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Study on mercury in airborne particulates from different functional areas of Jiaozuo City, China

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Abstract

Aim : A study was conducted with an aim to further understand the air environmental quality of Jiaozuo City, China and characteristics of pollutants by comparing the concentration of mercury in inhalable particles and dust of four functional areas.

Methodology : The sampling filter for inhalable particles was dried at 500°C for 2 hrs before use. The dust samples were dried and grinded, and to all the samples nitric acid and hydrofluoric acid was added for nitrification. The concentration of mercury was determined by Atomic Fluorescence Spectrometer after microwave digestion.

Results : The average concentration of mercury in inhalable particulate matter was 4.44 ng m^{-3} . was higher than the global background values. The concentration of mercury in the dust ranged from 0.45 to $1.51 \text{ } \mu\text{g g}^{-1}$ and the average value was $0.86 \text{ } \mu\text{g g}^{-1}$, which was 13.23 times higher than soil natural background value of mercury ($0.065 \text{ } \mu\text{g g}^{-1}$) in China. The analysis results of corresponding relationship indicated that mercury in inhalable particulate matter and dust originate from different sources.

Interpretation : The commercial and traffic activities may have a certain influence on the content of mercury in airborne particulates in Jiaozuo City.

Introduction

Atmospheric particulate matter and dusts are the main cause of air pollution, which exist widely. Particulates contain a variety of chemical elements, including organic and inorganic compounds. Trace elements are the most important among the inorganic compounds (Yap *et al.*, 2011; Nazir *et al.*, 2011). Mercury is a unique heavy metal element with variable oxidation states, and in atmosphere it mainly exist in gaseous phase which can be divided into gaseous elemental mercury-Hg⁰, reactive gaseous mercury -Hg²⁺ and particulate mercury (Liu and Luo, 2012; Xu *et al.*, 2015; Sun *et al.*, 2013). Mercury is highly toxic and harmful to human beings and environment, which has aroused great attention around the world (Li, 2010; Feng *et al.*, 2013; Olayan and Thomas, 2015; Matchavariani *et al.*, 2015). Generally, mercury in the environment originates from natural as well as anthropogenic source. Natural sources include volcanoes, forest fires, geothermal vents and evaporation from soil and water (Kim *et al.*, 2009). According to the United Nations Environment Programme, the anthropogenic mercury emission is about 1960 t, accounting for about 30% of the total amount of atmospheric mercury emissions (UNEP, 2013). Besides, anthropogenic emissions are mainly from coal combustion, which accounts for 45% of the total global anthropogenic emissions (Xu *et al.*, 2011). Currently, the production and consumption of coal in China is high as compared to other countries. The proportion of coal is more than three quarters in the total energy consumption, and coal-power plant is one of the most important source of mercury pollution in the urban environment. Unfortunately, this situation tends to last for a long period in the near future.

Rising mercury in the atmosphere has attracted the attention of Chinese government, and accordingly many studies on anthropogenic mercury emission have been conducted. Zhang *et al.* (2011) used the methods of emission factors and grey prediction to investigate the atmospheric mercury emissions from anthropogenic sources in the city of Chongqing. The results indicated an annual average increase of 16.20% from 2009 to 2015. Wu *et al.* (2017) predicted the atmospheric mercury emission of China's nonferrous metal smelting industry under different scenarios and analyzed the contribution of main emission abatement measures. Hui *et al.* (2017) developed the atmospheric mercury emission inventories for coal fire power plants, coal fire industrial boilers and coal fire residential stoves in 2010 and 2012, and predicted the atmospheric mercury emission from the coal combustion sector by 2020 and 2030 based on scenario analysis to evaluate the effectiveness of different control measures in the future. Jiaozuo City has a long history of being heavy industry. Its industrial layout is unreasonable, functional areas are sophisticated and urban infrastructure is poor, which makes the urban air pollution caused by anthropogenic factors more prominent.

In the present study, four typical urban functional areas were selected to collect the samples of inhalable particles and surface dust. The present study was conducted with an aim to

assess the atmospheric quality of Jiaozuo City and characteristics of pollutants by comparing the concentration of mercury in inhalable particles and dust of four functional areas, so as to provide a theoretical basis and practical significance for the city's environmental management and protection.

Materials and Methods

Study area and sample collection : The present study was conducted in Jiaozuo City, China. The climate of the study area is warm temperate continental monsoon with average temperature ranging between 12.8 to 14.9°C, annual precipitation of 603.5mm and frost-free period of 231 days. Four sampling sites in the city were selected (Fig.1) Coal fire power plant (Jiaozuo power plant), Commercial area (Wanfang Technical College), Traffic area (Jiaozuo tourist bus station) and Cultural area (Henan Polytechnic University).

In order to study the direct influence of atmospheric dust on human body, the dust was collected from 1.5-2 m near the surface. The sampling ensured no rain for seven days, and each sampling was done under similar climatic condition and the sampling time ranged from 48 to 68 hr. The dust on wooden doors and windows, surface of glass, roof platform, air conditioner shell and other non-paint wooden objects were swept into the polyethylene plastic bags. To make the samples representative, two batches of 80-100 g samples were collected in two or three streets every 1-2 m distance in each sampling area, and sieved uniformly to mix into a comprehensive sample. In total of four samples were collected from four functional areas. All the airborne particulate samples were collected with a TE-20-800 type 8 atmospheric sampler classification, manufactured by an American Tisch Company. The particle cutting aerodynamic equivalent particles size were 9.0, 5.8, 4.7, 3.2, 2.1, 1.1, 0.65 and 0.43 μm . The aerosol samples were collected on a 0.45 μm pore size quartz fiber membrane filters at 28.3 l·min⁻¹ flow-rate.

Sample analysis : Before sampling each filter was oven dried at 500°C for 2 hrs. After complete drying, the filters were weighted by a 1/10000 precision electronic weighing scales. The mass concentration of particulate matters were analyzed by gravimetric method. The dust samples were ground by agate mortar filtered with 200 mesh sieve and stored in sampling bags. The concentration of mercury was determined by Atomic Fluorescence Spectrometer a sample was randomly selected from every 10 samples and the sample was repeatedly measured for 4 times. The relative standard deviation was between 0.02%~5.19%, which indicated a high precision of the instrument.

Results and Discussion

The concentration of mercury in different size particles ranged from 0.54~2.30 ng m⁻³, 0.10~0.65 ng m⁻³, 0.14~0.37 ng m⁻³ and 0.15~0.65 ng m⁻³, respectively in coal fire power plant, traffic, commercial and cultural area, respectively (Fig. 2). The average

concentration of mercury in the fine particulate matter ($PM_{2.1}$) was 1.93 ng m^{-3} . The mercury concentrations were higher than those in typical urban polluted regions of Beijing (1.54 ng m^{-3}) (Chen *et al.*, 2007) and Beijing city (1.36 ng m^{-3}) (Wang *et al.*, 2004).

In the present study, the mercury concentration was higher in $PM_{5.8-10}$ and $PM_{2.1}$, as compared to other fractions particle

Fig. 1: Location of sampling areas in four different functional regions of Jiaozuo City, China

especially for $PM_{2.1}$. Similar result were reported during the study on the distribution of PM_{10} in traffic environment in Beijing (Qian *et al.*, 2011). The concentration of mercury increased with decrease in the particle size and appeared a peak in $1.1\text{--}2.1 \mu\text{m}$ in the typical areas of Beijing (Chen *et al.*, 2007).

The results showed that the mercury mass concentration was $0.0107 \mu\text{g m}^{-3}$, $0.0021 \mu\text{g m}^{-3}$, $0.0017 \mu\text{g m}^{-3}$, $0.0033 \mu\text{g m}^{-3}$, respectively for coal fire power plant, traffic area, commercial and cultural area, respectively. The total mass concentration of mercury in PM_{10} of coal fire power plant area was highest among all function areas followed by the cultural, traffic and commercial area. The total mass concentration of mercury in PM_{10} from coal fire power plant area was almost five times greater than other three regions. Burning fossil fuels was the main source of mercury in the atmosphere (Xu *et al.*, 2011; Wang *et al.*, 2004). The particulate matters produced by coal gas emission and transportation process in the coal fire power plant area consisted mercury, which may be the main reason why particle from this area had higher mercury concentration notably than others. The mass concentrations of mercury in PM_{10} from cultural area were higher than that from the traffic and commercial area. Soil mercury pyroelectric may release mercury in the atmosphere and the contents of atmospheric mercury were proportional to the temperature (Liu and Xiang, 2008; Li *et al.*, 2009). The soil and air temperature was high and the bare surface areas were large during summer sampling period in cultural area, which resulted in the release of mercury from soil in the atmosphere.

The concentration of mercury in Jiaozuo exceed the global background value ($0.001\text{--}0.086 \text{ ng m}^{-3}$) of mercury concentration in atmospheric suspended particulate matter (Keeler *et al.*, 1995) and the background value of global atmospheric mercury concentration ($1.5\text{--}2 \text{ ng m}^{-3}$) (Valente *et al.*,

Fig. 2 : Mass concentration of mercury on different diameters particles from four different functional regions of Jiaozuo City, China

2007; Kock *et al.*, 2005; Temme *et al.*, 2007). The study showed that average concentration of mercury during summer in inhalable particulate matter was 4.44 ng m^{-3} in Jiaozuo, which was close to the level of that in Guangzhou (4.16 ng m^{-3}) (Liu *et al.*, 2012) during same season, but higher than that in Ningbo (2.50 ng m^{-3}) (Wang *et al.*, 2017), Chongming Island (3.65 ng m^{-3}) (Li *et al.*, 2016) and JeJu Island (3.98 ng m^{-3}) (Nguyen *et al.*, 2010), respectively. All the above analysis showed that the concentration of mercury in inhalable particles in the study area was influenced by anthropogenic factors, and mercury in atmospheric particulates could easily combine with the fine particles.

According to the analysis results, the concentration of mercury in the surface dust in the studied area ranged from 0.45 to $1.51 \text{ } \mu\text{g g}^{-1}$ and the average concentration was $0.86 \text{ } \mu\text{g g}^{-1}$. The comparison of mercury concentration in the surface dust with other cities showed the level of mercury in the surface dust reported was similar to those reported for Baoji ($1.1 \text{ } \mu\text{g g}^{-1}$) (Chen *et al.*, 2011), but higher than the levels reported for Wuhu ($0.23 \text{ } \mu\text{g g}^{-1}$), Chongqing ($0.16 \text{ } \mu\text{g g}^{-1}$) and Beijing ($0.86 \text{ } \mu\text{g g}^{-1}$) (Fang *et al.*, 2009; Dai *et al.*, 2015; Li *et al.*, 2010) and 13.23 times higher than the natural background value of soil and mercury (Wei *et al.*, 1991). This reveals that mercury in the surface dust of the study areas in Jiaozuo can be affected by pollution factors.

The high to low order of mercury concentrations in the surface dust of different function areas was in the following order: commercial area, traffic area, coal fire power plant area and cultural area. Frequent human activities in the commercial and traffic area lead to larger flow rate of visitors and vehicles. The abnormal concentration of mercury may be due to mercury waste, vehicle emission. In addition, the particle size of the surface dust is fine and it can easily absorb heavy metals (Fang *et al.*, 2009; Zheng *et al.*, 2009).

Relevantly, Martin *et al.* (2017) and Gou *et al.* (2016) reported that inhalable particles and dust have different chemical compositions, and can exhibit a variety of characteristics of air pollution. The pollution elements present have different temporal and spatial distribution characteristics. In this study, four functional areas of inhalable particles and dust in mercury were obviously different. By comparative analysis, we could further understand the characteristics of pollution sources and pollution area in the air.

In the present study, the concentration order of mercury in inhalable particles was in the following order: coal fire power plant area>cultural area>traffic area>commercial district; and the concentration order of mercury on surface dust was in the following order: commercial district>traffic area>coal fire power plant area>cultural area. Comparative analysis revealed that among the coal fire power plant was present in high proportion in inhalable particulates, but in low proportion in surface dust. Particulate matter in commercial and traffic areas were mainly affected by the joint action of surface dust and exhaust emission. The concentration of mercury in commercial and traffic area was

Table 1 : Correlation coefficient between mercury level in PM_{10} , $\text{PM}_{2.5}$ and dust

	PM_{10}	$\text{PM}_{2.5}$	Dust
PM_{10}	Pearson Correlation		
	Sig. (2-tailed)		
	N		
$\text{PM}_{2.5}$	Pearson Correlation	.999(**)	
	Sig. (2-tailed)	.001	
	N	4	
Dust	Pearson Correlation	-.555	-.562
	Sig. (2-tailed)	.445	.438
	N	4	4

**Correlation is significant at 0.01 probability level (2-tailed)

lower than coal fire power plant and cultural area. Li *et al.* (2011) reported that the higher position above the ground, higher the concentration of metal element will show in atmospheric particulates, which indicated that human activities or industry emission was the main source of the element. This conclusion is roughly consistent with the results of this study.

In order to explore the correlation of mercury content between PM_{10} , $\text{PM}_{2.5}$ and dust in the study area, the present study analyzed the data and found that the mercury content in PM_{10} and $\text{PM}_{2.5}$ showed negative correlation with of dust. Furthermore, there was a significant correlation between PM_{10} and $\text{PM}_{2.5}$ (Table 1), which explains that higher the position from the ground, higher is the concentration of heavy metals in particulates. It is suggested that mercury in the surface dust and inhalable particles could be influenced by different pollution factors. According to the analysis presented above, presence of mercury in the atmospheric particulate matter in the urban air and near surface dust showed different spatio temporal distribution characteristics due to soil parent material, soil utilization mode and influence of different factors such as human activities. In addition, mercury had different distribution characteristics in different functional areas in the present study.

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