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Trace elements in $PM_{2.5}$ in Shandong Province: Source identification and health risk assessment

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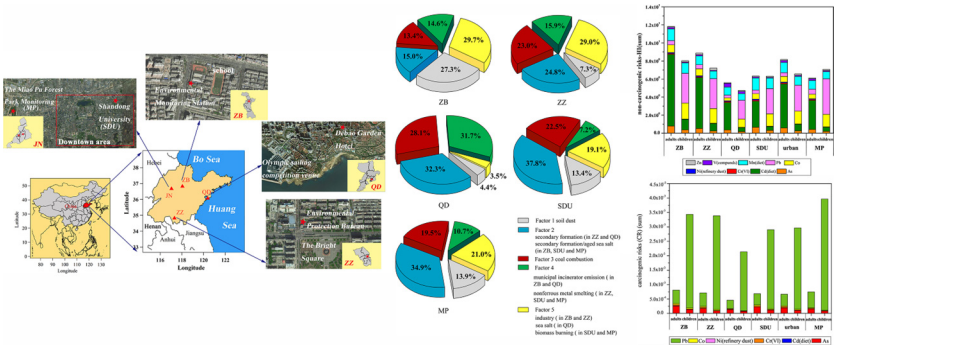
HIGHLIGHTS

- The temporal and spatial distributions of trace elements in $PM_{2.5}$ in Shandong
- Secondary formation, coal combustion and industry emissions were the main sources.
- Cd (diet) for adults, Pb and Co for children had non-carcinogenic risks.
- As and Pb existed carcinogenic risks, especially Pb for children.

GRAPHICAL ABSTRACT



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ABSTRACT

The chemical compositions in $PM_{2.5}$ in metropolitan areas have obtained lots of attentions, of which concerns of airborne trace elements are relatively lacking. Here, $PM_{2.5}$ samples were collected simultaneously in one year at four urban sites (Zibo (ZB), Zaozhuang (ZZ), Qingdao (QD) and Jinan (JN (Shandong University)), and a rural site (JN (Miaopu)) in Shandong province. 25 elements (Al, Na, Cl, Mg, Si, S, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Br, Sr, Cd, Ba and Pb) in $PM_{2.5}$ were measured by wavelength dispersive X-ray fluorescence spectrometer (WDXRF). Most trace elements (Al, Na, Cl, Mg, Si, Ca, Ti, Mn, Fe, Co, Ni, As, Se, Br, Cd, Ba and Pb) exhibited the highest levels at ZB and the lowest at QD. Meanwhile, they presented obvious seasonal variations with the highest concentrations in winter or spring and the lowest in summer. S and K were the most abundant elements in the area. In the non-crustal trace metal elements, Zn, Pb and Mn presented the highest concentrations. Positive matrix factorization (PMF) modeling revealed that secondary formation, coal combustion and industry emissions were the main sources in the region. The health risk assessments suggested that at the five sites Cd (diet) for adults, Pb and Co for children, and Mn (diet) for both adults and children (at ZB and SDU sites) had non-carcinogenic risks. As and Pb for adults and children existed carcinogenic risks, especially Pb for children. The sources of these elements with health risks were further explored. Notably, Cd, As and Pb should be paid special attention in the area due to their high concentrations in aerosol water exceeding the acceptable health risks, especially Pb.

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1. Introduction

Currently, China is suffering from severe air pollution, e.g. haze. The particulate matters not only influence the regional natural conditions and make climate change (Wang et al., 2013), but also have critical effects on human health under the high levels due to their deposition in the respiratory tract and reach the pulmonary alveoli (Makkonen et al., 2010; Moreno et al., 2011). The trace elements bonded with particulates are important components in the atmosphere. They are omnipresent in our surrounding and have a significant contribution to potential health on the lung under long-term environmental exposure owing to their high bioreactivity and toxicity (Betha et al., 2014; Han et al., 2015; Moreno et al., 2011; Pan et al., 2015). According to the International Agency for Research on Cancer (IARC) of WHO (World Health Organization), the carcinogenic compounds are classified into 5 groups: Group 1 (carcinogenic to humans), Group 2A (probably carcinogenic to humans), Group 2B (possibly carcinogenic to humans), Group 3 (not classifiable as to its carcinogenicity to humans), and Group 4 (probably not carcinogenic to humans). In that study, As, Cd, Ni and Cr (VI) belong to Group 1, and Pb, Co and Mn are included in Group 2A (Du et al., 2012; Han et al., 2015; Tian et al., 2015). These toxic elements associated with particulate matter have potential threat to human health through air pollutants inhalation, dermal contact and ingestion in the human bodies (Zhang et al., 2015). Especially for children heavy metal adsorptions from the digestion system and hemoglobin sensitivity to these metals are both much higher than those for adults due to children's body at early ages and lower body weight (Kurt-Karakus, 2012). Therefore, trace elements in particulates are a serious concern for human health.

The emissions of trace metals in the atmosphere in Asia are the highest all over the world along with the increasing demand for energy and developing industry (Nie et al., 2013; Pacyna and Pacyna, 2001). In China, the emissions of the trace elements including Cd, Pb, Cr and As rank within the top three in the worldwide (Pacyna and Pacyna, 2001). Shandong province (34°22.9′ N-38°24.01′ N, 114°47.5′ E-122°42.3′ E) lies in North China Plain, one of the major hazy areas in China (Cheng et al., 2011), adjacent to Korea and Japan across Bo Sea on the eastern coast of China (Fig. 1). As the conjunction joint of the Yellow River economic zone and the Bo Sea economic zone, here there is a high population density (620 person/km^2 in 2015) under the high growth rate of gross domestic product (GDP) (9.2%–10.0% in 2006–2015) mainly driven by the heavy industry (China Statistical Yearbook, 2007-2016). The population and economic rapid developments lead to higher energy demands, among which coal consumption accounted for over 70.0% of the total energy consumption in the province (Shandong Statistical Yearbook, 2008) as a result of 10.8% coal-fired power plants in China on only 1.6% national territorial area of Shandong (Shi et al., 2007). This resulted in the highest SO_2 (10%), NO_x (8.4%), PM_{10} (9.3%), $PM_{2.5}$ (9.1%) emissions on a provincial basis (Zhang et al., 2009). Particulate matter is generally primary pollutant in the province, accounting for 43% of the pollution burden (Environmental Status Bulletin of 2006, 2008). The mean concentration of PM_{10} and $PM_{2.5}$ in Shandong reached 180 μg/m^3 and 90 μg/m^3, respectively (Yao and Lu, 2014). Serious pollution not only influenced local economic growth, but also had adverse impacts on human health.

Significant progresses have been made in the area on the understanding of particulate characteristics, including the chemical compositions, i.e., water soluble ions, organic carbon and elemental carbons, and the source apportionments of particles by principal component analysis (PCA), chemical mass balance (CMB) and positive matrix factorization (PMF) models (Lai et al., 2016; Song et al., 2006; Yao et al., 2016; Zhang et al., 2015). Even the microscopic properties, the morphology and formation of the individual particles have been determined by transmission electron microscopy (TEM) (Li et al., 2016; Li et al., 2013). However, few researches investigate trace elements associated with $PM_{2.5}$ in the region. Only in individual city, e.g. Taian (Liu et al., 2016), Liaocheng (Zhang et al., 2017), Longkou (Zong et al., 2016) and Qingdao (Li et al., 2012), the levels and/or sources apportionment of elements in $PM_{2.5}$ were reported. While Ding et al. (2015) indicated that the serum levels of trace elements might be associated with breast cancer risk. Therefore, the research on trace elements in $PM_{2.5}$, including both sources and health risks, are still insufficient in the whole Shandong. Knowing where the trace elements come from, can we make

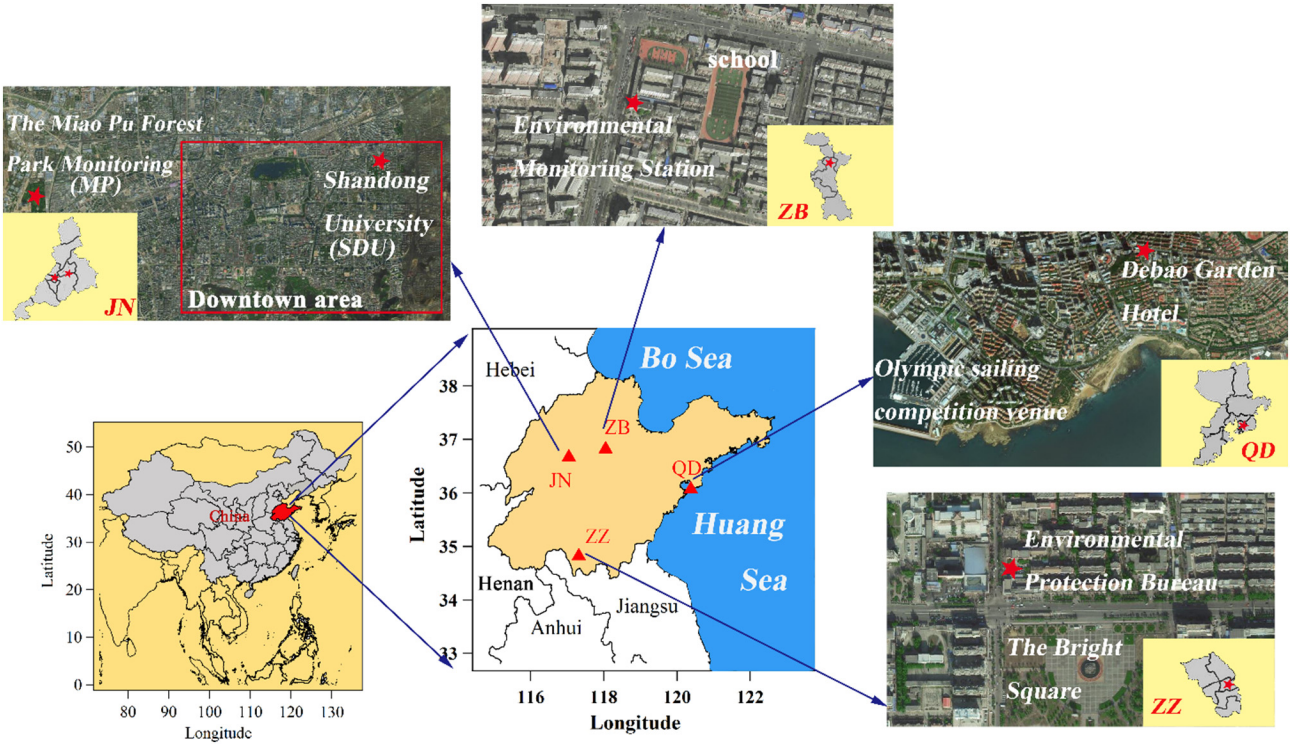

Fig. 1. Locations of sampling sites in Shandong Province.