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Chemical characterization and toxicity assessment of fine particulate matters emitted from the combustion of petrol and diesel fuels

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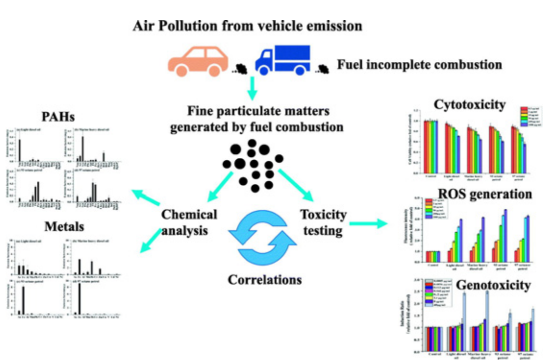
HIGHLIGHTS

- Chemical composition and toxicity of the FPMs emission from fuels combustion were analyzed.
- These FPMs have significant cytotoxicity, ROS activity, and genotoxicity.
- Significant correlations were found between PAHs and ROS-generating capacities.

GRAPHICAL ABSTRACT



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ABSTRACT

Fuel consumption is one of the major contributors to air pollution worldwide. Plenty of studies have demonstrated that the diesel and petrol exhaust fine particulate matters (FPMs) are associated with increases of various diseases. However, the influences of different fuel types and their chemical components on toxicity have been less investigated. In this study, four kinds of fuels that widely used in China were burned in a laboratory simulation, and the FPMs were collected and analyzed. Transmission electron microscopy showed that black carbon was mainly soot with a dendritic morphology. For light diesel oil, marine heavy diesel oil, 93 octane petrol and 97 octane petrol diesel oil, the emission factors of FPMs were 3.05 ± 0.29, 3.21 ± 0.54, 2.36 ± 0.33, and 2.28 ± 0.25 g/kg fuel, respectively. And the emission factors for the "16 US EPA" PAHs of FPM were 0.45 ± 0.20, 0.80 ± 0.22, 1.00 ± 0.20, and 1.05 ± 0.19 mg/g FPMs, respectively. Fe is the most abundant metal in these FPMs, and the emission factors of FPMs were 2.58 ± 1.70, 4.45 ± 0.11, 8.18 ± 0.58, and 9.24 ± 0.17 mg/g FPMs, respectively. We ranked the cytotoxicity of the FPMs emission from fuels combustion: marine heavy diesel oil > 97 octane petrol > 93 octane petrol > light diesel oil, and the genotoxicity of FPMs emission from fuels combustion: marine heavy diesel oil > light diesel oil > 93 octane petrol > 97 octane petrol. Significant correlations were found between PAH concentrations and reactive oxygen species (ROS) generation. Our results demonstrated that fuels exhaust FPMs have strong association with ROS activity, cytotoxicity and genotoxicity. These results indicated that fuels exhaust FPMs pose a potentially serious health, and emphasized the importance of assessing the health risks posed by the particulate pollutants in vehicle exhausts.

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1. Introduction

Traffic is the important source of air pollutions worldwide, and vehicle exhaust is one of the major contributors to the haze in urban environments (Morawska et al., 2008; EPA, 2004). The combustion of both diesel and petrol fuels that occurs in automobile engines produces combustion-derived fine particulate matters (FPMs) (Araujo and Nel, 2009; Watson and Chow, 2001). A great number of epidemiological, clinical, and toxicological studies have demonstrated that the FPMs from vehicle exhaust are associated with increases in asthma and cardiovascular, respiratory, and other diseases (Laskin et al., 2012; Loomis et al., 2013; Pope and Dockery, 2006).

Generally, diesel and petrol exhaust fine particles (DEFPs and PEFPs, respectively) are smaller than 2.5 μm (2.5 μm particulate matter, [$PM_{2.5}$]), and most of them are ultrafine particulate matters with dimensions of <0.1 μm (Donaldson et al., 2005). Owing to the small size, these DEFPs and PEFPs can be deposited in the alveolar and bronchiolar regions of the lung (Dockery, 2009). DEFPs and PEFPs also carry myriad toxic organic compounds and metals, only a very small fraction of which have been identified (Groenzin and Mullins, 2000). The small size and associated toxic components of DEFPs and PEFPs considerably increase their threat to public health (Durga et al., 2014).

Toxicity assessment complements the chemical analysis of FPMs because different effects are induced when various types of compounds are mixed, including the contributions of unidentified compounds, and these interactive effects must be taken into consideration because antagonistic and synergistic phenomena occur in complex mixtures (Arey, 2004; DeMarini et al., 2004; Singh et al., 2004). Toxic responses are also risk-scaled, for example, more potent chemicals will induce a more toxic response than less potent chemicals. Although the link between vehicle exhaust particles and their adverse effects on health is well established, the influence of different fuel types and their chemical components on their toxicity have been less investigated.

In this study, we explored the chemical components and toxicity of the emissions produced by the combustion of four typical petrol and diesel fuels widely-used in China. We systematically and experimentally investigated important heavy metals, 16 kinds of polycyclic aromatic hydrocarbons (PAHs) that are listed as priority controlled pollutants by the U.S. Environmental Protection Agency (U.S. EPA), and the cytotoxicity, oxidative stress, and genotoxicity of the FPMs emitted from the combustion of petrol and diesel fuels. To identify the chemical components that contribute to the toxicity of the emissions, we evaluated the correlations between the critical chemicals and the emission toxicity. Our results provide indications for the future use of petrol and diesel fuels, and emphasize the importance of assessing the health risks posed by the particulate pollutants in vehicle exhausts.

2. Material and methods

2.1. Combustion experiments and fine particulate matters sampling

A schematic overview of the combustion and sampling system was shown in Fig. S1·The particles were sampled with a wick burner under a semi-enclosed cylindrical glass cover with a sufficient particle-free air supply and a stainless-steel environmental chamber equipped with magnetic fans fixed at the bottom of the chamber ensured mixing of the chamber contents, as well as a set of sampling instruments (Fig. S1). This combustion chamber is similar with that in previous studies (Dobbins et al., 2006; Wilson et al., 2013). Each combustion experiment lasted for 10 min and the combustion experiments for each fuel type were conducted at least in triplicate. Three different sampling systems were used to collect the FPMs emitted from the combustion of the fuel oils. For the transmission electron microscopy (TEM) analysis, the particles were collected onto 300-mesh copper TEM grids coated with carbon film, using a single-stage cascade impactor and a jet nozzle with a 0.5 mm diameter at a flow rate of 1.0 L/min (Fu et al., 2012). The sampling periods ranged from 60 s and 300 s, depending on the particle loading. For the chemical analyses, the samples were collected with a Teflon filter (Whatman, UK) for the metal analysis and with a quartz fiber filter (Whatman, UK) for the analysis of organic carbon and PAHs (Alves et al., 2015; Cheng et al., 2010; Riddle et al., 2007). For the toxicity assessment, the particle samples were collected with a $PM_{2.5}$ impactor, then ground, sieved, and sterilized according to previous studies (Landis et al., 2007; Liacos et al., 2012).

2.2. Scanning transmission electron microscopy (STEM) and energy-dispersive X-ray spectrometer (EDS) analyses

Detailed information about the process used for the STEM analysis has been reported previously by our group (Fu et al., 2012). Briefly, the collected samples were examined with a JEOL-2100F field emission high-resolution transmission electron microscope (FE-HRTEM) equipped with an Oxford EDX and a STEM unit with a high-angle annular dark-field detector (HAADF). To ensure that the particles analyzed represented the whole size range, at least three areas were analyzed from the center and periphery of the sampling spot on each grid. The particle size was calculated using an ellipse that best fitted a particle outline, and the particle diameter was calculated as the arithmetic mean of the short and long axes of the ellipse, according to a previous study in our laboratory (Li and Shao, 2010). The elemental compositions were determined semi-quantitatively with EDS, which can detect elements heavier than carbon, according to previous studies in our laboratory (Fu et al., 2012).

2.3. PAHs analysis

After sampling, 16 PAHs, namely, naphthalene (Nap), acenaphthylene (Ace), acenaphthene (Acp), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benzo[*a*]anthracene (BaA), chrysene (Chry), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[1,2,3-c,d]pyrene (InP), dibenzo[*a*,*h*]anthracene (DahA), and benzo[*g*,*h*,*i*]perylene (BghiP) were extracted and identified with gas chromatography/mass spectrometry. The extraction and analytical procedures used for the PAHs were those used in a previous study in our laboratory (Zhang et al., 2011). One quarter of the filter was ultrasonicated twice with 25 mL (15 + 10 mL) of dichloromethane in a Branson™ Ultrasonic Cleaner for 30 min. The solution of each sample was evaporated to 2–3 mL on a rotary evaporator (Buchi, Switzerland) and was blow down to 1 mL under a gentle stream of nitrogen. To ensure high-quality data, quality assurance procedures were conducted. First, the recoveries of the PAHs were determined to test the availability of the analytical method. The average recovery efficiencies ranged from 85% to 99%, and the PAH concentrations were calculated from the recovery efficiencies. In general, laboratory blanks detection showed that no PAHs have been detected and the method detection limit (MDL) for PAH analysis by GC–MS was about 0.005 mg/L. The emission factors for the 16 PAHs were calculated and expressed based on the masses of the PAHs per unit mass (g) of the FPMs and per kg of fuel emitted during the combustion of petrol and diesel fuels.

2.4. Metals analysis

The elemental concentrations were determined with inductively coupled plasma optical emission spectrometry (ICP-OES; Atom Scan 2000, JarroU-Ash, USA). After the sampling filters were weighed, they were cut into pieces, placed into polytetrafluoroethylene digestion vessels, and digested with highly concentrated acids (HNO_3–HF–HCl) (Bi et al., 2014; Huang et al., 2016). When the particles and filters were completely digested, the digestion solutions were diluted to 20 mL with high-purity water (18 MΩ cm^{-1}) generated with a Milli-Q system (Millipore, Bedford, MA, USA). The solutions were analyzed with ICP-OES to determine the concentrations of various elements (As, Fe, Al,