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Research article

Distribution and sources of particulate mercury and other trace elements in $PM_{2.5}$ and PM_{10} atop Mount Tai, China

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ABSTRACT

The concentrations of particulate mercury (PHg) and other trace elements in $PM_{2.5}$ and PM_{10} in the atmosphere were measured at the summit of Mount Tai during the time period of 15 June – 11 August 2015. The average PHg concentrations were 83.33 ± 119.1 pg/m^3 for $PM_{2.5}$ and 174.92 ± 210.5 pg/m^3 for PM_{10}. Average concentrations for other trace elements, including Al, Ca, Fe, K, Mg, Na, Pb, As, Se, Cu, Cd, Cr, V, Mo, Co, Ag, Ba, Mn, Zn and Ni ranged from 0.06 ng/m^3 (Ag) to 354.33 ng/m^3 (Ca) in $PM_{2.5}$ and 0.11 ng/m^3 (Co) to 592.66 ng/m^3 (Ca) in PM_{10}. The average concentrations of PHg were higher than those at other domestic mountain sites and cities in other counties, lower than those at domestic city sites. Other trace elements showed concentrations lower than those at the domestic mountain sites. Due possibly to increased control of emissions and the proportion of new energy, the PHg and trace element concentrations decreased, but the PHg showed concentrations higher than those at the Mountain sites, this showed that the reasons was not only severely affected by anthropogenic emissions, but also associated with other sources. The concentration changed trend of the main trace elements indicated that PHg, trace elements and particle matters present positive correlation and fine particulate matter has a greater surface area which was conductive to adsorption of Hg and trace elements to particles. On June 19, June 27 and July 6, according to the peak of mercury and trace elements, we can predict the potential sources of these three days. The results of principal component analysis (PCA) suggested that, crustal dust, coal combustion, and vehicle emissions were the main emission sources of PHg and other trace elements in Mount Tai. The 24-h backward trajectories and potential source contribution function (PSCF) analysis revealed that air masses arriving at Mount Tai were mainly affected by Shandong province. Mount Tai was subjected to five main airflow trajectories. Clusters 1, 2, 3, and 5 represented four pathways for local and regional sources and cluster 4 originated long-distance transportation. Central Shandong was the main source regions of PHg, Pb, Se, As, Cu and Cd. Southeastern and northwestern Shandong province and northern Jiangsu province were the most polluted source regions of Mn, Zn, and Ni. The crustal elements Fe and Ca had similar distributions of potential source regions, suggested by the highest PSCF values in southeastern Shandong and northern Jiangsu.

1. Introduction

The 2010 Global Burden of Disease Study suggested that outdoor air pollution led to the 1.2 million premature deaths in China (Hu et al., 2014). Particulate matter (PM) pollution was considered as the leading air pollutant in most regions as it caused a decline in atmospheric visibility and can also increase the incidence of respiratory diseases and mortality risk (Association and Management, 1990; Chan et al., 1997; D'Ippoliti et al., 2003; Han et al., 2011; Jung et al., 2009). According to the air quality reports issued by the Ministry of environmental protection of China (MEP, 2015), The annual mean $PM_{2.5}$ (particles with aerodynamic diameters of up to

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2.5 μm) concentration was 50 μg/m^3 over the entire country of China in 2015, which were twice the World Health Organization (WHO) air quality guideline for daily $PM_{2.5}$ of 25 μg/m^3 (Roy and Singh, 2014). The number of days with $PM_{2.5}$ and PM_{10} (particles with aerodynamic diameters of up to 2.5 and 10 μm, respectively) as the primary pollutant accounted for 66.8% and 15%, respectively, of the number of severe and above pollution days. In 2016, 32 cities had severe pollution days and above for more than 30 days, most of them distributed in the North China Plain (NCP) (MEP, 2016). The NCP is a heavily polluted region high in $PM_{2.5}$ and PM_{10} concentrations.

Mount Tai is the highest peak in the central NCP. Sampling at the summit can obtain information on regional-scale atmospheric processes affecting transport pathways and potential sources in the NCP. Although the adsorption of particulate mercury (PHg) and other trace elements on PM were accounted for a small proportion of fine particle mass, its special physical and chemical properties can cause harm to human body. The PHg and trace elements have both natural and anthropogenic sources. Natural sources include volcanic eruptions, sandstorms, and crustal dust and rock weathering. Anthropogenic sources mainly include coal combustion, vehicle emissions, nonferrous metal mining and metallurgical smelting (Xu et al., 2015). The particles chemical composition can be used to understand their origins of particles. For example, crustal weathering has been identified as the main source of Al, Ca, Fe, K, Mg, and Na (Marcazzan et al., 2001; Al-Momani, 2003; Singh et al., 2002; Swietlicki et al., 1996; Lin et al., 2005). Coal combustion is a major source of Pb, Se, Cu, As and Cd (Bunt and Waanders, 2008; Li et al., 2012). For instance, Se is frequently used as a tracer since it can be present in fine particles resulting from high temperature combustion and be retained in the atmosphere over long distances (Husain et al., 2004). Mn, Zn, and Ni mainly originate from vehicle emissions (Monaci et al., 2000).

Previous studies on PHg and other elements in PM analyzed the concentrations of different forms of mercury (Zhu et al., 2014; Xu et al., 2015; Fu et al., 2011; Wang et al., 2006; Li et al., 2016; Marcazzan et al., 2001) or different trace elements (Al-Momani, 2003; Berg and Steinnesb, 1994) in PM, and their diurnal to seasonal variation in urban areas. They also examined size distributions of mercury or other trace elements in PM (Singh et al., 2002), potential sources (Li et al., 2015; Keeler et al., 1995), chemical and physical transformations and wet and dry deposition (Nie et al., 2017). However only a handful of studies were conducted at Mountain sites (Fu et al., 2008, 2011; Li et al., 2015). Fu et al. (2008) showed distinct seasonal variation in PHg concentrations at Mt. Gongga in Southwest China with 74.1 pg/m^3 in winter, 22.5 pg/m^3 in autumn, 15.3 pg/m^3 in spring and 10.8 pg/m^3 in summer, and regional and local coal combustion as the main source of PHg. Li et al. (2015) suggested that for Mount Lu in Central China, nonferrous metal mining and metallurgical smelting, coal combustion, crustal materials were main emission sources of trace elements. More studies on mountain sites are needed to quantify background concentrations of pollutants in more remote regions in China in order to obtain more complete knowledge of anthropogenic influence on large scale air quality.

There are currently two articles on the analysis of PM at Mount Tai. One of them is a study by Deng et al. (2011), which analyzed the concentrations of trace elements (Al, Ca, Fe, Mg, Zn, Na, S, BC, Ti, Sr, Mn, Cu, As, Cd, Co, Cr, Ni, Pb, P, V) and potential emission sources in $PM_{2.5}$ and total suspended particles (TSP) at Mount Tai spring (14 March–6 May) and summer (2–30 June) 2006 and spring (26 March–18 May) 2007. The other is a study by Zhao et al. (2017) charactering of water-soluble inorganic ions (WSIIs) in $PM_{2.5}$ at Mount Tai during summer 2014. This study was the first to examine PHg in $PM_{2.5}$ and PM_{10} at Mount Tai.

In this study, we quantified concentrations of PHg and other trace elements in $PM_{2.5}$ and PM_{10} and examined their relationships with meteorological variables at Mount Tai during summer 2015. In summer, the particulate matter was primarily comprised of fine particles. Some of which could be crustal dust carrying pollutants from up to hundred and thousand miles away in Northwest China area (Deng et al., 2011). Here, we investigated the impact of crustal dust on Mount Tai via gaseous species, trace elements and anthropogenic aerosol. The influence of PM and meteorological conditions on potential source regions of trace elements were evaluated by PHg and element concentration in $PM_{2.5}$ and PM_{10}.

2. Methodology

2.1. Site description

Mount Tai is the highest peak in the central NCP, eastern China (Fig. 1). As shown in Fig. 1, The NCP is the region with the most serious $PM_{2.5}$ concentrations during 2008–2010 in China. Samples were collected at the weather station, near the summit of Mount Tai (36°15′N and 117°6′E; 1534 m asl). The sampling site is in the open terrain, free from the influence of tall trees, buildings, and other obstacles. The area has no permanent population, but frequented by tourists, which could influence the site under certain wind conditions.

2.2. Sample collection

Samples of $PM_{2.5}$ and PM_{10} were collected using atmospheric samplers (Model TH-150AIII) with a medium flow rate of 100 L/min. Sample collection was conducted from 08:00 a.m. to 07:30 a.m. next day between 15th June and 11th August 2015. The filter was stabilized at 25 °C and 50 ± 5% relative humidity for 24 h and the samples were weighed three times before and after collection. Fifty-three valid ambient samples were analyzed in the laboratory.

2.3. Sample analysis

As PHg can be volatile and exists at low quantities, it was analyzed separately from the other trace elements. First, the teflon filter was placed in a 20 mL tall, brown borosilicate glass bottle with 5 mL of 0.5% bromine chloride to ensure that the entire filter was soaked in the solution at 25 °C for 24 h. Then 15 mL ultrapure water was added, and the sample was refrigerated to be tested. Before analysis, 0.1 mL of $NH_2OH{\cdot}HCl$ was added to remove the effect of halogens. The sample was then shaken and mercury levels were analyzed using an atomic fluorescence spectrometer (AFS -830). This method of analysis was used previously by Li et al. (2015).

The element analysis process (Talbot et al., 2011) was as follows: First, we cut the teflon filters into beakers and added the 7 mL mixed solution of nitric acid and hydrochloric acid in the shredded membrane. Then the small beaker with the solution was placed in the heating instrument, and reflux for 2 h at 100 °C, cooling. Prior to analysis, setting up the sample to be refrigerated. The concentrations of twenty trace elements (Ca, Na, K, Fe, Al, Mg, Zn, Pb, Mn, Cu, Ba, Ni, As, Cr, Se, V, Cd, Mo, Co, and Ag) were measured through inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500a). Helium was used as the internal standard substance to eliminate matrix interference based on the EPA 200.8 method.

2.4. Principal component analysis

Principal Component Analysis (PCA) was introduced by Hotelling (1933), who proposed a multivariate statistical method designed to lose less valid information by combining multiple