual) to really quantify, at least semi-quantitatively, what the contribution could be with respect to other sources. The only discussion I recall is on the elemental composition regarding tracers to distinguish different sources, but no discussion to quantify their potential contribution (also in what terms? Mass? Number? Something else?). So the "considerable potential contribution" statement is a vague ill-posed guess that is not really proven here as the data are presented. One could try to estimate the contribution by calculating for example estimated total contribution of GDI engines (by multiplying the fuel-based emission factor by the fraction of fuel consumed by GDI vehicles in the area) vs. the total particle burden in the region... or some other estimate exercise of this sort. Otherwise the sentence cannot be supported by the evidence provided.
- Lines 386-388: This is a clear example, among others, of biased representation (or

C5

lack of) previous work in this paper. There have been very many previous (definitely previous to 2017) studies showing that individual particles, including organics come internally mixed with other particles, so this statement is not very true (especially in terms of "recent") and biased in terms of literature discussion and citations.

- Lines 391-393: Again limited (poor) literature citation choice and a bit of simplistic view of the effect of internal mixing on aerosol radiation interaction (which I think is what the authors refer to here). Please improve the discussion and support it with some more reprehensive and balanced citations judiciously chosen from the large body of previous literature in the field. Another such example appears in lines 399 to 400.
- Lines 403 to 404: In this case, I would say the sentence is flatly untrue; there are plenty of studies on the optical properties of POA, and the authors should discuss them and cite them accordingly.
- Figure 2: As mentioned earlier, specify that this distribution is from the electron microscopy analysis, and overlap or add a side plot with the size distribution from the DMS500.
- Figure 4: For regulatory applications, it could be interesting to generate a similar figure but for estimated mass (or at least volume) fractions.

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