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Temporal variation of oxidative potential of water soluble components of ambient $PM_{2.5}$ measured by dithiothreitol (DTT) assay

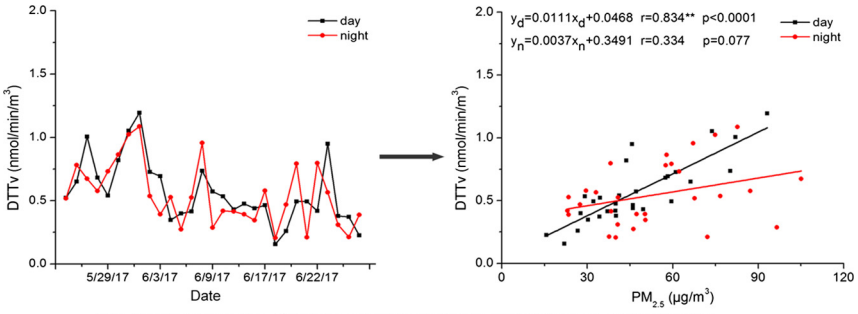
Jingpeng Wang [a], Xin Lin [b], Liping Lu [a,c], Yujie Wu [a,c], Huanxin Zhang [d], Qi Lv [a], Weiping Liu [a,c], Yanlin Zhang [b], Shulin Zhuang [a,e,*]

[a] College of Environmental and Resource Sciences, Zhejiang University, Hangzhou 310058, China
[b] Yale-NUIST Center on Atmospheric Environment, Nanjing University of Information Science and Technology, Nanjing 210044, China
[c] Research Center for Air Pollution and Health, Zhejiang University, Hangzhou 310058, China
[d] College of Marine Life Sciences, Ocean University of China, Qingdao 266003, China
[e] The First Affiliated Hospital College of Medicine, Zhejiang University, Hangzhou 310003, China

HIGHLIGHTS

- Ambient $PM_{2.5}$ showed significant seasonal variations in Hangzhou city in 2017.
- The annual mean of volume-normalized DTT activity (DTTv) was 0.62 ± 0.24 nmol/min/m^3.
- The mass-normalized DTT activity (DTTm) showed seasonal and diurnal variations.
- The oxidative potential (OP) of $PM_{2.5}$ was dominated mainly by the components rather than mass concentration.
- Secondary inorganic aerosol (SIA) species were correlated prominently with DTTv ($p < 0.05$).

GRAPHICAL ABSTRACT



The diurnal tendency of DTTv and its correlation with PM mass concentration.

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ABSTRACT

The exposure to ambient fine particulate matter ($PM_{2.5}$) can induce oxidative stress, contributing to global burden of diseases. The evaluation of the oxidative potential (OP) of $PM_{2.5}$ is thus critical for the health risk assessment. We collected ambient $PM_{2.5}$ samples in Hangzhou city, China for four consecutive quarters in the year 2017 and investigated the oxidation property of $PM_{2.5}$ components by the dithiothreitol (DTT) assay. The annual mean of ambient $PM_{2.5}$ mass concentrations in 2017 was 63.05 μg m^{-3} (median: 57.34, range: 6.67–214.33 μg m^{-3}) with the significant seasonal variations ranking as winter > spring > summer > autumn. Secondary inorganic aerosol (SIA) species including SO_4^{2-}, NO_3^- and NH_4^+ totally account for >50% of $PM_{2.5}$ mass. The annual mean volume-normalized DTT activity (DTTv) showed a relatively high value of 0.62 nmol/min/m^3 (median: 0.62, range: 0.11–1.66 nmol/min/m^3) and DTTv of four seasons was roughly at the same level, indicating a high annual exposure level of ambient $PM_{2.5}$. SIA species were correlated well with the corresponding DTTv and showed significant diurnal variations with strong or moderate correlations at day and weak correlations at night, suggesting strong secondary formation in daytime with contribution to the particulate OP. The annual mean mass-normalized DTT activity (DTTm) had a relatively low value of 6.39 pmol/min/μg (median: 5.63, range: 1.99–22.70 pmol/min/μg), indicating low intrinsic oxidative toxicity. The DTTm of four seasons ranked as autumn > winter > spring > summer, indicating seasonal variations of the DTT-active components. The $PM_{2.5}$

* Corresponding author.
E-mail address: shulin@zju.edu.cn (S. Zhuang).

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mass concentration is more related to exposure levels than intrinsic properties of components, while OP is determined by the components rather than $PM_{2.5}$ mass concentration. Our results provide an insight into reactive oxygen species-induced health risk of $PM_{2.5}$ exposure and decision for subsequent emission control.

1. Introduction

The fine particulate matter ($PM_{2.5}$) contributes significantly to the ambient air pollution occurred frequently in many regions and countries, arising public health concern. Exposure to $PM_{2.5}$ contributes to the global burden of diseases and $PM_{2.5}$ was ranked among the top five global mortality risk factors (Cohen et al., 2017). Numerous epidemiological studies have revealed the association between $PM_{2.5}$ and increasing morbidity and mortality from various diseases, including cardiovascular diseases, respiratory system disorders, and lung cancer (Cao et al., 2012; Sun et al., 2010). The exposure to fine PM with a 10-μg/m^3 elevation was reported to cause 8% to 18% increase in mortality risk for ischemic heart disease, dysrhythmias, heart failure, and cardiac arrest (Pope et al., 2004). The long-term residential exposure to air pollution with a ten-unit increase in $PM_{2.5}$ (μg/m^3) led to the increase of 1.29 (95% confidence interval = 0.95–1.76) for lung cancer incidence in eight Canadian provinces from 1994 to 1997 (Hystad et al., 2013). Although the underlying pathological mechanisms for these diseases are yet incompletely understood, $PM_{2.5}$-induced oxidative stress is considered one of the major biochemical pathways (Rui et al., 2016; Valavanidis et al., 2013). The exposure to $PM_{2.5}$ may stimulate cells to produce excessive reactive oxygen species (ROS) and disturb redox homeostasis, thereby triggering a cascade of downstream episodes such as systemic inflammation, DNA damage and cell death (Becker et al., 2005; Soberanes et al., 2009; Vattanasit et al., 2014; Xu et al., 2018).

The oxidative potential (OP) of $PM_{2.5}$ is a versatile indicator suitable for the evaluation of the oxidation property of PM components (Crobeddu et al., 2017; Perrone et al., 2016). Determination of ambient fine particulate OP can be used as a promising and integrative metric for preliminary screening. ROS were produced through transferring electron from cellular reductants, such as NADPH, to molecular oxygen (Kumagai et al., 1997; Liu et al., 2017). Dithiothreitol (DTT) was used reportedly as a substitute of cellular reductants and chemical components of $PM_{2.5}$ can catalyze electronic transport from DTT to oxygen, generating superoxide radical. The ambient particulate OP can be evaluated by the determination of DTT consumption rates (Cho et al., 2005; Kumagai et al., 2002). The measured volume-normalized DTT activities (DTTv) and mass-normalized DTT activities (DTTm) can facilitate to characterize the exposure-relevant levels and intrinsic properties of $PM_{2.5}$. This cell-free DTT assay is used frequently to measure the ambient particulate OP with the advantages of fast results reading and strong environmental controllability (Rattanavaraha et al., 2011; Vreeland et al., 2017).

The DTT assay can effectively assess the temporospatial variation trend of ambient particulate OP at long-term, low-dose exposure levels. Such seasonal and diurnal variation is crucial for the prediction of ROS-induced regional health risks. The significant seasonal variations of ambient particulate OP were observed in Atlanta and Los Angeles Basin that higher DTT activities occurred in colder months than in warmer periods (Fang et al., 2015b; Saffari et al., 2014). The significant diurnal variation was also observed in coastal cities of the Bohai Sea with higher DTT activities in summer nighttime than in daytime (Liu et al., 2018). The temporospatial variation of OP is contributed by different components. Certain metals and organic components were reported to be associated with ROS or OP (Fang et al., 2015a; Verma et al., 2015). Copper and manganese were the most DTT-active compounds among the soluble transition metals of $PM_{2.5}$, and quinones were moderately contributed to DTT loss (Charrier and Anastasio, 2012). Secondary organic aerosol was also reported to be a major contributor associated with DTT activity (Ma et al., 2018; Tuet et al., 2017). Secondary inorganic aerosol (SIA, including SO_4^{2-}, NO_3^- and NH_4^+) contributes a large amount of $PM_{2.5}$, however their relations to the OP of $PM_{2.5}$ need to be clarified (Jansen et al., 2014; Xu et al., 2017). SIA-abundance ambient PM may produce acidic environment, possibly inducing oxidative stress and inflammation through synergistic action with other components (e.g. transition metals) (Fang et al., 2017a). Therefore, it is necessary to emphasize the relation of SIA to ambient particulate OP, which is essential to better assess the adverse health effects of different $PM_{2.5}$ components.

In the present study, we collected ambient $PM_{2.5}$ samples in Hangzhou city, China for four consecutive quarters in 2017 and monitored online their components. The seasonal and diurnal variations of the exposure-relevant level and intrinsic properties of $PM_{2.5}$ were investigated by the measurements of DTTv and DTTm via the DTT assay. The relation of SIA to the observed OP of ambient $PM_{2.5}$ was further identified. Our results indicated ROS-induced external exposure risk at long term and deciphered the components contributing to the adverse effect. Such information is critical to the elucidation of downstream molecular mechanism and beneficial to subsequent emission control decisions.

2. Materials and methods

2.1. Site description and $PM_{2.5}$ sampling

The $PM_{2.5}$ samples were collected at the environmental monitoring station of Zhejiang University in Hangzhou city, China (30.31°N, 120.08°E). There is a two-way six-lane highway about 700 m away from the north side of the station and a construction site 500 m away from the west site. The combined influence of residential, traffic, and construction emissions makes it as a well representative of urban emission levels. The ambient air $PM_{2.5}$ sampler was put on the roof of the station, about 5 m above ground level without visible tall buildings and pollution source around. Ambient $PM_{2.5}$ samples were continuously collected in winter (December 20, 2016–January 20, 2017), spring (March 20, 2017 to April 20, 2017), summer (May 25, 2017–June 29, 2017), and autumn (October 9, 2017–November 11, 2017). The sampling collection last for 23 h each day during winter and spring, while in summer and autumn, sampling times for day and night samples were 09:00 to 20:30 h and 21:00 to 08:30 h, respectively. The meteorological data during sampling periods were listed in Table 1.

A high-volume sampler (model KC-1000, Laoshan Mountain Electronic Instrument Factory Co., Ltd.) equipped with a $PM_{2.5}$ selective-inlet head was employed to collect particles with the nominal flow rate of 1.05 m^3/min using prebaked 8 × 10 inch quartz fiber filters (Peta Instrument Co., Ltd., Guangzhou, China). The filters were equilibrated (72 h at 39% RH, room T) and weighed with a microbalance (100 μg precision) before and after the sampling and all sampled filters were immediately wrapped in aluminum foil and stored in the refrigerator at −20 °C until analysis. The blank filters were prepared by operating the sampler for 10s at the beginning