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

Min Cui[a], Yingjun Chen[a*], Yanli Feng[b*], Cheng Li[c], Junyu Zheng[c*], Chongguo Tian[d], Caiqing Yan[e], Mei Zheng[e]

[a] Key Laboratory of Cities' Mitigation and Adaptation to Climate Change in Shanghai (China Meteorological Administration), College of Environmental Science and Engineering, Tongji University, Shanghai, China

[b] Institute of Environmental Pollution and Health, School of Environmental and Chemical Engineering, Shanghai University, Shanghai, China

[c] Laboratory for Atmospheric Research and Environmental Simulation, School of Environment and Energy, South China University of Technology, Guangzhou, China

[d] Key Laboratory of Coastal Zone Environmental Processes and Ecological Remediation, Yantai Institute of Coastal Zone Research, Chinese Academy of Sciences, Yantai, China

[e] State Key Joint Laboratory of Environmental Simulation and Pollution Control, College of Environmental Sciences and Engineering, Peking University, Beijing, China

Correspondence to: Yingjun Chen (yjchentj@tongji.edu.cn)

Yanli Feng (fengyanli@shu.edu.cn)

Junyu Zheng (Zhengjunyu_work@hotmail.com)

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

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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≤100μm) ~~emissions in ambient PM~~ (Oanh et al., 2010, Zhang et al., 2015b). 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~~ implementation~~g~~ ~~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 inventor~~y~~ies 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., 2015b). 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 km^{-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. HoweverO, 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 tobecause of the complexity of real-world conditions, includingsuch 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 essentialimportant for studies of source apportionment, human health, and climate change studies. Primary PM emitted from diesel vehicles contains a variety of chemical compositionscomponents, such asincluding 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 compositions of PM emitted from diesel vehicles. Zhang et al. (2015b) 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 factorsEF_{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 compositions emitted from diesel vehiclesare still largely unknown, especially for organic matterscompounds, 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 compositions 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~~ werelisted ~~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~~.~~:

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

~~Where~~ 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~~ 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:

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

~~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:

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

~~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 ($\mathrm{BaP_{eq}}$)

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. $\mathrm{BaP_{eq}}$ is typically used to evaluate the carcinogenic risks associated with individual PAH (Mirante et al., 2013). The $\mathrm{BaP_{eq}}$ 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 $\mathrm{BaP_{eq}}$ 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 fFuel-based emission factors of PM for in excavator exhausts

The EF_{PM} values for excavators exhaust are are presented illustrated in Figure 3, with the detailed information shown given in Table S1S5. 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 widely range ind EF_{PM} values in this studyhere maycould 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 beis 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, fFuel quality hashad 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 fromreported 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 fromreported 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 toand China IV trucks and from between the China III truck toand China IV trucks in EF_{PM} were 63.53.5% and 65.65.6%, 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 toa result of poor driving conditions, that include i.e., low average speed and more volatilehighly varied speed in speed for those trucks (Figures S2 S3 and Figure S3S4). 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, tThe 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 S3S4).

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 byby a factor of 1.6 (Almeida et al., 2006); (2) EC; (3) water-soluble ionsWSIs; 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 forIn 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 S2S6. 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 carbonOC and EC in PM ranged from 70.2% to 90.61% (Liu et al., 2005). BBecause 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 profoundseen in Figure 6, which may and could be caused affected by a number of factors, (such asincluding 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.

PAHs ~~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 giveprovides references values for the isomer ratios of PAHs for in non-road diesel vehicle exhausts.

3.3.2 Particulate matter composition for individual diesel trucks

For diesel trucks, the total carbonaceous composition (OM+EC) were accounted for 44.0% (TE1), 27.9% (E2T2), 43.9% (E3T3), 51.6% (E4T4) and 46.3% (E5T5) 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 towas –the different OC and EC detection methods used in our studyfor 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. FurthermoreTherefore, 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 ofthe isomer ratios for diesel trucks were covered fromwere 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 Fluas/(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 exhausts

The average PM chemical component compositions of PM for excavator exhausts was are listed in Table 3. It appeared that cCarbonaceous 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 exhausts (5.14%) was higher than the proportions of the those for other organic matter typess (PAHs were : 0.098% while ; hopane and sterane were : 0.026%), and C20 and / C19 were was the most abundant maximum carbon in n-alkanes. For the parent PAHs, the emissions were dominated by Pry and Fluo, followed by Nap and Chry.

To compare our results with other studies, Table 3 summarizess the average source profiles of PM for in excavator exhaust ass 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 thatose 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 thePM for excavator exhausts 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 test~~s~~ 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 excavator~~s~~ 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~~

elementsrespectively). 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 PAHsBaP_{eq} levels for excavators and trucks were absolutely difference, although the average percentage of total PAHs average percentages ofin 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 datumconcentrations 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 controlled urgently.

Conclusions

This study reporteds 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 andor 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 emission~~s~~ 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 literaturesstudies, 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 identicalsimilar, 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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Table captions

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

Table 2 Diesel contents from excavators

Table 3 Comparison of average source profiles 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					
C37	0.018					
C38	0.025					
C39	0.031					
C40	0.003					
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 (a) and the correlation with sulfur contents (b)

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 BaPeq for parent PAHs in each tested excavators (A) and trucks (B)

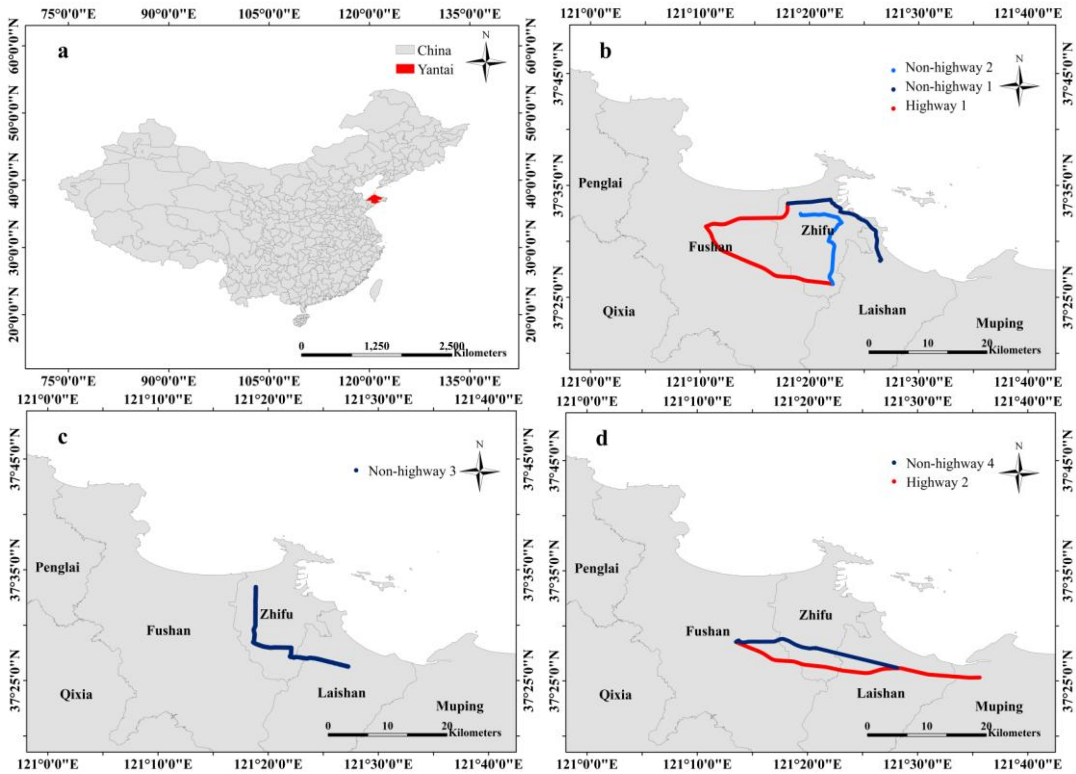

Figure 1The routes for diesel trucks; a was the site of Yantai; b was the route for China Ⅲ and China Ⅳ light-duty diesel trucks; c was the rout for China Ⅱ heavy-duty diesel truck ; d was route for China Ⅲ medium-duty and heavy-duty trucks

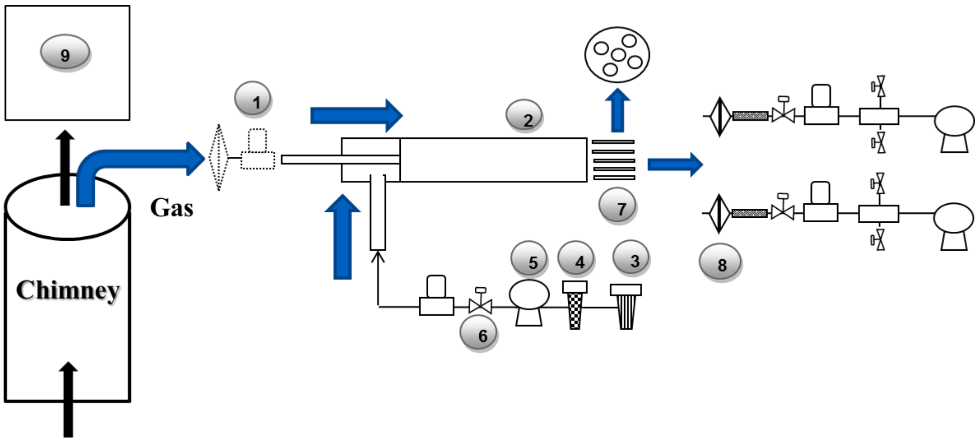

Figure 2 Particulate matter sampling system; 1 ~~was~~ is the flowmeter; 2 ~~was~~ is the dilute tunnel; 3 ~~was~~ is the filtrator; 4 ~~was~~ is the activated carbon; 5 ~~was~~ is the fan; 6 ~~was~~ is the valve; 7 ~~was~~ is the flow divider; 8 ~~was~~ is the filter membrane sampler; and 9 ~~was~~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

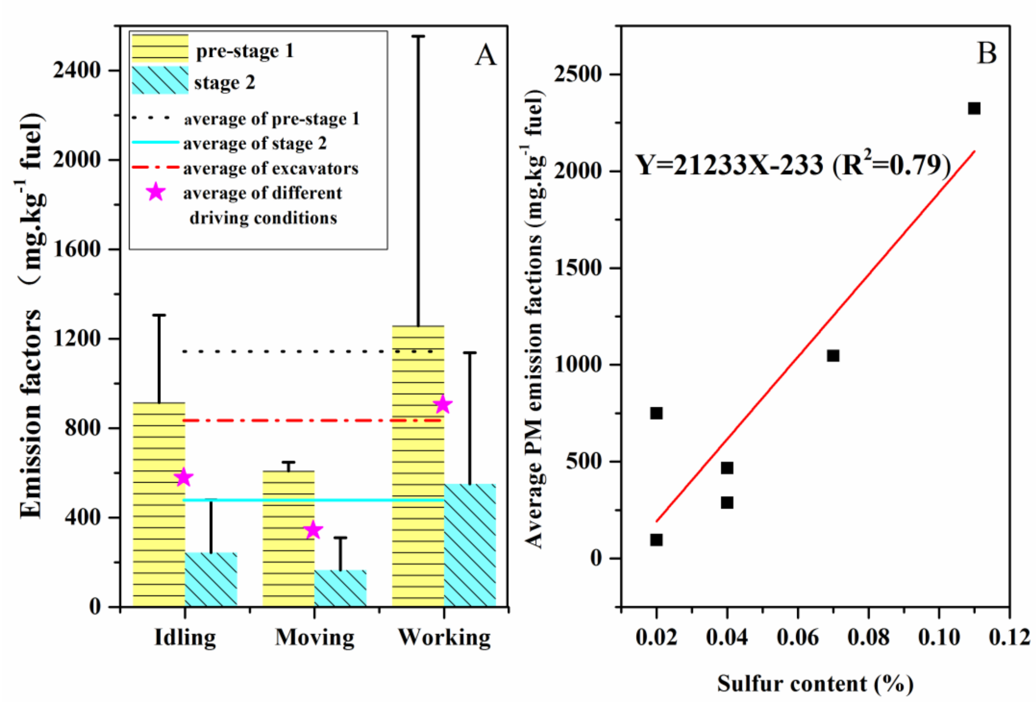

Figure 3 EF_{PM} for excavators with different operating operational modes and emission standards (A) and the correlation with sulfur contents (B)

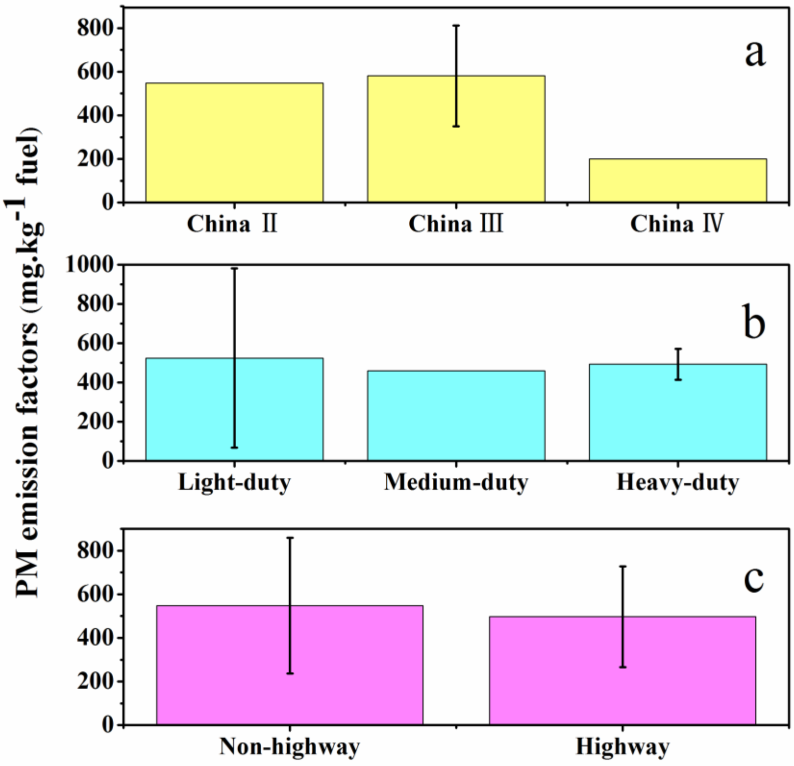

Figure 4 Diesel trucks EF_{PM} for different emission standards (a), vehicle sizes (b) and driving conditions (c)

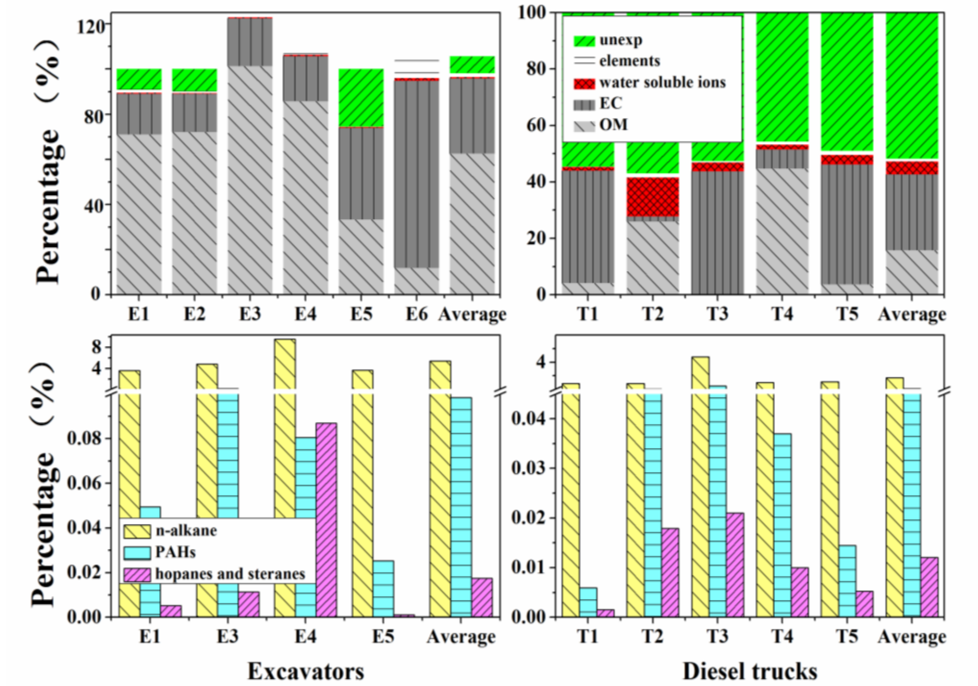

Figure 5 Compositional constituents of PM for individual vehicles (%)

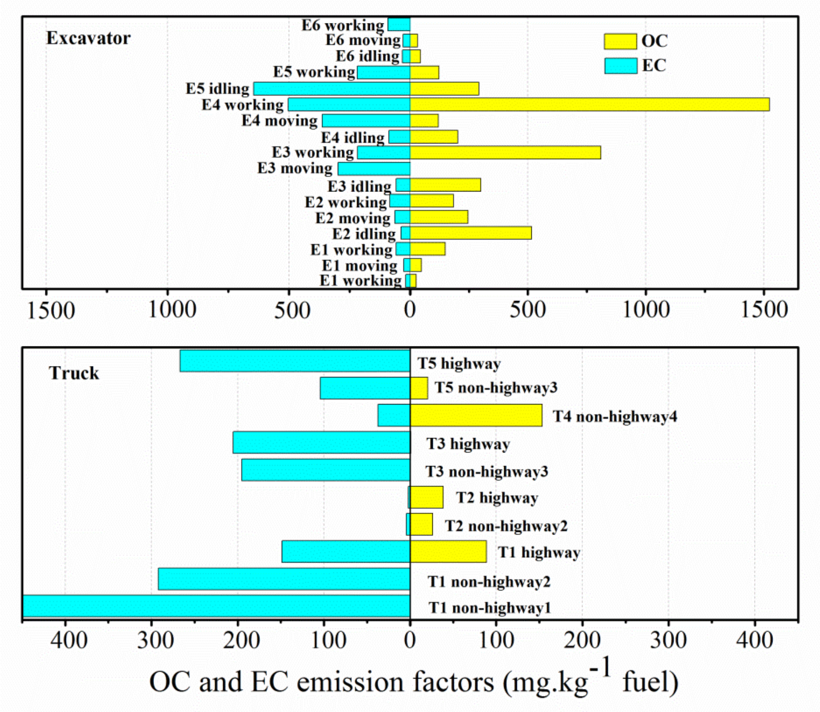

Figure 6 OC/EC ratios ~~under~~ in different ~~operating~~ operational modes and driving conditions for excavators and trucks

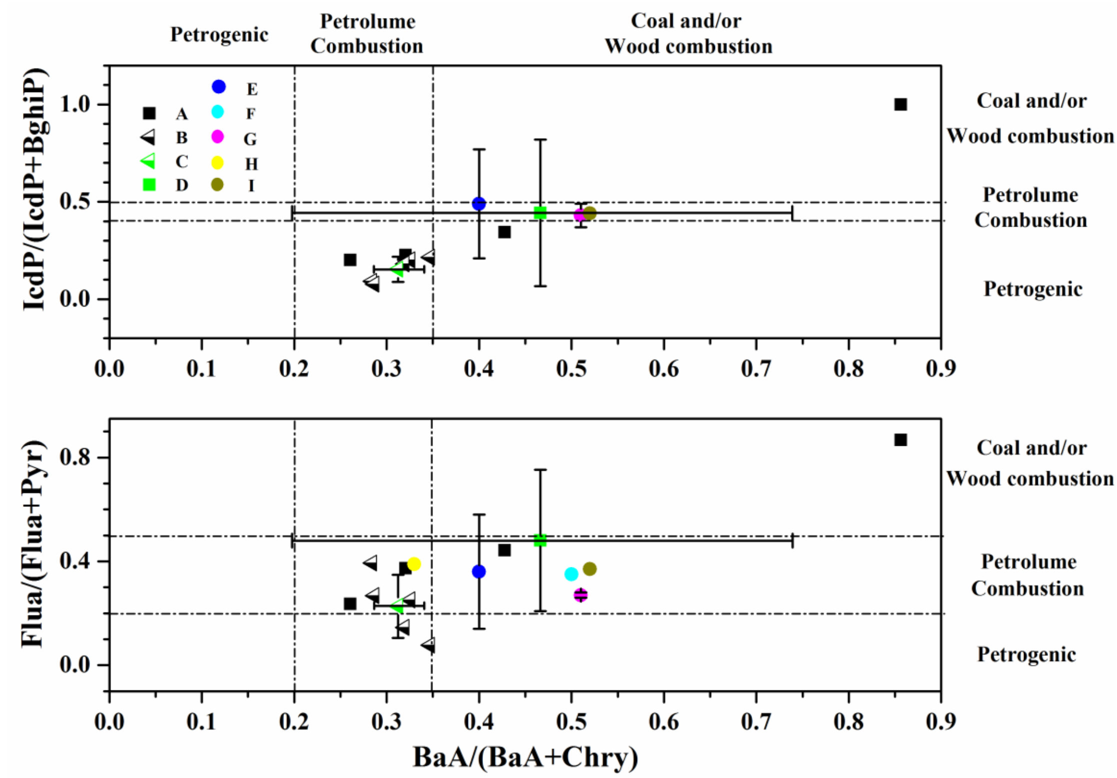

Figure 7 Cross plots for the ratios of BaA/(BaA+Chry) vs IcdP/(IcdP+BghiP) and BaA/(BaA+Chry) vs Flua/(Flua+Pry) and comparison with those from other diesel vehicle sources. A and B ~~were~~ are the isomer ratios of the PAHs ~~for~~ from the excavators and trucks~~-~~, respectively, tested in this study~~, 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)

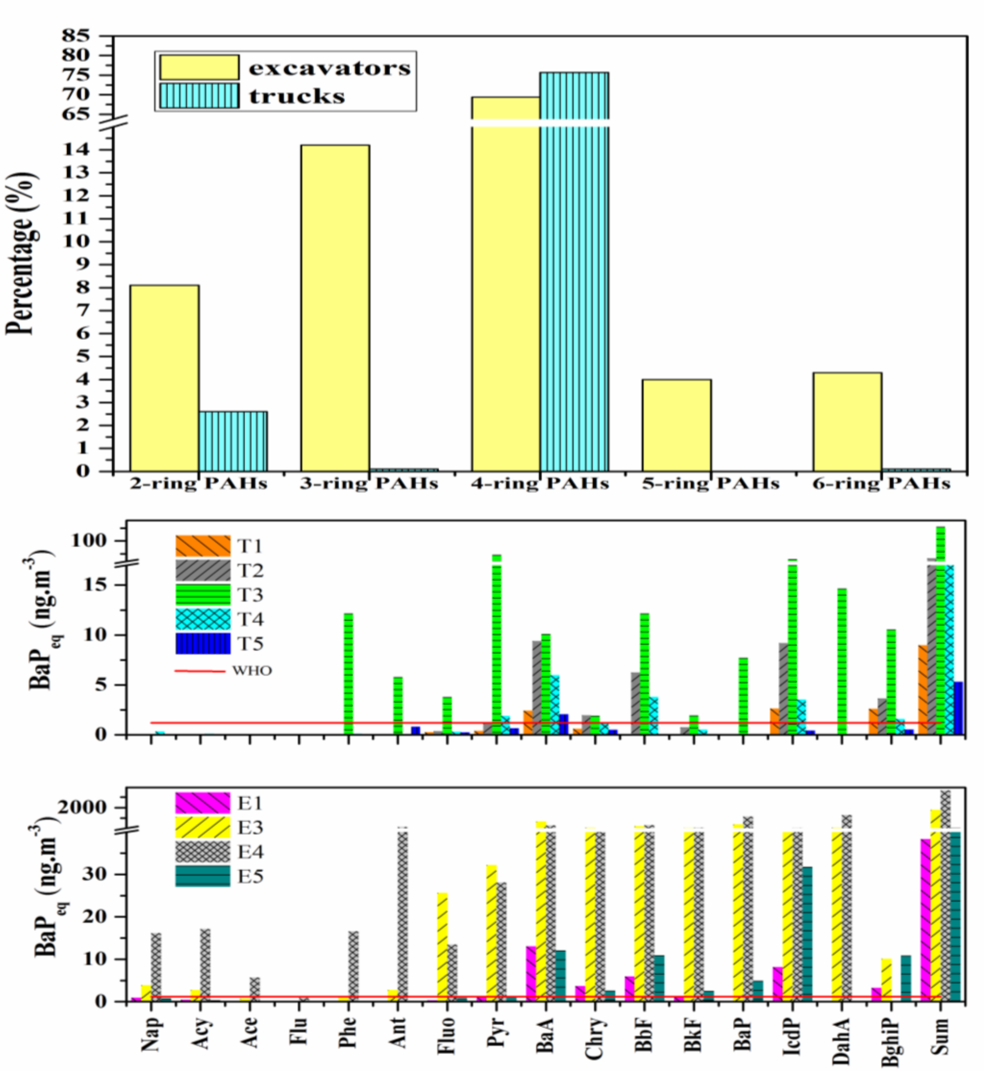

Figure 8 a) Percentages of each ring PAHs to total PAHs; BaPeq for parent PAHs in each tested trucks (~~A~~b) and excavators (~~B~~c)