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Characterization of the chemical composition of $PM_{2.5}$ emitted from on-road China III and China IV diesel trucks in Beijing, China

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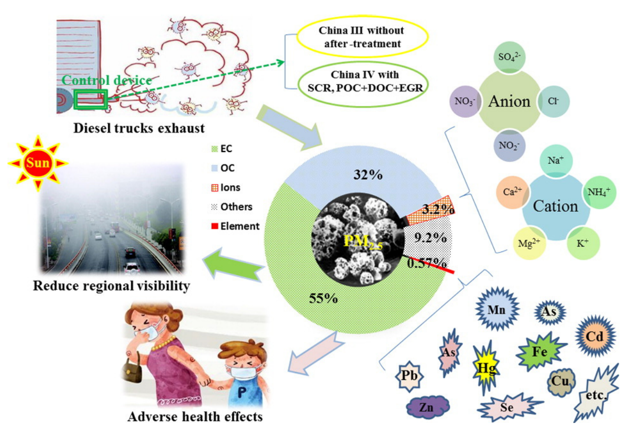
HIGHLIGHTS

- 18 China III and China IV diesel trucks operating in Beijing were monitored using a PEMS.
- EFs for the $PM_{2.5}$ chemical components of the tested diesel trucks were characterized.
- Carbonaceous compounds, WSIs and elements shared approximately 91% of the $PM_{2.5}$.
- The EC/OC of the China IV diesel trucks were higher than those of the China III diesel trucks.
- The EFs for the $PM_{2.5}$ and OC under non-highway were higher than those under highway.

GRAPHICAL ABSTRACT



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ABSTRACT

The composition of diesel exhaust fine particulate matter ($PM_{2.5}$) is of growing interest because of its impacts on health and climatic factors and its application in source apportionment and aerosol modeling. We characterized the detailed chemical composition of the $PM_{2.5}$, including the organic carbon (OC), elemental carbon (EC), water-soluble ions (WSIs), and elemental contents, emitted from China III and China IV diesel trucks (nine each) based on real-world measurements in Beijing using a portable emissions measurement system (PEMS). Carbonaceous compounds were the dominant components (totaling approximately 87%) of the $PM_{2.5}$, similar to the results (greater than 80% of the $PM_{2.5}$) of our previous study of on-road China III diesel trucks. In general, the amounts of individual component groups (carbonaceous compounds, WSIs, and elements) and $PM_{2.5}$ emissions for China IV diesel trucks were lower than those of China III diesel trucks of the same size, except for the WSIs and elements for the light- and medium-duty diesel trucks. The EC/OC mass ratios were strongly dependent on the emission standards, and the ratios of China IV diesel trucks were higher than those of China III diesel trucks of the same size. The chemical species in the $PM_{2.5}$ were significantly affected by the driving conditions. Overall,

Abbreviations: CARB, California Air Resources Board; CMB, chemical mass balance; DOC, diesel oxidation catalysts; DPM, diesel particulate matter; EC, elemental carbon; EFs, emission factors; EGR, exhaust gas recirculation; FTP, federal test procedure; GVWs, gross vehicle weights; HDDTs, heavy-duty diesel trucks; HHDDTs, heavy heavy-duty diesel trucks; HW, highway; IC, ion-chromatography; ICP-MS, inductively coupled plasma-mass spectroscopy; LDDTs, light-duty diesel trucks; MDDTs, medium-duty diesel trucks; MELs, mobile emission laboratory methods; MPS, Micro-proportional Sample System; MSAT, mobile source air toxic; NEDC, New European Driving Cycle; NHW, non-highway; OC, organic carbon; PAHs, polycyclic aromatic hydrocarbons; PEMS, portable emissions measurement system; PGE, platinum group elements; PM, particulate matter; POC, particle oxidation catalysts; R, Pearson's correlation coefficient; RSD, relative standard deviation; SCR, selective catalytic reduction; TC, total carbon; USEPA, United States Environmental Protection Agency; WSIs, water-soluble ions.

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the emission factors (EFs) of the $PM_{2.5}$ and OC under non-highway (NHW) driving conditions were higher than those under highway (HW) driving conditions, and the EC/OC mass ratios presented an increasing trend, with decreasing OC/$PM_{2.5}$ and increasing EC/$PM_{2.5}$ from NHW to HW driving conditions; similar trends were reported in our previous study. In addition, Pearson's correlation coefficients among the $PM_{2.5}$ species were analyzed to determine the relationships among the various chemical components.

1. Introduction

Fine particulate matter ($PM_{2.5}$, particulate matter with an aerodynamic diameter less than 2.5 μm) emitted from diesel vehicles has been a focus of concern due to its adverse effects on human health, including its role in cardiopulmonary disease and cancer (Chio et al., 2014; Ema et al., 2013; Oravisjärvi et al., 2014; Ristovski et al., 2012; Sydbom et al., 2001; US-EPA, 2002), as well as environmental problems, such as urban air pollution and global climate change (Maricq, 2007; Robert et al., 2007). In China, diesel vehicles have significantly contributed to urban air pollution of $PM_{2.5}$ (Huo et al., 2011; Wang et al., 2010). Chemical composition profiles of $PM_{2.5}$ in diesel exhaust particles are important for the source apportionment of urban aerosols (Balachandran et al., 2013; Gross et al., 2005; Kim and Hopke, 2004; Toner et al., 2008) and provide supporting data for aerosol modeling (Cheng et al., 2013; Song et al., 2006; Watson et al., 1997; Yu et al., 2013). Primary $PM_{2.5}$ emitted by diesel vehicles contains a variety of chemical constituents, such as elemental carbon (EC), organic carbon (OC), water-soluble ions (WSIs), elemental compounds, and unidentified compounds that are important in the study of diesel exhaust (Biswas et al., 2009; Cheng et al., 2010; Cheung et al., 2010; Chiang et al., 2012). The EC of diesel particulate matter (DPM) is formed through the pyrolysis of unburned fuels at high temperatures. DPM can reduce visibility due to the absorption of direct light by EC and light scattering (Eidels-Dubovoi, 2002; Eldering and Cass, 1996). The OC of DPM is produced as a result of incomplete fuel combustion and lubricant oil (Kittelson et al., 2006; Li et al., 2014). WSIs are produced by inorganic contaminants in the fuel and lubricating oil and engine wear, which could play a role in diesel exhaust nucleation (Kleeman et al., 2000; Sakurai et al., 2003; Toner et al., 2006; Yu, 2001). Elements are important constituents of DPM, although they only account for a small fraction of the $PM_{2.5}$ mass emissions (Lough et al., 2005).

Previously, tunnel testing, roadside tests and chassis dynamometer measurements were used to assess the particulate matter (PM) emitted from diesel vehicles (Biswas et al., 2008; Chellam et al., 2005; May et al., 2014; Ntziachristos et al., 2007; Kim Oanh et al., 2010; Park et al., 2003; Sharma et al., 2005). A number of studies have focused on the chemical composition of DPM using these methods. For example, Sharma et al. (2005) characterized particulate emissions in terms of the benzene soluble fraction and the metal and elemental and organic carbon contents from a mid-sized diesel engine operated under four different loads in India. Cheung et al. (2010) used a chassis dynamometer and assessed the chemical and oxidative potential of particulate exhaust emissions from a diesel vehicle with a three-stage oxidation system located in Greece. Chiang et al. (2012) measured the PM and its constituents, including the EC, OC, WSI species, elements and polycyclic aromatic hydrocarbons (PAHs) exhaust from six in-use, light-duty diesel vehicles by a chassis dynamometer following the driving pattern of the FTP-75 in Taiwan. Dallmann et al. (2014) measured the chemical composition of PM emitted by gasoline and diesel vehicles at a high temporal resolution through a highway tunnel in the San Francisco Bay area. In recent years, mobile emission laboratory methods (MELs) and a portable emissions measurement system (PEMS) have been developed. Mobile approaches have the potential to provide practical and highly effective methods of measuring emission characteristics directly from the tailpipe in real-world conditions (Cocker et al., 2004; Giechaskiel et al., 2014; Mamakos et al., 2013; Shah et al., 2004; Wang et al., 2012; Weiss et al., 2012). Shah et al. (2004) reported the EC, OC and PM emission rates of 11 heavy heavy-duty diesel trucks (HHDDTs) operating under real-world conditions by following the speed trace from the CARB HHDDT cycle using a MEL in Coachella, CA. In China, PEMS are gradually being implemented to measure the emission factors (EFs) of vehicles on the road (Huo et al., 2012; Liu et al., 2009; Wang et al., 2012; Zheng et al., 2015). However, research on the chemical composition of $PM_{2.5}$ emitted from on-road diesel trucks remains limited in China.

Recently, Zhang et al. (2015) characterized the chemical composition of $PM_{2.5}$ emitted from on-road pre-China I and China I heavy-duty diesel trucks using PEMS. We characterized the OC and EC in $PM_{2.5}$ emitted from on-road China III diesel trucks and investigated the influence of vehicle size and driving conditions according to distance and CO_2 emissions (Wu et al., 2015). However, the EFs for the detailed chemical components of $PM_{2.5}$ emitted from diesel trucks were not studied, and the tested vehicles included only China III diesel trucks. China IV emission standards for diesel trucks were adopted in Beijing in 2013 and were fully implemented in China in 2015. To attain the limits of the standard, exhaust gas recirculation (EGR), diesel oxidation catalysts (DOC), particle oxidation catalysts (POC), and selective catalytic reduction (SCR) were applied to China IV diesel trucks. Following our previous study, this study aims to characterize in greater detail the chemical composition of $PM_{2.5}$ emitted from on-road China III and China IV diesel trucks, including the OC, EC, WSIs, and elemental contents, and to analyze the impact of new emission control technologies on the chemical components of $PM_{2.5}$ emitted from diesel trucks. The EFs reported here can enrich the local database and be used for bottom-up emission inventories at urban and national scales where detailed fleet data are available.

2. Methodology

2.1. Sampling system

A PEMS based on our previous on-board emission system was used to collect data and samples (Huo et al., 2012; Wu et al., 2015; Yao et al., 2007; Yao et al., 2014). This system consists of four main parts: the SEMTECH EFM-2 tube, the SEMTECH DS mobile emission analyzer, the SEMTECH MPS (Micro-proportional Sample System; Sensors Inc., Ann Arbor, MI, USA), and the $PM_{2.5}$ sampling system. The SEMTECH EFM-2 tube was used to measure the flow of diesel exhaust, the SEMTECH DS was used to analyze the instantaneous concentration of gas emissions (CO_2, CO, HC, and NOx), the SEMTECH MPS was used to dilute the diesel exhaust with clean air, and the $PM_{2.5}$ sampling system was used to collect the samples. Additional detailed information on the system is provided in our previous study (Wu et al., 2015).

2.2. Vehicle fleet and test routes

The primary $PM_{2.5}$ emissions were measured from eighteen on-road diesel trucks in Beijing. The tested diesel trucks were divided into three groups based on their gross vehicle weights (GVWs): six light-duty diesel trucks (LDDTs, less than 4500 kg), six medium-duty diesel trucks (MDDTs, 4500–12,000 kg), and six heavy-duty diesel trucks (HDDTs, more than 12,000 kg). Half of the vehicles in each group were China III diesel trucks, and the other half were China IV diesel trucks. After-treatment devices for exhaust were not installed on the China III diesel trucks. For the China IV diesel trucks, POC, DOC, and EGR control devices