Response to editor' Comment on Manuscript: acp-2016-1038

Dear Editor,

We are thankful very much to you for the profound comments. We have made every effort to polish our English. The manuscript was revised by a specialist language service institution (Elsevier Language Editing) and we also asked a native English speaker to take a proof reading of the final version of the revised manuscript.

Best regards,

Dr. Yingjun Chen

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

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Abstract. With the rapid growth in the number of both non-road and on-road diesel vehicles, the adverse effects of particulate matter (PM) and its constituents on air quality and human health have attracted increasing attentions. However, studies on the characteristics of PM and its composition emitted from diesel vehicles are still scarce, especially ~~those performed in~~under real-world driving conditions. In this study, six excavators and five trucks ~~involving~~that provided a wide range of emission~~s~~ standards

and operation~~al~~ modes were tested, and PM emissions ~~as well as~~and their ~~its~~ constituents, including organic carbon (OC), elemental carbon (EC), water soluble ions (WSIs), elements, and organic species ~~such as~~like polycyclic aromatic hydrocarbons ~~(~~PAHs~~)~~, n-alkanes, and hopane~~s~~, and steranes were analyzed and characterized. The average emission factors for PM (EF_{PM}) from excavator and truck emissions were 829 ± 806 and 498 ± 234 mg ~~k~~g^{-1} fuel, respectively. EF_{PM} and PM constituents were significantly affected by fuel quality, operational mode, and emission standards. ~~S~~A significant correlation (R^2=0.79, p<0.01) was found between EF_{PM} for excavators and the sulfur contents in fuel. The highest average EF_{PM} for working excavators was 904 ± 979 mg ~~k~~g^{-1} fuel~~,~~ ~~–~~as a ~~because of the~~ high~~er~~ engine load required in this mode. From pre-stage 1 to stage 2, the average EF_{PM} for excavators decreased by 58%. For trucks, the average non-highway EF_{PM} at ~~(~~548 ± 311 mg ~~k~~g^{-1} fuel~~,)~~ was higher than the highway EF_{PM} ~~(~~at 497 ± 231 mg ~~k~~g^{-1} fuel~~)~~. ~~Meanwhile~~Moreover, the reduction ratess~~~~ were 63.5% and 65.6% when ~~switching~~ switched from China II and III to China IV standards, respectively. Generally, the PM composition emitted from excavators was dominated by OC (39.2 ± 21.0%) and EC (33.3 ± 25.9~~%), –~~%); ~~while~~ PM from trucks was dominated by EC (26.9 ± 20.8%), OC (9.89 ± 12%), and WSIs (4.67 ± 5.74%). The average OC/EC ratios for idling and working excavators were ~~3~~ three to ~~4~~ four times higher than those for moving excavators. Although the EF_{PM} for excavators and trucks ~~were~~ was reduced with the constraint of regulations, the element fractions for excavators increased from 0.49% ~~(~~in pre-stage 1~~)~~ to 3.03% in ~~(~~stage 2~~)~~, and the fraction of WSIs for the China IV truck was ~~6-fold~~six times higher than the average value of all other-level trucks.~~.~~ Furthermore, as compared with other diesel vehicles, wide ranges were found for excavators ~~in~~ of 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) (0.24-0.87)~~ were found for excavators~~, which might be a result of the complex characteristics of the excavator operation~~al~~ modes. ~~Through c~~A comparison of our results with those in the literature revealed that~~s,~~ on-board measurement~~s~~ data ~~could~~ more accurately reflect actual ~~real~~

conditions~~ better~~. Although the fractions of the 16 priority PAHs in PM from the excavator and truck emissions were similar, the equivalent concentrations of total benzo[a]pyrene~~,~~ of excavators were 31 times than that for trucks, implying that more attention should be paid to non-road vehicle emissions.

Keywords

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

1. Introduction

Particulate matter (PM) emitted from diesel vehicles has significant adverse effects on air quality, human health, and global climate change~~,~~ and therefore merits close examination (Aggarwal et al., 2015, 2016). Previous studies have found that diesel vehicle exhaust is a major source of ambient fine PM emissions ($D_p \leq 2.5$ μm) (Oanh et al., 2010, Zhang et al., 2015a). For ~~instance~~example, vehicle exhaust was reported to contribute almost 30% of ambient $PM_{2.5}$ in ~~9~~ nine Chinese cities ~~of China~~ in 2015 (MEP 2016). The International Agency for Research on Cancer (IARC) ~~found~~ reported that exposure to diesel exhaust ~~could~~ can cause lung cancer (IARC 2012). Adar et al. (2015) surveyed more than 25 million children and ~~reported~~ concluded that a disproportionate ~~occurrence~~ number of cases of respiratory disease ~~had been~~were caused ~~when~~ by breathing polluted air from diesel school buses ~~by a survey for more than 25 million children~~. Moreover, ~~Nearly~~ nearly 34% of element carbon (EC) emissions, a major contributor to current global warming and poor human health, ~~comes~~ originates from off-road diesel vehicle emissions in the ~~USA~~ United States (USEPA 2015).

The numbers of on-road and non-road diesel vehicles have increased considerably in China~~,~~ and have caused severe environmental problems. On-road diesel vehicles can be classified into 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 diesel is not the primary fuels used for these vehicles ~~were not diesel~~. The number of on-road diesel vehicles increased from 11.0 million in 2009 to 32.8 million in 2015, ~~while~~ meanwhile, 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). ~~Based on the~~According to China's vehicle environmental management annual report for 2015 (MEP 2016), 0.56 million tons of $PM_{2.5}$ were emitted from on-road mobile sources, ~~of which~~ 90% of which ~~resulted~~ originated from on-road diesel vehicle emissions (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) compiled an emission inventory for non-road equipment ~~(~~, including agricultural equipment, river/ocean-going vessels, locomotives, and commercial airplanes~~,)~~ and found that ~~there were~~ 349 Gg of PM were emitted from non-road vehicles in China during 2012. Construction equipment ~~contributed as~~was the largest non-road diesel vehicles emission source. Zhang et al. (2010) reported that PM_{10} emitted from construction equipment in the Pearl River Delta (PRD) region accounted for 26.5% of the total emissions from non-road vehicles in 2006. The ~~number of~~ construction equipment in use increased from 1.97 million to 5.85 million between 2006 and 2012 (CCCMIY 2013). Furthermore, excavators, ~~as~~ one of the most abundant types of construction equipment (Figure S1), ~~excavators~~ contributed almost 65% of the PM emissions from construction equipment (Li et al., 2012).

In order to control PM emission pollution from diesel vehicles, China began to implement emission standards in early 2001 for light ~~duty diesel vehicles~~ and heavy-duty diesel vehicles (SEPA et al., 2001). These standards were tightened in the subsequent 12 years, from the China I to China V standards. Although the China V emission standard for on-road diesel vehicles has been formulated, insufficient diesel fuel quality has ~~slows~~ slowed their implementation (Yue et al., 2015). In addition, the

China IV emission standards for on-road diesel vehicles ~~are~~ have not been~~not~~ fully implemented. Moreover, the ~~Implementation~~ implementation timeline of emission standards for non-road diesel vehicles has lagged behind that ~~of the~~for on-road diesel vehicles. China implemented two emission standards for new non-road diesel engines, stage 1 and stage 2 in 2007 and 2009, respectively. However, the first implementation in China was ~~7~~ seven years later than that ~~of~~ in the United States (USEPA 2003, SEPA et al., 2007). The pollution emission limits for on-road and non-road diesel vehicles are given in Tables S1 and S2.

EF_{PM} is an important parameter in the compilation of emission inventories 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). Most of the EF_{PM} from trucks ~~have~~ has been measured by using tunnel and dynamometer tests, which ~~could~~ cannot be used to~~not~~ evaluate influential factors for real world PM emissions from a single truck ~~in real-world conditions~~ (Alves et al., 2015b, Mancilla et al., 2012, Pio et al., 2013). Although several studies have measured real world PM emissions from trucks by using on-board tests ~~in real-world conditions~~ (Wu et al., 2016, Wu et al., 2015, Zhang et al., 2015a), the data should be updated frequently (Huo et al., 2012) because EF_{PM} emitted from trucks could change along with improved emission standards. In addition, ~~the~~ data of real world EF_{PM} emitted from non-road diesel vehicles ~~in real-world conditions~~ ~~is~~ are scarce in China. In 2014, the Ministry of Environmental Protection of the People's Republic of China ~~had~~ issued a report titled "Technical guide for the preparation of a single source emission inventory of atmospheric fine particulate matter." However, no measured data of EF_{PM} for non-road vehicles ~~could be~~are referred in this technical guide, ~~especially~~ particularly for construction machinery (6 g km^{-1} ~~were~~ was predicted for uncontrolled standards) (MEPPRC 2014). ~~Until now~~Thus far, ~~there was~~ only one study in China, by Fu et al. (2012), ~~had~~ provided EF_{PM} of 12 excavators using a portable emission measurement system (PEMS) under different operation~~al~~ modes. On-board measurements need to be expanded to improve the localization of EF_{PM} for non-road diesel vehicles in China as soon as possible owing to~~, because of~~ the

complexity of real-world conditions, including the lagging diesel quality and changing emission standards.

Analysis of the chemical composition of PM is essential for source apportionment, human health, and climate change studies. Primary PM emitted from diesel vehicles contains a variety of chemical components~~,~~ including organic carbon (OC), elemental carbon (EC), water soluble ions (WSIs), elements, and organic species such as n-alkanes, polycyclic aromatic hydrocarbons (PAHs), and hopanes and steranes). Several previous field studies have focused on the chemical composition of PM emitted from diesel vehicles~~,~~; however, specific characteristics of PM emitted from diesel vehicles and its composition are still largely ~~are great~~ unknown, ~~especially~~ particularly for organic compounds. Zhang et al. (2015a) characterized $PM_{2.5}$ compositions (OC, EC, WSIs and elements) 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 OC, EC, WSIs, and element contents. In 2012, Fu et al. (2012) were the first~~ly~~ to test~~ed~~ 12 excavators using on-board test in China, ~~but~~ although only the optically-based EF_{PM} was given.

In this study, PM emitted from on-road and non-road diesel vehicles was measured to (I) test the ~~emission factors of PM~~real world EF_{PM} for excavators and trucks ~~in real-world conditions;~~ , (II) identify influential factors on the emitted PM and its composition, and (III) characterize the chemical components present in the emitted PM. Although the study results required substantial effort, ~~it~~ they provide~~d~~ valuable information for developing ~~of~~ effective control policies to reduce PM emissions from excavators and trucks.

Experimental

2.1 Diesel vehicle and operation~~al~~ mode selection

In this study, six excavators and five trucks were selected to cover a wide range of emission standards, manufacturers and engine loads. Detailed information for the selected excavators and trucks is shown in Table 1. As shown in Figure S2, the increase in the annual production of excavators, from 70,000 to 85,000, did not change substantially between 2007 and 2009 ~~(an increase from 70,000 to 85,000),~~

when the ~~during~~ stage 1 non-road vehicle emission standard was implemented. Therefore, excavators produced ~~with~~ during pre-stage 1 and stage 2 emission standards were chosen for this study. ~~Based on~~On the basis of the China national standard (SEPA 2007), excavators are divided into five types according to their power rating. The excavators in this study were categorized into three types including ~~as~~ low (0-75 kw), medium (75-130 kw) or high (130-560 kw) power under different emission standards. Three operational modes were selected for the excavators to reflect actual use conditions, ~~including~~ such as idling, moving and working. Further descriptions of these three modes can be found in Fu et al. (2012). 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 sampling times during idling, moving, and working were 41.7, 24.0, and 28.5 min, respectively.

Three types of diesel trucks were selected in this study, including one China II standards truck, three China III standards trucks, and one China IV standards truck. The China III trucks included one of each light-duty, medium-duty, and heavy-duty diesel truck. ~~Based on~~On the basis of the traffic rules and driving conditions for on-road diesel trucks, routes were predesigned for the test trucks in Yantai, Shandong province, ~~of~~ China (Figure 1). Because different trucks ~~drove~~ drive on different routes, the selected routes in this study were divided into non-highway and highway categories. The selected routes for China III and China IV light-duty trucks included non-highway 1, non-highway 2, and highway 1 at~~.~~ ~~The~~ lengths of ~~these three roads were~~ 19, 35 and 17 km, respectively. The route chosen for the China II heavy-duty truck (yellow label) was non-highway 3, ~~which was~~at length of 25 km ~~long~~. The routes chosen for China III medium-duty and heavy-duty trucks included non-highway 4 and highway 2 at~~. The~~ lengths of ~~these two roads were~~ 47 and 23 km, respectively. The detailed velocity and road grade information for all of the tested routes are shown in Figures S3 and 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 owing to~~,~~ ~~which was due to the~~ incomplete~~ness of some~~ monitoring

data (e.g. the data of CO_2 and CO concentrations missing). As shown in Tables S3 and S4, the variability for the same operation~~al~~ mode was considered to be acceptable ~~in test times~~. Some actions were required to reduce the uncertainty. For example, we combined sampling filters for the repeated experiments for T1 and T3 to ~~carry out~~conduct organic compound analysis.

2.2 On-board emission measurement system

The on-board emission measurement system was designed and constructed by our research group (Figure 2). A description of the on-board emissions test system was given in our previous report (Zhang et al., 2015b). Briefly, this system consists of two main components: a Photon II analyzer, which was used to analyze the flue gas (HC, CO, CO_2, SO_2, and NOx), and a PM sampling system (TSP sampler). Although we used TSP sampler to collect PM ($D_p \leq 100$ μm) in this study, ~~mainly~~ most of the PM collected in this experiment was considered as fine particles~~. Because~~ because almost all of the particles emitted from engine combustion are fine ~~particles~~ (An et al. 2011). The PM sampling system consisted of a dilution system followed by five exhaust channels. Two channels were connected to PM samplers, and the other three were blocked. Before sampling, the emission measurement system was ~~put~~ installed on a truck and was connected to the excavator exhaust tube by a stainless steel pipe. ~~The~~ This system ~~had~~ showed clear improvements over other on-board instruments, such as PEMSs and FPS4000 (Zheng et al., 2015)~~;~~, moreover, it has ~~with~~ better portability and ~~better~~ a stronger ability to collect filter samples for further chemical analysis in the laboratory. The results in this study present~~ed~~ 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 significant effect on PM emissions from vehicles (Cui et al., 2016, Liang et al., 2005, Zhang et al., 2014). ~~Since~~ Because ~~the~~ various fuels are used in excavators ~~were various~~ and always ~~with~~ have poor qualities, all of the corresponding fuels from each of the tested excavators were collected ~~to have~~for quality analysis. The results ~~of fuel quality analysis~~ are given in Table 2. ~~Comparing~~ A

comparing of the diesel quality used in this study with the standards for non-road vehicles (GB 252-2015) (SEPA et al., 2015) revealed~~, it was found~~ that the sulfur content~~s~~ in most types of ~~the~~ diesel~~s~~ used in this study (200-1100 ppm) were higher than ~~which~~ allowed by GB 252-2015 (<350 ppm). Additionally, the sulfur content in the diesel used by E4 was 1100 ppm, which ~~was~~ is ~~much~~ significantly higher than that used in the other excavators. Furthermore, the ash content of the diesel used by E4 was 4.16%, which is about 420 times ~~higher~~ than the limit given by GB 252-2015.

2.3.2 PM and chemical composition analysis

Quartz-fiber filters were used to collect PM samples for PM weight measurement and chemical analysis~~.~~; ~~And~~ the weight losses of these filters ~~could be~~were neglected through strict sampling processes. All of the filters were weighed before and after sampling to determine the PM mass concentrations. Before each ~~weighing~~measurement, the filters were balanced at 25 ℃ and 40% relative humidity for 24 h. Each filter was weighed three times. WSIs were analyzed using ion chromatography (Dionex ICS3000, Dionex Ltd., Sunnyvale, California, United States~~America~~) following the method of Cui et al. (2016). Elements analysis was performed using inductively coupled plasma mass spectromet~~er~~ry (ICP-MS; ELAN DRC II type, Perkin Elmer Ltd., Hong Kong) (Cui et al. 2016).

Because the organic compounds on the filters were insufficient for quantification, we merged filters of different operation~~al~~ modes or driving routes based on the proportion of sampling time during each mode or route for analyzing the PM characteristic ~~of PM~~ for each diesel vehicle. 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}) and were ultrasonically extracted ~~two times~~twice in 30 mL of a 1:1 mixture of hexane and dichloromethane for 10 min. All extracts from each sample were combined, filtered and concentrated to approximately 0.5 mL.

Organic species including n-alkanes, PAHs, and hopanes and steranes were analyzed using GC-MS (Agilent 7890A GC-5975C MS) with a DB-5MS column ~~(~~of ~~length~~ 30 m in length, an inner diameter of ~~×~~ ~~i.d.~~ 0.25 mm, and ~~×~~ thickness of 0.25

μm~~)~~. The following GC operating program was ~~as follows~~used: 60 ℃ for 4 min, then increase 5 ℃ min^{-1} to 150 ℃ with 2 min static time , and finally ~~then~~ increase 3 ℃ min^{-1} to 306 ℃ with a 20 min static time. The GC had an injector temperature of 290 ℃, an injector volume of 2 μL, ~~helium~~ He carrier gas, and a gas flow rate of 1.2 mL min^{-1}. The electron impact (EI) mode at 70 eV and selected-ion-monitoring (SIM) mode were selected to determine the concentrations of PAHs, hopanes, and steranes. For organic matter, blank samples and recovery rates (66.7-128% for five surrogates) were measured. The blank concentrations were subtracted from the sample concentrations. The final concentrations of organic matters were not corrected for the recoveries.

The PM chemical constituents analyzed in this study were OC; EC; WSIs including~~:~~ SO_4^{2-}, NO_3^-, Cl^-, and NH_4^+; elements including~~:~~ Na, Mg, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, and Pb~~)~~; n-alkanes including~~:~~ C12 to C40; the ~~sixteen~~ 16 USEPA priority PAHs ~~of~~ including naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (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); Hopanes and steranes including~~:~~ 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-based emission factors

Fuel-based emission factors were calculated using the carbon mass balance formula:

$$EF_i = \frac{\Delta X_i}{\Delta CO_2} \cdot \frac{M_i}{M_{CO_2}} \cdot EF_{CO_2} \quad (1)$$

where EF_i and EF_{CO_2} (g kg^{-1} fuel) are the emission factors for species *i* and CO_2, respectively~~,~~; ΔX_i and ΔCO_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:

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

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 emission rate was calculated as:

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

where C_F (g C kg^{-1} fuel) represents the mass of carbon in 1 kg 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.

2.4.2 Average fuel-based emission factors for excavators and trucks

The average fuel-based emission factor for each excavator in each relevant operation~~al~~ mode was calculated as:

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

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

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

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

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

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. Instead, BaP_{eq} is typically used to evaluate the carcinogenic risks associated with individual PAH (Mirante et al., 2013), which ~~was~~ is calculated as:

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

where PAH_i is the measured concentration of an individual PAH for excavator *i*, and *PEF* is the potency equivalence factor for that PAH obtained from Wang et al. (2008).

3. Results and discussion

3.1 Fuel-based PM emission factors for excavator exhaust

The EF_{PM} values for excavator exhaust are illustrated in Figure 3, with detailed information given in Table S5. The maximum ~~fuel-based PM emission factor~~EF_{PM} was 37 times ~~higher~~ than the minimum value. In general, the average EF_{PM} for different excavators ranged from 96.5 to 2323 mg ~~k~~kg^{-1} ~~f~~fuel, with an average of 829 ± 806 mg ~~k~~kg^{-1} fuel. The EF_{PM} values of the excavators reported by Fu et al. (2012) ~~were~~are within the range of EF_{PM} values in this study. The wide range of EF_{PM} values here could be ~~due~~ attributed to ~~the~~ differences in emission standards for the excavators. ~~The excavators~~Those tested by Fu et al. (2012) included stage 1 and stage 2 emission standards, ~~while~~ whereas the excavators in this ~~studies~~ study were used with the emissions standards of pre-stage 1 and stage 2.

The EF_{PM} measured for pre-stage 1 excavators during idling, moving and working were 914 ± 393, 609 ± 38 and 1258 ± 1295 mg ~~k~~kg^{-1} fuel, respectively, whereas those for stage 2 excavators were~~, it was~~ 243 ± 236, 165 ± 144 and 551 ± 587 mg ~~k~~kg^{-1} fuel, respectively. That is, the EF_{PM} for the stage 2 excavators under idling, moving and working modes was~~, were~~ reduced by 73%, 73% and 56% compared ~~to~~ with the pre-stage 1 excavator, respectively, ~~while~~ and the average EF_{PM} for the excavator decreased by 58% from pre-stage 1 to stage 2. EF_{PM} ~~could~~ can be influenced by many factors. In this study, the EF_{PM} ~~range~~ for excavators with different power ratings ranged from 96.5 (35 kw) to 2323 (110 kw) mg ~~k~~kg^{-1} fuel~~,~~; however, the correlations between EF_{PM} and engine power (~~See~~ Figure S5) were weak. Additionally, fuel quality, emission standards and operation~~al~~ mode significantly influenced the EF_{PM}. Given

that there is no government supervision of diesel used for non-road vehicles, the reduction ~~of~~ in average EF_{PM} from pre-stage 1 to stage 2 could be ~~mainly~~ attribute mainly to both the different emission standards and diesel quality. As shown in Table S5, the average EF_{PM} from E5 to E6 with the same fuel quality but different emission standards was reduced 87.1%. Similarly, EF_{PM} was reduced 38.2% from E2 to E1~~.~~, ~~From~~ which ~~it indicated~~ indicates that emission standards have significant impacts on EF_{PM}. ~~Likewise~~Similarly, the average EF_{PM} for E3, E1 and E6 ~~that~~ were under the same emission standard decreased with improvement ~~of~~ in fuel quality, which ~~suggesting~~ suggests the influence of diesel quality. As shown in Figure 3, good correlation ($R^2 = 0.79$, $P < 0.01$) was found between the average EF_{PM} for excavators and sulfur contents in fuels, which ~~was~~is consistent with the results reported by Yu et al. (2007). Furthermore, the EF_{PM} for the various excavators varied significantly under different operation~~al~~ modes. Specifically, working excavators exhibited the highest EF_{PM}, which was more than double the values for idling and moving excavators. The average EF_{PM} for excavators ~~were~~ was 578 ± 467 while idling, 343 ± 264 while moving, and 904 ± 979 mg kg^{-1} fuel while working. Excavators under the working mode produced the highest average EF_{PM}, which might be ascribed to ~~that~~ the higher engine load ~~caused~~ causing a lower air-fuel ratio ~~and thus prompted PM production~~.

3.2 Fuel-based PM emission factor for trucks

The EF_{PM} for all measured trucks varied from 176 to 951 mg kg^{-1} fuel. The maximum EF_{PM} for trucks was three times ~~higher~~ than the minimum value. The average EF_{PM} for the tested diesel trucks was 498 ± 234 mg kg^{-1} fuel, which is ~~Consistent~~ consistent with that reported by Wu et al. (2016) (range: 95.6-1147 mg kg^{-1} fuel; average: 427 mg kg^{-1} fuel). The average real world EF_{PM} values of diesel trucks ~~under real-world conditions~~ with different emission standards, vehicle sizes, and driving patterns ~~were~~ are given in Figure 4. The measured EF_{PM} for China II, China III, and China IV diesel trucks varied from 200 to 548 mg kg^{-1} fuel. The EF_{PM} for the China II truck measured in this study ~~was~~ is lower than that reported by Liu et al. (2009) (910-2100 mg kg^{-1} fuel). The average EF_{PM} values for light-duty, medium-duty and heavy-duty diesel trucks were 524 ± 457, 459, and 492 mg kg^{-1} fuel,

respectively. The average EF_{PM} values for trucks under non-highway and highway driving patterns were 548 ± 311 and 497 ± 231 mg ~~k~~g^{-1} fuel, respectively. As shown in Figure 4, reductions of EF_{PM} from the China II to China IV trucks, and those from the China III to China IV trucks were 63.5% and 65.6%, respectively. The diesel used for trucks was assumed to ~~have~~ be identical in quality ~~because of~~owing to strict diesel quality regulations ~~of~~ for on-road trucks. Therefore, the reductions of EF_{PM} for different trucks could be ~~mainly~~ attributed mainly to the improvements in ~~the~~ emission standards. Of particular note was that the EF_{PM} values for China III and light-duty diesel trucks were higher than ~~the values~~those for the other corresponding trucks. The reason might be the results of poor driving conditions, i.e., low average speed and highly varied speed (Figures S3 and S4). The same tendency is ~~apparent~~ shown in Figure 4, ~~with~~ in which diesel trucks driving on ~~the~~ non-highways (average speed of 28.5 km ~~h~~$^{-1}$) emitted more PM than ~~that~~ those driving on ~~the~~ highways (average speed of 60.7 km ~~h~~$^{-1}$). The road grade further affected the EF_{PM} of the on-road diesel trucks. For example, the EF_{PM} for T5 driving on ~~the~~ highways was lower than ~~those~~ that for T1 driving on ~~the~~ highways owing to the~~, because of~~ lower road grade for T5 (Figure S4).

3.3 Particulate matter composition for individual diesel vehicles

Four types of constituents were considered for reconstituting the PM mass in this study: (1) organic matter, which was calculated by multiplying the corrected OC by a factor of 1.6 (Almeida et al., 2006); (2) EC; (3) WSIs; and (4) elements. The reconstituted masses for the excavator sample~~r~~s were 74.7-123% of the measured mass, ~~while~~ whereas the reconstituted masses for the diesel truck samples were only 43.2-54.4% of the measured mass (Figure 5). In addition to the uncalculated components, this discrepancy might be ~~due~~ attributed to a distribution error between OC and EC by using TOR, droplet effects, or oxides when only metal elements were ~~only~~ considered.

3.3.1 Particulate matter composition for individual excavator

The chemical composition of PM for each excavator is shown in Figure 5 and Table S6. For each excavator, the carbonaceous component (OM+EC) was the dominant

species~~,~~. ~~which~~ This is consistent with results of a previous study by Liu et al. (2005), who reported that the proportions of OC and EC in PM ranged from 70 to 91% (Liu et al., 2005). Because the OC/EC ratio is also used to identify the source of atmospheric particulate pollution, further assessment was performed on ~~the OC/EC~~these ratios in different operation~~al~~ modes for each excavator (Figure 6). The average OC/EC ratios during idling, moving, and working were 1.57, 0.57, and 2.38, respectively. The OC/EC ratio during idling was higher than 1 because soot is rarely~~hardly~~ generated at low temperatures and fuel-rich zones. These results ~~were~~ are also consistent with those ~~from~~ of Liu et al. (2005). Furthermore, Liu et al. (2005) reported that the OC/EC ratios decreased with an increase in load ~~increasing~~ for non-road engines. Although the trend of OC/EC ratios from idling (low load) to moving (medium load) ~~was~~ are consistent with those reported by Liu et al. (2005), the OC/EC ratio under working (high load) was higher than those under idling and moving, which ~~was accorded~~agrees with the results reported by Zhang et al. (2014). As shown in Figure 6, the differences between the OC/EC ratios for different excavator operation~~al~~ modes were significant~~,~~ and could have been ~~be~~ affected by ~~a number of~~numerous factors such as~~,~~ ~~including~~ transient working conditions, diesel sulfur content, and extensive OC sources (Cocker et al., 2004, Liu et al., 2005, Ruiz et al., 2015).

As shown in Figure 5, the WSIs and element~~s~~ fractions ranged from 0.335 to 1.21% and from 0.163 to 7.50%, respectively, for all excavators. The total proportion of WSIs and elements to PM was the highest in excavator E6, followed by excavator E1. Generally, the total proportion of WSIs and elements to PM in the exhaust from excavator E6 was 4 to 14 times ~~higher~~ than the corresponding proportions in the exhaust from the other excavators. Sulfate and nitrate were the main WSIs (79.1-90.0% of WSIs) for almost all of the excavators, except for E1, in which the proportion of Cl^- (67.2%) was the highest (Table S6). Fe, Ca, Na, Mg, and K were the relatively dominant elements, except for E4, which ~~that~~ showed Fe, Zn, and Cu ~~were~~ as the most abundant elements. Wang et al. (2003) reported that the concentrations of the crustal elements ~~of~~ Fe, Ca, and Mg accounted for 50% of the total elements in diesel fuel, which ~~were~~ was significantly higher than anthropogenic elements emitted from diesel

vehicle engines. ~~That~~ This result is consistent with the results ~~from~~ of our study. Similarly, we supposed that diesel was the dominant source for these elements because the sampling tube was placed directly on the tailpipe. In addition, ~~it was different from other excavators that~~ Zn and Cu were also abundant elements for E4, which was different from other excavators. Lin et al. (2015) found that Zn and Cu ~~were~~ originated from lubricating oil~~,~~ except for that used in brake linings. Therefore, we supposed that diesel and lubricating oil combustion were ~~probably~~ likely the main sources of the element~~s~~ emitted from E4 (produced in 2004). Furthermore, the element~~s~~ fractions for the two excavators manufactured in 2013 (1.42% for E1, 7.50% for E6 and 4.09 mg ~~k~~g^{-1} fuel for E1 and 7.24 mg ~~k~~g^{-1} fuel for E6) were higher than those for the other excavators (4.10, 1.71, 8.73, 1.56 mg ~~k~~g^{-1} fuel for E2, E3, E4, and E5). This indicates that elements emissions were deteriorating and that more stringent control technology should be developed to avoid adverse health effects from the total element~~s~~ composition of PM in the exhaust.

The n-alkanes, PAHs, and hopane~~s~~ and steranes fractions in the exhaust from the excavators were 3.6 to 9.6%, 0.03 to 0.24%, and 0.001 to 0.09% respectively. Liang et al. (2005) characterized diesel ~~particulate matter~~PM emitted from non-road engines using a dynamometer test and found that n-alkanes accounted for 0.83% of PM, which ~~was~~ is lower than the proportion found in this study. ~~It was~~This result could be attributed to the ~~possibly caused by the~~ low sulfur diesel fuel ~~they used~~ and the different sampling methods they used. In contrast to the fractions of WSIs and elements, the fractions of n-alkanes, hopane and steranes were the highest in excavator E4, ~~while~~ whereas the fraction of PAHs was the highest for the exhaust from E3. E4 had poorer diesel quality compared with E3, which ~~might be the reason for~~could explain high n-alkane, hopane and steran~~e~~s concentrations. Similarly, Rogge et al. (1993) found that n-alkanes, hopane and steranes were mostly derived from the incomplete combustion of fuel and lubricant oil. However, we speculate~~d~~ that the PAHs in this study were affected by combustion conditions (i.e., combustion temperature) ~~in this study,~~ because E3, with the stage 2 standard, had better performance and superior fuel quality. PAH isomer ratios have been widely used to

conduct source apportionment 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 had the best potential to distinguish between natural and anthropogenic sources. For the excavators, the ranges of the ratios 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 the PAHs in excavator exhaust obtained in this study ~~were~~ are similar to those ~~from~~ reported by Liu et al. (2015). The E4 excavator ~~had~~ showed obvious differences in the ratios of BaA/(BaA+Chry), IcdP/(IcdP+BghiP), and Flua/(Flua+Pry) ~~to~~ from those ~~from~~ for the other excavators. 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~~.~~, which are ~~These were~~ different from the ranges for fuel combustion defined by Yunker et al. (2002). The ratios of PAHs emitted from diesel vehicles reported by Yunker et al. (2002) mainly referred ~~to~~ those from on-road diesel vehicles. However, the operation~~al~~ mode and fuel quality for non-road diesel vehicles are more complicated than those for on-road diesel vehicles. Therefore, the results obtained in this study could provide references values for the isomer ratios of PAHs in non-road diesel vehicle exhaust.

3.3.2 Particulate matter composition for individual diesel trucks

For diesel trucks, the total carbonaceous composition (OM+EC) accounted for 44.0% (T1), 27.9% (T2), 43.9% (T3), 51.6% (T4) and 46.3% (T5) of PM, ~~that are all~~which are all lower than the values reported in previous studies (Chow et al., 2011, Wu et al., 2015). One of the main reasons ~~was inferred as~~is the ~~different~~ differences in OC and EC detection methods used in our study. Through a comparison of National Institute for Occupational Safety and Health (NIOSH) and Interagency Monitoring of Protected Visual Environments (IMPROVE) protocols,~~Through a comparison of~~ two common thermal-optical methods ~~(NIOSH and IMPROVE)~~ of OC and EC analysis for 333 $PM_{2.5}$ samples collected by Cheng et al. (2011), it was found that NIOSH-defined EC was up to 80% lower ~~(up to 80%)~~ than that of defined by IMPROVE. The IMPROVE thermal-optical method was used in this study, which

could have caused under-valuation of OC. Except for T2 and T4 trucks, almost all of the OC/EC ratios for diesel trucks calculated in this study were lower than 1, which is consistent with conclusions from previous studies (Figure 6). The OC/EC ratios for T2 during highway and non-highway driving were 5.64 and 15.5, respectively, which might be an ~~result~~ effect of the ~~effect by~~ China IV emission standard. ~~Other~~ A different study also found that modern diesel passenger cars (Euro 4 and Euro 5) had high OC/EC ratios (Alves et al. 2015b). The OC/EC ratio for T4 while driving on ~~the~~ non-highways was 4.10, which might ~~be~~ have been caused by the low driving ~~aped~~speed (the driving speed was zero for the first 500s for T4 as shown in Figure S3). ~~Because~~ Cheng et al. (2015) ~~have~~ reported that the OC/EC ratios were substantially ~~above~~ higher than 1 under idling or with low load~~. .And the driving speed was zero for the first 500 seconds for T4 as shown in Figure S3.~~

The sum of WSIs and element~~s~~ fractions was lower than 5% of the PM for all of the diesel trucks~~,~~ except for T2, which is consistent with the results of Zhang et al. (2015a). SO_4^{2-} was the most abundant ion for trucks T2 and T5, ~~while~~ whereas NO_3^- was the most abundant ion for trucks T1, T3 and T4. For T2, WSIs (13.8%) ~~were~~ was the most significant PM component ~~of PM~~, followed by OC, which was 4 to 10 times higher than other trucks (Table S6). ~~The main reason was inferred as that~~This occurred likely because T2 ~~was~~ is a China IV diesel vehicle with well-controlled combustion conditions, which ~~leading~~ leads to more water emissions, which in turn accelerates the transformation from the gas phase to WSIs (e.g., the transformation of SO_2 to SO_4^{2-}). As ~~can be seen~~shown in Table S6, Fe was the most abundant element for trucks T1 and T5, ~~while~~ whereas Ca was the most abundant element for trucks T2, T3, and T4. The total element fraction of T2 (China IV) was 16 times higher than that of T1 (China III). Although the EF_{PM} for diesel trucks decreased with stricter emission standards, the WSIs and element contents increased ~~instead~~. It is well known that sulfate and nitrate are major precursors of acid rain, and elements emitted by diesel engines also have significant adverse health effects on humans. Thus ~~and~~ attention needs to be paid to this phenomenon.

The n-alkanes, PAHs, hopane and steranes fractions were 0.85- 4.78%, 0.01-0.54%

and 0.002- 0.024%, for the trucks. As shown in Table S6, C20 was the most abundant n-alkane in PM from T1, T2 and T4, ~~while~~ whereas that from T3 and T5 was C19 ~~was the most abundant n-alkane of T3 and T5~~. ~~And t~~The most abundant species of PAHs was pyrene. N-alkanes, PAHs, hopanes and steranes accounted for the highest proportions of PM ~~for~~ in the exhaust from T3, which might ~~be~~ have been affected by many factors, including differences in the engine power rating, complex reactions in the engine (combustion processes and pyrolysis reactions related to temperature, humidity, etc.), and driving conditions. As shown in Figure 7, the isomer ratios for diesel trucks were 0.28-0.35 for BaA/(BaA+Chry), 0.08-0.22 for IcdP/(IcdP+BghiP) and 0.08-0.39 for Flua/(Flua+Pry), with averages of 0.31 ± 0.03, 0.15 ± 0.06 and 0.23 ± 0.12, respectively. These results are similar to ~~results~~ those reported by Schauer et al. (1999).

3.4 Average chemical composition of PM emitted from diesel vehicles

3.4.1 Average chemical composition of PM in excavator exhaust

The average PM chemical compositions for excavator exhaust are listed in Table 3. Carbonaceous matter was the dominant component, ~~and accounted~~ accounting for 72.5% of the PM for excavators; ~~whereas~~ furthermore, OC was the most abundant species (39.2%) for PM. The total element fraction was the second largest group, ~~and contributed~~ contributing 1.76% of the PM. Of the elements, emissions were dominated by Fe at 46.3%. In addition, the proportion of n-alkanes in the PM from excavator exhaust (5.14%) was higher than ~~the proportions~~that of the other organic matter types (PAHs were 0.098% while hopane and sterane were 0.026%) and C20/C19 was the most abundant n-alkane. For parent PAH, the emissions were dominated by pyrene and fluoranthene, followed by naphthalene and chrysene.

Table 3 summarizes the average source profiles of PM in excavator exhaust as derived in this study, as well as ~~ones~~ those 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 ~~was~~ is consistent with that for a marine engine, ~~while~~ whereas the element fraction 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). The EC fraction of PM in the excavator exhaust ~~was~~ is higher than that reported by Sippula et al. (2014), which might be the result of a lower metal fraction in the excavators used for their study. The proportion~~s~~ of n-alkanes measured in this study ~~were~~ was significantly higher than those emitted from a marine engine (~~4~~four-fold) and non-road generator (~~6~~six-fold) in ~~another~~ a different study (Liang et al., 2005), which could be the result of different aliphatic compounds in the diesel fuels (Sippula et al., 2014). For the marine engine and non-road generators, C22 and C17 were the most abundant n-alkane species. PAHs were dominated by ~~phenanthalene~~ phenanthrene for a marine engine and fluoranthene for non-generators, which ~~was~~ is different from the result obtained for the excavators. This ~~could~~ indicates that the PM emitted from different types of non-road diesel vehicles has varying source profiles based on the operational conditions.

3.4.2 Average source profile of PM for trucks

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

In comparison, the total carbon emissions in this study ~~were~~ is lower than those in previous studies, whereas the WSIs and elements fractions ~~were~~ are higher (Alves et al., 2015a, Cui et al., 2016, Schauer et al., 1999, Wu et al., 2016). Several factors ~~could~~ have influenced these differing results~~, including~~ such as fuel quality, driving condition, engine parameters (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 element in studies using on-road tests and tunnels, which is similar to our results, ~~while~~ whereas Zn and Na were dominant ~~in~~ elements ~~from~~ in the results obtained by a dynamometer. Therefore, the results obtained from the real world conditions (on-road

tests and tunnels) ~~were~~ are different from those obtained in a laboratory. For organic matter, the proportion of PAHs, hopanes and steranes to PM ~~were~~ are consistent with the results ~~from~~ of Schauer et al. (1999) and Cui et al. (2016). ~~As i~~In this study, the most abundant ~~in~~ n-alkane was C20, as measured by Schauer et al. (1999), and pyrene was the most abundant PAH, as reported by Cui et al. (2016). Thus, the average profile of PM for on-road diesel trucks is relatively stable and consistent across studies.

3.5 Comparison of source profile between excavators and trucks

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

When we compared the average percentages of chemical components in PM for excavators with those for trucks, several differences were found. In general, the carbonaceous (95.9%) and elements (1.76%) fractions for excavators were higher than those for diesel trucks (42.8% and 0.94%, respectively). As shown in Figure 8, the structures of different ring PAHs in the exhaust from excavators and trucks varied sharply, ~~especially~~ particularly for 5 and 6-ring PAHs~~, .~~ ~~although~~ However, the average percentage of total PAHs in the PM ~~were~~ was consistent between the excavators and trucks. ~~Due~~ Owing to their lipophilicity, 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 BaPeq for the excavators was 31 times ~~larger~~ than that for the diesel trucks. Almost all of the ~~parent~~

PAH BaP_{eq} values calculated in this study for trucks and excavators ~~were~~ are higher than the concentrations that cause 1/10000 of the carcinogenic risk, according to the World Health Organization (WHO). ~~Due~~ Owing to the adverse environmental effects and health hazards caused by carbonaceous compositions, elements, and PAHs, the PM emissions from excavators require urgent control.

Conclusions

This study reported the characteristics of PM source profiles for excavators and the EF_{PM} values for exhaust from excavators and trucks with different emission standards and those used in different operation~~al~~ modes, or road conditions ~~were obtained~~. 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. The highest average EF_{PM} for working excavators ~~that are working~~ (904 ± 979 mg kg^{-1} fuel) might be the result of higher engine load, which causes ~~causing~~ lower air-fuel ratios. The average EF_{PM} for the tested diesel trucks with different emission standards and vehicle sizes under different driving conditions was 498 ± 234 mg kg^{-1} fuel. The average EF_{PM} for excavators was decreased by 58% from pre-stage 1 to stage 2. Moreover, the reductions in EF_{PM} from the China II to the China IV truck and from the China III to the China IV truck were 63.5 and 65.6%, respectively. This ~~indicating~~ indicates that the improvements ~~of~~ in the emission standards and fuel quality for diesel trucks and excavators have significant~~ly~~ effects ~~one~~ on the reduction of PM emissions. It should be noticed that the EF_{PM} for China III and light-duty diesel trucks were higher than those for the other trucks, which could be a result of poor driving conditions that include~~d a~~ low average and highly variable speeds. For each excavator, the carbon component (OM+EC) was the dominant fraction and accounted for approximately 74.1-123% of the PM. The average ranges of 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 exhaust from E4, which might to the result of poor fuel quality and the vehicle's age. Additionally, the element fractions for the two

excavators produced in 2013 (E1 (1.42%) and E6 (7.50%)) were higher than those of other excavators, which might indicate that the element~~s~~ emissions control deteriorated and that more stringent control technology should be developed. For excavators, the ranges of the ~~ratios~~ BaA/(BaA+Chry), IcdP/(IcdP+BghiP) and Flua/(Flua+Pry)~~,~~ ratios 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) accounted for 44.0% (T1), 27.9% (T2), 43.9% (T3), 51.6% (T4) and 46.3% (T5) of PM. For T2, WSIs (13.8%) were the most significant fraction of PM after OC, and ~~it was~~were higher than those for the other trucks by a factor of 4 to 10. The n-alkanes, PAHs, hopane and steranes fractions ranged from 0.85 to 4.78%, 0.01 to 0.54% and 0.002 to 0.024% for trucks, respectively. In comparison with the results from other studies, the characteristics of the average source profiles for different types of non-road diesel vehicles varied sharply, ~~while~~ whereas those for on-road diesel vehicles, showed more stability. Although the PAH~~s~~ fractions for the excavators and trucks were similar, the total BaP_{eq} that was used to evaluate the carcinogenic risk was 31 times ~~greater~~ for excavators than for trucks.

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References

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

An, P. Z., Sun, W. C., Li, G. L., Tan, M. Z., Lai, C. J. and Chen, S. B.: Characteristics of particle size distributions about emissions in a common-rail diesel engine with biodiesel blends. 2nd International Conference on Challenges in Environmental Science and Computer Engineering. Q. Zhou. Amsterdam, Elsevier Science Bv. 11:1371-1378 2011

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, Environmental Science. 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 $PM_{2.5}$ 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 Science 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, Atmospheric Chemistry and Physics. 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 nn-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 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	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 ℃)(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

5

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 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	

8 n.d. = not detected

Figure captions

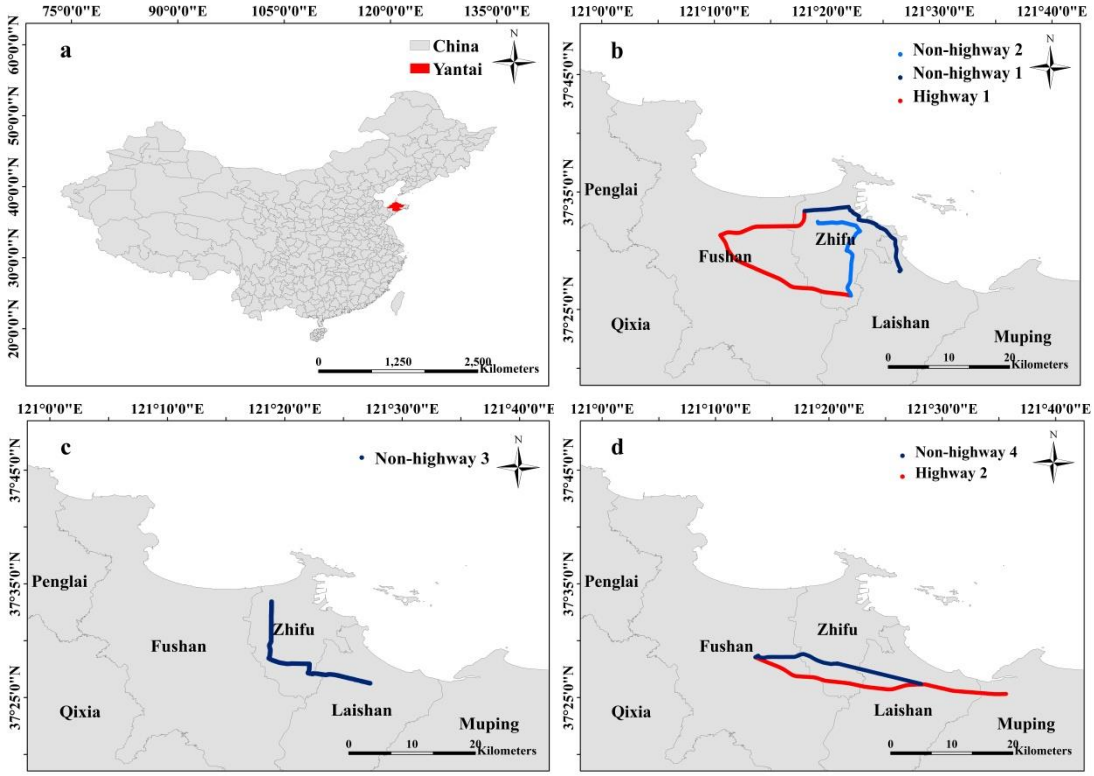

Figure 1The routes for diesel trucks; a was the site of Yantai; b was the route for China III and China IV light-duty diesel trucks; c was the rout for China II heavy-duty diesel truck; d was route for China III medium-duty and heavy-duty trucks

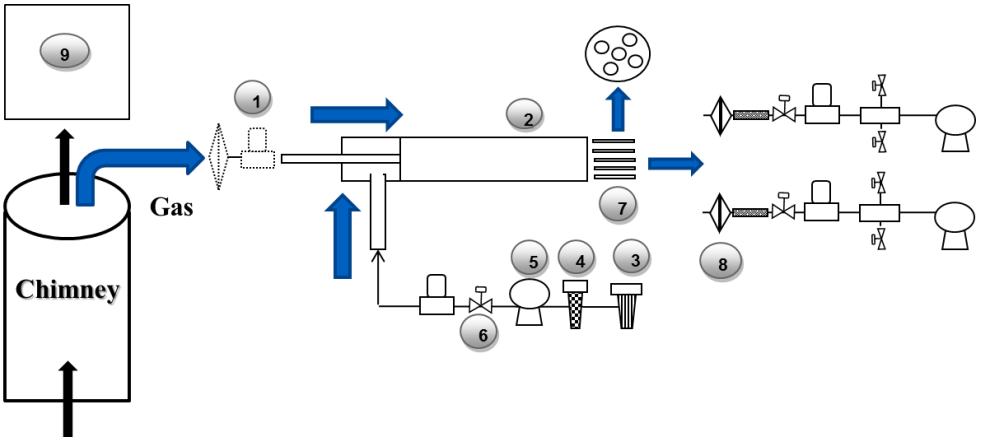

Figure 2 Particulate matter sampling system; 1 is the flowmeter; 2 is the dilute tunnel; 3 is the filtrator; 4 is the activated carbon; 5 is the fan; 6 is the valve; 7 is the flow divider; 8 is the filter membrane sampler; and 9 is the exhaust analyzer

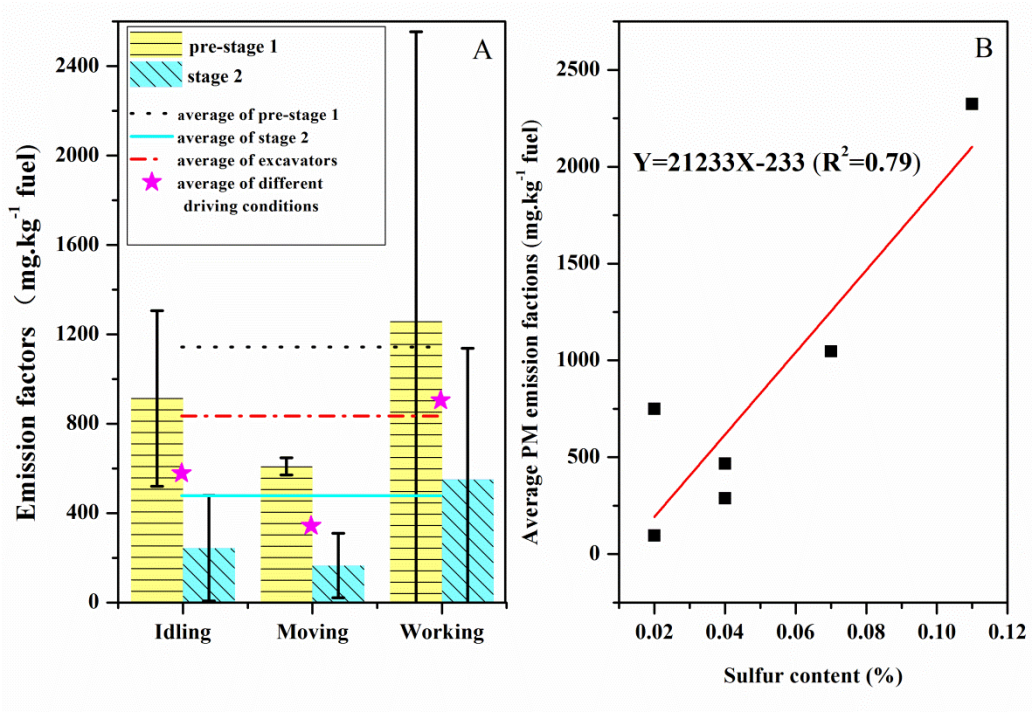

Figure 3 EF_{PM} for excavators with different operational modes and emission standards (A) and the correlation with sulfur contents (B)

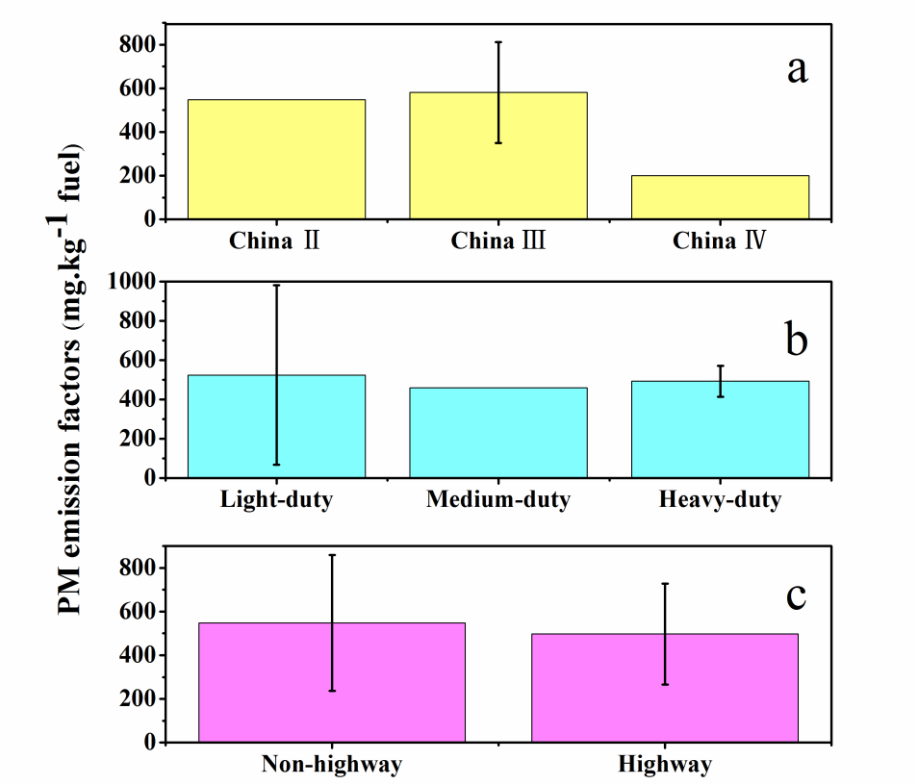

Figure 4 Diesel trucks EF_{PM} for different emission standards (a), vehicle sizes (b) and driving conditions (c)

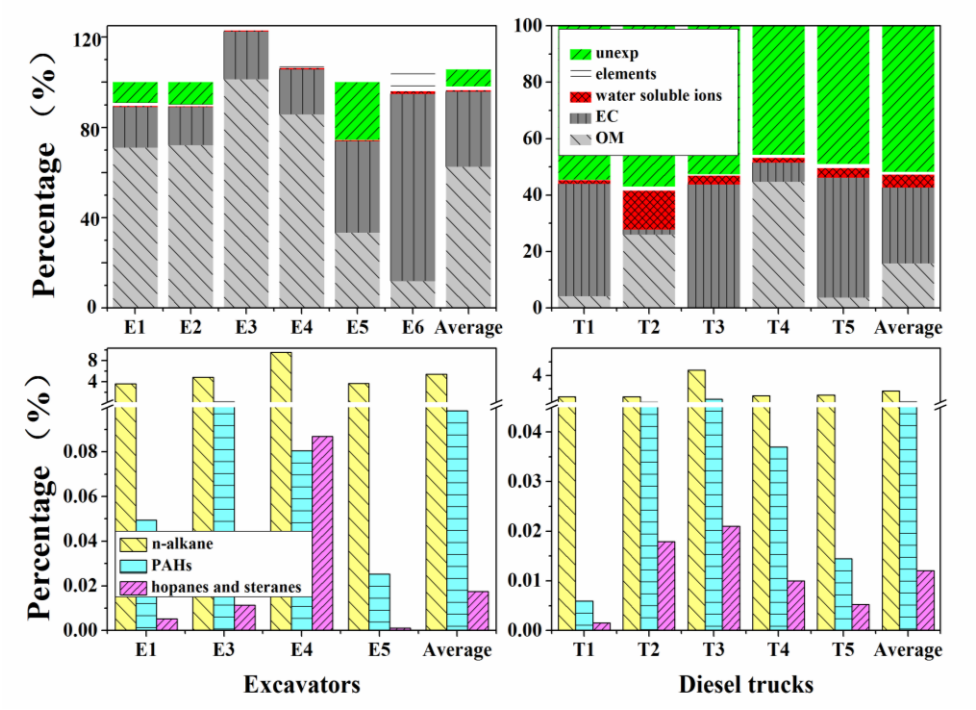

Figure 5 Compositional constituents of PM for individual vehicles (%)

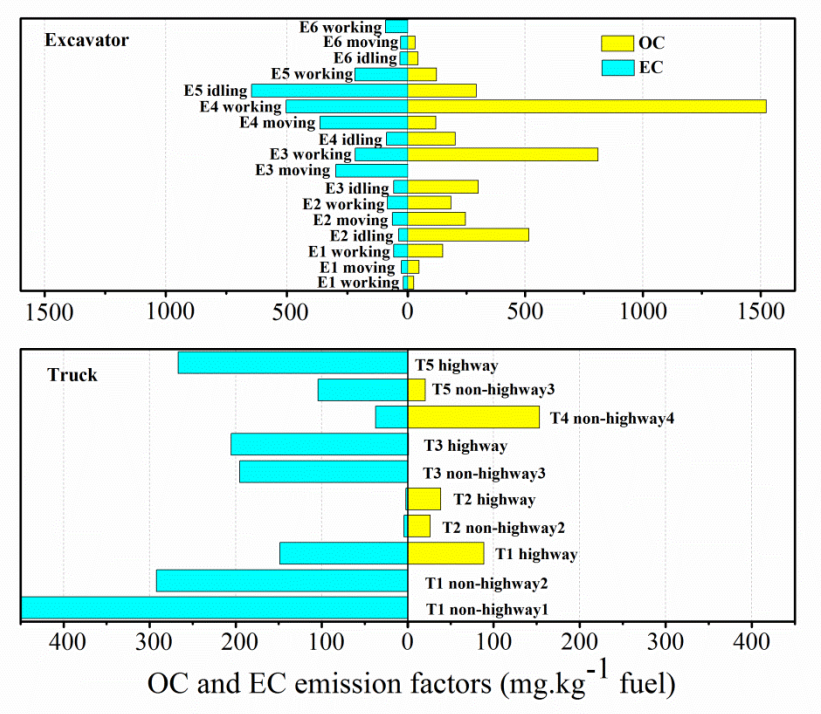

Figure 6 OC/EC ratios in different operational modes and driving conditions for excavators and trucks

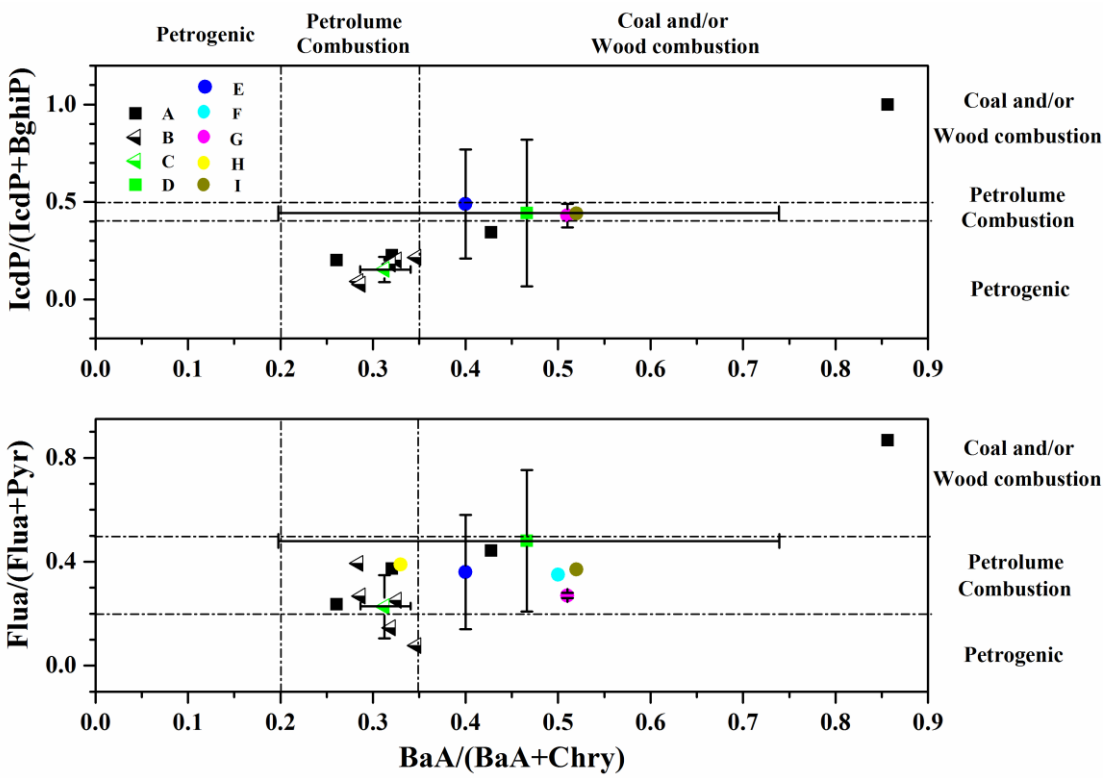

Figure 7 Cross plots for the ratios of BaA/(BaA+Chry) vs IcdP/(IcdP+BghiP) and BaA/(BaA+Chry) vs Flua/(Flua+Pry) and comparison with those from other diesel vehicle sources. A and B are the isomer ratios of the PAHs from the excavators and trucks, respectively, tested in this study; C and D are the average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

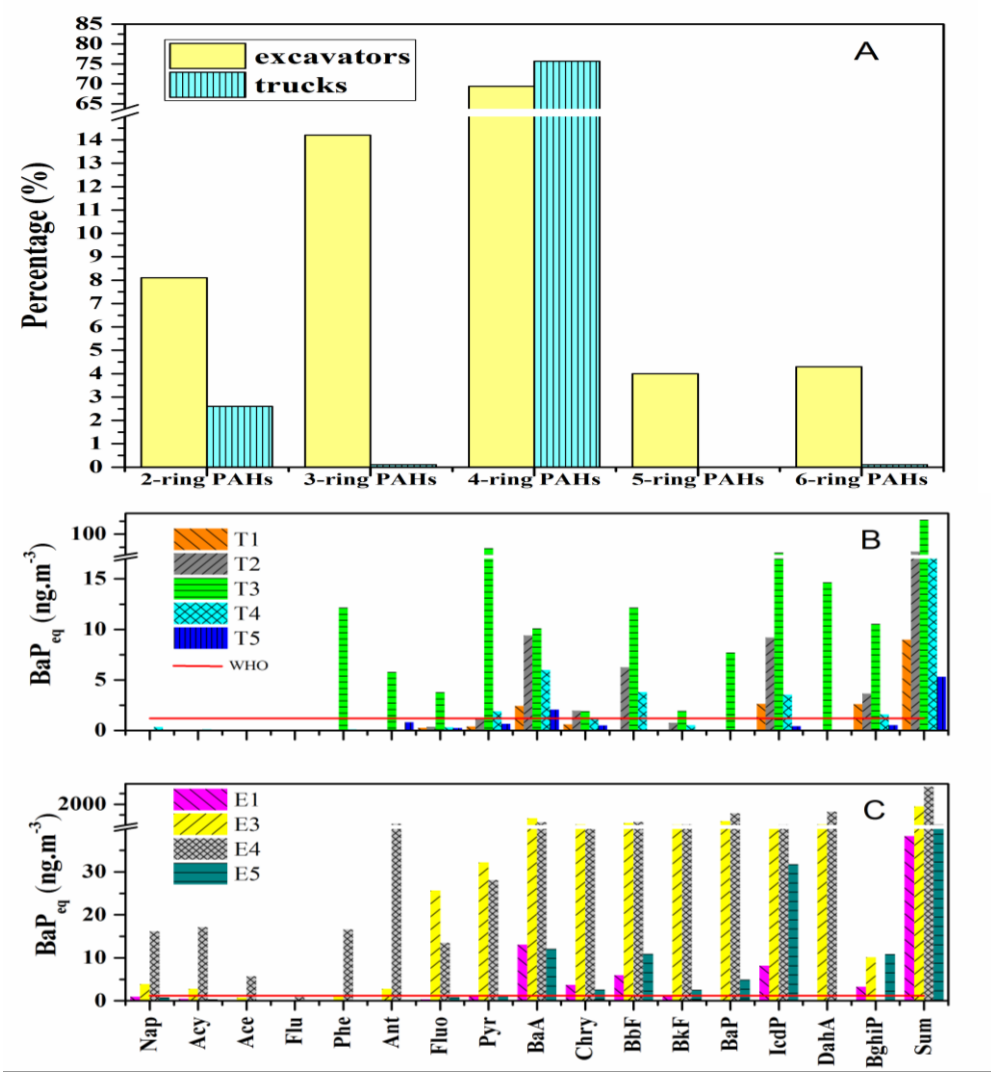

Figure 8 Percentages of each ring PAHs to total PAHs (A); BaPeq for parent PAHs in each tested trucks (b) and excavators (c)