**Response to Reviewers' Comments on Manuscript: acp-2016-1038**

Dear Editor,

We are thankful very much to you and the anonymous reviewers for the profound comments and suggestions. We have revised this manuscript accordingly. Listed below are our point-by-point responses (blue) to each reviewer's comments (black) in the revised manuscript with marks. In addition, we would like to ask you if we can add Yanli Feng as another corresponding author, considering contributions to experiment instruction and manuscript revised?

Best regards,

Dr. Yingjun Chen

**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 9 line 16-23**).

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 |
| | non-highway 2 | 21.3 | 16.1 | / | 3.68 |
| Heavy duty-China II | 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 9 line 3-6**), 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 9 line 6-7**).

(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 14 line 19-21**).

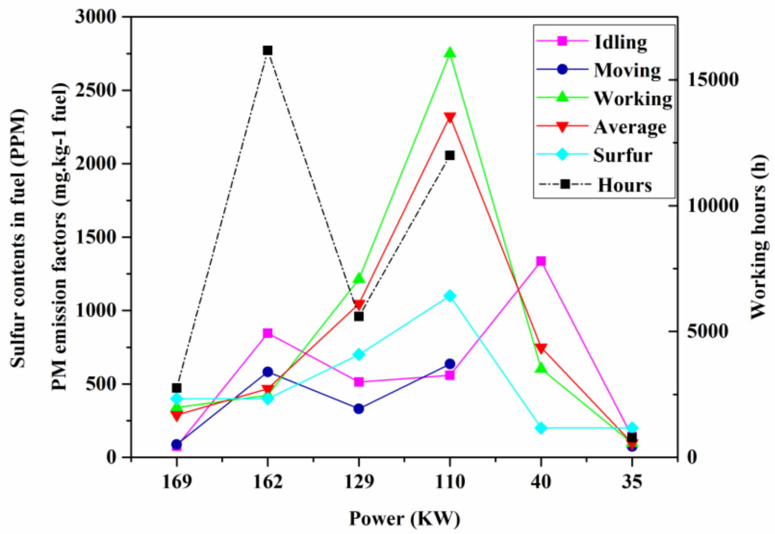


Figure S5 PM emission factors for different power excavators

**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 10 line 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 11 line 28**).

**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 16 line 15**).

**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 16 line 23**).

**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). One 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 18 line 6-17**).

**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 the contribute to a very small fraction of the emissions.

**Response:** Thanks for the comment. The average emission factors of elemental were 5.66 $mg \cdot kg^{-1}$ for E1+E6 and 4.02 $mg \cdot kg^{-1}$ for E2+E3+E4+E5, and were mentioned in the revised manuscript (**Page 18 line 19-20**).

**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 19 line 1-13**).

**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 22 line 1**).

**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 22 line 27-28**).

**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 24-25; Page 3 line 1-3**).

**Comments#16:** Define BaPeq in the abstract 3.

**Response:** Thanks for the comment. The $BaP_{eq}$ has been defined in the revised abstract (**Page 3 line 9**).

**Comments#17:** P3, L 7: define PM. Throughout the paper indicate what size PM refers to (PM1, PM2.5, etc).

**Response:** Thanks for the comment. PM referred to total suspended particulate ($D_p \leq 100$ μm) in this study. We have remarked in the revised manuscript (**Page 3 line 23**).

**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 16 line 23**).

**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 26 line 25**)

**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 40; Page 42; Page 43; Page 44; Page 46**).

**Comments#21:** Fig 7: what do the errors bars represent? Unclear form 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 46**).

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

**Response:** Thanks for the comment. The annotations wre 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.

**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 9 line 25-26; Page 10 line 5-10**). 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.

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 9 line 16-23**).

**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 18 line 6-17; Page 19 line 1-13; Page 25 line 1-11**). 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 24-26**).

**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 28**).

**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 6 line 1-30;Page 7 line 1-6)**.

**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 8 line 24-28**). 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 km·h$^{-1}$) , 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 10 line 26-29**).

**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 13 line 18-30**).

**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 9 line 16-23; Page 20 line 14-17**).

**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 20 line 13-14**). As shown in table 1, 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 mg $kg^{-1}$ 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.

Table 1 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 |

**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 12 line 4-11**). 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 18 line 6-17; Page 19 line 1-13; Page 25 line 1-11**).

**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 13 line 19-30; Page 25 line 1-11**).

**References:**


Adlakha, J., Singh, P., Ram, S.K., Kumar, M., Singh, M.P., Singh, D., Sahai, V., Srivastava, P.: Optimization of conditions for deep desulfurization of heavy crude oil and hydrodesulfurized diesel by Gordonia sp. IITR100, Fuel. 184: 761-769 2016

Chow, J.C., Lowenthal, D.H., Chen, L.W.A., Wang, X.L., Watson, J.G.: Mass reconstruction methods for PM2.5: a review, Air Quality Atmosphere and Health. 8(3): 243-263 2015

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 $PM_{2.5}$ 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

Gilardoni, S., Russell, L.M., Sorooshian, A., Flagan, R.C., Seinfeld, J.H., Bates, T.S., Quinn, P.K., Allan, J.D., Williams, B., Goldstein, A.H., Onasch, T.B., Worsnop,

D.R.: Regional variation of organic functional groups in aerosol particles on four US east coast platforms during the International Consortium for Atmospheric Research on Transport and Transformation 2004 campaign, Journal of Geophysical Research-Atmospheres. 112(D10): 11 2007
Japar, S.M., Szkarlat, A.C., Gorse, R.A., Heyerdahl, E.K., Johnson, R.L., Rau, J.A., Huntzicker, J.J.: comparison of solvent-extraction and thermal optical carbon analysis-methods-application to diesel vehicle exhaust aerosol, Environmental Science & Technology. 18(4): 231-234 1984
Kasper, M., Sattler, K., Siegmann, K., Matter, U., Siegmann, H.C.: The influence of fuel additives on the formation of carbon during combustion, Journal of Aerosol Science. 30(2): 217-225 1999
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
Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T.: Sources of fine organic aerosol. 2. Noncatalyst and catalyst-equipped automobiles and heavy-duty diesel trucks, Environmental Science & Technology. 27(4): 636-651 1993
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
Sánchez, N.E., Millera, Á., Bilbao, R., Alzueta, M.U.: Polycyclic aromatic hydrocarbons (PAH), soot and light gases formed in the pyrolysis of acetylene at different temperatures: Effect of fuel concentration, Journal of Analytical and Applied Pyrolysis. 103: 126-133 2013
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
Wang, Y.F., Huang, K.L., Li, C.T., Mi, H.H., Luo, J.H., Tsai, P.J.: Emissions of fuel metals content from a diesel vehicle engine, Atmospheric Environment. 37(33): 4637-4643 2003
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

# Measurement ~~on~~ of PM and its chemical composition~~s~~ ~~for~~ in real-world emissions from non-road and on-road diesel vehicles

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**Abstract.** With ~~increasing population~~ the rapid growth in the number of both non-road and on-road diesel vehicles, the adverse effects of particulate matter (PM~~-~~) and its ~~compositions~~ constituents ~~(~~such as elemental carbon (EC), and p~~P~~olycyclic aromatic hydrocarbons (PAHs)~~)~~, on air quality and human health ~~get~~ have been receiving ~~more and more~~ increasing attention. However, studies on the characteristics of PM and its composition which~~s~~ emitted from diesel vehicles are scarce, particularly

those ~~measured under~~ performed in real-world conditions~~, are scarce~~. In this study, six excavators and five trucks~~,~~ involving a ~~wide~~ range of emissions standards and ~~working in different operating~~ operational modes~~,~~ were tested to characterize PM constituents, ~~of PM (~~including organic carbon (OC), EC, water soluble ions (WSIs), elements, and organic species such as PAHs, n-alkanes, hopanes, and steranes~~)~~. The average emission factors ~~of~~ for PM ($EF_{PM}$) ~~for~~ from excavators and trucks were 829 ± 806 and 498 ± 234 mg $kg^{-1}$ fuel, respectively, which ~~are~~ are similar to values found in ~~comparable with~~ other studies. ~~However,~~ $EF_{PM}$ was significantly affected by fuel quality, ~~operating~~ operational mode,~~s~~ and emission standards. ~~High~~ A significant correlation ($R^2$=0.79, p<0.01) ~~existed~~ was found between the $EF_{PM}$ for excavators and the sulfur contents in fuel. The highest average $EF_{PM}$ ~~under~~ for working ~~mode for~~ excavators was 904 ± 979 mg $kg^{-1}$ fuel, ~~due to~~because of the high engine load required in this mode. ~~under this mode~~. From pre-stage 1 to stage 2 emission standards, the average $EF_{PM}$ for excavators ~~with different emission standards~~ decreased by 58%. ~~Similarly, f~~For trucks, the average non-highway $EF_{PM}$ under ~~non-highway condition~~ (548 ± 311 mg $kg^{-1}$ fuel) was higher than ~~those under~~the highway ~~condition~~ $EF_{PM}$ (497 ± 231 mg $kg^{-1}$ fuel). Meanwhile, the reductions when switching from China II and ~~China~~ III to China IV standards were 6~~3.5~~3.5% and 6~~5.6~~5.6%, respectively. Generally, the PM composition~~s~~ emitted from excavators was dominated by OC (39.2~~%~~ ± 21.0%) and ~~,~~ EC (33.3~~%~~ ± 25.9%), ~~and~~ while PM ~~for~~ from trucks~~,~~ ~~PM~~ was dominated by EC (26.9~~%~~ ± 20.8%), OC (9.89~~%~~ ± 12%), and WSIs (4.67~~%~~ ± 5.74%). Several differences ~~of~~ in composition~~s~~ were observed among the various ~~operating~~ operational modes, emission standards, and fuel ~~quality~~qualities. The average OC/EC ratios ~~under idling and working modes~~ for idling and working excavators were 3 ~~and~~ to 4 times higher than those ~~in~~ for moving ~~modes~~excavators. Although the $EF_{PM}$ for excavators and trucks was reduced by the constraint of~~with~~ stringent emission standards, the ~~fractions of~~ elemental fractions~~s~~ for excavators ranged from 0.49~~%~~ to 3.03% from pre-stage 1 to stage 2,~~;~~ and the fraction of WSIs for the China IV truck was 6-~~-~~fold higher than ~~those from~~they were for the other trucks. Furthermore, as compared with ~~the results from~~ other diesel vehicles, wide ranges ~~of~~

in the ratios of benzo[a]anthracene/(benzo[a]anthracene+chrysene) (0.26-0.86), indeno[1,2,3-cd]pyrene/( indeno[1,2,3-cd]pyrene +benzo[ghi]perylene) (0.20-1.0) and fluoranthene/(fluoranthene+pyrene)~~BaA/(BaA+Chry) (0.26-0.86), IcdP/(IcdP+BghiP) (0.20-1.0) and Flua/(Flua+Pry)~~ (0.24-0.87) ~~for excavators~~ were found for excavators, which ~~may~~ might be ~~attributed to~~a result of the complex characteristics –of excavator ~~operating~~ operational modes ~~for excavators~~. ~~Although~~ Similar fractions of the ~~total~~ 16 priority –PAHs (as identified by the U.S. Environmental Protection Agency) were found in the exhaust from the ~~for~~ excavators and trucks ~~were similar,~~. ~~the~~ The equivalent concentrations of total ~~of~~ benzo[a]pyrene, ~~BaPeq that~~ which ~~was~~ were used to evaluate ~~the~~ carcinogenic risk, ~~was~~ were 31 times higher for excavators than ~~those~~ they were for trucks~~. Therefore~~, implying that more attention should be paid to non-road vehicle~~’s~~ emissions.

**Keywords**

Diesel vehicles; excavators; trucks; PM; chemical composition; ~~impact~~ influential factors

**Copyright statement**

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## 1. Introduction

Particulate matter (PM) emitted from diesel vehicles ~~have~~ has significant~~ly~~ adverse ~~impacts~~ effects on air ~~pollution~~quality, human health, and global climate change, ~~,~~ and therefore merit close ~~should be examined~~ examination ~~closely~~ (Aggarwal et al., 2015, 2016). ~~Many~~ Previous studies have reported that diesel vehicle~~s~~ exhaust ~~was~~ is ~~a~~ a major source of ambient PM emissions ($D_p \le 100$ μm) ~~emissions in ambient PM~~ (Oanh et al., 2010, Zhang et al., 2015a). For ~~example~~instance, ~~it is reported that~~ vehicle exhaust was reported to ~~contributed~~ contribute to almost 30% of ambient PM emissions ~~in ambient PM~~ in 9 cities ~~in~~ of China in 2015 (MEP 2016). The International Agency for Research on Cancer (IARC) found that exposure to diesel exhaust causes lung cancer (IARC 2012). ~~It is~~ Adar et al. (2015) reported that more than 25 million children breathe polluted air on ~~diesel school~~ diesel school buses, which then ~~causing~~ causes a disproportionate occurrence of ~~adverse~~ respiratory disease~~ health (Adar et al., 2015)~~. Nearly 34% of element carbon (EC)~~ emission~~ emissions, a major contributor to current global warming and poor human health, ~~accounts~~ comes ~~for nearly 34%~~ from off-road diesel vehicles in the USA (USEPA 2015).

The ~~populations~~ numbers of on-road and non-road diesel vehicles ~~have~~ have increased considerably in China, and have contributed to ~~especially for non-road diesel vehicles, causing~~ severe emissions ~~situation~~problems. On-road diesel vehicles can be classified as light-duty, medium-duty, and heavy-duty trucks. Non-road diesel vehicles mainly include construction machinery and agricultural equipment (MEP 2014). Airplanes, trains, and vessels are not included as non-road diesel vehicles in this study, because the primary fuels used for these vehicles does not include diesel. ~~According to reports, t~~The number of on-road diesel vehicles increased from 11.0 million in 2009 to 32.8 million in 2015,~~,~~ ~~and the number of~~ while the number of non-road diesel vehicles increased from 20.6 million in 2006 to 33.6 million in 2012 (CCCMIY et al., 2013, MEP 2016). ~~According to~~ Based on the China vehicle environmental management annual report for 2015 (MEP 2016), 0.56 million tons of PM were emitted from on-road mobile sources and ~~almost higher~~ more than 90% of PM ~~came~~ resulted from on-road diesel vehicle~~s~~ emissions ~~in 2015~~(Figure S1). However, pollutants emitted from non-road diesel vehicles should not be neglected. In 1991, The U.S. Environmental Protection Agency (USEPA) published a report indicating that PM emitted from non-road diesel vehicles was significantly higher than that emitted from on-road diesel vehicles (USEPA 1991). Wang et al. (2016) estimated ~~the~~ an emission inventory ~~from~~ for non-road equipment (including agricultural equipment, river/ocean-going vessels, locomotives, and commercial airplanes) and found that there ~~are~~ were 349 ~~thousand tons~~Gg of PM emissions from non-road vehicles in China ~~in~~ during 2012. Construction equipments was the largest source of PM emissions from non-road diesel vehicles. ~~According to~~ Zhang et al. (2010) reported that ~~Pearl River Delta (PRD) region's~~ PM emissions from construction ~~instruments~~equipment in the Pearl River Delta (PRD) region ~~significantly~~ accounted for 26.5% of the total emission from non-road vehicles in 2006. ~~As a~~An important type of non-road diesel vehicle, the number of construction ~~instruments~~ equipment in China increased from 1.97 million to 5.85 million between ~~during~~ 2006 ~~to~~ and 2012 ~~in China~~ (CCCMIY 2013). ~~According to Zhang et al. (2010) Pearl River Delta (PRD) region's PM emission from construction instruments significantly accounted for 26.5% of the total non-road vehicles in 2006.~~ Furthermore, ~~As~~ as one of the most abundant types of construction ~~instruments~~ equipment (Figure S1), excavators contribute ~~diesel consumption and PM emission from excavators were 7450 and 34.8 thousand tons in 2007~~almost ~~–~~65% of the PM emissions from construction equipment (Li et al., 2012).

In order to control ~~diesel vehicles~~ PM emissions pollution from diesel vehicles, China ~~has~~ began~~started~~ to implement emission standards ~~early~~ in early 2001 for light-duty diesel vehicles and heavy-duty diesel vehicles (SEPA et al., 2001). ~~Those~~ These standards ~~have been~~were tightened in the subsequent 12 years, from the China I to China V standards ~~in 12 years~~. Although emission standards for on-road diesel vehicles ~~were~~ were formulated ~~to~~ in China V, insufficient diesel fuel quality slows their ~~retards~~ implementationg ~~of emission standards~~ (Yue et al., 2015). In addition, ~~The~~ the China IV emission standards for on-road diesel vehicles are not fully implemented ~~until now~~. ~~Compared with on-road diesel vehicles, t~~The ~~implementing~~ implementation timeline ~~for~~ of emission standards for non-road diesel vehicles has lagged behind that of the on-road diesel vehicles. China ~~has~~ implemented two emission standards for new non-road diesel engines, stage 1 and stage 2~~,~~ in 2007 and 2009, respectively. ~~Furthermore~~However, this first ~~implemented~~ implementation ~~time~~ in China was 7 years later than implementation in the ~~compared with the~~ USA (USEPA 2003, SEPA et al., 2007). The pollution emissions limits for on-road and non-road diesel vehicles are given in Tables S1 and S2.

~~The fundamental work of~~ $EF_{PM}$ ~~that~~ is an important parameter in the compilation of emission inventories~~y~~ for on-road and non-road diesel vehicles in China. However, the foundational work towards quantifying $EF_{PM}$ is relatively weak and contains large uncertainties (Huang et al., 2011). ~~Recently, m~~Most of the $EF_{PM}$ from trucks have been measured using tunnel and dynamometer tests, which do not allow for evaluating influential factors for PM emissions from a single truck in real-world conditions (Alves et al., 2015b, Mancilla et al., 2012, Pio et al., 2013). ~~used in emission inventory research came from developed countries.~~ Several studies have measured PM emissions from trucks using on-board tests in real-world conditions (Wu et al., 2016, Wu et al., 2015, Zhang et al., 2015a). Because the $EF_{PM}$ emitted from trucks could change along with improved emission standards, data should be updated frequently (Huo et al., 2012). ~~Wang et al. (2016) estimated emission inventory from non-road equipment and suggested that real-world measurements of emissions for non-road equipment are desperately needed. Along with increasingly serious environmental problems, PM emission from on-road diesel vehicles has been taken seriously in China. There are considerable studies about on-road tests to study PM from on-road diesel vehicles. Wu et al. (2015) tested 17 in-use diesel trucks in Beijing using a portable emission measurement system (PEMS) and calculated the~~ ~~$EF_{PM}$~~ ~~of those vehicles. Moreover, Zhang et al. (2015b) measured the real world PM emission factors from in-use HDDTs using PEMS.~~ In addition, the data for $EF_{PM}$ emitted from non-road diesel vehicles ~~on~~ in real-world conditions ~~was~~ is scarce in China. ~~To our knowledge, dynamometer test was used by most of studies to research non-road vehicle emission (Liang et al., 2005, Liu et al., 2015, Pietikainen et al., 2015). Liu et al. (2015) measured the PAH and nitro-PAH emission from non-road diesel engine, which was conducted utilizing the dynamometer test cycles required by U.S. EPA Tier 4 Final standards.~~ In 2014, the Ministry of Environmental Protection of the People's Republic of China had issued "Technical guide for the preparation of a single source emission inventory of atmospheric fine particulate matter." However, no measured baseline for emission factors of PM from non-road vehicles, especially construction machinery (6 g ~~k~~m$^{-1}$ were predicted for uncontrolled standards) could be found in this technical guide (MEPPRC 2014). Until now, there was only one study in China by Fu et al. (2012), who provided $EF_{PM}$ for ~~tested~~ 12 excavators using portable emission measurement system (PEMS) ~~PEMS to determine PM emission factors under~~ for different ~~working~~ operational modes. ~~However~~O~~,~~n-board measurements need to be expanded to improve localization of $EF_{PM}$ for non-road diesel vehicles in China as soon as possible, ~~due to~~because of the complexity of real-world conditions, including~~such as~~ lagging diesel quality and changing emission standards~~,~~. ~~the on-board measurements need to be expanded to improve localization of $EF_{PM}$ for on-road and non-road diesel vehicles in China as soon as possible.~~

Analysis of the ~~Chemical~~ chemical composition of PM ~~constituents~~ ~~are~~ is essential~~important~~ for ~~studies of~~ source apportionment, human health, and climate change studies. Primary PM emitted from diesel vehicles contains a variety of chemical ~~compositions~~components, ~~such as~~including organic carbon (OC), elemental carbon (EC), water soluble ions (WSIs), elements, and organic species ~~(~~such as n-alkanes, polycyclic aromatic hydrocarbons (PAHs), hopane and sterane). Several previous field studies ~~have~~ have focused on chemical composition~~s~~ of PM emitted from diesel vehicles. Zhang et al. (2015a) characterized PM compositions (OC, EC, WSIs and elements) ~~emission~~ emitted from heavy-duty diesel trucks (HDDTs). Wu et al. (2016) reported the detailed chemical composition of $PM_{2.5}$ emitted from China III and China IV diesel trucks, including the ~~organic carbon (~~OC~~)~~, ~~elemental carbon (~~EC~~)~~, ~~water soluble ions (~~WSIs~~)~~, and element contents~~, emitted from China III and China IV diesel trucks~~. In 2012, Fu et al. (2012) tested 12 excavators in the first on-board test for excavators in China, but ~~to determine~~ only optically-based ~~PM emission factors~~$EF_{PM}$ were given~~, which was the first on-board test for excavators in China. However,~~. Therefore, the specific characteristics of PM emitted from diesel vehicles and its composition~~s~~ ~~emitted from diesel vehicles~~are still largely unknown, especially for organic ~~matters~~compounds~~, are lacking~~.

In this study, PM ~~and its composition~~ emitted from on-road and non-road diesel vehicles ~~were~~ was measured ~~in order~~ to (I) test emission factors of PM for excavators and trucks ~~under~~ in real-world conditions; (II) identify influential ~~impact~~ factors ~~of~~ on the emitted PM and its composition~~s~~ ~~for non- and on-road diesel vehicles;~~, and (III) characterize chemical components ~~of~~ present in the emitted PM ~~from excavators and trucks~~. ~~Our~~ The study results required substantial effort ~~results of this study could~~

and provide valuable information for use in the development of effective control policies ~~and~~ for reducing~~e~~ PM emissions from excavators and trucks.

**Experimental**

**2.1 Diesel vehicle~~s~~ and operational mode~~s~~ selection**

In this study, six excavators and five trucks were selected to cover a wide range of emission standards, manufacturers and engine loads. ~~The d~~Detailed information ~~of~~ for the selected excavators and trucks ~~is~~ is shown in Table 1. ~~The tested excavators were divided into two groups based on their emission standards: three pre-stage 1 excavators and three stage 2 excavators.~~ As shown in Figure ~~S1~~S2, the annual production~~s~~ of excavators ~~have~~ did not change~~d~~ substantially ~~much in~~ between 2007 and ~~-~~2009 (an increase from 70,000 to 85,000 excavators), during which stage 1 non-road vehicle emission standard~~s~~ was implemented~~, varying from 70,000 to 85,000 pieces of excavators~~. Therefore, excavators ~~conducted~~ produced with pre-stage 1 and stage 2 emission standards were chosen ~~in~~ for this study. Based on China national standard (SEPA 2007), excavators ~~can~~ are~~be~~ divided into five types ~~(0<P<8 kw; 8<P<18 kw; 18<P<37 kw; 37<P<75 kw; 75<P<130 kw; 130<P<560 kw)~~ according to ~~the~~ their ~~rated~~ power rating~~(P)~~. ~~Thus, each type of e~~The excavators were categorized for this study ~~divided by~~ by emission standards and were rated as~~included~~ low (0-75 kw), medium (75-130 kw) ~~and~~ or high (130-560 kw) ~~excavators~~power~~, which represent the low, medium and high power excavators, respectively~~. As a way to ~~For~~ reflect~~ing~~ ~~the real~~actual ~~operation~~ use environments, three ~~operating~~ operational modes were selected for the excavators~~ were selected~~ ~~-~~idling~~ mode~~, moving ~~mode~~ and working~~ mode, respectively~~. Further descriptions of these three modes can be found ~~were listed~~ in Fu et al. (2012).~~ In this study~~ In addition, consistent sampling times for the different modes were not strictly required in this study, as long as sufficient amounts of PM were collected to conduct the subsequent chemical analysis. The average ~~average duration consumption~~ sampling times ~~in~~ during idling, moving, and working ~~were~~ were 41.7, 24.0, and 28.5 min~~utes~~, respectively.

~~For diesel trucks, there were three~~ Three types of diesel truck~~s~~ were selected ~~according to emission standards,~~ one China II standards truck, three China III standards trucks, and one China IV standards truck. ~~Practicality~~The~~, just~~ China III trucks ~~contained~~ included ~~three trucks including~~one each of light-duty, medium-duty, and heavy-duty diesel trucks. Based on the traffic ~~control measures~~rules and driving conditions ~~of~~ for on-road diesel trucks, ~~different~~ predesigned routes were chosen for ~~different emission standards and size~~ testing the trucks in Yantai, Shandong province ~~in~~ of China (Figure 1). Because different trucks drove on different routes, the selected routes in this study were divided into non-highway and highway categories. The selected routes ~~chosen~~ for China III and China IV light-duty trucks included ~~arterial road~~non-highway 1 ~~(non-highway 1)~~, ~~secondary road (non-highway 2)~~ non-highway 2 and highway 1. The lengths of ~~those~~these three roads were 19, 35 and 17 km, respectively. The route chosen for the China II heavy-duty truck (yellow label) was ~~special used for "yellow label car" (non-highway 3)~~ non-highway 3 which was 25 km. The routes chosen for China III medium-duty and heavy-duty trucks included non-highway 4 ~~4~~ and highway 2. The lengths of ~~those~~ these ~~–~~two roads were 47 and 23 km, respectively. The detailed velocity and road grade information for all of the tested routes ~~were~~ are shown in Figures S3~~2~~ and ~~S3~~S4. Although repeated tests were conducted for some vehicles, it should be noted that only one set of integral data was selected for further discussion, due to the incompleteness of some monitoring data (e.g. $CO_2$ and CO concentrations). As shown in Tables S3 and S4, the variability in test times for the same operational mode was considered acceptable. Some actions were required to reduce the uncertainty. For example, we combined sampling filters for the repeated experiments for vehicles T1 and T3 to carry out organic compound analysis.

**2.2 On-board emission measurement system**

The on-board emission measurement system was ~~self-~~designed and ~~combined~~ constructed in our ~~our~~ laboratory (Figure 2). ~~The~~ A description of the ~~used~~ on-board emissions test system was given ~~by~~ in our ~~previous~~ previous ~~study~~ report (Zhang et al., 2015b). Briefly, this system ~~has~~ consists of two main ~~functional parts~~components~~,~~: ~~including~~ a Photon II analyzer, which was used to analyze the ~~for~~ flue gas (HC, CO, $CO_2$, $SO_2$, and NOx) ~~analyzer~~ , and a PM ~~sampler~~ sampling system. The PM sampling~~er~~ system ~~consists~~ consisted of a dilution system followed by~~, and~~ five exhaust channels~~ behind this dilution system~~. Two channels were connected ~~with~~ to PM samplers, and the other~~s~~ three were blocked. ~~When~~ Before ~~sample~~ sampling the PM ~~–~~emitted from an excavator~~s~~, the emission measurement system was put on a truck and connected to the excavator~~s~~ exhaust tube ~~via~~ by a stainless steel pipe. The system showed clear improvements over other on-board instruments, such as PEMSs and FPS4000 (Zheng et al., 2015), with better portability and the ability to collect filter samples for further chemical analysis in the laboratory. The results presented here include the first dataset from on-board measurement of non-road diesel vehicle exhaust in China.

**2.3 Chemical analysis**

2.3.1 Fuel quality analysis

Fuel quality has a ~~great~~ large effect on PM emissions from vehicles (Cui et al., 2016, Liang et al., 2005, Zhang et al., 2014). ~~Due to~~Since the ~~poor~~ fuel ~~quality~~ used in excavators is often of poor quality, diesel~~s~~ ~~for~~ was collected from each of the tested excavators ~~were collected~~ and ~~–~~analyzed~~tested~~. The results ~~for~~ of fuel quality analysis ~~were~~ are ~~shown~~ given in Table 2. Comparing the diesel quality used in this study with the diesel quality standards for non-road vehicles (GB 252-2015) (SEPA et al., 2015), it was found that ~~most of~~ the sulfur contents in most ~~in~~ of the diesels used in this study (200-1100 ppm) were higher than ~~those~~ allowed ~~in~~ by GB 252-2015 (<350 ppm). Additionally, the sulfur content in the diesel used by E4 was 1100 ppm, which was much higher than ~~those~~ that ~~in diesel~~ used ~~for~~ in the other excavators. Furthermore, the ash content of ~~E4's~~ diesel used by E4 was 4.16%, about~~ therefore~~ 420 times higher than the limit ~~value~~ given ~~in~~ by GB 252-2015.

2.3.2 PM and chemical composition analysis

Quartz-fiber filters were used for collecting the PM samples because the weight losses of these filters could be neglected through strict sampling processes, and quartz-fiber filters could be used for both the PM weight measurement and chemical analysis. The ~~The quartz-fiber~~ filters were weighed before and after sampling to determine the collected PM mass concentrations~~ of PM~~. Before each weighing, the filters were balanced at 25 °C and 40% relative humidity for 24 h. ~~The~~ Each filter~~s~~ ~~were~~ was weighed ~~there~~ three times~~ before and after sampling to insure that the error for each measurement was as low as possible~~. WSIs were analyzed ~~by~~ using ion chromatography (Dionex ICS3000, Dionex Ltd., America) following the method of Cui et al. (2016). Elements ~~was~~ analysis was performed~~analyzed~~ using inductively coupled plasma~~ coupled~~ ~~with~~ mass spectrometerry (ICP-MS~~,~~; ELAN DRC II type, Perkin Elmer Ltd., Hong Kong).

Because the ~~there was not enough~~ organic matter~~s~~ on each filter was insufficient for quantification, we ~~combined~~ merged filters from different ~~operating~~ operational modes or driving routes ~~filters~~ for ~~analysis~~ analyzing ~~for~~ each diesel vehicle~~s~~ ~~according~~ based ~~to~~ on the proportion of sampling~~tested~~ time during each mode or route. Quartz filter samples were spiked with internal standards (including acenaphthene-$d_{10}$, benzo[a]anthracene-$d_{12}$, pyrene-$d_{10}$, coronene-$d_{12}$, cholestane-$d_4$, *n*-C15-$d_{32}$, *n*-C20-$d_{42}$, *n*-C24-$d_{50}$, *n*-C30-$d_{58}$, *n*-C32-$d_{66}$, *n*-C36-$d_{74}$) ~~were~~ and ultrasonically extracted two times in 30 ~~ml~~ mL of a 1:1 mixture of hexane and dichloromethane for 10 min. All extracts ~~for~~ from each sample were combined, filtered and concentrated to ~~∼~~approximately 0.5 ~~ml~~mL.

Organic species including n-alkanes, PAHs, hopane and sterane were analyzed using GC-MS (Agilent 7890A GC-5975C MS)~~ equipped~~ with a DB-5MS column (length 30 m × i.d. 0.25 mm × thickness 0.25 μm). The GC operating program was as ~~following~~follows: 60 °C ~~with static time of~~for 4 min, increase 5 °C min$^{-1}$ to 150 ℃ with 2 min static time ~~to 150 °C with static time of 2 min~~, then ~~ramped~~ increase 3 ℃ min$^{-1}$ to 306 °C ~~at rate of 3 °C min$^{-1}$~~ with a 20 min static time~~ of 20 min;~~. and The GC ~~conditions:~~ had an injector temperature ~~was~~ of 290 °C, injector volume of ~~injector was~~ 2 μL, helium carrier gas~~ was helium~~, and gas flow rate of ~~gas was~~ 1.2 ~~ml~~mL min$^{-1}$. The electron impact (EI) mode at 70 eV and selected-ion-monitoring (SIM) mode were selected to ~~determining~~ determine concentrations of PAHs, hopane, and sterane. For organic matter~~s~~, ~~the~~ blank samples and recovery rates (66.7%-128% for five surrogates) were measured. The blank concentrations were subtracted from the sample concentrations.

The PM ~~Chemical~~ chemical constituents ~~of PM~~ analyzed in this study ~~are~~ were~~listed as follows:~~ OC~~,~~; EC~~,~~; WSIs: $SO_4^{2-}$, $NO_3^-$, $Cl^-$, $NH_4^+$; ~~Elements~~elements: Na, Mg, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Pb); n-alkanes: C12 to ~~-~~C40; the sixteen USEPA priority PAHs of ~~:~~ naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorine (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fluo), pyrene (Pyr), benzo [a]anthracene (BaA), chrysene (Chry), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IcdP), dibenz[a,h]anthracene (DahA) and benzo[ghi]perylene (BghiP); Hopane and sterane: ABB-20R-C27-Cholestane (ABB), AAA-20S-C27-Cholestane (AAA), 17A(H)-22,29,30-Trisnorhopane (Tm), 17A(H)-21B(H)-30-Norhopane (30AB), and 17A(H)-21B(H)-Hopane (29AB).

## 2.4~~.~~ Data processing

### 2.4.1 Fuel~~ed~~-based emission factors

Fuel~~ed~~-based emission factors were calculated ~~by~~ using the carbon ~~-~~mass balance formula~~.~~:

$$EF_i = \frac{\Delta X_i}{\Delta CO_2} \cdot \frac{M_i}{M_{CO_2}} \cdot EF_{CO_2} \tag{1}$$

~~Where~~ where $EF_i$ and $EF_{CO_2}$ (g kg$^{-1}$ fuel) are the emission factors for species $i$ and $CO_2$, respectively, $\Delta X_i$ and $\Delta CO_2$ (mol m$^{-3}$) are the background-corrected concentrations of species $i$ and $CO_2$, respectively, and $M_i$ and $M_{CO_2}$ (g mol$^{-1}$) represent the molecular weights of species $i$ and $CO_2$, respectively.

The $CO_2$ emission factors ($EF_{CO_2}$) were calculated as:

$$EF_{CO_2} = R_{FG} \cdot c(CO_2) \cdot M_{CO_2} \tag{2}$$

~~Where~~ where $c(CO_2)$ (mol m$^{-3}$) is the molar concentration of $CO_2$, and $R_{FG}$ (m$^3$ kg$^{-1}$ fuel) represents the flue gas emission rate.

The flue gas emissions were calculated as:

$$R_{FG} = \frac{C_F}{c(C_{CO}) + c(C_{CO_2}) + c(C_{PM})} \tag{3}$$

~~Where~~ where $C_F$ (g C kg$^{-1}$ fuel) represents the mass of carbon in 1 kg of diesel fuel, and c($C_{CO}$), c($C_{CO_2}$), and c($C_{PM}$) (g C m$^{-3}$) represent the flue gas mass concentrations of carbon as CO, $CO_2$, and PM, respectively~~ in the flue gas, respectively~~.

2.4.2 Average fuel-based emission factors for excavators and trucks

The average fuel-based emission factor for each excavator ~~under~~ in each relevant ~~different operating~~ operational mode~~s~~ was calculated ~~by follows~~as:

$$\mathrm{EF}_{i,j} = \sum EF_{\mathrm{i,j,g}} \times P_{j,\mathrm{g}} \quad (4)$$

~~Where~~ where $EF_{i,j}$ (g kg$^{-1}$ fuel) is the average emission factor of species $i$ ~~for~~ from excavator $j$, $EF_{i,j,g}$ (g kg$^{-1}$ fuel) ~~represents~~ is the emission factor of species $i$ ~~for~~ from excavator $j$ ~~under~~ in mode $g$ ~~mode~~, and $P_{j,g}$ (%) is the proportion of activity time (Fu et al., 2012) for excavator $j$ in mode ~~under~~ g ~~mode~~.

The average fuel-based emission factor for each truck ~~under~~ in different driving conditions was calculated ~~by follows~~as:

$$\mathrm{EF}_{i,j} = \sum EF_{\mathrm{i,j,s}} \times P_{j,s} \quad (5)$$

~~Where~~ where $EF_{i,j}$ (g kg$^{-1}$ fuel) is the average emission factor ~~of~~ for species $i$ ~~for~~ from ~~excavator~~ truck $j$, $EF_{i,j,s}$ (g kg$^{-1}$ fuel) ~~represents~~ is the emission factor of species $i$ for ~~excavator~~ truck $j$ ~~under~~ in driving condition s~~, condition,~~ and $P_{j,s}$ *(%)* is the proportion of activity time for truck $j$ ~~under~~ in driving condition s ~~condition~~.

2.4.3 Benzo[a]pyrene equivalent concentration ($BaP_{eq}$)

The various PAHs have a wide range of carcinogenic risks. Therefore, it is not accurate to evaluate the harmful effects of PAHs on human health using the total combined mass concentration. $BaP_{eq}$ is typically used to evaluate the carcinogenic risks associated with individual PAH (Mirante et al., 2013). The $BaP_{eq}$ was calculated as:

$$\mathrm{BaP_{eq}} = \sum \mathrm{PAH_i} \times \mathrm{PEF} \quad (6)$$

where $PAH_i$ is the measured concentration of an individual PAH for excavator $i$, and *PEF* is ~~Because of different carcinogenic risks for each PAH, the $BaP_{eq}$ for parent PAHs were given. The BaPeq was calculated by multiplication of the measured concentrations by the respective~~ the potency ~~equivalent~~ equivalence factor ~~(PEF)~~

~~(Mirante et al., 2013). The PEF values were~~ for that PAH obtained from Wang et al. (2008).

### 3. Results and discussion

#### 3.1 ~~Particulate matter f~~Fuel-based emission factors of PM ~~for~~ in excavator exhaust~~s~~

The $EF_{PM}$ values for excavator~~s~~ exhaust ~~are~~ are ~~presented~~ illustrated in Figure 3, with ~~the~~ detailed information ~~shown~~ given in Table ~~S1~~S5. The maximum fuel-based PM ~~fuel-based~~ emission factor was ~~almost~~ 37 times higher than the minimum ~~under different operating modes for different vehicles~~. In general, the average $EF_{PM}$ for different excavators ranged from 96.5 to 2323 mg kg$^{-1}$ fuel, with an average of 829 ± 806 mg kg$^{-1}$ fuel. The $EF_{PM}$ values of excavators reported by Fu et al. (2012) were within the range of $EF_{PM}$ in this study ~~but in a narrower range~~. The ~~reason for the more~~ wide~~ly~~ range in~~d~~ $EF_{PM}$ values ~~in this study~~here ~~may~~could ~~be~~ ~~that the~~ be due to the difference in the selection of excavators emission standards. The excavators selected by Fu et al. (2012) included stage 1 and stage 2 emission standards, while ~~this~~ our study tested excavators with pre-stage 1 and stage 2 emission standards. ~~Therefore, the range of $EF_{PM}$ in this study may reflect the general excavator's PM emission situation in China.~~

$EF_{PM}$ ~~could be~~is affected by many factors. In this study, the $EF_{PM}$ range for excavators with different power ratings was 96.5 (35 kw) to 2323 (110 kw) mg kg$^{-1}$ fuel, but the correlations between $EF_{PM}$ and engine power (See Figure S5) were weak. Conversely, fuel quality, emission standard and operational mode significantly affected the $EF_{PM}$. ~~Some variation characteristics about the $EF_{PM}$ values due to the different fuel quality, emission standards and operating modes were summarized as follows. Firstly, f~~Fuel quality ~~has~~had a ~~great~~ large impact on $EF_{PM}$ for the excavators. As shown ~~from~~ in Figure 3, a ~~high~~ significant correlation ($R^2$ = 0.79, P < 0.01) was found between the average emission factors for excavators and the fuel sulfur contents ~~in fuel~~, which ~~is~~ is consistent with ~~the~~ results ~~studied from~~reported by Yu et al. (2007).

The ~~Secondly,~~ $EF_{PM}$ also decreased with stricter ~~enhancing of~~ emission standards for the excavators. The $EF_{PM}$ measured ~~$EF_{PM}$~~ for pre-stage 1 excavators ~~under~~ during idling, moving and working ~~conditions~~ were 914 ± 393~~ mg kg$^{-1}$ fuel~~, 609 ± 38 ~~mg kg$^{-1}$ fuel~~ and 1258 ± 1295 mg kg$^{-1}$ fuel, respectively, whereas~~.~~ ~~The EF$_{PM}$ for stage 2 excavators under idling, moving and working conditions~~ for stage 2, they were 243 ± 236~~ mg kg$^{-1}$ fuel~~, 165 ± 144~~ mg kg$^{-1}$ fuel~~ and 551 ± 587 mg kg$^{-1}$ fuel, respectively. ~~Compared to pre-stage 1,~~ The EF$_{PM}$ of the stage 2 excavators were reduced by 73%, 73% and 56% ~~in~~ from the pre-stage 1 values under idling, moving and working modes, respectively. The average EF$_{PM}$ for excavators of different emission standards decreased by 58% from pre-stage 1 to stage 2, suggesting the effectiveness of the emissions control policy.

~~Lastly,~~ The EF$_{PM}$ varied sharply between different ~~operating~~ operational modes for the various excavators. Specifically, ~~excavators under~~ working excavators ~~modes have~~ had the highest EF$_{PM}$, which ~~is~~ was ~~higher~~ more than double the values for ~~other operating modes~~idling and moving excavators~~ by more than 1 fold~~. The average EF$_{PM}$ for excavators ~~under different driving conditions~~ were 578 ± 467~~ mg kg$^{-1}$ fuel~~ ~~(~~while idling~~)~~, 343 ± 264 ~~mg kg$^{-1}$ fuel~~while ~~(~~moving~~)~~ and 904 ± 979 mg kg$^{-1}$ fuel while ~~(~~working~~), respectively~~. ~~The~~ Working mode produced the highest average EF$_{PM}$, which ~~under working mode~~ might be because the ~~attributed to~~ higher engine load, ~~which causes~~ caused a lower air-fuel ratio~~s~~ and thus ~~then~~ prompted ~~the~~ PM production.

### **3.2 ~~Particulate matter f~~Fuel~~ed~~-based emission factors of PM for trucks**

The EF$_{PM}$ for all measured trucks ~~under different driving patterns~~ varied from 176 ~~mg kg$^{-1}$ fuel~~ to 951 mg kg$^{-1}$ fuel. The maximum EF$_{PM}$ for trucks was three times more than the minimum. ~~There were just tripled in PM emission factors for trucks from maximum to minimum.~~ The average EF$_{PM}$ for the tested diesel trucks was 498 ± 234 mg kg$^{-1}$ fuel. In comparison, Wu et al. (2016) reported an average EF$_{PM}$ for diesel trucks of 427 ~~(95.6-1147 mg kg$^{-1}$ fuel)~~ mg kg$^{-1}$ fuel (95.6-1147 mg kg$^{-1}$ fuel) ~~and it is~~, which was ~~within~~ similar~~the same~~ to the range for~~as~~ our results.

~~Besides, The~~ The average EF$_{PM}$ of diesel trucks ~~for~~ with different emission standards and ~~,~~ vehicle sizes ~~and~~ while using different driving patterns were provided under real-world conditions (Figure 4). The measured EF$_{PM}$ for China II, China III, and China IV diesel trucks varied from 200~~ mg kg$^{-1}$ fuel~~ to 548 mg kg$^{-1}$ fuel. The EF$_{PM}$ for the China II truck measured in this study was lower than ~~the results obtained from~~reported by Liu et al. ~~(910-2100 mg kg$^{-1}$ fuel)~~ (2009) (910-2100 mg kg$^{-1}$ fuel). The average EF$_{PM}$ for light-duty, medium-duty and heavy-duty diesel trucks were 524 ± 457~~ mg kg$^{-1}$ fuel~~, 459, ~~mg kg$^{-1}$ fuel~~ and 492 mg kg$^{-1}$ fuel, respectively. The average EF$_{PM}$ ~~of~~ for trucks under non-highway and highway driving patterns were 548 ± 311 ~~mg kg$^{-1}$ fuel~~ and 497 ± 231 mg kg$^{-1}$ fuel, respectively. As shown in Figure 4 ~~shows~~, reductions ~~of~~ in the measured EF$_{PM}$ between the ~~for~~ China II ~~truck to~~and China IV trucks and ~~from~~ between the China III ~~truck to~~and China IV trucks ~~in EF$_{PM}$~~ were 6~~3.5~~3.5% and 6~~5.6~~5.6%, ~~which indicated~~ indicating that improvements ~~of~~ in the emission standards for diesel trucks ~~significantly decreased~~ could significantly reduce PM emissions. ~~It should be noticed~~ Of particular note was that the EF$_{PM}$ for China III and light-duty diesel trucks were higher than the values for the other corresponding trucks. The reason ~~may~~ might be ~~attributed to~~a result of poor driving conditions, ~~that include~~ i.e., low average speed and ~~more volatile~~highly varied speed ~~in speed for those trucks~~ (Figures ~~S2~~ S3 and ~~Figure S3~~S4). The ~~Same~~ same tendency is apparent in ~~could be seen from~~ Figure 4, ~~that~~ with diesel trucks ~~emitted~~ emitting more PM while driving on the ~~under~~ non-highway ~~condition~~ (average speed of~~:~~ 28.5 km h$^{-1}$) than while driving on the ~~those under~~ highway ~~condition~~ (average speed of ~~:~~ 60.7 km h$^{-1}$). ~~Furthermore, t~~The road grade further ~~was an another aspect effected~~ affected the EF$_{PM}$ of the on-road diesel trucks. For example, the EF$_{PM}$ for T5 ~~under~~ driving on the highway ~~road~~ was lower than those for T1 driving on the highway, because of ~~bigger~~ lower road grade for T5 ~~under highway road than those for T1~~ (Figure ~~S3~~S4).

### 3.3 Particulate matter composition for individual diesel vehicles

Four types of constituents were considered for reconstituting PM mass~~,~~ in this study: (1) organic matter, which was estimated by multiplying the corrected OC ~~by~~by a factor of 1.6 (Almeida et al., 2006); (2) EC; (3) ~~water soluble ions~~WSIs; and (4) elements. The reconstituted masses for ~~each~~ the excavator samplers ~~was~~ were 74.7-123% of the measured mass, while the reconstituted masses for the diesel truck ~~sampler~~ samples ~~was~~ were only 43.2-54.4% of the measured mass (Figure 5). ~~Except~~

~~for~~In addition to uncalculated components, this discrepancy ~~may~~ might be ~~attributed to uncertainties in the weighing process~~ due to a distribution error from OC and EC, moisture effects, or metal oxidation ~~(Dai et al., 2015)~~.

3.3.1 Particulate matter composition for individual excavator

The chemical composition of PM for each excavator ~~was~~ is shown in Figure 5 and Table ~~S2~~S6. For each excavator, the carbonaceous component (OM+EC) was the dominant species, which is ~~consisted~~ consistent with results from a previous study by Liu et al. (2005)~~from a non-road diesel generator that had found~~, who reported that the proportions of ~~organic and element carbon~~OC and EC in PM ranged from 70~~.2%~~ to 9~~0.6~~1% (Liu et al., 2005). B~~Because~~ecause the OC/EC ratio is also used to identify the source of atmospheric particulate pollution, ~~deeper~~ further assessment was performed on the ~~discussion about~~ OC/EC ratios ~~under~~ in different ~~operating~~ operational modes for each excavator ~~was conducted~~ (Figure 6). The average OC/EC ratios ~~for~~ during idling, moving, and working ~~modes~~ were 1.57, 0.57, and 2.38, respectively. The OC/EC ratio ~~under~~ during idling was ~~higher~~ greater than 1 because soot hardly generated at low temperatures ~~hardly~~ and fuel-rich zone. These results were consistent those in ~~, which is similar to the research done by~~ Liu et al. (2005). Furthermore, Liu et al. (2005) reported that the OC/EC ratios decreased with an ~~increasing~~ increase in the load for non-road engines~~load~~. However, this trend was not ~~couldn't be~~ observed in this study. The OC/EC ratio was 2.38 ~~under~~ while working ~~mode~~, and ~~increasing~~ increased ~~again~~ with ~~load~~ increasing load, which was consistent with the results reported by ~~from~~ Zhang et al. (2014). As shown in Figure 6, the differences between ~~Large~~ OC/EC ratios ~~differences~~ for ~~excavators under~~ different excavator ~~operating~~ operational modes were profound~~seen in Figure 6~~, ~~which may~~ and could be ~~caused~~ affected by a number of factors, ~~(such as~~including transient working conditions, diesel sulfur content, and extensive OC sources ~~for OC)~~ (Cocker et al., 2004, Liu et al., 2005, Ruiz et al., 2015).

As shown ~~from~~ in Figure 5, ~~WISs~~ WSIs and elements fractions ranged from 0.335~~%~~ to 1.21% and from 0.163~~%~~ to 7.50%, respectively, for all excavators. The total ~~sum~~ proportion of ~~WISs~~ WSIs and elements to PM was the highest in excavator E6, followed by excavator E1. Generally, the total ~~sum~~ proportion of ~~WISs~~ WSIs and elements to PM in exhaust from excavator ~~E1~~ E6 was 4 to 14 times higher than the corresponding proportions in exhaust from the other excavators. Sulfate and nitrate were the main ~~WISs~~ WSIs (79.1~~%~~-90.0% of ~~WISs~~WSIs) for almost all of the excavators, except for E1, in which ~~while~~ the proportion of $Cl^-$ ~~of WISs for excavator E1~~ (67.2%) was the highest (Table ~~S2~~S6). Fe, Ca, Na, Mg, and K were relatively dominant ~~in~~ elements, ~~but~~ except for E4 ~~excavator~~, Fe, Zn, and Cu were the most abundant elements. Wang et al. (2003) reported that the concentrations of the crustal elements Fe, Ca, and Mg that account for 50% of the total elements in diesel fuel were significantly higher than anthropogenic elements emitted from diesel vehicle engines, which is consistent with the results from our study. Similarly, diesel was the dominant source for these elements because the sampling tube was placed directly on the tailpipe. ~~It may be attributed to that Zn is known from oil additives and Cu usually emitted from wear debris (Lin et al., 2015, Wu et al., 2015). Table 1 and Table 2 showed that excavator E4 produced in 2004 and the diesel quality used was poor, resulting in high Zn and Cu emission.~~ The abundance of Fe, Zn, and Cu in the exhaust of E4 could have been affected by E4 being used to transport ironstone. ~~Besides~~In addition, the elements fractions for the two excavators ~~produced~~ manufactured in 2013 (~~E1 (~~1.42% for E1~~)~~, ~~and~~ , ~~E6 (~~7.50% for E6~~)~~ and 5.66 mg $kg^{-1}$ for E1 + E6) were higher than those for the other excavators (a total of 4.02 mg $kg^{-1}$ for E2, E3, E4, and E5)~~, which may~~ . This indicates that elements emissions ~~was~~ were deteriorating and more stringent control technology should be developed to avoid ~~the total elements~~ adverse health effects from the total elements composition of PM in the exhaust.

~~In addition, t~~The n-alkanes, PAHs, hopane and steranes fractions in exhaust from the excavators were ~~ranged from~~ 3.6~~%~~ to 9.6%, ~~from~~ 0.03~~%~~ to 0.24%, and ~~from~~ 0.001~~%~~ to 0.09% ~~for excavators,~~ respectively. Liang et al. (2005) characterized diesel particulate matter emitted from non-road engines using a dynamometer test and found that n-alkanes accounted for 0.83% of PM, which was lower than the proportion found in ~~results obtained from~~ this study~~.~~, ~~The main reasons are the~~ possibly because they used low sulfur diesel fuel and different sampling methods~~used in Liang's study~~

~~and different methods used in obtained the PM~~. ~~Contrary~~ In contrast to ~~what was observed~~the fractions ~~for~~ of ~~WISs~~ WSIs and elements, ~~Figure 5 showed that~~ the fractions of n-alkanes, hopane and steranes ~~fractions~~ were the highest in excavator E4, while the fractions of PAHs ~~fraction~~ was the highest ~~in~~ for the exhaust from ~~excavator~~ E3. In a comparison of the fuel quality between E3 and E4, E4 had poorer diesel quality, which might be the reason for high n-alkane, hopane and steranes concentrations. Similarly, ~~It was said by~~ Rogge et al. (1993) found that n-alkanes, ~~PAHs,~~ hopane and steranes ~~are~~ were mostly derived from the incomplete combustion of fuel and lubricant oil. ~~— By comparing the differences between fuel quality and performance of excavators, it could be deduced that n-alkanes, hopane and steranes were influenced by fuel quality and~~However, we speculated that –PAHs ~~was~~ were affected by combustion conditions (i.e., combustion temperature) in this study, because E3, with the stage standard, had better performance and superior fuel quality. PAH ~~s~~ isomer ratios ~~have~~ have been widely used to ~~distinguish~~ conduct~~the~~ source apportionment ~~in~~ for environmental receptors (such as sediments) (Liu et al., 2012). Yunker et al. (2002) found that the ratios of the principal masses of PAH 178, 202, 228 and 276 ~~parent PAHs~~ ~~have~~ had ~~a~~ the best potential to distinguish between natural and anthropogenic sources. For the excavators, the ratios ranges ~~of ratios of~~for BaA/(BaA+Chry), IcdP/(IcdP+BghiP), and Flua/(Flua+Pry) were 0.26-0.86, 0.20-1.0, and 0.24-0.87, respectively, with averages of 0.47 ±0.27, 0.44 ±0.38, and 0.48 ±0.27, respectively (Figure 7). The average ratios of PAHs ~~for~~ in excavator exhaust~~s~~ obtained in this study were similar ~~with~~ to ~~that~~ those from Liu et al. (2015)~~ reported for non-road diesel engines~~. The E4 excavator~~s~~ had a clear ~~showed an~~ obvious difference in the ratios of BaA/(BaA+Chry), IcdP/(IcdP+BghiP), and Flua/(Flua+Pry) to those from ~~between~~ the other excavators~~ tested in this study~~. The isomer ratios of BaA/(BaA+Chry), IcdP/(IcdP+BghiP) and Flua/(Flua+Pry) for E4 were 0.86, 1.0 and 0.87, respectively~~, and it were~~. These were different ~~with~~ from the ranges for fuel combustion defined by Yunker~~s~~ et al. (2002). The ratios of PAHs emitted from diesel vehicles reported by Yunker~~s~~ et al. (2002) mainly referred to those from on-road diesel vehicles. However, the ~~operating~~ operational mode~~s~~ and fuel quality for non-road diesel vehicles ~~are~~ are more complicated than those ~~from~~ for on-road diesel vehicles. Therefore, ~~the results~~ in this study ~~could give~~provides references values for the isomer ratios of PAHs ~~for~~ in non-road diesel vehicle exhaust~~s~~.

3.3.2 Particulate matter composition for individual diesel trucks

For diesel trucks, the total carbonaceous composition (OM+EC) ~~were~~ accounted for 44.0% (T~~E~~1), 27.9% (~~E2~~T2), 43.9% (~~E3~~T3), 51.6% (~~E4~~T4) and 46.3% (~~E5~~T5) of PM, which ~~is~~ are all lower than ~~those~~ the values reported in previous studies (Chow et al., 2011, Wu et al., 2015) because of ~~. The reason may be mainly attributed to~~was ~~–~~the different OC and EC detection methods used in our study~~for organic carbon and elements carbon~~. Cheng et al. (2011) collected 333 $PM_{2.5}$ samples and analyzed OC and EC by two common thermal-optical methods (NIOSH and IMPROVE). They~~and~~ found that NIOSH-defined EC was lower (up to 80%) than that defined by IMPROVE. The IMPROVE thermal-optical method was used in this study~~was IMPROVE~~, which would cause under valuation ~~make content~~ of OC~~under evaluated~~.~~–~~ Except for the T2 and T4 trucks, ~~Almost~~ almost all of the OC/EC ratios for diesel trucks ~~under different driving conditions~~ calculated in this study were lower than 1, which ~~was~~ is consistent with~~the~~ conclusions from previous studies (Figure 6)~~, except for the T2 and T4 trucks~~. The OC/EC ratios for T2 ~~under~~ during highway and non-highway driving ~~conditions~~ were 5.64 and 15.5, respectively,~~.~~ ~~This result may be attributed to~~ which may be a result of the China IV emission standard for.~~T2 (China IV).~~ Alves et al. (2015b) reported that modern diesel passenger cars (Euro 4 and Euro 5) ~~exhibit~~ have high OC/EC ratios. As shown in Figure S3, the driving speed for T4 was zero for the first 500 seconds. Cheng et al. (2015) reported that the OC/EC ratios were substantially above 1 while idling or with low load. ~~Furthermore~~Therefore, the OC/EC ratio for T4 while driving on the ~~under~~ non-highway ~~condition~~ was 4.10, which ~~may~~ might ~~be~~ have been caused by the low driving speed. ~~Cheng et al. (2015) reported that the OC/EC ratios were substantially above unity at idling and low load. As shown from Figure S2, the driving speed for T4 was zero in 500 seconds before driving.~~

The ~~Sum~~ sum of ~~WISs~~ WSIs and elements fractions were lower than 5% for the exhaust from ~~almost~~ all of the diesel trucks, except for that from T2~~ truck~~, which is consistent with the results ~~gained from~~in Zhang et al. (2015a). $SO_4^{2-}$ was the most abundant ion~~s~~ for trucks T2 and T5, while $NO_3^-$ was the most abundant ion~~s~~ for trucks T1, T3 and T4. For T2~~ diesel truck~~, ~~WISs~~ WSIs (13.8%) ~~was~~ were the most significant component of PM, ~~after~~ followed by OC, ~~and~~ which ~~it~~ was ~~higher by a factor of~~ 4 to 10 times higher than it was for the ~~those in~~ other trucks (Table ~~S2~~S6). T2 ~~truck is~~ was a China IV diesel vehicle ~~and~~ with well-controlled combustion conditions ~~caused~~ leading to more water emissions, which accelerates the ~~translation~~ transformation from the gas phase to ~~WISs~~ WSIs (~~such as~~e.g., ~~$SO_2$ translate~~ the transformation of $SO_2$ to $SO_4^{2-}$). As ~~we~~ can~~can~~ be seen ~~from~~ in Table ~~S2~~S6, Fe was the most abundant element for trucks T1~~, T3~~ and T5, while Ca was the most abundant ~~the most abundant element~~ for trucks ~~T1~~T2, T3, and T4. The total element fraction of T2 (China Ⅳ) was 16 times higher than that of T1 exhaust (China Ⅲ)~~Compared with elements fractions in T2 (China IV) and T1 (China III) trucks, fractions changed from 0.09% (T2) to 1.5% (T1)~~. Although the ~~PM emission factors~~$EF_{PM}$ for diesel trucks decreased with stricter emission standards, the ~~WISs~~ WSIs and element~~s~~ contents ~~increasing~~ increased ~~along with promoting the emission standards for diesel trucks~~. ~~In consideration of~~Because acid rain is ~~causing~~ caused by sulfate and nitrate and adverse health effects are caused by elements, ~~great~~ attention ~~should~~ needs to be ~~pay~~ paid to this phenomenon.

The n-alkanes, PAHs, hopane and steranes fractions ~~ranged from~~were 0.85~~% to~~-4.78%, ~~from~~ 0.01~~% to~~-0.54% and ~~from~~ 0.002~~% to~~- 0.024%, for the trucks~~, respectively~~. As shown in Table ~~S2~~S6, C20 was the most abundant ~~species in~~ n-alkane~~s~~ ~~for truck~~in exhaust from T1, T2 and T4, while C19 was the most abundant n-alkane in exhaust from ~~species for truck~~ T3 and T5. ~~For~~ Of the PAHs, the most ~~notable~~ abundant species was ~~Pyrene~~pyrene~~, which was substantially higher than all other PAHs for all trucks~~. ~~The proportions of n~~N-alkanes, PAHs, hopane and steranes accounted for the highest proportions ~~to~~ of PM ~~were highest~~ for the exhaust from ~~truck~~ T3, ~~which~~ and ~~may~~ might be affected by many factors, ~~such as~~including differences in the engine ~~rate~~ power rating, complex reactions in the engine (combustion process and pyrolysis reactions related to temperature, humidity, etc.), and driving conditions. As shown ~~from~~ in Figure 7, ~~scatters of~~the isomer ratios for diesel trucks ~~were covered from~~were 0.28~~ to ~~-0.35 for BaA/(BaA+Chry), ~~from~~ 0.08~~ to~~ -0.22 for IcdP/(IcdP+BghiP) and~~ from~~ 0.08~~ to ~~-0.39 for Flua~~s~~/(Flua+Pry), with averages of 0.31 ± 0.03, 0.15 ± 0.06 and 0.23 ± 0.12, respectively. ~~There~~ These ~~were~~ are similar to ~~the~~ results ~~from~~ reported by Schauer et al. (1999).

**3.4 Average chemical ~~constituent~~ composition of PM emitted from diesel vehicles**

3.4.1 Average chemical composition ~~constituent~~ of PM ~~for~~ in excavator exhaust~~s~~

The average PM chemical ~~component~~ compositions ~~of PM~~ for excavator exhaust~~s~~ ~~was~~ are listed in Table 3. ~~It appeared that c~~Carbonaceous matter ~~was~~ was the dominant component and accounted for 72.5% of the PM for excavators, whereas OC was the most abundant species (39.2%) for PM. ~~Total~~ The total element fraction was the second largest group and contributed 1.76% of PM. ~~For~~ Of the elements, ~~the~~ emissions ~~was~~ were ~~obviously~~ dominated by Fe at~~ which accounted for~~ 46.3%~~ of the elements~~. In addition, ~~Table 3 showed that~~ the proportion of n-alkanes in PM ~~for~~ from excavator exhaust~~s~~ (5.14%) was higher than the proportions of the ~~those for~~ other organic matter types~~s~~ (PAHs were ~~:~~ 0.098% while ~~;~~ hopane and sterane were ~~:~~ 0.026%)~~,~~ and C20~~ and~~ / C19 ~~were~~ was the most abundant ~~maximum carbon in~~ n-alkane~~s~~. For~~ the~~ parent PAH~~s~~, ~~the~~ emissions were dominated by Pry and Fluo, followed by Nap and Chry.

~~To compare our results with other studies,~~ Table 3 summarizes~~s~~ the average source profiles of PM ~~for~~ in excavator exhaust as~~s~~ derived in this study, as well as ~~those~~ ones previously reported by others for comparison. As shown in Table 3, the average fraction of total carbonaceous components for the excavators tested in this study ~~are~~ was consistent with that~~ose~~ for a ~~measured from~~ marine engine, while the element fraction ~~of elements~~ was lower than that for a marine engine (Sippula et al., 2014). Iron oxide is recognized as a catalyst and can promote soot burnout during combustion processes (Kasper et al., 1999).~~It is said that oxidation of soot was enhanced during increasing of transition metals for diesel engines (Kasper et al., 1999).~~ The EC fraction of PM in the~~PM~~ ~~for~~ excavator exhaust~~s~~ was higher than that

~~those from~~ reported by Sippula et al. (2014), which ~~may~~ might ~~be attributed~~ be the result of a ~~to~~ lower metal fraction in the excavators used for their study. ~~Comparing results from this study with other references showed that t~~The proportions of n-alkanes measured in this study ~~is~~ were significantly higher than those emitted from a marine engine (4-fold) and non-road generator~~s~~ (6-fold) in another study ~~(Liang et al., 2005).~~ (Liang et al., 2005), which could be the result of ~~The reason may be attributed to~~ different ~~contents of~~ aliphatic compounds ~~existing~~ in the diesel fuels ~~used for those non-road vehicles~~ (Sippula et al., 2014). For the marine engine and non-road generators, C22 and C17 were the most abundant n-alkane species. ~~in n-alkanes, respectively.~~ PAHs ~~emission was~~were dominanted by Phe for a marine engine and Fluo for non-generators, which was different ~~with~~ from the result obtained ~~from~~ for the excavators. This could indicate that the PM emitted from different types of non-road diesel vehicles has ~~various~~ varying source profiles ~~because of~~ based on the ~~diverse~~ operational conditions.

3.4.2 Average source profile of PM for trucks

As shown in Table 3, ~~average emission of~~ PM from trucks was dominated by carbonaceous matter (36.8%),~~;~~ ~~and~~ followed by ~~WISs~~ WSIs (4.67%) and elements (0.941%). For individual species, sulfate and nitrate were the most abundant ~~in water soluble ions~~WSIs, and Fe was the most abundant ~~dominated in~~ element~~s~~. Moreover, for organic matter~~s~~, the average proportions of n-alkanes, PAHs, hopanes and steranes ~~was~~ were 1.73%, 0.130%, and 0.011%, respectively. C20 was the ~~maximum~~ most abundant ~~carbon in~~ n-alkane~~s~~, and the ~~emission of~~ PAHs ~~was~~ were dominated by Pry.

In comparison, ~~emission of~~ total carbon emissions ~~from~~ in this study ~~was~~ were lower than those in previous studies, whereas~~,~~ the ~~WISs~~ WSIs and elements fractions were ~~relatively~~ higher ~~than results obtained from other research groups~~ (Alves et al., 2015a, Cui et al., 2016, Schauer et al., 1999, Wu et al., 2016). ~~There are s~~Several ~~reasons~~ factors could have influenced these differing ~~be used to explain the~~ results, including fuel quality, driving condition, ~~parameters of~~ engine parameters~~s~~ (fuel injection timing, compression ratio, and fuel injector design) and experimental methods (Sarvi et al., 2008a, Sarvi et al., 2008b, Sarvi et al., 2009, Sarvi et al., 2010).

As shown in Table 3, Fe was the dominant ~~in~~ element~~s~~ ~~from~~ in ~~results measured by~~studies using on-road tests and tunnels, which ~~was~~ is similar ~~with~~ to our results, while Zn and Na were dominant in elements from results obtained by a dynamometer. Therefore, the results obtained from real world (on-road tests and tunnels) were different from those obtained in a laboratory. ~~on-road test and tunnels measured in real world would reflect real PM emission better.~~ For organic matter~~s~~, the proportion of PAHs, hopane and sterane to PM were consistent with the results from Schauer et al. (1999) and Cui et al. (2016). ~~Similar with~~As in this study, the ~~maximum carbon~~most abundant in n-alkane~~s~~ was C20 as measured by Schauer et al. (1999), and Pyr was the most abundant ~~species in~~ PAH~~s~~ reported by Cui et al. (2016). Thus, the average profile of PM for on-road diesel trucks ~~was~~ is relatively stable and consistent across studies.

### 3.5 ~~Comparing average s~~Source profile~~s~~ comparison ~~from~~ for excavators ~~with those from~~and trucks

~~Compared with the average $EF_{PM}$ of excavators and diesel trucks obtained in this study, a~~Average $EF_{PM}$ for excavators (836 ± 801 mg kg$^{-1}$ fuel) was higher than ~~those~~ that for diesel trucks (498 ± 234 mg kg$^{-1}$ fuel). ~~The~~ This result ~~was~~ is understandable because ~~state~~ the operations for excavators ~~were~~ are more transient than those for trucks. Sarvi et al. (2010) reported that particulate matter ~~emission~~ emitted from diesel engines was typically low during steady state operation. Although the average $EF_{PM}$ of excavators was higher than that ~~emitted from~~of trucks, the average $EF_{PM}$ of the stage 2 excavators was 477 mg kg$^{-1}$ fuel, which was lower than those for the ~~that emitted by~~ China II and China III trucks. Thus, appropriate regulations formulated for non-road diesel vehicles could improve their PM emissions ~~situation~~.

When we compared the average percentages of chemical components in PM for excavators with those for trucks, ~~we found that there were some~~several differences were found~~between excavators and trucks~~. In general, the carbonaceous ~~composition~~ (95.9%) and elements (1.76%) fractions for excavators were higher than those for diesel trucks (42.8% ~~for carbonaceous composition~~ and 0.94%, ~~for~~

~~elements~~respectively). As shown in Figure 8, ~~the~~ the structures of different ring PAHs in the exhaust from excavators and trucks varied sharply, especially for 5 and 6-ring PAHs~~BaP$_{eq}$ levels for excavators and trucks were absolutely difference~~, although the average percentage of total PAHs ~~average percentages of~~in the PM were consistent between the excavators and trucks. ~~Almost all of the parent PAHs's BaP$_{eq}$ calculated in this study for trucks and excavators were higher than the datum from WHO that concentration caused 1/10000 risk of carcinogenic.~~ Due to their lipophicity, high molecular weight (5+6 ring) PAHs are considered to be more harmful to human health than the other PAHs. For further distinction, BaPeq was used in this study. The range of total BaPeq for trucks was 5.32 (T5) to 155 (T3) ng m$^{-3}$, while for excavators, the range of total BaPeq was 38.3 (E1) to 3637 (E4) ng m$^{-3}$. Moreover, the total~~-~~ average ~~of~~ BaPeq for the excavators was 31 ~~fold~~ times larger than that ~~of those~~ for the diesel trucks. Almost all of the parent PAHs~~'s~~ BaP$_{eq}$ values calculated in this study for trucks and excavators were higher than the ~~datum~~concentrations ~~from WHO that concentration~~ caused 1/10000 of the ~~risk of~~ carcinogenic risk~~.~~, according to the World Health Organization (WHO). Due to the ~~some~~ adverse environmental effects and health hazards caused by ~~for~~ carbonaceous composition, elements, and PAHs, the PM emissions from excavators require urgent ~~should be~~ control~~led urgently~~.

**Conclusions**

This study report~~s~~ed the characteristics of PM source profiles for excavators and the EF$_{PM}$ ~~trucks. Above all, PM emission factors~~ values for exhaust ~~emitted~~ from excavators and trucks with different emission standards and used ~~under~~ in different ~~operating~~ operational modes, ~~emission standards and~~or road conditions were obtained. The ~~The~~ EF$_{PM}$ for different excavators ranged from 96.5 to 2323 mg kg$^{-1}$ fuel, with an average of 810 mg kg$^{-1}$ fuel and showed a high correlation ($R^2$=0.79, P<0.01) with the fuel sulfur contents~~ in the fuel~~. The highest average EF$_{PM}$ for excavators that are ~~in~~ working ~~mode~~ (904 ± 979 mg kg$^{-1}$ fuel) might be ~~attributed to~~ the result of higher engine load~~, which~~ ~~caused~~ causing lower air-fuel ratios. The average EF$_{PM}$ for the tested diesel trucks ~~of~~ with different emission standards and vehicle sizes under different driving conditions was 498 ± 234 mg kg$^{-1}$ fuel. The average EF$_{PM}$ for excavators with different emission standards ~~excavators~~ decreased by 58% from pre-stage 1 to stage 2. Moreover, the reductions in $EF_{PM}$ from the China II ~~truck~~ to the China IV truck and from the China III ~~truck~~ to the China IV truck ~~in $EF_{PM}$~~ were 63.5~~%~~ and 65.6%, respectively~~.~~, ~~Those~~ ~~I~~indicat~~e~~ing that improvements ~~of~~ to the emission standards for diesel trucks and excavators have significantly decreased PM emissions ~~significantly~~. It should be noticed that the $EF_{PM}$ for China III and light-duty diesel trucks were higher than those for the other ~~corresponding~~ trucks~~.~~, which ~~The~~ could be a result of ~~reasons may be attributed to~~ poor driving conditions that included a low average and highly variable speed ~~and more volatile in speed for those trucks~~. For each excavator, the carbon component (OM+EC) was ~~the~~ dominant ~~species~~ fraction and accounted for approximately 74.1-123% of the PM. The average ranges of ~~WISs~~WSIs, elements, n-alkanes, PAHs, hopane and sterane fractions for each excavator were 0.335~~%~~-1.21%, 0.163~~%~~-7.50%, 3.6~~%~~-9.6%, 0.03~~%~~-0.24% and 0.001~~%~~-0.09%, respectively. In contrast to the other excavators, Zn and Cu were the second and third most abundant elements in ~~excavators~~ exhaust from E4, which ~~may~~ might ~~be attributed to~~to the result of poor fuel quality and the ~~old vehicles~~ vehicle age. ~~Besides~~Additionally, the element~~s~~ fractions for the two excavators produced in 2013 (E1 (1.42%) and E6 (7.50%)) were higher than other excavators, which ~~may~~ might indicate that elements emissions control ~~was deteriorating~~ deteriorated and more stringent control technology should be developed ~~to avoid the total elements adverse health effects~~. For excavators, the ranges of the ratios ~~of~~ BaA/(BaA+Chry), IcdP/(IcdP+BghiP) and Flua/(Flua+Pry) were 0.26-0.86, 0.20-1.0 and 0.24-0.87, respectively, with average of 0.47 ± 0.27, 0.44 ± 0.38 and 0.48 ± 0.27, respectively. For diesel trucks, the total carbonaceous composition (OM+EC) ~~were~~ accounted for 44.0% (~~E1~~T1), 27.9% (~~E2~~T2), 43.9% (~~E3~~T3), 51.6% (~~E4~~T4) and 46.3% (~~E5~~T5) of PM. For T2 ~~diesel truck~~, ~~wSIs~~ WSIs (13.8%) ~~was~~ were the most significant ~~component~~ fraction of PM after OC, and it was higher than those ~~in~~ for the other trucks by~~, within~~ a factor of 4 to 10. The n-alkanes, PAHs, hopane and steranes fractions ranged from 0.85~~%~~ to 4.78%, ~~from~~ 0.01~~%~~ to 0.54% and ~~from~~ 0.002~~%~~ to 0.024% for trucks, respectively. In comparison with the results from other

~~literatures~~studies, the characteristics of the average source profiles for different types of non-road diesel vehicles varied sharply, while those for on-road diesel vehicles, ~~those characteristics~~ showed more stability. Although the PAHs fractions ~~of PAHs~~ for the excavators and trucks were ~~identical~~similar, the total ~~of~~ $BaP_{eq}$ that was used to evaluate the carcinogenic risk was 31 times greater for excavators than ~~fold of those~~ for trucks.

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## Reference

Adar, S.D., D'Souza, J., Sheppard, L., Kaufman, J.D., Hallstrand, T.S., Davey, M.E., Sullivan, J.R., Jahnke, J., Koenig, J., Larson, T.V., Liu, L.J.S.: Adopting Clean Fuels and Technologies on School Buses. Pollution and Health Impacts in Children, American Journal of Respiratory and Critical Care Medicine. 191(12): 1413-1421 2015

Aggarwal, P., Jain, S.: Impact of air pollutants from surface transport sources on human health: A modeling and epidemiological approach, Environment International. 83: 146-157 2015

Almeida, S.M., Pio, C.A., Freitas, M.C., Reis, M.A., Trancoso, M.A.: Approaching $PM_{2.5}$ and $PM_{2.5-10}$ source apportionment by mass balance analysis, principal component analysis and particle size distribution, Science of the Total Environment. 368(2-3): 663-674 2006

Alves, C.A., Barbosa, C., Rocha, S., Calvo, A., Nunes, T., Cerqueira, M., Pio, C., Karanasiou, A., Querol, X.: Elements and polycyclic aromatic hydrocarbons in exhaust particles emitted by light-duty vehicles, Environmental Science and Pollution Research. 22(15): 11526-11542 2015a

Alves, C.A., Lopes, D.J., Calvo, A.I., Evtyugina, M., Rocha, S., Nunes, T.: Emissions from Light-Duty Diesel and Gasoline In-Use Vehicles Measured on Chassis Dynamometer Test Cycles, Aerosol and Air Quality Research. 15(1): 99-116 2015b

CCCMIY (Compiling committee of China machinery industry yearbook), CCMA (China Construction Machinery Association): China construction machinery industry yearbook. China Machine Press Place, Published, 2013.

Chen, F., Hu, W., Zhong, Q.: Emissions of particle-phase polycyclic aromatic hydrocarbons (PAHs) in the Fu Gui-shan Tunnel of Nanjing, China, Atmospheric Research. 124: 53-60 2013

Cheng, M.T., Chen, H.J., Young, L.H., Yang, H.H., Tsai, Y.I., Wang, L.C., Lu, J.H., Chen, C.B.: Carbonaceous composition changes of heavy-duty diesel engine particles in relation to biodiesels, aftertreatments and engine loads, Journal of Hazardous Materials. 297: 234-240 2015

Cheng, Y., Zheng, M., He, K.-b., Chen, Y., Yan, B., Russell, A.G., Shi, W., Jiao, Z., Sheng, G., Fu, J., Edgerton, E.S.: Comparison of two thermal-optical methods for the determination of organic carbon and elemental carbon: Results from the southeastern United States, Atmospheric Environment. 45(11): 1913-1918 2011

Chow, J.C., Watson, J.G., Lowenthal, D.H., Antony Chen, L.W., Motallebi, N.: $PM_{2.5}$ source profiles for black and organic carbon emission inventories, Atmospheric Environment. 45(31): 5407-5414 2011

Cocker, D.R., Shah, S.D., Johnson, K.C., Zhu, X.N., Miller, J.W., Norbeck, J.M.: Development and application of a mobile laboratory for measuring emissions from diesel engines. 2. Sampling for toxics and particulate matter, Environmental Science & Technology. 38(24): 6809-6816 2004

Cui, M., Chen, Y., Tian, C., Zhang, F., Yan, C., Zheng, M.: Chemical composition of $PM_{2.5}$ from two tunnels with different vehicular fleet characteristics, Science of The Total Environment. 550: 123-132 2016

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 $PM_{2.5}$ 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

Fu, M.L., Ge, Y.S., Tan, J.W., Zeng, T., Liang, B.: Characteristics of typical non-road machinery emissions in China by using portable emission measurement system, Science of the Total Environment. 437: 255-261 2012

Huang, C., Chen, C.H., Li, L., Cheng, Z., Wang, H.L., Huang, H.Y., Streets, D.G., Wang, Y.J., Zhang, G.F., Chen, Y.R.: Emission inventory of anthropogenic air pollutants and VOC species in the Yangtze River Delta region, China, Atmospheric Chemistry and Physics. 11(9): 4105-4120 2011

Huo, H., Yao, Z.L., Zhang, Y.Z., Shen, X.B., Zhang, Q., He, K.B.: On-board measurements of emissions from diesel trucks in five cities in China, Atmospheric Environment. 54: 159-167 2012

IARC (International Agency for Research on Cancer): Diesel Engine Exhaust Carcinogenic, 2012.

Kasper, M., Sattler, K., Siegmann, K., Matter, U., Siegmann, H.C.: The influence of fuel additives on the formation of carbon during combustion, Journal of Aerosol Science. 30(2): 217-225 1999

Li, D.l., Wu, Y., Zhou, Y., Du, X., Fu, L.x.: Fuel Consumption and Emission Inventory of Typical Construction Equipments in China, Huanjing Kexue. 33(2): 518-524 2012

Liang, F.Y., Lu, M.M., Keener, T.C., Liu, Z.F., Khang, S.J.: The organic composition of diesel particulate matter, diesel fuel and engine oil of a non-road diesel generator, Journal of Environmental Monitoring. 7(10): 983-988 2005

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

Liu, H., He, K.B., Lents, J.M., Wang, Q.D., Tolvett, S.: Characteristics of Diesel Truck Emission in China Based on Portable Emissions Measurement Systems, Environmental Science & Technology. 43(24): 9507-9511 2009

Liu, L.-Y., Wang, J.-Z., Wei, G.-L., Guan, Y.-F., Zeng, E.Y.: Polycyclic aromatic hydrocarbons (PAHs) in continental shelf sediment of China: Implications for anthropogenic influences on coastal marine environment, Environmental Pollution. 167: 155-162 2012

Liu, Z.F., Lu, M.M., Birch, M.E., Keener, T.C., Khang, S.J., Liang, F.Y.: Variations of the particulate carbon distribution from a nonroad diesel generator, Environmental Science & Technology. 39(20): 7840-7844 2005

Liu, Z.G., Wall, J.C., Ottinger, N.A., McGuffin, D.: Mitigation of PAH and Nitro-PAH Emissions from Nonroad Diesel Engines, Environmental Science & Technology. 49(6): 3662-3671 2015

Longhin, E., Gualtieri, M., Capasso, L., Bengalli, R., Mollerup, S., Holme, J.A., Ovrevik, J., Casadei, S., Di Benedetto, C., Parenti, P., Camatini, M.: Physico-chemical properties and biological effects of diesel and biomass particles, Environmental Pollution. 215: 366-375 2016

Mancilla, Y., Mendoza, A.: A tunnel study to characterize $PM_{2.5}$ emissions from gasoline-powered vehicles in Monterrey, Mexico, Atmospheric Environment. 59(0): 449-460 2012

MEP (Ministry of Environmental Protection of China): The technical guide for compiling primary source emission inventories of atmospheric fine particulate matter, 2014.

MEP (Ministry of Environmental Protection of China): China vehicle environmental management annual report, 2016.

MEPPRC (Ministry of Environmental Protection of the People’s Republic of China): Technical guide for the preparation of a single source emission inventory of atmospheric fine particulate matter**:** 12, 2014.

Mirante, F., Alves, C., Pio, C., Pindado, O., Perez, R., Revuelta, M.A., Artinano, B.: Organic composition of size segregated atmospheric particulate matter, during summer and winter sampling campaigns at representative sites in Madrid, Spain, Atmospheric Research. 132: 345-361 2013

Oanh, N.T.K., Thiansathit, W., Bond, T.C., Subramanian, R., Winijkul, E., Paw-armart, I.: Compositional characterization of $PM_{2.5}$ emitted from in-use diesel vehicles, Atmospheric Environment. 44(1): 15-22 2010

Pietikainen, M., Valiheikki, A., Oravisjarvi, K., Kolli, T., Huuhtanen, M., Niemi, S., Virtanen, S., Karhu, T., Keiski, R.L.: Particle and NOx emissions of a non-road diesel engine with an SCR unit: The effect of fuel, Renewable Energy. 77: 377-385 2015

Pio, C., Mirante, F., Oliveira, C., Matos, M., Caseiro, A., Oliveira, C., Querol, X., Alves, C., Martins, N., Cerqueira, M., Camões, F., Silva, H., Plana, F.: Size-segregated chemical composition of aerosol emissions in an urban road tunnel in Portugal, Atmospheric Environment. 71(0): 15-25 2013

Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T.: Sources of fine organic aerosol. 2. Noncatalyst and catalyst-equipped automobiles and heavy-duty diesel trucks, Environmental Science & Technology. 27(4): 636-651 1993

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

Sarvi, A., Fogelholm, C.-J., Zevenhoven, R.: Emissions from large-scale medium-speed diesel engines: 1. Influence of engine operation mode and turbocharger, Fuel Processing Technology. 89(5): 510-519 2008a

Sarvi, A., Fogelholm, C.J., Zevenhoven, R.: Emissions from large-scale medium-speed diesel engines: 2. Influence of fuel type and operating mode, Fuel Processing Technology. 89(5): 520-527 2008b

Sarvi, A., Kilpinen, P., Zevenhoven, R.: Emissions from large-scale medium-speed diesel engines: 3. Influence of direct water injection and common rail, Fuel Processing Technology. 90(2): 222-231 2009

Sarvi, A., Zevenhoven, R.: Large-scale diesel engine emission control parameters, Energy. 35(2): 1139-1145 2010

Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T.: Measurement of emissions from air pollution sources. 2. C-1 through C-30 organic compounds from medium duty diesel trucks, Environmental Science & Technology. 33(10): 1578-1587 1999

SEPA (State Environmental Protection Administration), SAQSIQ (State Administration for Quality Supervision and Inspection and Quarantine): Limits and measurement methods for emissions from light-duty vehicles (I), 2001.

SEPA (State Environmental Protection Administration), SAQSIQ (State Administration for Quality Supervision and Inspection and Quarantine ): Limits and measurements methods for exhaust pollutants from diesel engines of non-road mobile machinery (I, II), 2007.

SEPA (State Environmental Protection Administration), SAQSIQ (State Administration for Quality Supervision and Inspection and Quarantine): General diesel fuels, 2015.

Shah, S.D., Ogunyoku, T.A., Miller, J.W., Cocker, D.R.: On-road emission rates of PAH and n-alkane compounds from heavy-duty diesel vehicles, Environmental Science & Technology. 39(14): 5276-5284 2005

Sippula, O., Stengel, B., Sklorz, M., Streibel, T., Rabe, R., Orasche, J., Lintelmann, J., Michalke, B., Abbaszade, G., Radischat, C., Groger, T., Schnelle-Kreis, J., Harndorf, H., Zimmermann, R.: Particle Emissions from a Marine Engine: Chemical Composition and Aromatic Emission Profiles under Various Operating Conditions, Environmental Science & Technology. 48(19): 11721-11729 2014

USEPA (Environmental Protection Agency): Nonroad Engine and Vehicle Emission Study, 1991.

USEPA (Environmental Protection Agency): Air Radiation. Reducing Air Pollution from Nonroad Engines, http://www.epa.gov/otaq/cleaner-nonroad/f03011.pdf.

USEPA (Environmental Protection Agency) Evaluation of Black Carbon Emission Reductions from Mining Trucks in Russia: The Case of the Murmansk Region, 2015.

Wang, F., Li, Z., Zhang, K.S., Di, B.F., Hu, B.M.: An overview of non-road equipment emissions in China, Atmospheric Environment. 132: 283-289 2016

Wang, X., Wang, W., Liu, X., Cheng, H., Xu, X.: Health risk assessment of PAHs in fine particulate matter in Beijing Environmental chemistry. 2008

Wang, X.W., Song, C.L., Lv, G., Song, J.N., Li, H., Li, B.: Evolution of in-cylinder polycyclic aromatic hydrocarbons in a diesel engine fueled with n-heptane and n-heptane/toluene, Fuel. 158: 322-329 2015

Wang, Y.-F., Huang, K.-L., Li, C.-T., Mi, H.-H., Luo, J.-H., Tsai, P.-J.: Emissions of fuel metals content from a diesel vehicle engine, Atmospheric Environment. 37(33): 4637-4643 2003

Wu, B., Shen, X., Cao, X., Yao, Z., Wu, Y.: Characterization of the chemical composition of PM2.5 emitted from on-road China III and China IV diesel trucks in Beijing, China, Science of The Total Environment. 551–552: 579-589 2016

Wu, B., Shen, X., Cao, X., Zhang, W., Wu, H., Yao, Z.: Carbonaceous composition of $PM_{2.5}$ emitted from on-road China III diesel trucks in Beijing, China, Atmospheric Environment. 116: 216-224 2015

Yu, W., Yang, T.: Formation mechanism and technical measured for reducing emissions of diesel engine, Transportation energy saving and environmental protection. 2007

Yue, X., Wu, Y., Hao, J.M., Pang, Y., Ma, Y., Li, Y., Li, B.S., Bao, X.F.: Fuel quality management versus vehicle emission control in China, status quo and future perspectives, Energy Policy. 79: 87-98 2015

Yunker, M.B., Macdonald, R.W., Vingarzan, R., Mitchell, R.H., Goyette, D., Sylvestre, S.: PAHs in the Fraser River basin: a critical appraisal of PAH ratios as indicators of PAH source and composition, Organic Geochemistry. 33(4): 489-515 2002

Zhang, L.J., Zheng, J.Y., Yin, S.S., Peng K., Zhong, L.J., : Development of Non-road Mobile source Emission inventory for the Pearl River Delta Region, Environmental Scence 31(4): 886-891 2010

Zhang, Y., Yao, Z., Shen, X., Liu, H., He, K.: Chemical characterization of $PM_{2.5}$ emitted from on-road heavy-duty diesel trucks in China, Atmospheric Environment. 122: 885-891 2015a

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 2015b

Zhang, Z.-H., Balasubramanian, R.: Influence of butanol–diesel blends on particulate emissions of a non-road diesel engine, Fuel. 118: 130-136 2014

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

**Table captions**

Table 1 Specifications ~~of~~ for the tested excavators and trucks

Table 2 Diesel contents from excavators

Table 3 Comparison of average chemical constituents of PM for different diesel vehicles

Table 1 Specifications of tested excavators and trucks

| ID | ~~manufacturers~~Manufacturers | Model years | Emission standards | Powers (kw) | Total weights (kg) | Displacements (L) | Working hours (h) | Mileages (km) |
|---|---|---|---|---|---|---|---|---|
| E1 | Volvo | 2013 | stage 2 | 169 | 30,500 | 7.1 | 2,751 | / |
| E2 | Hitachi | 2007 | pre-stage 1 | 162 | 30,200 | 9.8 | 16,166 | / |
| E3 | Sany | 2012 | stage 2 | 128 | 22,900 | / | 5,598 | / |
| E4 | Doosan | 2004 | pre-stage 1 | 110 | 22,000 | 8.1 | 12,000 | / |
| E5 | Doosan | 2007 | pre-stage 1 | 40 | 5,250 | 2.8 | / | / |
| E6 | Komatsu | 2013 | stage 2 | 35 | 5,300 | 2.4 | 780 | / |
| T1 | Futian | 2010 | China III | 68 | 4,495 | 2.6 | / | 100,238 |
| T2 | JAC | 2014 | China IV | 88 | 4,495 | 2.8 | / | / |
| T3 | Futian | 2011 | China III | 70 | 11,190 | 3.9 | / | 99,000 |
| T4 | Chunlan | 2002 | China II | 125 | 15,480 | / | / | / |
| T5 | JAC | 2011 | China III | 105 | 15,590 | 4.3 | / | 130,000 |

Table 2 Diesel contents from excavators

| ID | E1 | E2 | E3 | E4 | E5 | E6 | GB 252-2015 |
|---|---|---|---|---|---|---|---|
| Gross thermal value (MJ/kg) | 45.1 | 45.1 | 45.3 | 45.3 | 45.3 | 45.3 | / |
| Net thermal value (MJ/kg) | 42.4 | 42.4 | 42.7 | 42.8 | 42.6 | 42.5 | / |
| Kinematic viscosity (20 °C)($mm^2/s$) | 4.23 | 4.23 | 3.89 | 4.16 | 4.60 | 4.39 | 3.00-8.00 |
| Moisture (%) | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | / |
| Ash content (%) | 0.04 | 0.04 | 0.05 | 4.16 | 0.03 | 0.05 | 0-0.01 |
| C (%) | 86.3 | 86.3 | 86.4 | 86.8 | 85.9 | 85.9 | / |
| H (%) | 11.6 | 11.6 | 11.5 | 11.2 | 12.0 | 12.1 | / |
| O (%) | 1.99 | 1.99 | 2.01 | 1.85 | 2.07 | 1.86 | / |
| N (%) | 0.05 | 0.05 | 0.05 | 0.04 | 0.06 | 0.05 | / |
| S (ppm) | 400 | 400 | 700 | 1100 | 200 | 200 | <350 |

n.d. = not detected

Table 3 Comparison of average chemical constituents of PM for different diesel vehicles (%)

| Vehicle types | Excavators | Trucks | Trucks | Medium-duty trucks | Diesel vehicles | Light-duty ~~Diesel~~ diesel engines | Marine engine | Non-road generator |
|---|---|---|---|---|---|---|---|---|
| Methods | On-road | | On-road | Dynamometer | Tunnel | Dynamometer | Dynamometer | Dynamometer |
| Reference | This study | | (Wu et al., 2016) | (Schauer et al., 1999) | (Cui et al., 2016) | (Alves et al., 2015b) | (Sippula et al., 2014) | (Liang et al., 2005) |
| **EC** | 33.3 | 26.9 | 55.3 | 30.8 | 39.5 | 69.9 | 14.1 | |
| **OC** | 39.2 | 9.89 | 31.8 | 19.7 | 27.2 | 12.7 | 60.0 | |
| **Ions** | 0.614 | 4.67 | 1.49 | 1.96 | 11.7 | 0.638 | | |
| $NH_4^+$ | 0.044 | 0.215 | 0.188 | 0.730 | 2.06 | 0.005 | | |
| $Cl^-$ | 0.098 | 0.110 | 0.247 | | 1.06 | 0.115 | | |
| $NO_3^-$ | 0.278 | 1.08 | 0.529 | 0.230 | 3.81 | 0.459 | | |
| $SO_4^{2-}$ | 0.193 | 3.27 | 0.529 | 1.00 | 4.80 | 0.059 | | |
| **Elements** | 1.76 | 0.941 | 0.493 | 0.200 | 12.8 | 0.069 | 3.17 | |
| Na | 0.245 | 0.047 | | | 0.287 | 0.041 | 0.564 | |
| Mg | 0.106 | 0.079 | | | 1.71 | 0.008 | 0.422 | |
| K | 0.197 | 0.028 | | | 0.872 | 0.002 | 0.671 | |
| Ca | 0.241 | 0.211 | | 0.030 | 5.69 | 0.017 | 1.01 | |
| Ti | 0.008 | 0.011 | 0.145 | | 0.206 | 0.0001 | 0.005 | |
| V | 0.001 | 0.000 | 0.001 | | 0.008 | | 0.044 | |
| Cr | 0.035 | 0.039 | 0.011 | 0.010 | 0.013 | | 0.010 | |
| Mn | 0.013 | 0.009 | 0.002 | 0.010 | 0.064 | | 0.006 | |

Continued Table 3

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Fe | 0.815 | 0.276 | 0.247 | 0.050 | 3.71 | 0.0003 | 0.138 | |
| Co | 0.001 | 0.005 | 0.0002 | 0.010 | 0.002 | | 0.006 | |
| Ni | 0.015 | 0.006 | 0.002 | nd | | | 0.016 | |
| Cu | 0.042 | 0.107 | 0.004 | 0.010 | 0.013 | | 0.130 | |
| Zn | 0.027 | 0.111 | 0.076 | 0.070 | 0.213 | 0.0001 | 0.130 | |
| Pb | 0.011 | 0.010 | 0.005 | 0.010 | 0.008 | | 0.013 | |
| **Alkanes** | 5.14 | 1.73 | | 0.222 | | | 1.37 | 0.831 |
| C12 | 0.003 | 0.020 | | | | | | 0.003 |
| C13 | 0.003 | nd | | | | | | 0.006 |
| C14 | 0.019 | 0.0003 | | | | | | 0.020 |
| C15 | 0.057 | 0.013 | | 0.001 | | | | 0.056 |
| C16 | 0.201 | 0.062 | | 0.005 | | | | 0.116 |
| C17 | 0.107 | 0.144 | | 0.003 | | | | 0.265 |
| C18 | 0.587 | 0.215 | | 0.002 | | | 0.049 | 0.148 |
| C19 | 0.777 | 0.308 | | 0.002 | | | 0.120 | 0.126 |
| C20 | 0.977 | 0.311 | | 0.052 | | | 0.260 | 0.074 |
| C21 | 0.516 | 0.290 | | 0.022 | | | | 0.014 |
| C22 | 0.769 | 0.143 | | 0.028 | | | 0.264 | 0.001 |
| C23 | 0.349 | 0.099 | | 0.025 | | | 0.177 | 0.001 |
| C24 | 0.245 | 0.061 | | 0.022 | | | 0.128 | 0.001 |
| C25 | 0.197 | 0.032 | | 0.014 | | | 0.083 | 0.0004 |

Continued Table 3

| | | | | | | |
|---|---|---|---|---|---|---|
| C26 | 0.119 | 0.016 | 0.019 | | 0.075 | |
| C27 | 0.031 | 0.009 | 0.014 | | 0.056 | |
| C28 | 0.023 | 0.004 | 0.011 | | 0.058 | |
| C29 | 0.013 | 0.002 | 0.003 | | 0.046 | |
| C30 | 0.007 | 0.001 | | | 0.025 | |
| C31 | 0.010 | 0.002 | | | 0.017 | |
| C32 | 0.010 | 0.001 | | | 0.007 | |
| C33 | 0.010 | 0.00001 | | | 0.002 | |
| C34 | 0.010 | 0.0004 | | | | |
| C35 | 0.013 | 0.00004 | | | | |
| C36 | 0.016 | nd | | | | |
| C37 | 0.018 | nd | | | | |
| C38 | 0.025 | nd | | | | |
| C39 | 0.031 | nd | | | | |
| C40 | 0.003 | nd | | | | |
| **PAHs** | 0.098 | 0.130 | | 0.251 | 0.021 | 0.021 |
| Nap | 0.008 | 0.001 | | 0.014 | | 0.0004 |
| Acy | 0.005 | 0.0003 | | 0.006 | | 0.0002 |
| Ace | 0.001 | 0.00004 | | 0.001 | | 0.0003 |
| Flu | 0.002 | 0.0001 | | | | 0.001 |
| Phe | 0.005 | 0.021 | | 0.007 | | 0.008 |
| Ant | 0.001 | 0.001 | | 0.002 | | 0.0004 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Fluo | 0.026 | 0.010 | | | 0.027 | | 0.009 | 0.002 |
| Pyr | 0.028 | 0.088 | | | 0.052 | | 0.008 | 0.007 |
| BaA | 0.007 | 0.001 | | | 0.014 | | 0.001 | 0.0005 |
| Chry | 0.008 | 0.002 | | | 0.025 | | 0.003 | 0.0005 |
| BbF | 0.002 | 0.001 | | | 0.016 | | | 0.0003 |
| BkF | 0.001 | 0.0001 | | | 0.003 | | | 0.0002 |
| BaP | 0.0004 | 0.00001 | | | 0.009 | | | 0.0004 |
| IcdP | 0.001 | 0.00002 | | | 0.013 | | 0.0004 | 0.001 |
| DahA | 0.000 | 0.001 | | | 0.001 | | | 0.0002 |
| BghiP | 0.003 | 0.004 | | | 0.062 | | 0.0003 | 0.0003 |
| **Hopane, sterane** | 0.026 | 0.011 | | 0.014 | 0.167 | | 0.143 | |
| ABB | 0.001 | 0.0005 | | 0.0004 | 0.007 | | | |
| AAA | 0.002 | 0.001 | | 0.001 | 0.006 | | | |
| Tm | 0.001 | 0.001 | | 0.001 | 0.014 | | 0.012 | |
| 30AB | 0.011 | 0.005 | | 0.006 | 0.065 | | 0.069 | |
| 29AB | 0.011 | 0.004 | | 0.006 | 0.075 | | 0.061 | |

n.d. = not detected

**Figure captions**

Figure 1The routes for diesel trucks

Figure 2 Particulate matter sampling system

Figure 3 $EF_{PM}$ for excavators with different ~~operating~~ operational modes and emission standards and the correlation with sulfur contents

Figure 4 Diesel trucks $EF_{PM}$ for different emission standards, vehicle sizes and driving conditions

Figure 5 PM compositional constituents for individual vehicles

Figure 6 OC/EC ratios ~~under~~ in different ~~operating~~ operational modes and driving conditions for excavators and trucks

Figure 7 Cross plots for the ratios of BaA/(BaA+Chry) vs IcdP/(IcdP+BghiP) and BaA/(BaA+Chry) vs Flua/(Flua+Pry) and comparison with those from other diesel vehicle sources.

Figure 8 Percentages of each ring PAHs to total PAHs; BaPeq for parent PAHs in each tested trucks and excavators

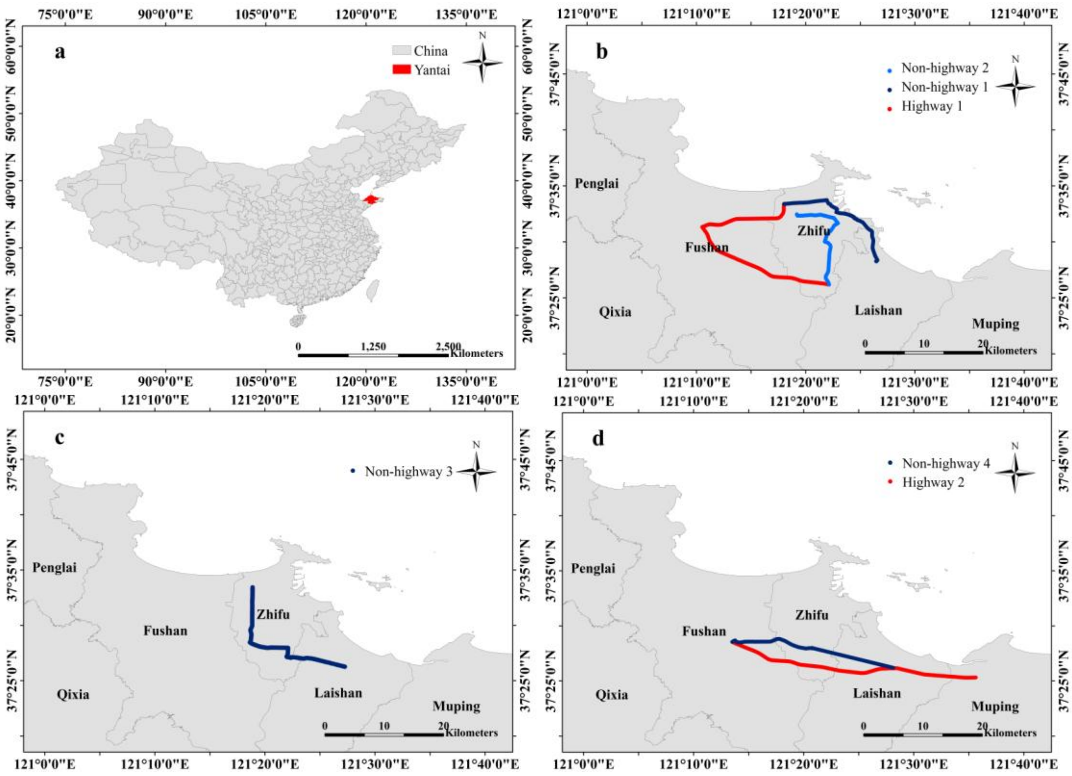


Figure 1The routes for diesel trucks; a was the site of Yantai; b was the route for China Ⅲ and China Ⅳ light-duty diesel trucks; c was the rout for China Ⅱ heavy-duty diesel truck ; d was route for China Ⅲ medium-duty and heavy-duty trucks

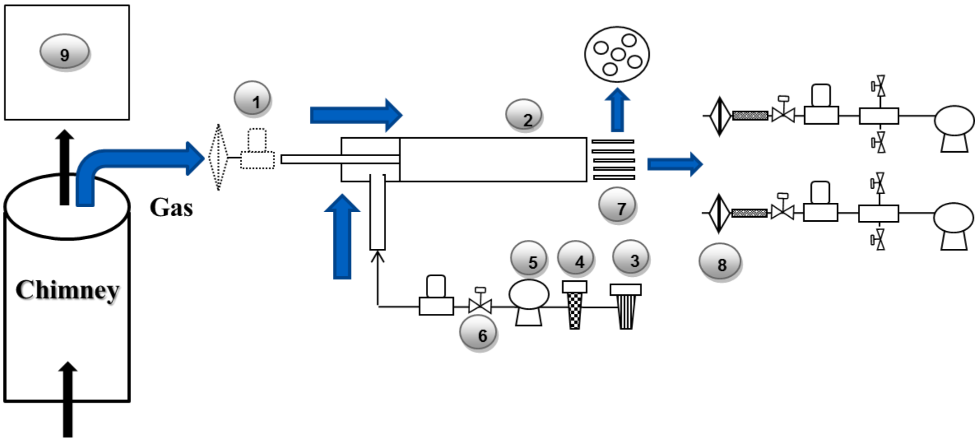


Figure 2 Particulate matter sampling system; 1 ~~was~~ is the flowmeter; 2 ~~was~~ is the dilute tunnel; 3 ~~was~~ is the filtrator; 4 ~~was~~ is the activated carbon; 5 ~~was~~ is the fan; 6 ~~was~~ is the valve; 7 ~~was~~ is the flow divider; 8 ~~was~~ is the filter membrane sampler; and 9 ~~was~~ is the exhaust analyzer

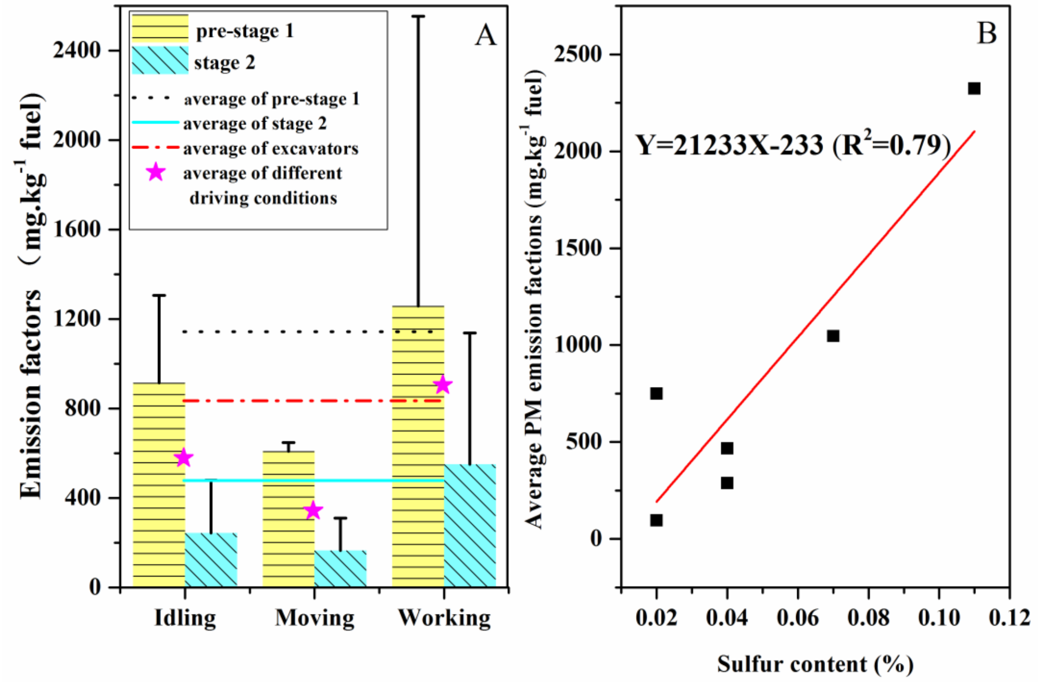


Figure 3 $EF_{PM}$ for excavators with different ~~operating~~ operational modes and emission standards (A) and the correlation with sulfur contents (B)

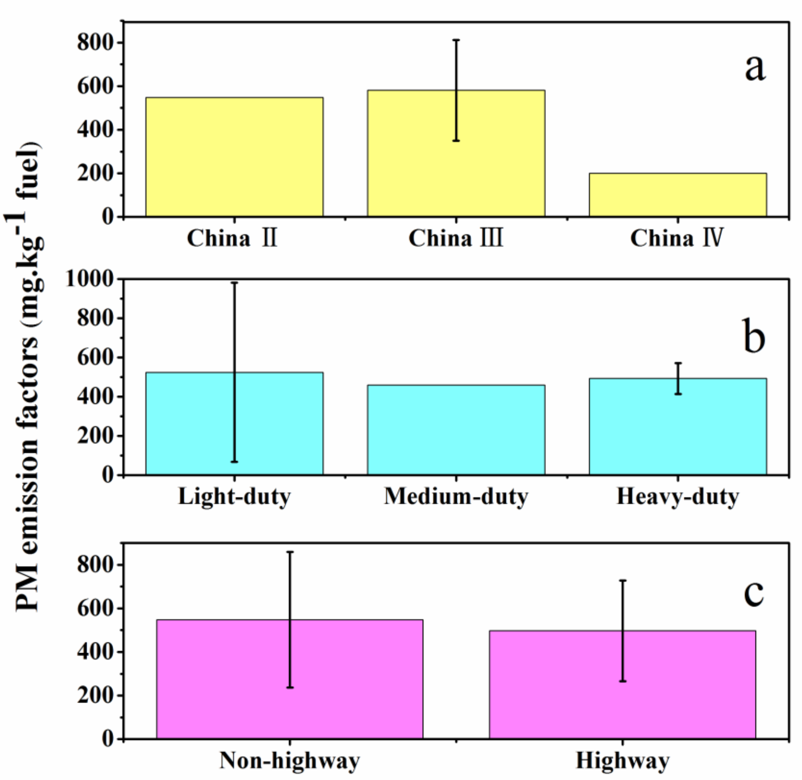


Figure 4 Diesel trucks $EF_{PM}$ for different emission standards (a), vehicle sizes (b) and driving conditions (c)

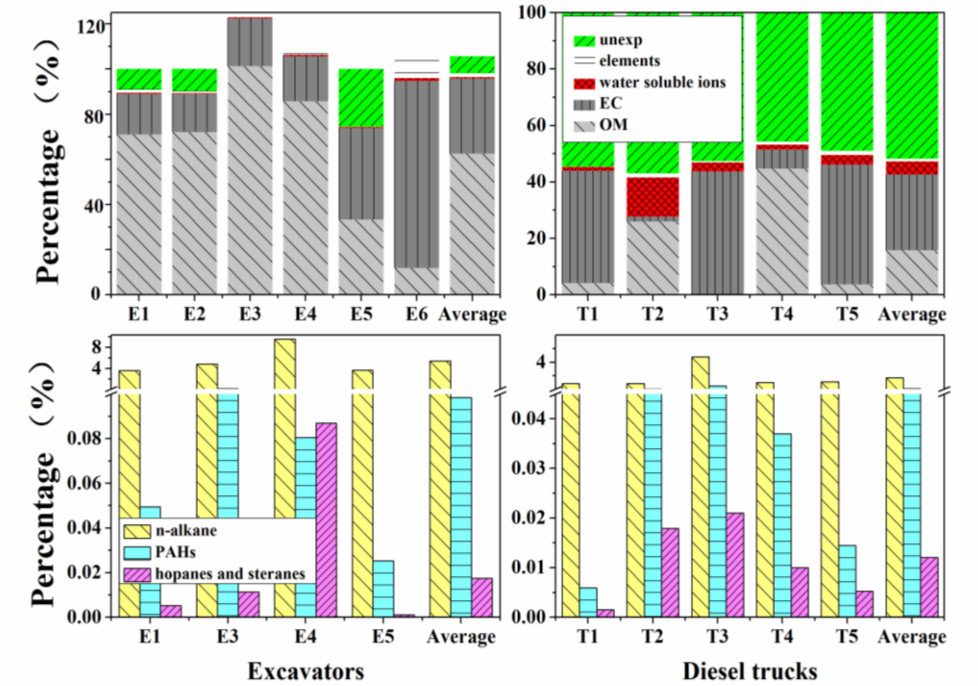


Figure 5 Compositional constituents of PM for individual vehicles (%)

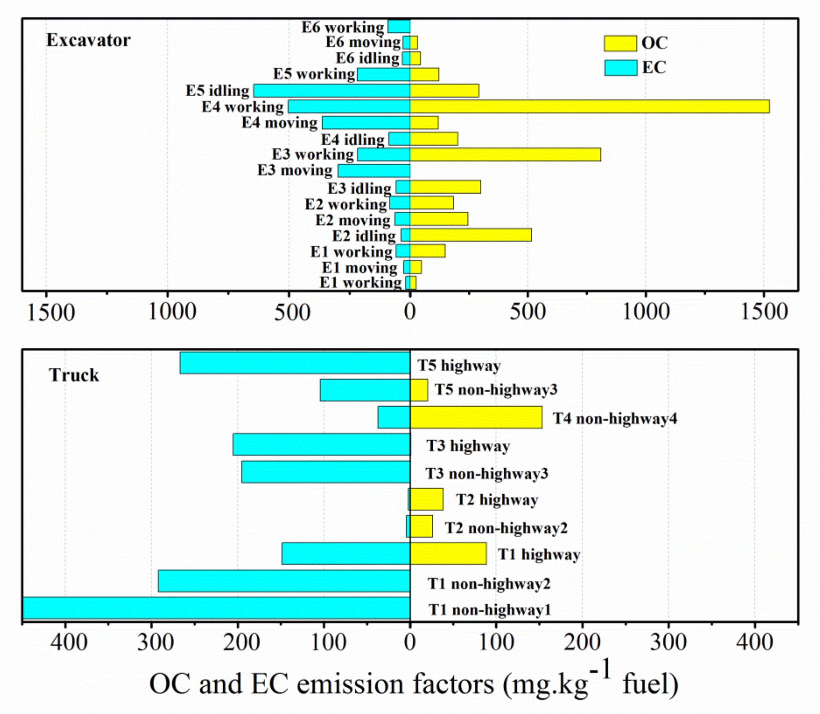


Figure 6 OC/EC ratios ~~under~~ in different ~~operating~~ operational modes and driving conditions for excavators and trucks

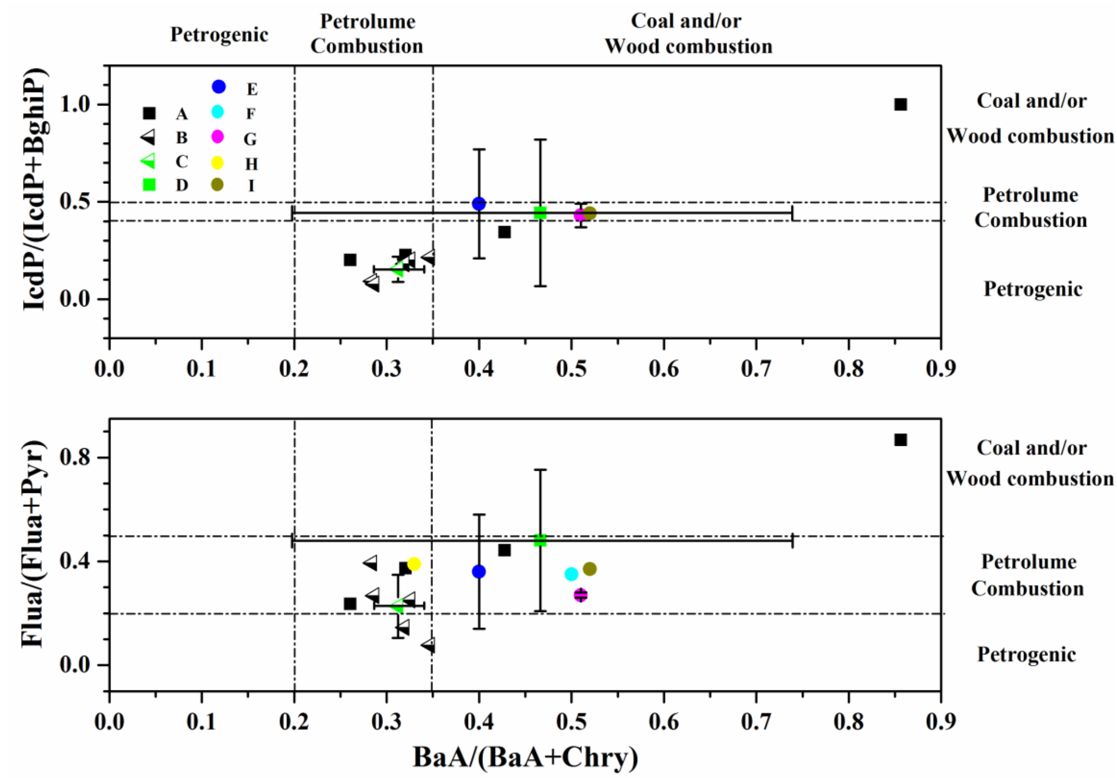


Figure 7 Cross plots for the ratios of BaA/(BaA+Chry) vs IcdP/(IcdP+BghiP) and BaA/(BaA+Chry) vs Flua/(Flua+Pry) and comparison with those from other diesel vehicle sources. A and B ~~were~~ are the isomer ratios of the PAHs ~~for~~ from the excavators and trucks, respectively, tested in this study~~, respectively~~; C and D ~~were~~ are the average isomer ratios of PAHs for trucks and excavators tested in this study; E, F, G, H, I ~~were~~ are results obtained from Liu et al. (2015), Wang et al. (2015), Shah et al. (2005), Schauer et al. (1999), Chen et al. (2013)

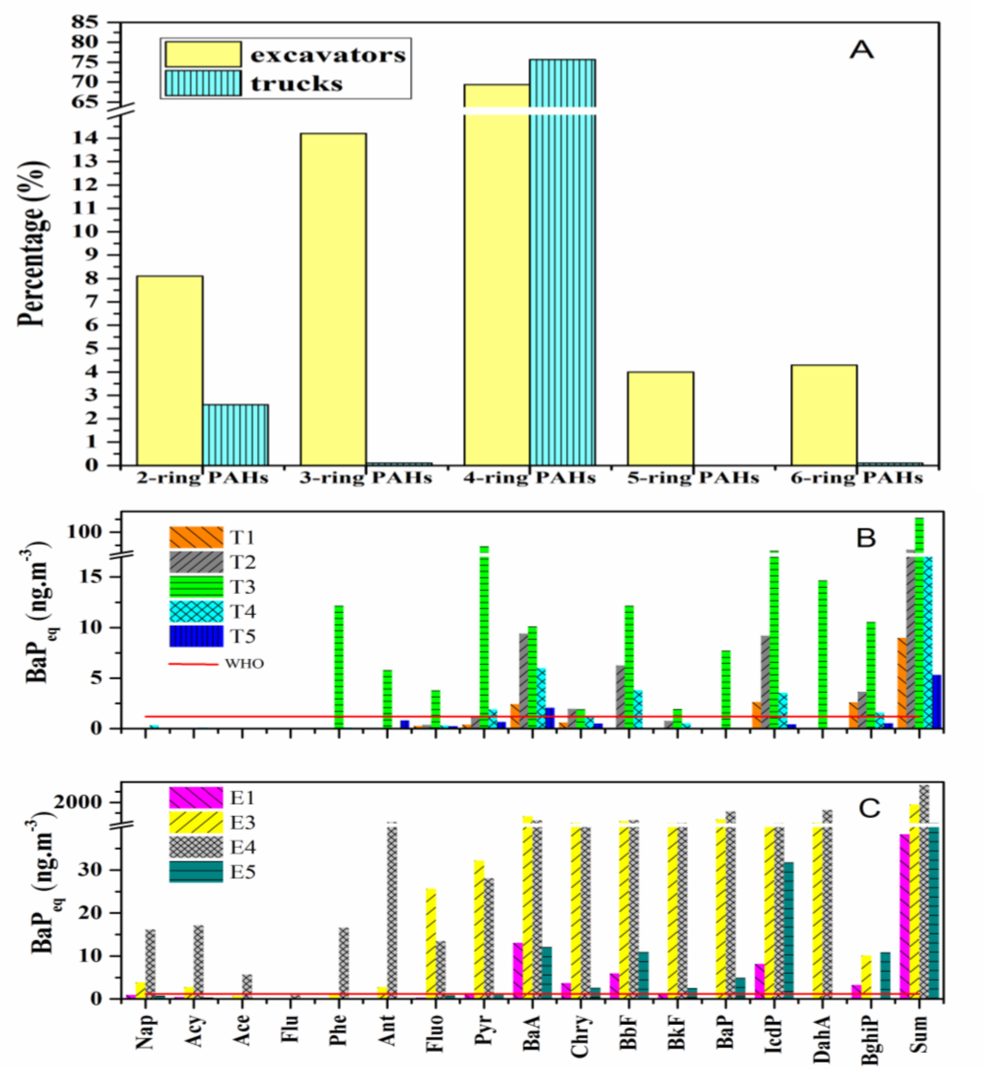


Figure 8 Percentages of each ring PAHs to total PAHs (A); BaPeq for parent PAHs in each tested trucks (B) and excavators (C)

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