

Response to Referee' Comments on Manuscript: acp-2016-1038

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Title: Measurement of PM and its chemical composition in real-world emissions from non-road and on-road diesel vehicles

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Referee #1

General comments

The manuscript by Cui et al. summarizes emissions measurements from multiple generation diesel excavators and trucks under different operating and driving conditions. These types of measurements are unique in China and much needed. The paper is well organized, but it needs a thorough edit as many words, verbs, etc are not used correctly or are missing. Below I highlight the technical weaknesses, minor clarifications, and instances where sentences are confusing and need to be rephrased. I approve publishing the paper after these concerns are addressed.

Response: Thanks for the reviewer's positive approval. Clarifications have been provided and confusing sentences were rephrased in the revised manuscript.

Comments #1: (1) One of the weaknesses of this work is that each truck/excavator was tested only once. Thus it's unknown how representative these results are and how variability in the measurements affect the observed emission factors. I doubt that duplicate runs can now be carried out; however, the authors should at least mention and address this weakness. (2) Another weakness is that driving conditions of the trucks were not similar (as shown in Figure S2); since driving conditions and engine load can have significant impacts on the emission factors, how can the results be interpreted in a unified manner? This should also be addressed in the discussion and conclusion sections. (3) Related to this is the variety of the engines tested in this work for both excavators and trucks. For example for excavators, engine powers span a range of 35-169 KW and total weights and engine displacements also vary a lot. On one hand, it's good to have a sampling pool of various engine types/sizes. On the

other hand, these difference should be kept in mind and referred to when comparisons are made throughout the paper.

Response: Thanks for the reviewer's constructive suggestions. This major question was divided into three questions and we would provide a personal response to your comments, separately.

(1) We appreciate the review's comment. Indeed, we are also attached importance to the weaknesses of tested time in this study. However, given the difficulties of field measurements and some important parameters missing in the links of repeat tests, only one relative complete test was chosen for further discussion. In order to evaluate the variability, we had conducted some repeats for individual vehicles, and the results were presented in Tables S3 and S4 in the revised supporting information. As shown in Tables S3 and S4, the variability in test times for the same operational mode was considered acceptable. Moreover, we combined some repeat tests for organic matter analysis for T1 and T2, which could reduce the uncertainty. We confirmed that the weaknesses of repeatability existed in this study, and mentioned this weakness in the revised manuscript (**Page 7 line 19-26**).

Table S3 Pollutants mass concentrations emitted from E4 in three idling repeat tests

	O ₂ ^a (%)	CO ₂ ^a (%)	CO ^a (ppm)	NOx ^a (ppm)	PM (mg m ⁻³)	OC (mg m ⁻³)	EC (mg m ⁻³)
1	16.2	3.4	309	453	11.9	4.3	1.9
2	16.3	3.4	257	457	14.6	6.1	2.9
3	16.3	3.4	262	445	14.4	6.8	2.5
SD	0.08	0.01	28.6	5.68	1.55	1.26	0.53

a: the datum were presented on other unpublished research

Table S4 PM mass concentrations emitted from trucks in some repeat tests (mg m⁻³)

Trucks	Roads	1	2	3	SD
Light duty-China III	non-highway 1	15.0	16.2	/	0.87
	highway 1	19.8	30.6	/	7.67
Heavy duty-China II	non-highway 2	21.3	16.1	/	3.68
	non-highway 3	7.87	6.11	6.69	0.89
Medium duty-China III	non-highway 4	11.0	10.3	/	0.49
	highway 2	8.79	17.1	/	5.85
Heavy duty-China III	non-highway 4	5.29	9.56	6.99	2.15
	highway 2	10.6	7.42	/	2.24

(2) Thanks. As mentioned in the revised manuscript (**Page 7 line 8-10**), different emission standards diesel trucks must run on different roads, which was restricted by traffic rules. For example, “yellow label car” can only run on the particular road and is not allowed running on the highway and arterial road. Therefore, different routes were chosen for different trucks. Although driving conditions of the trucks were not similar shown in Figure S2, the different characteristics of velocity on the highway and non-highway were obviously. Therefore, we just discussed highway and non-highway routes in this study. We have addressed this weakness and interpreted the unified manner in the revised manuscript(**Page 7 line 11-12**).

(3) Thanks for the comment. As we could seen from Figure S5 in the revised supporting information, the average EF_{PM} was less affected by engine power. It was regretful that the sample size in this study seemed not enough to reflect the impact from engine power. Thus, we just gave EF_{PM} of different engine power in the revised manuscript, and didn’t discuss in-depth (**Page 11 line 27-29**).

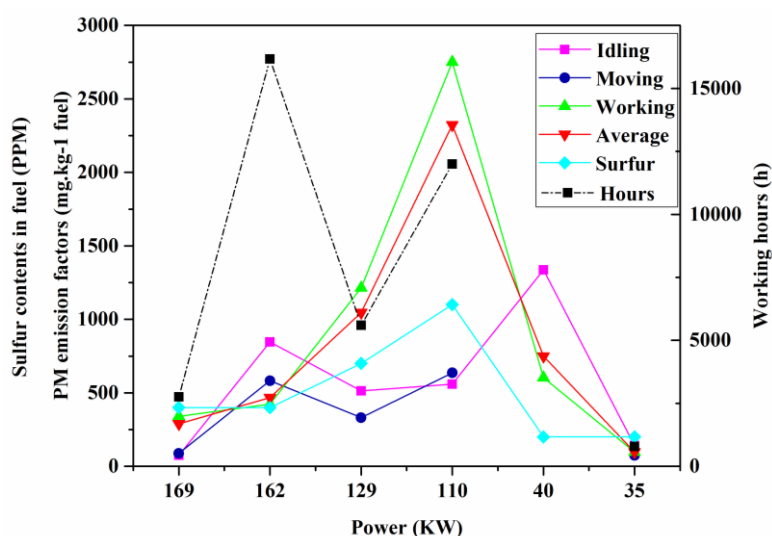


Figure S5 PM emission factors for different power excavators

Comments #2: For readers who are not familiar with the standards in China, it will be useful to have a table where major particulate and gaseous emissions of each generation standard for trucks/excavators are listed.

Response: Thanks for the suggestion. The major particulate and gaseous emissions of each generation standard for trucks/excavators were listed in Tables S1 and S2 (Supporting information).

Comments #3: P7, L23: Although mentioned in Table 2, please indicate in the text the average (or range of) sulfur content of the fuels as well as the limit of GB 252-2015.

Response: Thanks. The range of sulfur content and limit of GB 252-2015 have been added in the revised manuscript (Page 8 line 19-20).

Comments #4: P8, L22: what recovery % for each species were achieved?

Response: Thanks for the comment. The recoveries of five surrogates have been added in the revised manuscript (Page 9 line 21).

Comments #5: P12, L3: It seems the trucks with China II and China III standards had similar PM emission factors. Why is that so? Do these standards pose similar levels for PM? or is it that the trucks tested don't necessarily represent the standard? or is this an instance where results from a single measurement from a truck are uncertain?

Response: We appreciate the review's comment. As we discussed in the manuscript, the most important reason causing this result was different driving conditions for those two trucks. Due to heavy pollutions from China II trucks, traffic laws regulate that China II trucks are forbidden to drive on city center and only allowed to drive on some remote parts of the city, while the roads for China III trucks are always jammed. For evaluating the emission from trucks in the real world, we shouldn't neglect the driving conditions to discuss trucks itself. However, we confirmed that the number of measurement was shortage in this study, and we will lucubrate in the future.

Comments #6:P12, L7: unclear what "more volatile" means here

Response: Thanks for the comment. "more volatile" refers to highly varied speed (Page 13 line 11).

Comments #7:P12, L11-13: It doesn't make sense that trucks driven on road with higher grade have lower emissions. Please clarify.

Response: Thanks. There was wrong with expression and we have modified in the revised manuscript (Page 13 line 16).

Comments #8:P. 12, L17: what's the justification for using OM/OC=1.6 for such fresh emissions? How will the result change if a lower factor, more representative of fresh emissions, is used?

Response: Thanks for the reviewer's constructive suggestions. Chow et al (2015) showed that a conversion factor used to transform OC to OM was ranged from 1.2 to 2.6, depending on the extend of OM oxidation. Fresh aerosols from different sources had different values, such as 1.4 and 1.6 for diesel engine (Gilardoni et al., 2007, Japar et al., 1984) and 1.7 for biomass burning (Chow et al., 2015). Therefore, we assumed the conversion factor is 1.6 in this study.

Comments #9:P13, L9, P14,L2: it is mentioned that diesel sulfur content affected OC/EC. It is unclear to me how fuel sulfur can affect emission of organic compounds and soot. Please explain.

Response: We appreciate the review's comment. According to references, we assumed that the formation of organic compounds and soot was obviously affected by diesel sulfur content in two points. On the one hand, organosulfurs constituted up to

62% of the total sulfur content in diesel (Adlakha et al., 2016). Organic compounds existing in diesel were removed simultaneously by process of desulfurization. Therefore, emissions of organic compounds and soot generated by hydrogen abstraction/acetylene addition were reduced (Sánchez et al., 2013). On the other hand, sulfuric acid, the nucleating agent in diesel particle formation, generated by sulfur in diesel (Ruiz et al., 2015). These nucleating agents might provide a place for organic compounds condensation and reaction.

Comments #10:P13, L11-26: It is unclear what the elemental emissions are stemming from: the fuel or bad conditions of the engine or the lubricating oil? Please explain. For example, L22, it is mentioned that diesel quality used in E4 was poor. Was the fuel also tested for elemental content? Were Cu and Zn higher in this fuel as well?

Response: Thanks for the comment. Although diesel quality was analyzed in this study, many elemental contents were below the method detection limit. Wang et al. (2003) reported that the concentrations of Fe, Ca and Mg accounted for 50% of the total elements in diesel fuel. Thus, the possible source of elements was diesel, while Cu and Zn were affected by sampling environments for E4. The detail information could be seen in the revised manuscript (**Page 14 line 23-30**).

Comments#11:P13, L23, P14, L4-5, P17, L24-26: Authors mention that % of elemental composition in E1 and E6 was higher. How did absolute concentrations or emission factors of the elements compared for these two vs. the others? Since % values depend on concentrations of other components as well, I don't think they're as relevant to be mentioned, especially since they contribute to a very small fraction of the emissions.

Response: Thanks for the comment. The average emission factors of elemental were $5.66 \text{ mg}\cdot\text{kg}^{-1}$ for E1+E6 and $4.02 \text{ mg}\cdot\text{kg}^{-1}$ for E2+E3+E4+E5, and were mentioned in the revised manuscript (**Page 15 line 2-3**).

Comments#12:P14, L7-10: It is unclear how the authors concluded that alkane/hopane/steranes were influenced by fuel quality and PAHs by combustion. Please explain and clarify.

Response: Thanks for the comment. N-alkanes, hopanes and steranes fractions were the highest in excavator E4, while PAHs fraction was the highest in excavator E3.

Comparing the fuel quality between E3 and E4, E4 had a poorer diesel quality, which might be the main reason for high n-alkane, hopanes and steranes. Similarly, it was said by Rogge et al. (1993) that n-alkanes, hopanes and steranes were mostly derived from incomplete combustion of fuel and lubricant oil. However, we speculated that PAHs was affected by combustion conditions (e.g. combustion temperature) in this study, due to E3's better performance (stage 2) and relatively superior fuel quality. The distinct explanation was added in the revised manuscript (**Page 15 line 12-21**).

Comments#13: P16, L11: Please explain what reactions in the engine authors refer to.

Response: Thanks for the comment. The description of reactions was provided in the revised manuscript (**Page 17 line 19-20**).

Comments#14: P17, L3: Is it really that presence of metals oxidizes soot?! or do the metals enhance combustion and reduce formation of soot?

Response: We appreciate the review's comment. It was said by Kasper et al., (1999) that the action of iron oxide was recognized as a catalyst and burnout rate of soot could promote during combustion process. Therefore, we inferred that metals may enhance combustion of soot. The corresponding expression was added in the revised manuscript (**Page 18 line 10-12**).

Comments#15: Acronyms of PAHs should not be used in the abstract.

Response: Thanks, the acronyms of PAHs have been changed to full names (**Page 1 line 24; Page 2 line 25-27**).

Comments#16: Define BaP_{eq} in the abstract 3.

Response: Thanks for the comment. The BaP_{eq} has been defined in the revised abstract (**Page 3 line 1**).

Comments#17: P3, L 7: define PM. Throughout the paper indicate what size PM refers to (PM₁, PM_{2.5}, etc).

Response: Thanks for the comment. PM referred to total suspended particulate (D_p≤100 μm) in this study. We have remarked in the revised manuscript (**Page 3 line 12**).

Comments#18: P12, L12: consider using "higher road grade".

Response: Thanks. We agree with the reviewer's suggestion and changed the word as suggested (**Page 13 line 16**).

Comments#19: P20, L3: Do authors mean excavators rather than diesel truck here or should E1, E2,... be T1, T2, etc?

Response: Thanks. The E1,E2... have changed to T1,T2....(**Page 21 line 15-16**)

Comments#20: Figures: Axis labels are all too small and need to be modified for better quality figures.

Response: Thanks for the advice. Axis labels in Figure 1, 3, 4, 5 and 7 were modified in the revised manuscript (**Page 35; Page 37; Page 38; Page 39; Page 41**).

Comments#21:Fig 7: what do the errors bars represent? Unclear from the caption what the difference between A-B and C-D symbols are.

Response: Thanks for the comment. A and B are isomer ratios of PAHs for excavators and trucks tested in this study, respectively; C and D are average isomer ratios of PAHs for trucks and excavators tested in this study. The vertical and horizontal errors bars represent the standard deviation of values shown in vertical and horizontal axis, respectively (**Page 41**).

Comments#22:Fig. S3. What are the crosses and dashed lines in these box and whisker plots?

Response: Thanks for the comment. The annotations were shown in the revised Figure S4 (**Supporting information**).

Comments#23:Sentences needing to be rephrased: 1. P3, L 13-15 2. P4, L18-20 3. P7, L12-14 4. P12, L1-3 5. P. 13, L19-20 6. P.18, L3-4.

Response: Thanks for the comment. We have made every effort to polish our English and asked a native English speaker to take a proof reading of the revised manuscript.

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