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Characteristics and oxidative potential of atmospheric $PM_{2.5}$ in Beijing: Source apportionment and seasonal variation

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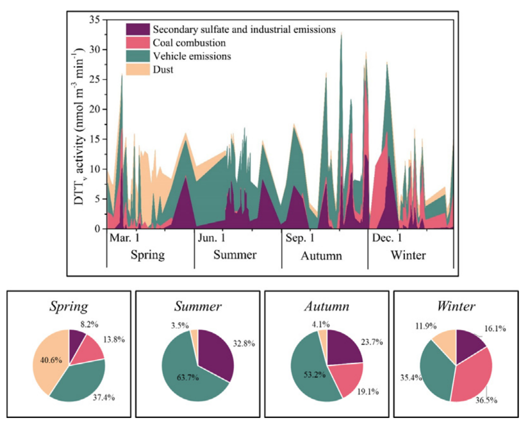
HIGHLIGHTS

- Seasonal changes in $PM_{2.5}$, compositions and DTT-based ROS activity occurred.
- Averaged DTT_m and DTT_v reached the peak values during the local summer.
- PMF model using the annual data categorized the main sources of $PM_{2.5}$ in Beijing.
- A sensitivity sequence of DTT_v activity to the known sources was established.

GRAPHICAL ABSTRACT



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ABSTRACT

$PM_{2.5}$ (particulate matter with the aerodynamic diameter $D_p < 2.5$ μm) was hypothesized to generate reactive oxygen species (ROS) and induce oxidative stress associated with inflammation and cardiovascular diseases. In the current study, $PM_{2.5}$ concentrations, water-soluble ions and elements, carbonaceous components and ROS activity characterized by the dithiothreitol (DTT) assay were determined for the $PM_{2.5}$ samples collected in Beijing, China, over a whole year. Source apportionments of $PM_{2.5}$ and DTT activity were also performed. The mean ± standard deviation of $PM_{2.5}$, DTT_m (mass-normalized DTT activity) and DTT_v (volume-normalized DTT activity) were 113.8 ± 62.7 μg·m^{-3}, 0.13 ± 0.10 nmol·μg^{-1}·min^{-1} and 12.26 ± 6.82 nmol·m^{-3}·min^{-1}, respectively. The seasonal averages of DTT_m and DTT_v exhibited peak values during the local summer. Organic carbon (OC), NO_3^-, SO_4^{2-}, NH_4^+ and elemental carbon (EC) were the dominant components in the constituents tested. Higher concentrations of carbonaceous components occurred in autumn and winter compared with spring and summer. Based on the positive matrix factorization model (PMF), the simulation results of source apportionment for $PM_{2.5}$ in Beijing, obtained using the annual data, identified the main categories as follows: dust, coal combustion, secondary sulfate and industrial emissions, vehicle emissions and secondary nitrates. Most detected constituents exhibited significantly positive correlations with DTT_v ($p < 0.01$). The results corresponding to multiple linear regression (MLR) between DTT_v activity and source contribution to $PM_{2.5}$ manifested the sensitivity sequence of DTT_v activity for the resolved sources as vehicle emissions > secondary sulfate and industrial emissions > coal combustion > dust.

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Capsule: Based on a descending sequence of relative contribution, the diagnostic sources of DTT_v activity in $PM_{2.5}$ from Beijing included primarily vehicle emissions, secondary sulfates and industrial emissions, coal combustion, and dust.

1. Introduction

Numerous studies have indicated exposure to $PM_{2.5}$ (particulate matter with the aerodynamic diameter $D_p < 2.5$ μm) could exert adverse impacts on human health, such as the development of respiratory diseases and increased morbidity and mortality of cardiovascular illnesses (Atkinson et al., 2014; Hamra et al., 2014; Pui et al., 2014; Kim et al., 2015; Lu et al., 2015; Du et al., 2016). In addition, other investigations have suggested that particulate matter (PM) could induce oxidative stress and then inflammation (Araujo and Nel, 2009; Cachon et al., 2014; Li et al., 2015). For example, oxidative stress was usually associated with inflammations of airways and systems, cardiopulmonary diseases and lung cancers (Bhatt and Dransfield, 2013; Valavanidis et al., 2013). The corresponding hypothesized mechanisms of PM-induced oxidative stress involved the direct production of reactive oxygen species (ROS) by PM and the potential for stimulating cells to produce ROS (Ayres et al., 2008). A recent report showed that fine PM could penetrate across the barrier-damaged skin and produce ROS-triggered inflammation (Jin et al., 2018).

Considering the chemical nonspecificity of PM and a complex interplay of multiple factors, e.g., particle size, composition, source and season (Sarnat et al., 2008; Dergham et al., 2015), only the mass concentration could not account for the negative effects on human health. Compared with $PM_{2.5}$, dithiothreitol (DTT) activity exhibited a much stronger correlation with the emergency treatment of asthma and congestive heart failure (Bates et al., 2015), which provided further epidemiological evidence to elucidate the mechanisms of oxidative stress. For instance, using a DTT assay to quantify the oxidative potential (OP) of particle sources from California during two seasons, Charrier et al. (2015) found submicron fine particles (0.17 μm ≤ D_p ≤ 1.0 μm) usually had greater mass-standardized OP with respect to ultrafine PM (D_p ≤ 0.17 μm), and the source contribution of OP was dominated by vehicle emissions. Another study in California (Shirmohammadi et al., 2015) indicated both vehicular abrasion and resuspension of road dust correlated with the activity of water-soluble ROS, while vehicular abrasion provided a significant contribution to water-insoluble ROS activity. In addition, Verma et al. (2014) evaluated the potential of water-soluble fractions in atmospheric fine aerosols from the southeastern United States to produce ROS and identified the main ROS-related sources.

Many studies focused on the source apportionment of $PM_{2.5}$ in China, due to frequent and severe particulate air pollution in recent years (Wang et al., 2015; Yang et al., 2016; Huang et al., 2017). However, to date, studies concerning the oxidation activity of atmospheric particles are still inadequate. Liu et al. (2014) investigated the oxidative potential and inflammatory effects of ambient air pollution sources in Beijing, but the results were limited to the local spring. Beijing, as the capital of China, is characterized by a high-density population and heavy traffic jams, and the local atmospheric conditions are strongly affected by the typical monsoon climate and intensive human activities, such as large-scale centralized heating in winter, all of which prompted the deterioration of air quality (Batterman et al., 2016; Wang et al., 2018). In this case, the situation in a single season could not reflect the noticeable seasonal variations in a whole year (Yang et al., 2015). Therefore, additional research is required.

The primary objectives of the current study are to determine the main emission sources of $PM_{2.5}$ and DTT activity accounting for air OP in Beijing and to localize the different source regions responsible for the relative contribution probability (probabilistic influences) for $PM_{2.5}$ in Beijing. In brief, a continuous sampling of atmospheric $PM_{2.5}$ was performed for a full year in Beijing. The particle mass concentration, concentrations of water-soluble ions, elements and carbonaceous components, and ROS activity (by DTT assay) of the collected $PM_{2.5}$ samples in different seasons were determined. Then, the positive matrix factorization (PMF) model combined with multiple linear regression (MLR) were applied for source apportionment of $PM_{2.5}$ and corresponding DTT activity. The acquired results may be beneficial for the accurate source diagnosis and seasonal variations of $PM_{2.5}$ and related ROS activity (OP) in Beijing. Accordingly, these results may be conducive to promulgating and implementing the cost-effective abatement measures on PM emissions for improving air quality.

2. Materials and methods

2.1. Sampling site and sample collection

From May 2015 to April 2016, the $PM_{2.5}$ samples for an entire year were collected at a campus site at Peking University (116°19′17″E, 39°59′53″N) in Beijing. Based on the national standard methods for $PM_{2.5}$ (HJ93-2013), the flow rate of the air impact sampler (Type 2034, Laoying, Qingdao, China) was assigned at 100 $L\cdot min^{-1}$, equipped with a quartz microfiber filter (diameter = 90 mm, QAT-UP, Pall, USA), and all of the filters were baked at 650 °C for 4 h before use. Fig. S1 in the Supplementary Materials illustrates the number of samples for each month. After the exclusion of lost or damaged samples, a total of 147 ambient $PM_{2.5}$ samples were collected, including intensive (high-frequency) sampling in January (a typical heating period) and July (a representative nonheating period) and regular sampling in other months. To further investigate the diurnal variation, each daily sampling in January was scheduled with two intervals of daytime and nighttime, and each daily sampling in July was divided into three intervals of morning, noon and night. Each daily sample collection in January and July covered a 48-h consecutive sampling, during which the air $PM_{2.5}$ samples corresponding to the same time interval were collected using the same quartz microfiber filter for adequate mass loadings. In addition, each regular sampling by the active air sampler in the rest months lasted for 24 h. Based on the climatic characteristics and local heating periods in Beijing, the season division was set as spring (March, April and May), summer (June, July and August), autumn (September, October and November) and winter (December, January and February). All the filter samples were carefully sealed in aluminum foil and saved in a −20 °C refrigerator.

2.2. Quantitative determinations

2.2.1. Mass concentration of $PM_{2.5}$

Before weighing, all of the filters were adapted to the conditions in a weighing chamber maintained at 22 ± 2 °C and 35 ± 2% of humidity. The gravimetric analysis was performed using a digital balance with an accuracy of 1×10^{-5} g (XS105, Mettler Toledo, Switzerland). Each filter was weighed at least three times, and the acceptable deviations among the replicates were $<5 \times 10^{-5}$ g. The mass difference before and after sampling divided by the sampled air volume was employed to calculate the mass concentration of $PM_{2.5}$.