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Filterable $PM_{2.5}$, Metallic Elements, and Organic Carbon Emissions from the Exhausts of Diesel Vehicles

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ABSTRACT

Urban air pollution in the form of fine particulate matter ($PM_{2.5}$) poses a substantial health threat to humans via inhalation. To assess the risks from this pollutant, we characterized and quantified the $PM_{2.5}$ exhausted by 15 diesel vehicles ranging from 28,306 to 883,374 km (an average of 525,854 km) in total mileage and varying in manufacturer and model year (1988–2005). We applied inductively coupled plasma optical emission spectroscopy (ICP-OES) to analyze the metallic element constituents, among which Ca composed the largest fraction, followed by Zn, Al, K, Fe, Mg, and Cr. Measuring the carbonaceous content, we found that the total carbon (TC), the organic carbon (OC), and the elemental carbon (EC) accounted for 3461 μg m^{-3}, 1410 μg m^{-3}, and 2051 μg m^{-3} of the $PM_{2.5}$ concentration, respectively. Of the metallic elements, Ca exhibited the highest emission factor (EF), between 45.3 and 259 μg L-$fuel^{-1}$ (with an average of 132 μg L-$fuel^{-1}$), whereas Zn and Cr displayed the lowest ones, averaging 13.1 μg L-$fuel^{-1}$ and 1.91 μg L-$fuel^{-1}$, respectively. We also investigated the relationship between the concentrations and the EFs of the metallic elements in the exhaust, and the ambient $PM_{2.5}$ composition. These results help illustrate the contribution of $PM_{2.5}$ emitted by diesel vehicles to metallic element concentrations in the natural environment and the consequent risks.

Keywords: $PM_{2.5}$ emissions; Diesel vehicles; Metallic element concentrations; Organic carbon emissions.

INTRODUCTION

The pollution control for fine particulate matter ($PM_{2.5}$) is one of the greatest challenging air pollution issues and is the highest priority pollutant in Kaohsiung City, Taiwan (Lin *et al.*, 2015; Li *et al.*, 2016). Vehicles, especially in-use diesel vehicles and heavy-duty diesel trucks (HDDTs), are considered to be among the main contributors of atmospheric $PM_{2.5}$ (Liu *et al.*, 2018b). Nonetheless, this issue indicates that we lack a comprehensive understanding of vehicular PM emission proportions in terms of the exact pollutant constituents. This information would be beneficial for defining secondary PM formation models, emission inventories (Agarwal *et al.*, 2017; Al Hanai *et al.*, 2019), and most importantly to examine the health effects of $PM_{2.5}$ exposure (Badaloni *et al.*, 2017; Chartres *et al.*, 2019).

In order to examine automobile emissions, a chassis dynamometer testing system and on-board testing system with a portable emissions measurement system (PEMS) have been employed as universal standard methods (Zhang *et al.*, 2015), and a large number of studies on vehicle emissions have been conducted in Taiwan's neighboring countries such as China, among others, in recent years (Liang *et al.*, 2019). Thus far, most of the studies on vehicle emission or quantifications have been centered around emission rate measurements, while the scope of the research on chemical characterization of $PM_{2.5}$ emissions has been inadequate. Presently, there are insufficient studies on the investigation of filterable $PM_{2.5}$, metallic element concentrations, and organic carbon emissions of $PM_{2.5}$ from the exhausts of diesel vehicles with diverse accrued mileages, particularly for in-use diesel vehicles in Taiwan (Tsai *et al.*, 2018) although several researchers established the emission inventories for black carbon (BC), organic carbon (OC), and other constituents found in $PM_{2.5}$ using SPECIATE (U.S. EPA, 2014) and recounted BC emission factors (EFs) (Campagnolo *et al.*, 2019). Numerous preliminary findings related to these factors have been conducted in other countries. For example, Jaiprakash *et al.* (2017) described the emissions from a diesel engine exhaust in India to determine the concentrations of metallic elements and the EC/OC component, and Oanh *et al.* (2010) developed a PM profile for the purpose of in-use diesel vehicles in Thailand with the help of a chassis dynamometer.

Carbon is among the most significant elements found in atmospheric PM, which constitute approximately 20–60%

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of $PM_{2.5}$ concentration and largely occurs in a form of OC and EC. Atmospheric EC is released straight from primary anthropogenic sources (Cui *et al.*, 2017), whereas OC could be emitted right from sources like primary particulates and secondary OC that may be produced from the products of chemical reactions in the atmosphere with low vapor pressure. OC and EC in PM play crucial roles with respect to the impact of global climate change, visibility degradation, and human health (Cui *et al.*, 2017).

$PM_{2.5}$ has a large surface area that has a potential to act as a transporter of metallic elements and heavy metals (Jiang *et al.*, 2019; Sun *et al.*, 2019), bacteria, and viruses that can also be inhaled by human beings via the respiratory system and absorbed by the alveoli or can enter other organs through lung aeration, possibly causing asthma, bronchitis, and cardiovascular diseases (Liu *et al.*, 2016; McGuinn *et al.*, 2019). Many studies on heavy metals as constituents of $PM_{2.5}$ and their sources have been carried out in different parts of the world, including China, Denmark, Turkey and France (Hvidtfeldt *et al.*, 2019; Jiang *et al.*, 2019).

It has been reported by various studies that exposure to metallic elements is associated with lower lung function and higher occurrence of respiratory symptoms and diseases in adults (Achilleos *et al.*, 2017; Akinwunmi *et al.*, 2017). Suvarapu and Baek (2017) examined both the concentrations of metallic elements in $PM_{2.5}$ (external) and in blood (internal) by acquiring inclusive statistics about metallic elements and examined the effect of metallic elements on human respiratory health (Grivas *et al.*, 2018). Metallic elements have been proven to have harmful effects on human body, and metallic components attached to $PM_{2.5}$ particles are typically found in the $PM_{2.5}$ concentrations (Pervez *et al.*, 2018). Thus, metallic elements can pose harmful effects on the heart, lungs, and nervous system, and people who are exposed to toxic metallic elements over a long period of time may be affected (Yang e*t al.*, 2019; Zhao *et al.*, 2019). In this case, children are the most vulnerable to toxic environments and may suffer from learning disabilities, memory and attention deficits, or psychiatric complications (Lee *et al.*, 2018; Liu *et al.*, 2018a; 2019). As detailed in the introduction, metallic elements in the natural environment constitute the source of many diseases, and their concentrations and quantity impact health in different ways (Zhang *et al.*, 2018; Turap *et al.*, 2019). In Huzhou, China, Peng *et al.* (2017) assessed the significant source of contributions to $PM_{2.5}$-bound metallic elements (i.e., Cr, Co, Ni, As, Cd, and Pb) and the order was as follows: secondary sources > vehicle sources > soil dust > coal combustion > cement dust. Cr is typically released to air mainly through processes of combustion and through metal industries. Other sources of Cr into the environment include cement-manufacturing plants (cement contains chromium), chromium-containing asbestos linings, chromium-based automotive catalytic converter pollution, and tobacco smoke (ATSDR, 2012; Tchounwou *et al.*, 2012; Ryder *et al.*, 2020). Cr exposure has been associated with various health conditions such as nasal septum atrophy and cancer (ATSDR, 2012).

To address this issue, we conducted a comprehensive study of filterable $PM_{2.5}$, metallic element concentrations in $PM_{2.5}$ and organic carbon emissions from the exhausts of 15 in-use diesel vehicles. The ultimate goal for this study was to investigate and determine the concentrations of metallic element concentrations in $PM_{2.5}$ and organic carbon emissions from the exhausts of diesel vehicles by performing a chemical analysis (Fe, Zn, Mg, K, Ca, Al and Cr) with the help of inductively coupled plasma optical emission spectroscopy (ICP-OES).

MATERIALS AND METHODS

Sampling Methods

Diesel Vehicle Details and Engine Testing

15 vehicles were tested in this study to determine the concentrations of metallic elements in $PM_{2.5}$ and organic carbon emissions from the exhausts of different diesel vehicle groups. The detailed specifications of each sampled vehicle, including the brand, model year, fuel type, displacement (L), emission standard classification, and mileages (km), among indices, are shown in Table 1. The samples from the 15 tested diesel vehicle exhausts were collected at a constant, measured flow rate, suitable for the precise testing procedure for the interval corresponding to the period of each mode of the driving phase. Therefore, an emissions rate (g kWh^{-1}) for each technique of the cycle was achieved. The descriptions of the 15 experimented diesel vehicles are presented in Table 1, while the schematic illustration of the experiment is presented in Fig. 1.

Sampling Procedures for Pollutant Emissions

The emissions from gaseous pollutants, mainly $PM_{2.5}$, in the tail-pipe exhaust were monitored and observed online by using a portable, convenient gaseous pollutant analyzer (Sprint V4; Telegan). This specific analyzer has a detection limit of 0–5000 ppm, with a 1 ppm resolution and a reaction time of 30 seconds. During the experiments, the air samples were uninterruptedly extricated from the tail exhaust pipe, and a fraction of the sample was directed to the analyzer to identify the pollutants of significance. The controller unit, which serves as a convenient, flexible gauging device for spot checks and on-site measurements, was furnished with a probe socket and a coordinated disproportion pressure test. The inclusive range of tests makes it appropriate for precise measurements of temperature, humidity, pressure, velocity, current, and voltage. The device provides consistent precision and reliability in research facility measurements in a compact unit that can quantify five significant constituents (PM_{10} and $PM_{2.5}$, O_3, SO_2, NO_2/NO_x, and CO) in the field. It provides a quicker response time than the prevailing models and therefore, it is 20% lighter. Preparation interval has also been reduced by half to enable portable measurement. This instrument is simple to operate, and features are of innovative design that secures the unit from shocks and vibrations, features that improve its practicality in the field.

Sampling Procedures and Particle Component Analysis for EC/OC

During this study, Pallflex Tissuquartz filters (2500QAT-UP) were employed for the collection of particles, where

Table 1. Details of diesel vehicles tested for $PM_{2.5}$ emissions and metallic element concentrations in $PM_{2.5}$.

Number	Items				
	Manufacturer Brand	Model Year	Emission Standard Classification	Displacement (L)	Cumulative Mileage (km)
M1	Hino	1999	Euro 2	12.9	828,440
M2	Hino	1999	Euro 2	6.5	425,117
M3	Hino	2000	Euro 3	7.7	325,357
M4	Fuso	1994	Euro 1	11.1	651,818
M5	CMC Motor	2005	Euro 4	7.5	419,002
M6	CMC Motor	1988	Pre-Euro	3.3	432,211
M7	CMC Motor	2005	Euro 4	2.8	541,318
M8	Isuzu	1988	Pre-Euro	6.5	639,426
M9	Hino	1993	Euro 1	6.5	761,672
M10	Mitsubishi	1992	Pre-Euro	16.0	28,306
M11	Nissan	1994	Euro 1	18.0	492,874
M12	CMC Motor	1996	Euro 2	7.5	883,374
M13	Mitsubishi	1997	Euro 2	12.0	466,443
M14	Isuzu	2000	Euro 3	7.8	711,332
M15	Mitsubishi	1997	Euro 2	12.0	643,118

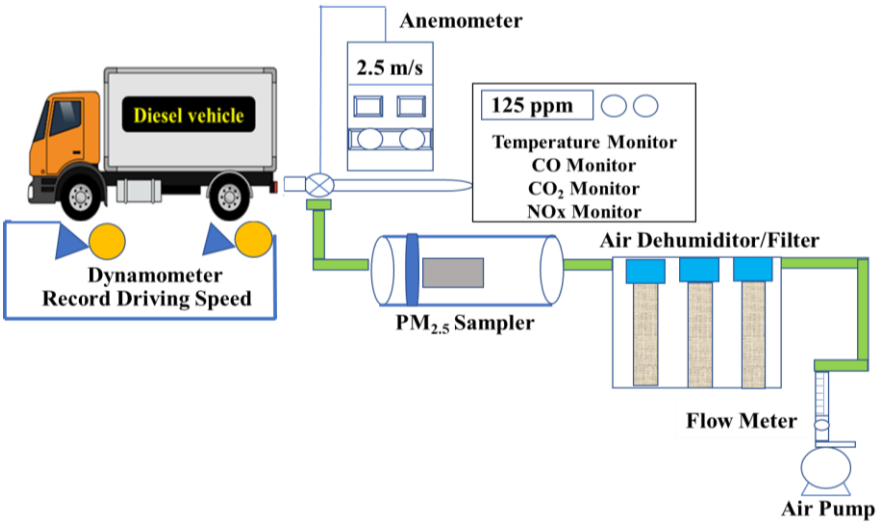

Fig. 1. Schematic diagram of the experimental system.

each 0.5 cm^2 filter area was used to quantify OC and EC. These filters were pre-baked at a temperature of 550℃ for 6 h to remove all adsorbed OC before use. Both OC and EC were measured and estimated with a Thermal/Optical Carbon Analyzer (Model 2001A; Desert Research Institute, USA) in accordance with the NIOSH 5040 procedures (Zhang *et al.*, 2015). With the organic vapor adsorption on the quartz filters, particularly volatile OC can produce positive artifacts. Further discussion on procedures and particle component analysis used in this section is provided in Section S3 of the supplementary material.

Analytical Procedures

Measurements for Atmospheric Metallic Element Concentration

The PM and further pollutant samples along with blank quartz fibers were analyzed to determine the chemical and isotopic composition via absorption. The $PM_{2.5}$ samples along with the two blank quartz fiber filters were absorbed with a mixed acid of HF + $HClO_4$ (10:1) and HNO_3 for 24 h in securely sealed Teflon flasks on a hot plate at a temperature of 120℃. In order to minimize the chance of contamination, all acids used for the absorption process and solution procedure were comprised of ultra-pure reagents, and Milli-Q water was applied in their preparation and dilution. The removal of the supernatant was performed through filtration (0.45 µm membrane filter paper), acidified with a few drops of nitric acid and stored at 4℃. The chemical analyses and determination of metallic element concentrations of Fe, Zn, Mg, K, Ca, Al, and Cr were carried out with ICP-OES (Optima 7000 DV; PerkinElmer, USA).

RESULTS AND DISCUSSION

$PM_{2.5}$ Concentration and Emission Factor

As presented in Table 2, the $PM_{2.5}$ concentration (µg m^{-3})

from 15 diesel vehicles was found to be in the range of 3875–16,458 μg m^{-3}. The $PM_{2.5}$ released from the exhausts of these 15 diesel vehicles was the main component, accounting for an average concentration of 7927 μg m^{-3} of $PM_{2.5}$. The emission $PM_{2.5}$ factors from the diesel exhausts (μg L-$fuel^{-1}$) were analyzed as indicated in Table 2. $PM_{2.5}$ concentrations within the diesel exhausts of the 15 diesel vehicles ranged from 3875–16,458 μg m^{-3}, with the 1169–3881 μg L-$fuel^{-1}$ emission factors accounting for the concentration average of 7927 μg m^{-3} and 2006 μg L-$fuel^{-1}$, as detailed in Table 2. It was further found that the concentration of $PM_{2.5}$ from the vehicle exhausts was affected by various factors, including vehicle type, fuel quality, vehicle model year, and the vehicle speed (Jhang *et al.*, 2018; Lin *et al.*, 2019). The results further showed that the diesel vehicle emissions had a significant effect on the $PM_{2.5}$ concentration. This additionally implied that $PM_{2.5}$ emission factors could be minimized once the emission control policies are enforced by authorities like the Kaohsiung City Government. That is, the $PM_{2.5}$ emission factors would in all probability be reduced to 20.4% through enforcement of Taiwanese EPA emission standards (TEPA, 2020).

Carbonaceous Components in $PM_{2.5}$

The largest quantity of $PM_{2.5}$ under this investigation came from EC and OC. The organic material comprised several components such as engine lubrication oil, unburned fuel, and low levels of incomplete combustion and pyrolysis products (Chen *et al.*, 2019). The OC portion varied from 426–2870 μg m^{-3} (with a concentration average of 1410 μg m^{-3}), as shown in Table 3. The EC fraction ranged from 824–5570 μg m^{-3} (with a concentration average of 2051 μg m^{-3}), while TC had a fraction ranging from 1250–8390 μg m^{-3} (with a concentration average of 3461 μg m^{-3}). However, some of the 3461 μg m^{-3} of TC was attributed to metallic species and other $PM_{2.5}$ constituents such as the concentrations of the most common three major inorganic ions in $PM_{2.5}$, SO_4^{2-} (sulfate), NO_3^- (nitrate), and NH_4^+, (ammonium) (Jiang *et al.*, 2019). Fig. 2 shows the main chemical components (contribution of carbonaceous components) in the $PM_{2.5}$ emitted from different diesel vehicle groups in raw exhausts under most driving conditions. It was observed from the

Table 2. $PM_{2.5}$ emission concentration (μg m^{-3}) and emission factor (μg L-$fuel^{-1}$).

Parameter	Diesel engine (n = 15)			
	Range	Mean	SD	RSD (%)
Concentration (μg m^{-3})	3875–16,458	7927	3563	44.9
Emission factor (μg L-$fuel^{-1}$)	1169–3881	2006	650	32.4
Fuel consumption (L min^{-1})	0.5–1.3	0.804	0.269	33.5

*SD = standard deviation; RSD = relative standard deviation.

Table 3. Carbon concentration in $PM_{2.5}$ emitted from diesel engines (μg m^{-3}).

Component	Diesel engine (n = 15)			
	Range	Mean	SD	RSD (%)
EC	824–5570	2051	1340	65.3
OC	426–2870	1410	885	62.8
TC	1250–8390	3461	2010	58.1

*SD = standard deviation; RSD = relative standard deviation.

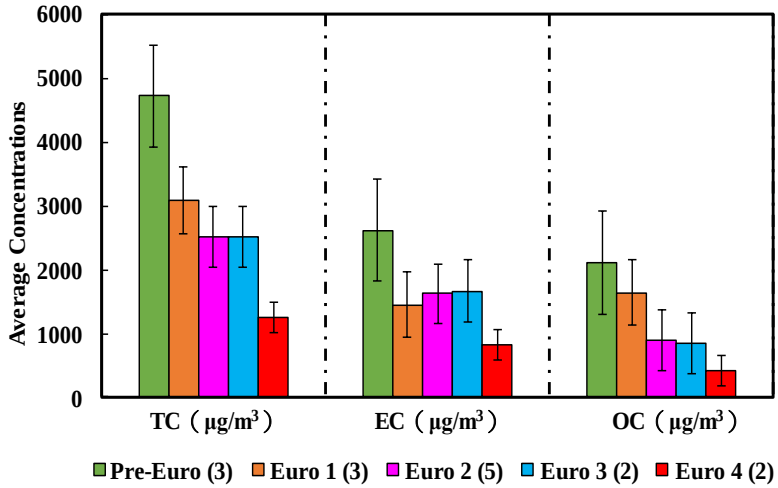

Fig. 2. Main chemical components (contribution of carbonaceous components) in the $PM_{2.5}$ emitted from different diesel vehicle groups.

results that the contribution of carbonaceous components in $PM_{2.5}$ resulted in an increase in TC, particularly in the case of the diesel vehicles that fell into the pre-Euro group, which contributed over 4500 μg m^{-3} of the carbonaceous components in the $PM_{2.5}$, followed by diesel vehicles in the Euro 1 group, with a fraction contribution of approximately 3000 μg m^{-3} of carbonaceous components in the $PM_{2.5}$. Diesel vehicles in the Euro 2 and Euro 3 groups contributed an equal fraction of more than 2500 μg m^{-3} of carbonaceous components in the $PM_{2.5}$ from raw exhaust under the various driving conditions. The diesel vehicles in the Euro 4 group contributed less than 1250 μg m^{-3} of TC. The EC contribution of carbonaceous components in the pre-Euro diesel vehicles group was observed to be less than 2700 μg m^{-3}, whereas the diesel vehicles in the Euro 1 group had an EC contribution of less than 1600 μg m^{-3}, and the diesel vehicles in the Euro 2 and Euro 3 groups had EC contributions of more than 1600 μg m^{-3}. The EC contribution from diesel vehicles in the Euro 4 group continued to decline, with less than 850 μg m^{-3}. The OC contribution was also observed to be declining in all pre-Euro to Euro 4 diesel vehicle groups, ranging from 2100 to 425 μg m^{-3}.

Jhang *et al.* (2018) found that the driving cycle, engine age, and engine technology are the major issues causing mass emissions of PM from diesel engines, which was similarly projected to have a substantial effect on the chemical composition of PM emissions from diesel engines. A study by Wei *et al.* (2019) on ship emissions clearly validated that engine operating condition has a substantial effect on the fraction of EC to OC in the $PM_{2.5}$ emissions from diesel-fueled vehicle working over an FTP cycle. It is clear that more investigations of this nature are needed for a better understanding of the components that influence the proportion of EC to TC in the emissions from diesel engines. Right now, only a restricted measurement of EC-OC for diesel engine emissions exists that exploits the NIOSH and IMPROVE EC-OC measurement techniques (Shen *et al.*, 2018).

Zhang *et al.* (2014) reported a reduction of 32.8% from the pre-EURO (an average of 60.3%) to the EURO 1 (an average of 60.7%) EC emission factors that follows the implementation of progressively stricter emission criteria. Zhang *et al.* (2009) quantified $PM_{2.5}$ emissions from HDDTs and attained carbonaceous emission factors. They revealed that the proportions of BC and OC in $PM_{2.5}$ were 43% and 37%, respectively. Shah *et al.* (2004) reported an EC proportion of 66% and OC portion of 21% in their international diesel PM emission inventory, whereas Oanh *et al.* (2010) found EC to be 47% using chassis dynamometer testing method. The EC proportion in our study was marginally higher than in these investigations, yet at the same time was within the same range. In relation to these results, the proportion of EC in our test was approximately 18.3–40% higher for the diesel vehicles.

Variations in Metallic Elements Concentrations and Contribution (%) in $PM_{2.5}$

The different metallic element concentrations and contributions (%) in $PM_{2.5}$ under this investigation are reported in Table 4. The average concentration of metallic elements (MEs) in $PM_{2.5}$ was recorded as follows: Ca had the highest concentration of 450.7 μg m^{-3}, followed by Mg with the second highest average concentration of 264.9 μg m^{-3}, Al with a concentration average of 153.3 μg m^{-3}, and K with a concentration average of 87.3 μg m^{-3}. The lowest concentration of metallic elements in the $PM_{2.5}$ emissions (μg m^{-3}) was observed in Cr, with an average concentration of 6.8 μg m^{-3}, followed by Fe, with an average concentration of 40.7 μg m^{-3} and Zn, with a concentration average of 42.3 μg m^{-3}. In all the metallic element concentrations in $PM_{2.5}$ (μg m^{-3}), the average contribution of total MEs emitted from the 15 tested vehicles was 1046 μg m^{-3}. A total of 7 metallic elements in $PM_{2.5}$ emitted from the 15 different vehicles were categorized. With the inclusive quantified metallic elements on the particle-bounded of $PM_{2.5}$ concentrations (total concentration), the highest metallic element concentration in $PM_{2.5}$ emitted from the 15 diesel vehicles was found to be in Ca (42.0%), followed by Zn, which accounted for an average of 25.0%, while Al had a contribution of 15.0%, and Cr accounted for an average of 0.695% as the smallest amount, as shown in Fig. 3.

Our results are compatible to the findings in the earlier literature by Ojekunle *et al.* (2018) who reported that metallic elements such as Cd, Cr, Ca, Cu, and Zn all had higher concentrations in $PM_{2.5}$ exceeding the WHO criteria in a study conducted in Nigeria. In a study conducted in Great Britain by Harrison *et al.* (2012), nearly one third of the Zn in atmospheric dust was ascribed to vehicle emissions such as diesel vehicles. Comparably, a related study was carried out by Liu *et al.* (2018a) on the priority of emission control

Table 4. Metallic element concentrations in $PM_{2.5}$ emitted from diesel engine exhaust (μg m^{-3}).

Element	Diesel engine (n = 15)			
	Range	Mean	SD	RSD (%)
Fe	22.1–73.5	40.7	23.0	56.5
Mg	135–441	264.9	77.6	29.3
Zn	9.87–87.9	42.3	22.4	52.9
K	16.3–118.9	87.3	38.9	44.6
Ca	286–928	450.7	205.7	45.6
Al	130–282	153.3	49.4	32.2
Cr	2.89–13.3	6.8	3.5	51.5
Total ME concentration	–	1046	–	–

*SD = standard deviation; RSD = relative standard deviation.

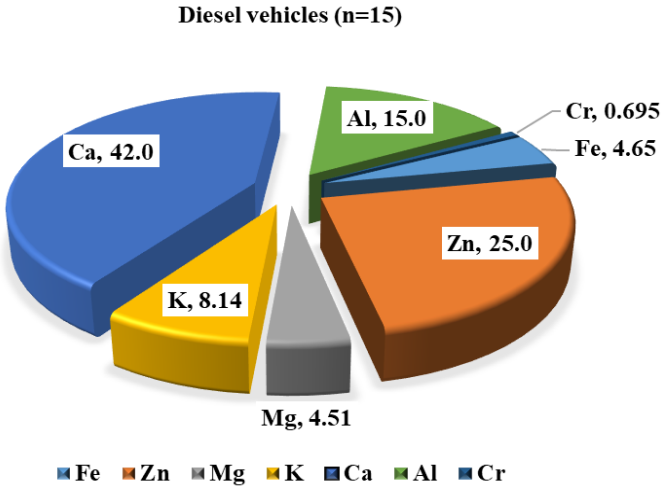

Fig. 3. Different metallic element contributions (%) in $PM_{2.5}$ emitted from diesel engines.

for $PM_{2.5}$-bound metallic elements in different seasons in Beijing, China. They found that Ca was among the metallic elements identified in annual trends in higher emitted levels, among which resuspended dust modeled the uppermost mass contribution to $PM_{2.5}$-bound metallic elements. The origin of metallic species and their effects on human health at concentrations of up to 30 atmospheric metallic elements can be determined concurrently in a single ICP analysis with an accuracy of ±5 ppm (Watson, 2010). Most of the metallic species commonly found in used oil come from two main sources: the lubricant additive package and that due to wear on engine components (Watson, 2010). Lubricant-derived ash is generally comprised of oxides, sulfates, and phosphates of Zn, Ca, and Mg (Suvarapu and Baek, 2017).

It is commonly acknowledged that there is no standard method for regulating the worn metal content in lubricating oils, although this evidence is believed to be beneficial by most vehicle operators and owners (Watson, 2010). There are two techniques commonly used for the determination of worn metallic species in lubricating oils (Cheng *et al.*, 2018). Spectrometric analysis provide data on the composition of metallic species emanating from wear on engine components and/or from contamination and also from the metallic salts' concentration in additives. The additive elements in % m/m are as follows: Ca, Mg, P, Zn, Mo, B, and N. Their concentrations may be used to classify the lubricant in use or to suggest that the lubricant tested has been mixed or contaminated by another type, grade, or brand (Watson, 2010). Worn metal elements (in ppm) such as Fe, Cr, Al, Pb, Sn, or Cu might signify wear on bearings, piston rings, cylinder liners, and other engine components. Contamination (in ppm) by MEs such as Na, V, Al, Si, Cl, and Ni are potential indicators of fuel-derived contamination. Mg, B, Cl, and Na can be extracted from seawater; Na and B can be extracted from cooling water, and Si can also be found in dust. However, Si can also be present as an anti-foam additive (Watson, 2010).

In this study, the atmospheric metallic elements found in our investigation included Fe, Zn, and Cr. These MEs have been found to have substantial effects on human health. Fang *et al.* (2010), in their study on metallic element pollution in Asia from 2000–2007, found that exposure to individual MEs can be a source of various risks for humans. For instance, extended exposure to Fe may result in the development of pneumoconiosis, whereas prolonged exposure to Zn may result in arteriosclerosis, hypertension, and heart disease. Remarkably, they also found out that Cr(VI) is carcinogenic and has the potential to cause asthma, liver damage, and nasal septum perforation (Banu *et al.*, 2017; Oruko *et al.*, 2020). Furthermore, Ni may lead to nasal and lung cancer (Cheng *et al.*, 2018). In most cases, Cr species can get into the eukaryotic system and cause impulsive reactions with the intracellular reductants, for instance, ascorbate and glutathione, causing the short-lived intermediates Cr(V) and/or Cr(IV), free radicals and the Cr(III) end-product (Oruko *et al.*, 2020). The short-lived intermediates within the cytoplasm are oxidized to Cr(VI), which quickly combines with complexes of DNA proteins and alters their usual physiological roles (Sun *et al.*, 2019) as well as destroying their DNA (Oruko *et al.*, 2020), which has genotoxic and mutagenic consequences. These also block important functional groups, displace other metal ions or change the active conformation of biological molecules (Torkmahalleh *et al.*, 2013; Nigam *et al.*, 2015) which contributes to hepatic damage and pulmonary obstruction and causes skin inflammation, gastrointestinal problems leading to ulcer formation. Moreover, Cr(VI) species has the potential to accumulate in the placenta and damage the fetus development, leading to birth defects and a decrease in reproductive health (Banu *et al.*, 2017; Oruko *et al.*, 2020). Moreover, Cu can lead to health conditions such as interstitial fibrosis, lung cancer, and pulmonary granuloma, whereas exposure to Cd and Pb may cause anemia, blood poisoning, and itai-itai disease.

These MEs may enter the coastal waters and estuaries, thereby causing devastating problems that could result in extensive harm to the life and activities of aquatic organisms and can also cause huge fatality. Thus, the accumulation of MEs in marine ecosystems becomes a significant global concern. MEs contamination might have devastating impacts

on the ecological balance of the environment and negatively affect the diversity of aquatic organisms (Oruko *et al.*, 2020). Among animal species, fishes are the inhabitants that cannot escape from the harmful effects of these ME pollutants (Banu *et al.*, 2017; Oruko *et al.*, 2020).

Wang *et al.* (2019) studied the health risk assessment of $PM_{2.5}$ in students' dormitories at a university in China and found that MEs in the dormitory comprised a mixture of coal combustion and emissions from industrial activities (71.21%), vehicle exhaust and traffic-related road dust (11.26%), and fugitive emissions ascribed to student activities (5.68%). They also found that $PM_{2.5}$-related MEs caused a greater health risk to college students in the dormitory environment than in the outdoor environment. However, the exposure environments of the college students in this study were limited to dormitory and outdoor environments. Wei *et al.* (2019) emphasized that exposure to pollutant emissions may result in the development of myopia. They highlighted that prolonged exposure to $PM_{2.5}$ and NO_x may intensify ocular surface soreness and later, retinal inflammation, which in turn increases the risk of development of myopia.

EFs of Atmospheric Metallic Species of $PM_{2.5}$

Table 5 presents the emission factors for atmospheric metallic species of $PM_{2.5}$ from the exhausts of diesel engine vehicles. The emission factors for the investigated atmospheric metallic species from 15 different diesel vehicles vary, as shown in Table 5. The results revealed that Ca was the highest EF among all the investigated atmospheric metallic elements. It was found within the range of 45.3–259 μg L-fuel^{-1} (with an average of 132 μg L-fuel^{-1}). It was further observed that Mg and Al were the second highest emission factors, in the range of 76.8 μg L-fuel^{-1} and 48.6 μg L-fuel^{-1}, respectively. Zn and Cr had the lowest EFs, accounting for a mean average of 13.1 μg L-fuel^{-1} and 1.91 μg L-fuel^{-1}, respectively, as detailed in Table 5. These results were similar to the EF values for each atmospheric metallic element in $PM_{2.5}$ found in other studies (Zhang *et al.*, 2015; Liu *et al.*, 2019) as well as in preceding studies led by Cheng *et al.* (2010) and Zhang *et al.* (2018).

The uncertainties (i.e., standard deviations) as a result of uncontrolled elements such as random errors in sampling and analysis, model year, and vehicle manufacture (brand), are presented in the results for the average of $PM_{2.5}$ compositions (Tables 2 and 3). One of the limitations of our study was that the small tested fleet of 15 diesel vehicles was arbitrarily selected, and only a limited number of diesel fuel vehicles was sampled under this investigation. Another limitation is we did not conduct a comprehensive analysis of chemical composition to determine the concentration of secondary inorganic aerosols (SIAs), such as SO_4^{2-} (sulfate), NO_3^- (nitrate), and NH_4^+, (ammonium) in $PM_{2.5}$. This study has not quantified the speciation fraction of Cr and thus cannot determine the toxicity of Cr emitted from the vehicle exhausts. This is another significant limitation of this study. In order to minimize these uncertainties, this study recommends that adequate representative vehicles should be tested. Furthermore, better knowledge of the vehicle details, such as engine model, the use for each vehicle (such as delivery van, cargo truck, bus, tractor, construction equipment, etc.) emissions control technologies, and a more rigorous experimental design with a large fleet of vehicles are necessary for future research.

CONCLUSION

We measured the metallic elements in vehicle-emitted $PM_{2.5}$ to evaluate their relationship with the ambient $PM_{2.5}$ composition. We found, in descending concentration, Ca, Zn, Al, K, Fe, Mg, and Cr in the emitted $PM_{2.5}$, which was sampled from the exhaust of 15 diesel-powered vehicles. Additionally, the carbonaceous content in the emitted $PM_{2.5}$ was primarily composed of EC, which exhibited a concentration of 824–5570 μg m^{-3}, averaging 2051 μg m^{-3}; the OC and the TC ranged from 426 to 2870 μg m^{-3} (averaging 1410 μg m^{-3}) and from 1250 to 8390 μg m^{-3} (averaging 3461 μg m^{-3}), respectively. Among the metallic elements, Ca displayed the highest EF (45.3–259 μg L-fuel^{-1}, averaging 132 μg L-fuel^{-1}), and Zn and Cr exhibited the lowest ones (averaging 13.1 μg L-fuel^{-1} and 1.91 μg L-fuel^{-1}, respectively). Because metallic elements are present in the ambient $PM_{2.5}$, the health effects arising from exposure to these substances should be given top priority. Although several of these elements are mainly associated with the combustion of heavy fuel oil, others, including Zn, Fe, K, and Cr, can be linked to non-exhaust traffic emissions. As studies have thus far focused on the filterable $PM_{2.5}$, organic carbon, and $PM_{2.5}$-bound metallic elements emitted in the exhaust of currently used diesel vehicles, further investigation is needed to obtain data about additional vehicle types or

Table 5. Metallic element emission factors in $PM_{2.5}$ from the exhaust of diesel engines (μg L-fuel^{-1}).

Element	Diesel engine (n = 15)			
	Range	Mean	SD	RSD (%)
Fe	7.27–28.4	14.3	7.56	52.8
Mg	39.9–129	76.8	26.9	35.0
Zn	3.70–20.4	13.1	5.42	41.4
K	6.93–43.0	24.2	11.6	47.8
Ca	45.3–259	132	62.4	47.5
Al	19.1–94.3	48.6	22.0	45.2
Cr	0.940–3.60	1.91	0.89	46.5
Total	–	311	–	–

*SD = standard deviation; RSD = relative standard deviation.

models powered by other fuels in order to confirm the heavy-metal concentrations in fine particulate matter. Furthermore, health risk assessments should be conducted to determine the potential risks of carcinogenic heavy metals in this fraction.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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