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Interactive comment on "Measurement on PM and its chemical compositions for real-world emissions from non-road and on-road diesel vehicles" *by* Min Cui et al.

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Response to Referee' Comments on Manuscript: acp-2016-1038 Manuscript Number: acp-2016-1038 Title: Measurement of PM and its chemical composition in real-world emissions from non-road and on-road diesel vehicles Authors: Min Cui, Yingjun Chen, Yanli Feng, Cheng Li, Junyu Zheng, Chongguo Tian, Caiqing Yan, Mei Zheng Corresponding authors: Yingjun Chen, Yanli Feng, Junyu Zheng

Referee #2: General comments Cui et al. present data from measurements of particulate matter emissions and composition from real-world testing of a suite of on- and non-road diesel vehicles. They find that PM emissions, while variable, exhibit trends with fuel quality and emissions standard. Although these data add to the literature and

will eventually help build more realistic emissions inventories for China, I do not recommend publication of this version of the manuscript in ACP. I have two major comments and numerous minor comments.

Response: Thanks very much for the comments. We have revised this manuscript carefully, and please find our detailed responses below.

Major comments: Fit: The manuscript, in my opinion, does not fit the research foci of publications typically accepted in ACP and I wonder if another journal would offer a better fit for this research.

Response: Thanks. But the authors disagree with this comment and consider ACP as the best journal of high quality to publish our precise measurement data. On the one hand, in recent years, PM emission from diesel vehicles drawn more and more attention in China, due to severe air pollution. However, the great uncertainty existing in PM from diesel vehicles exhausts makes those field datum very precious. Although our research is preliminary, as far as we know, this manuscript is the first on-board research in China that focused on PM chemical constituents from on-road and non-road diesel vehicles exhaust. The results of this study could provide basic data for air quality assessment and establishment of emission standard. Therefore, we chose ACP, one of the most influential journals in atmospheric fields, to publish our results for obtaining broader attention. On the other hand, the main subject areas for ACP comprise atmosphere modeling, field measurements, remote sensing, and laboratory studies of gases, aerosols, etc. Nowadays, several researches about emission factors and characteristics of PM from diesel engine have been published in ACP (Dai et al., 2015, Dallmann et al., 2014, Lin et al., 2015, Zhang et al., 2015). Therefore, this manuscript is fit to publish in ACP, because of general implications for source apportionment and health assessment.

Comments #1:No new methods/instruments were used that make the data novel.

Response: Thanks. We have added some descriptions about the progressiveness of

methods/instruments used which made the data novel in the present study in the revised manuscript (Page 8 line 26-27; Page 9 line 4-9). Briefly, the portable on-board emission measurement and dilute sampling system which was designed and manufactured in our laboratory has good performance (Zhang et al., 2015), and has obvious advancement compared with other on-board instrument for vehicles such as PEMS and FPS4000 (Zheng et al., 2015) by the portability and capability of filter sample collection for further PM chemical analysis in the laboratory. Furthermore, the present result was the first set data of on-board measurement for non-road diesel vehicle exhaust in China.

Comments #2:The measurements were performed on a very small cross-section and are not necessarily representative of the on- and off-road fleet in China. The small sample size, small cross-section, and large variability do not suggest large shifts/trends in emissions (or at least make them hard to observe).

Response: Thanks. We admitted that the sample size in this study was small, but wide ranges of vehicle types (including different emission standards and engine powers) were considered in this study. Furthermore, the most important purpose in this manuscript was to analyze the chemical constituents of PM from diesel vehicles exhausts, which needed a heavy workload (Page 7 line 3-5). Actually, we had selectively conducted some repeated experiments in this study to evaluate those variability and the results were shown in Tables S3 and S4 (Supporting information). As shown in the Tables S3 and S4, the variability was considered acceptable. Because there were some parameters missing in the field measurement, we decided to select an completed test for calculating the emission factors and combine the repeated filters to reduce this uncertain for some diesel vehicles. In the future work, we would increase the sample size to ensure the datum stability after this first attempt (Page 7 line 18-25).

Comments #3:Comparisons with literature data are not very insightful. While the data add to the literature in terms of quantifying emission factors of PM from a modern set of vehicles under real-world conditions, the scientific contributions in this research effort

are lean. The data need to be published but this journal may not be the right target.

Response: Thanks. The purpose and the greatest contribution of this study were established characteristics of PM and its constituents emitted from trucks and excavators using on-board measurements. In China, diesel vehicles are facing imperfect emission standards and messy diesel quality, especially for non-road diesel vehicles. The knowledge relative to the characteristics of PM emission from those diesel vehicles was slim to none. It was extremely difficult to collect literature data and compare with results obtained in this study, due to lacking of researches for characteristics of PM and its constituents by on-board tests. Following the reviewer's suggestion, we have made more interpretation among the comparison in the revised manuscript (Page 14 line 23-30; Page 15 line 12-21; Page 19 line 30; Page20 line 1-9). Finally, we chose ACP to publish our results for obtained broader attention from the perspective of the importance of the datum.

Comments #4:Writing: The quality of technical communication is very poor. This suggests one or all of the following: (a) the first author was rushed to write and submit this manuscript, (b) the senior authors have not read through this manuscript, (c) the authors place no emphasis on clear and effective communication. The manuscript needs to be significantly improved by the senior authors to meet the expectations of an English language publication in a high impact journal. If the manuscript is not heavily edited for English, this would be reason enough for rejecting the manuscript from publication. Here are a few examples from just the first few pages: a. Page 1, line 24: 'involving wide-range emission standards' b. Page 2, line 11: 'PM compositions emitted from excavators dominated' c. Page 2, line 23: 'the complex of operating modes' d. Page 3, line 7: 'diesel vehicles exhaust is a major source of emissions in ambient PM' e. Page 3, line 9: '30% of emissions in ambient PM' f. Page 3, line 18: 'causing severe emission situation' g. Page 3, line 23: 'almost higher than 90% of PM came from on-road diesel vehicles emission' h. Page 3, line 27: '349 thousand tons PM emission' i. Page 5, line 23: 'organic matters'? j. Page 5, line 26: 'impact factors of PM'; what does

that mean?

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 final version of the revised manuscript. a 'Involving wide-range emission standards' was changed to 'involving a range of emission standards. b 'PM compositions emitted from excavators dominated' was changed to 'PM composition emitted from excavators was dominated'. c 'The complex of operating modes' was changed to 'the complex characteristics of excavator operational modes'. d 'Diesel vehicles exhaust is a major source of emissions in ambient PM' was changed to 'Diesel vehicles exhaust is a major source of ambient PM emissions'. e '30% of emissions in ambient PM' was changed to '30% of ambient PM emissions'. f 'Causing severe emission situation' was changed to 'and have contributed to severe emissions problems'. g 'Almost higher than 90% of PM came from on-road diesel vehicles emission' was changed to 'more than 90% of PM resulted from on-road diesel vehicle emissions'. h '349 thousand tons PM emission' was changed to '349 Gg of PM emissions'. i 'Organic matters' was changed to 'organic compounds'. j 'Impact factors of PM' was changed to 'influential factors of PM'.

Minor Comments:

Comments #1: Emissions standards: It might be worthwhile to describe the on-road and off-road emissions standards (e.g., Stages and China) and their emissions limits for PM (and other pollutants too) at the beginning of the manuscript through a Table. This would help orient the reader and also allow easy comparison with the EPA and EURO standards.

Response: Thanks. We have added the on-road and off-road emission standards in the revised manuscript (Supporting information).

Comments #2: Page 2, line 9: Did vehicle exhaust contribute to 30% of the PM concentrations or emissions? Unclear; please clarify.

Response: Thanks. We have modified the unclear place in the revised manuscript (Page 3 line 9-10).

Comments #3: Page 4, line 3: construction equipment might be better word

Response: Thanks for the advice. Following the reviewer's suggestion, we have changed the expression in revised manuscript (Page 4 line 12).

Comments #4: Page 3, line 16 to page 4, line 5: It might be better if the number of vehicles, fuel consumption and PM emissions in China were represented through a table or figure, alongside the relative importance of trucks and excavators to justify the use of those vehicle types in this research.

Response: Thanks for the advice. The figure S1 was added in the revised supporting information (Supporting information).

Comments #5: Page 4, line 18 to page 5, 10: The authors have only cited other people's work but have not paraphrased their findings. Hence, it is unclear what the gaps and motivation for this work is.

Response: Thanks. We have rephrased the correspond contents in revised manuscript (Page 5 line 7-25).

Comments #6: Page 6, line 19: I did not understand how the duration of the different modes were determined. Also, what torque-speed ratings do the idling, moving, and working mode correspond to?

Response: Thanks. The time of sampling under different modes was not strictly required, as long as assured enough contents of PM to conduct chemical analysis. We have clarified it in the revised manuscript (Page 7 line 1-5). Actually, the basis of selecting those modes were not according to torque-speed ratings. The idling mode refers to engine keeps running at low speed (about 600-800 rpm), but not moving or working. The moving mode refers to that excavator moves at low speed (below 3-5 kmÂůh-1) , but the bucket is not unload. The working mode refers to that bucket scoops the soil,

then moves to another location and scoops again.

Comments #7: Page 7, line 28: Why did the researchers use quartz-fiber filters? My understanding is that the fibers can tear off during handling and bias the gravimetric measurement. Do the authors mean Teflon-coated quartz fiber filters?

Response: Thanks. We used quartz-fiber filters for gravimetric measurements in this study. The quartz-fiber filters losses could be neglected. Because the filters were parceled by aluminum foil after sampling to avoid filters tearing off, and the PM weight of error in quartz-fiber and Teflon filters could acceptance. In adition, quartz-fiber filters were selected to measure PM weight for consistent with those used in the chemical analysis. We have added the reasons in the revised manuscript (Page 8 line 25-28).

Comments #8: Section 2.4.3: The BaPeq method needs to be discussed in detail for the reader to follow the calculation.

Response: Thanks. The detailed BaPeq method was added in the revised manuscript (Page 11 line 8-16).

Comments #9: Section 3.1: What fraction of the improvement between pre-stage 1 and stage 2 can be attributed to better quality fuel as opposed to the emission standard?

Response: Thanks. We supposed that the fuel quality rather than the emission standards has a more great impact on PM constituents. Although the threshold (total emission) was set in non-road emission standards, constitutes of PM haven't regulated in these standards. Furthermore, it was said that sulfur in fuel translates to sulfuric acid which is the nucleating agent in diesel nanoparticle formation (Ruiz et al., 2015). After sulfuric acid nucleation particles formation, the organic compounds (volatile and low volatile) condense on it. Similarity, the soot was also influenced by this nucleating agent (Schneider et al., 2005). Considering the limit of sample size of our study, it was difficult to calculate the influence of the fuel quality and the emission standards on PM constituents separately. In our future study, we will continue to focus on this complex

issue.

Comments #10: Section 3.2: Given that there was only one China IV truck, how confident are the authors in their assessment that China IV trucks are better compared to the China III trucks. Similarly, is the China II truck any different than the China III trucks. Can the authors comment on how the small sample size could affect their conclusion?

Response: We appreciated this question. Actually, China IV truck is extremely rare, because few trucks could reach this emission standards in China. Therefore, we just found only one truck of China IV to conduct experimental. Furthermore, through comparing our results with references and assessing repeatability in the test results, we considered that our conclusions were credible. The detail explanations were added in the revised manuscript (Page 7 line 22-26; Page 16 line 20-21).

Comments #11: Section 3.3: Is the lack of a mass closure on the PM filter a result of using a quartz-fiber filter for gravimetric analysis?

Response: Thanks. We have replied in the comment 7, using quartz-fiber filter was not the main reason caused poor mass closure. The main reasons might be distribution error from OC and EC, water effect and metal oxidation. As mentioned in the revised manuscript, the distribution error from OC and EC by using IMPROVE could highly affect the results of mass closure (Page 16 line 17-19). As shown in Table R1, emission factors of OC was lower than those of n-alkanes for T3, which indicated that the OC content was underestimated. For example, emission factors of OC increased to 85.0 mgÂůkg-1 fuel, the mass closure would almost increase by 10%, correspondingly. For T2, the thick moisture was trapped in the filter, which could increase PM weighing error.

Comments #12: Pry, Fluo etc.: Repeatedly, the authors have used abbreviated names to refer to various PM species. Using the full name of the species might improve readability.

Response: Thanks for the comment. The full name of the individual PAH was displayed

in the revised manuscript (Page 9 line 26-30). But, considering the concise expression, we also used abbreviated names in the part of discussion.

Comments #13: Sections 3.3, 3.4 and 3.5: The authors have compared the PM composition data amongst the excavators and trucks and to literature data. However, it was hard for me to glean anything meaningful from all those comparisons and the ensuing discussion. I recommend that the authors spend some more time trying to make the interpretation more palatable to the reader.

Response: We appreciate the review's comment. We also want to do it, but the maneuverability was poor. It is extremely difficult to collect literature data and compare with results obtained in this study, due to lacking of researches for characteristics of PM and its constituents by on-board tests, especially for non-road diesel engine. Based on our purpose in this manuscript, we presented three parts for further discussion. In section 3.3, we tried to interpret difference in characteristics of PM emission between individual diesel vehicles tested in this study. In section 3.4, we tried to combine our results with those from other references to find some consensus. In section 3.5, through comparing the differences in characteristic of PM emission between excavators and trucks, we emphasized the PM emission difference of two types of vehicles. Following the reviewer's suggestion, we have made more interpretation between the comparison in the revised manuscript (Page 14 line 23-30; Page 15 line 12-21; Page 19 line 30; Page 20 line 1-9).

Comments #14: Page 18, line 26 to page 19, line 2: The health relevant calculations, comparisons, and following discussion were too hard to follow and seemed like they were added to the manuscript as an afterthought.

Response: Thanks. The carcinogenic risks of PAHs emitted from trucks and excavators were the important indicators to evaluate emission situation for those two diesel vehicles. We have enhanced the expression in the revised manuscript (Page 11 line 8-16; Page 19 line 30; Page20 line 1-9).

References: Dai, S., Bi, X., Chan, L.Y., He, J., Wang, B., Wang, X., Peng, P., Sheng, G., Fu, J.: Chemical and stable carbon isotopic composition of PM2.5 from on-road vehicle emissions in the PRD region and implications for vehicle emission control policy, Atmospheric Chemistry and Physics. 15(6): 3097-3108 2015 Dallmann, T.R., Onasch, T.B., Kirchstetter, T.W., Worton, D.R., Fortner, E.C., Herndon, S.C., Wood, E.C., Franklin, J.P., Worsnop, D.R., Goldstein, A.H., Harley, R.A.: Characterization of particulate matter emissions from on-road gasoline and diesel vehicles using a soot particle aerosol mass spectrometer, Atmospheric Chemistry and Physics. 14(14): 7585-7599 2014 Lin, Y.C., Tsai, C.J., Wu, Y.C., Zhang, R., Chi, K.H., Huang, Y.T., Lin, S.H., Hsu, S.C.: Characteristics of trace metals in traffic-derived particles in Hsuehshan Tunnel, Taiwan: size distribution, potential source, and fingerprinting metal ratio, Atmospheric Chemistry and Physics. 15(8): 4117-4130 2015 Ruiz, F.A., Cadrazco, M., López, A.F., Sanchez-Valdepeñas, J., Agudelo, J.R.: Impact of dual-fuel combustion with n-butanol or hydrous ethanol on the oxidation reactivity and nanostructure of diesel particulate matter, Fuel. 161: 18-25 2015 Schneider, J., Hock, N., Weimer, S., Borrmann, S., Kirchner, U., Vogt, R., Scheer, V.: Nucleation Particles in Diesel Exhaust: Composition Inferred from In Situ Mass Spectrometric Analysis, Environmental Science & Technology. 39(16): 6153-6161 2005 Zhang, F., Chen, Y., Tian, C., Li, J., Zhang, G., Matthias, V.: Emissions factors for gaseous and particulate pollutants from offshore diesel engine vessels in China, Atmos. Chem. Phys. Discuss., 15(17): 23507-23541 2015 Zheng, X., Wu, Y., Jiang, J.K., Zhang, S.J., Liu, H., Song, S.J., Li, Z.H., Fan, X.X., Fu, L.X., Hao, J.M.: Characteristics of On-road Diesel Vehicles: Black Carbon Emissions in Chinese Cities Based on Portable Emissions Measurement, Environmental Science & Technology. 49(22): 13492-13500 2015

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-1038/acp-2016-1038-AC2-supplement.zip

Table R1 Mass closure on the PM filter for trucks

Species	Units	T1	T2	T3	T4	T5
OC	mg/kg fuel	22.4	32.7	0.64	153	10.3
EC	mg/kg fuel	337	3.61	200	37.4	186
OM	mg/kg fuel	35.9	52.3	1.02	245	16.5
Water soluble ions	mg/kg fuel	12.0	27.7	14.5	8.80	14.6
Elements	mg/kg fuel	0.77	2.95	2.15	6.34	6.62
N-alkanes	mg/kg fuel	7.19	1.79	4.72	26.2	4.87
PAHs	mg/kg fuel	0.05	0.11	0.17	2.94	0.06
Hopane and sterane	mg/kg fuel	0.01	0.03	0.05	0.12	0.02
PM	mg/kg fuel	847	200	459	548	436
Mass balance	%	46	43	49	54	53

Fig. 1. Table R1 Mass closure on the PM filter for trucks