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Original article

Pollution characteristics of atmospheric particulate mercury near a coal-fired power plant on the southeast coast of China

Jinyu Gao, Hao Wang, Wu Cai, Jingting Wu, Yunfeng He*

College of Environmental and Resource Science, Zhejiang University, Hangzhou 310058, Zhejiang Province, China

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ABSTRACT

PM_{2.5} and PM₁₀ samples were collected in a rural area from December 2014 to August 2015 near a coal-fired power plant on the southeast coast of China. The total mercury concentrations in PM_{2.5} ranged from 116.2 to 1070.9 pg/m³ and in PM₁₀ from 173.4 to 1456.8 pg/m³, with averages of 309.5 ± 94.9 and 387.5 ± 133.5 pg/m³, respectively. These were much higher than those found in many foreign cities. The concentrations of mercury in PM_{2.5} and PM₁₀ varied seasonally, with the highest average concentration in winter. This was probably due to the seasonal variation of meteorological conditions and concentration of particulates. The highest mass-based concentration of mercury in PM_{2.5} and PM₁₀ was observed in spring. This result might be related to the prevailing sea winds in spring. The spatial variation of particulate mercury conformed to the concentration distribution characteristics of pollutant discharged from coal-fired power plant elevated source at the downwind area. The seasonal variation in the concentrations of the four mercury species (including exchangeable particulate mercury: EXPM, HCl-soluble particulate mercury: HPM, elemental particulate mercury: EPM, and residual particulate mercury: RPM) were all identical to the seasonal variation of total particulate mercury (TPM) in PM_{2.5} and PM₁₀ (winter > spring > summer). Owing to the variation of meteorological conditions, the proportion of different mercury species in TPM followed different seasonal trends. In contrast to RPM, the percentage of both HPM and EPM in PM_{2.5} was higher than in PM₁₀.

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1. Introduction

Mercury acts as a bioaccumulative neurotoxin and can exist in a gaseous state. Atmospheric mercury emitted from pollution sources could be transported long distances and deposited in remote places (Fitzgerald et al., 1998; Rasmussen, 1994; Sorensen et al., 1994). Atmospheric mercury exists primarily in three forms: gaseous elemental mercury (Hg⁰), divalent mercury (Hg²⁺), and particulate mercury (HgP). The different forms of atmospheric mercury have different physical and chemical properties that affect their transportation, deposition, and toxicity. Hg⁰ is stable and has a residence time of 0.5–2 yr in the atmosphere (Hall, 1995; Schroeder and Munthe, 1998). In contrast, Hg²⁺ and HgP are more easily removed from the atmosphere through dry and wet deposition

(Landis et al., 2002; Lee et al., 2001; Lindberg and Stratton, 1997), although these two types represent less than 5% of atmospheric mercury (Ames et al., 1998). A fraction of the mercury in particulates might form from conversion of gaseous mercury (Lu and Schroeder, 2004; Poissant et al., 2005; Xiu et al., 2005; Zhu et al., 2014). Considering the percentage total gaseous mercury (Hg⁰ and Hg²⁺) accounts, the mercury in particulates formed from conversion must be considered. Acting as a substrate for adsorption and chemical transformation of gaseous mercury, particulates play an important role in the biogeochemical processes of mercury in the atmosphere. HgP has a relatively shorter residence time in the atmosphere, and this kind of atmospheric mercury easily enters terrestrial and aquatic ecosystems through wet or dry deposition. The deposition velocity of particulates is related to their size (Lestari et al., 2003; Zhang et al., 2001). In addition, particle size has a significant influence on the adsorption of mercury on particulates (Kim et al., 2012). To sum up, particle size has an effect on the transformation and deposition of atmospheric particulate mercury. More than 60% of the mercury in inhalable particulates (PM₁₀) was

* Corresponding author. Tel./fax: +86 0571 88982898.

E-mail address: yfhe@zju.edu.cn (Y. He).

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concentrated in PM_{2.5} (Keeler et al., 1995; Tsai et al., 2003; Xiu et al., 2005). Moreover, PM_{2.5} can easily reach the alveoli of the lungs, thereby leading to severe damage to human health. Therefore, the study of mercury in PM_{2.5} and PM₁₀ has important implications.

In 2010, 1960 tons of mercury was emitted into the atmosphere because of human activities. Artisanal and small-scale gold mining, combustion of fossil fuels, and non-ferrous metal production accounted for 37, 24.7, and 15.5% of total mercury emissions, respectively. Moreover, 16.1% of the emissions came from coal combustion in power plants (AMAP/UNEP, 2013). Trace mercury in coal will be emitted to the atmosphere when the coal is burning (Clarke, 1993). China is regarded as the largest contributor of mercury in the world (Pacyna et al., 2010). In 1999, 487 million tons of raw coal were consumed in China and 33.6% of the mercury (68.0 tons) emitted from coal combustion came from power plants (Jiang et al., 2006; Streets et al., 2005). During the last decades, more and more coal-fired power plants in China have been equipped with air pollution devices that can remove part of the mercury emitted from a power plant. After cleaning, 19–72% of mercury (predominantly Hg⁰) in flue gas was emitted to the atmosphere (Wang et al., 2010). However, because of greater coal consumption in coal-fired power plants, the emission of mercury into the atmosphere from the coal-fired power plants in China increased to 132 tons in 2007 (Tian et al., 2011). Few investigations on particulate mercury near coal-fired power plants have been conducted over the past two decades in China. Instead, researchers have focused on HgP in big cities such as Beijing, Shanghai, Changchun, and Guiyang. Owing to coal combustion and other human activities, the atmospheric particulate mercury concentrations in these cities are high (Fang et al., 2001; Fu et al., 2011; Wang et al., 2006b; Xiu et al., 2005). Considering the seriously polluted condition, more studies about HgP should be conducted in industrial or urban areas in China. Different mercury species (with different properties) in atmospheric particulates, such as HgCl₂, HgS, HgO, and HgSO₄ would exhibit different behaviors. However, to the best of our knowledge, few studies providing speciation analysis of mercury in atmospheric particulates have been conducted.

In this study, we selected an area near a coal-fired power plant on the southeast coast of China to investigate atmospheric particulate mercury. We determined the concentrations of mercury in PM_{2.5} and PM₁₀ from December 2014 to August 2015, characterized their temporal and spatial variations, and subsequently analyzed the factors affecting the HgP concentrations. As the last step, the speciation of particulate mercury was determined to study the temporal variation of four mercury species.

2. Materials and methods

2.1. Site description and sampling

Particulate samples were collected from a rural area on the southeast coast of China near a coal-fired power plant. The prevailing winds in the study area were northwesterly in winter, and shifted to east in spring and summer. In this study, PM_{2.5} and PM₁₀ samples were collected from nine sampling spots (A, B, C, D, E, F, G, H, I), the exact locations of which are shown in Fig. 1.

Sampling campaigns were performed from December 2014 to January 2015 (winter), March 2015 to May 2015 (spring), and June 2015 to August 2015 (summer). Every month during this period, nine days were chosen to collect PM_{2.5} and PM₁₀ simultaneously using medium-volume samplers (Laoying 2034, China) with a flow rate of 100 L/min. That is, at every sampling spot, we took one day to collect samples including a PM_{2.5} sample and a PM₁₀ sample every month. In this study, a total of 160 samples (PM_{2.5} and PM₁₀)

were collected using quartz filters. Prior to sampling, the filters were heated at 500 °C for five hours to remove the mercury in them. Each sampling process lasted for 24 h. After sampling, all filters were packed, sealed in plastic bags, and stored in a freezer until use.

2.2. Analysis

2.2.1. Total mercury concentration

A quarter of each filter was digested in a 25-mL tube using a fresh mixture of HCl and HNO₃ (1:3, v/v) in a water bath at 95 °C (Qiu et al., 2006). During the digestion, BrCl was added to oxidize all the forms of mercury to Hg²⁺. After digestion, deionized water was used for dilution and then laid aside for 24 h. Then, NH₂OH·HCl was added to reduce the surplus oxidant. This was followed by the addition of SnCl₂ to convert all Hg²⁺ to Hg⁰. Finally, the total mercury concentration was determined by cold vapor atomic fluorescence spectrometry (CVAFS).

2.2.2. Organic carbon and elemental carbon

The filters sampled in December, March, April, and June were analyzed to obtain the content of organic carbon (OC) and elemental carbon (EC). A 0.5 cm² punch from a quarter of each filter was used for analysis by a DRI Model 2001 Thermal/Optical Carbon Analyzer following the IMPROVE thermal/optical reflectance (TOR) method. This method produced data for four OC fractions (OC1, OC2, OC3, and OC4) at temperatures of 120, 250, 450, and 550 °C in a non-oxidizing (He) atmosphere. A pyrolyzed carbon fraction (OP) was determined from the time that O₂ was added to the analysis atmosphere, until the reflectance attained its original value. Data for three EC fractions (EC1, EC2, and EC3) were produced at 580, 740, and 840 °C, respectively, in a 2% O₂/98% He (v/v) atmosphere. The IMPROVE protocol defines OC as OC1 + OC2 + OC3 + OC4 + OP and EC as EC1 + EC2 + EC3-OP. The detailed information was shown in previous study (Cao et al., 2003).

2.2.3. Mercury speciation analysis

The four mercury species in particulates were analyzed following sequential extraction procedures (Xiu et al., 2009). Four steps were needed to determine exchangeable particulate mercury (EXPM), HCl-soluble particulate mercury (HPM), elemental particulate mercury (EPM), and residual particulate mercury (RPM). EXPM exists in the atmospheric particulates unstably and has high potential for transportation and transformation. The main constituent of EXPM is HgCl₂. HPM refers to active mercury that is divalent, and is mainly composed of HgO and HgSO₄. EPM exists as Hg⁰. RPM is inert with stable properties. It consists of insoluble mercury compounds such as HgS and organic mercury (Wang et al., 2003; Xiu et al., 2009). Mercury speciation was determined for the particulate samples of December, March, and June to represent winter, spring, and summer, respectively.

Step 1: A quarter of each filter was put in a 50-ml plastic centrifuge tube to which 15 mL of 0.1 mol/L CaCl₂ solution was added. Then the tubes were placed in an ultrasound bath for 30 min, and then centrifuged at 3500 r/min for 10 min. The aqueous phase in the centrifuge tubes was transferred to 25-mL colorimetric tubes to determine the content of EXPM using the CVAFS method.

Step 2: A mixture of 20 mL of 1 mol/L HCl and 0.5 mL of 1% CuSO₄ was added to every centrifugal tube left after Step 1. Next, the tubes were placed in an ultrasound bath for 30 min; then centrifuged at 3500 r/min for 10 min. The aqueous phase in the centrifuge tubes was transferred to 25-mL colorimetric tubes to determine the content of HPM using the CVAFS method.

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