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$_{2.5}$ emitted 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 recouet 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%...
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 et 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., 2018). 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 primary sources > soil dust > secondary sources > vehicle source > cement dust. 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).

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 practicability 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 | 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.

Analytical Procedures

Measurements for Atmospheric Metallic Element Concentration

The PM$_{2.5}$ and further pollutant samples along with blank quartz fiber filters 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°C. 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°C. 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

![Graph](image)

**Table 2.** PM\(_{2.5}\) emission concentration (µg m\(^{-3}\)) and emission factor (µg L-fuel\(^{-1}\)).

| Parameter                  | Diesel engine (n = 15) |
|----------------------------|------------------------|
| Concentration (µg m\(^{-3}\)) | Range: 3875–16,458    | Mean: 7927   | SD: 3563 | RSD (%): 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) |
|-----------|------------------------|
| EC        | Range: 824–5570        | Mean: 2051  | SD: 1340 | RSD (%): 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 0.69% as the smallest amount, as shown in Fig. 3.

Our results are compatible with the findings in the 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

| Element | Range | Diesel engine (n = 15) |
|---------|-------|----------------------|
|         |       | Range | Mean | SD | RSD (%) |
| Fe      | 22.1–73.5 | 40.7 | 23.0 | 76.5 |
| Mg      | 135–441  | 264.9 | 77.6 | 29.3 |
| Zn      | 9.87–87.9 | 42.3 | 22.4 | 59.2 |
| 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.*
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 redoxants, 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$_2$$^\text{2-}$ (sulfate), NO$_x^-$ (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

| Element | Range   | Diesel engine (n = 15) | SD | RSD (%) |
|---------|---------|------------------------|----|---------|
| Fe      | 7.27–28.4 | 14.3                   | 7.36 | 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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