



# Particulate mercury in ambient air in Shanghai, China: Size-specific distribution, gas–particle partitioning, and association with carbonaceous composition<sup>☆</sup>

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## ARTICLE INFO

### Article history:

Received 16 November 2017

Received in revised form

23 March 2018

Accepted 25 March 2018

### Keywords:

Deposition

Gas–particle partition

Mercury

Size-distribution

Shanghai

## ABSTRACT

Mercury (Hg) has a complex atmospheric transformation cycle and acts as a global pollutant. Size-specific particle bound mercury (PBM) was implemented in different functional (industrial, urban and suburban) areas in Shanghai, China. The total concentration of 13-staged PBM (rang of 0.01–18.0 μm) varied of 99.0–611 pg/m<sup>3</sup>, with an average value of 318 ± 144 pg/m<sup>3</sup>. The Gaoqiao petrochemical industry (GQPI) site showed the highest concentrations, whereas the suburban Shanghai Jiao Tong University (SJTU) displayed the lowest. The PBM in nucleation, accumulation and coarse modes were 7.63–96.7, 69.5–455, and 9.43–176 pg/m<sup>3</sup>, respectively, and the fractions of 0.56–1.00 and 0.32–0.56 μm were the two most abundant. Both OC and EC displayed unimodal distribution patterns (peak of 0.56–1.00 μm) at GQPI, while bimodal distributions were observed at urban and suburban sites. Statistically positive correlations between the overall PBM and the corresponding PM and carbonaceous compounds ( $r = 0.38 - 0.54$ ,  $p < 0.01$ ), indicating their similar origins and OC/EC enhanced gaseous mercury forming PBM. The gas–particle partition model predicted gaseous oxidized mercury (GOM) were 253 ± 133, 237 ± 122, and 257 ± 144 pg/m<sup>3</sup> for GQPI, SAES and SJTU, respectively. The particle proportions of divalent mercury in the fraction of 0.32–1.00 μm were substantial (>80%), but smaller (<50%) for nucleation and coarse modes. The fraction of 9.90–18.00 μm occupied nearly 50% of the overall dry deposition fluxes of mercury. These finding highlight the emissions from different mercury and OC/EC origins, caused different size-specific distributions of PBM, which further affect their gas-particle partitioning and dry deposition of mercury species.

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## 1. Introduction

In view of its toxicity and bioaccumulation in the food chain, mercury (Hg) exposure in humans and wildlife has attracted increasing concern globally (Driscoll et al., 2012). Mercury is a naturally occurring metal, which can be directly emitted from volcanoes, wild forest fires, and geothermal vents. Anthropogenic

emissions of mercury account for ~64% (5207 tons/a) of the global budget of mercury (Tian et al., 2010; Huang et al., 2017; Streets et al., 2017), including coal combustion, waste incineration, and industrial exhausts from, for example, non-ferrous metals smelting, cement, iron/steel production, and chlor-alkali plants. The major mercury species in non-combustion processes is Hg<sup>0</sup>, while combustion processes (e.g., coal use, waste incineration, and wild fires) emit mercury primarily as divalent mercury (Hg<sup>2+</sup>). Landis et al. (2007) measured mercury levels in a tunnel and found that light duty vehicles were an important source of mercury species, and gasoline vehicles were ~4 times higher than diesel fuel vehicles. The average mercury concentration in crude oil ranged from 1 to

<sup>☆</sup> This paper has been recommended for acceptance by Prof. W. Wen-Xiong.

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10 ppm, and was mainly elemental and ionic mercury (Lan et al., 2015). It is estimated that ~56% of the total global anthropogenic mercury is currently emitted from East Asia (Mason and Pirrone, 2009; Streets et al., 2017).

Mercury is found in atmospheric, terrestrial, aquatic and marine ecosystems; however, the atmosphere is the foremost pathway for mercury emissions (Driscoll et al., 2012). Once emitted into the atmosphere (mostly as  $\text{Hg}^0$ ), mercury species can be transformed via redox chemistry, e.g., photooxidation, heterogeneous, and homogeneous reactions (Subir et al., 2011, 2012; Moore et al., 2014; Bargagli, 2016). Three major inorganic forms of mercury exist in the atmosphere, including gaseous elemental mercury (GEM,  $\text{Hg}^0$ ), gaseous oxidized mercury (GOM) and particle bound mercury (PBM). GOM and PBM together are called reactive mercury (RM), which possibly consists of  $\text{HgCl}_2$ ,  $\text{HgO}$ ,  $\text{HgBr}_2$ , and  $\text{Hg}(\text{OH})_2$ , but the exact composition is still unknown. The deposition velocity, scavenging capacity by precipitation and interactions with terrestrial surfaces are different for different mercury species (Mason and Pirrone, 2009; Blackwell and Driscoll, 2015). GEM is relatively inert with a low deposition velocity ( $\sim 0.1 \times 10^{-5}$  cm/s), which makes it the dominant species in the atmosphere (>90%) and a global pollutant through long range transport (Lin and Pehkonen, 1999; Gonzalez-Raymat et al., 2017). In contrast, GOM (0.5–6 cm/s) and PBM (0.02–2.0 cm/s) can be efficiently removed regionally downwind of the source via wet/dry deposition, resulting in atmospheric removal which is close to their corresponding emission sources or formation areas (Lin and Pehkonen, 1999; Zhang et al., 2009; Subir et al., 2011). Particle size is a crucial factor that controls the atmospheric residence time of PBM, with smaller size aerosols be removed less efficiently. Moreover, the size-specific distributions of PBM are dependent on their physical-chemical properties, gas–particle partitioning, and photochemical reactions (Chen et al., 2016). Thus, characterization of the size-specific distribution of PBM is crucial in understanding its formation, assessing its possible environmental fate, and offering appropriate management.

Carbonaceous compounds, usually divided into organic carbon (OC) and elemental carbon (EC), make up a significant fraction of PM, e.g., accounting for 30%–50% of  $\text{PM}_{2.5}$  mass (Feng et al., 2009; Vodička et al., 2015). EC is emitted into atmosphere as a primary aerosol due to the incomplete combustion of fossil fuels, e.g. vehicles, industry, and domestic space heating. However, OC can be both primary aerosols formed from combustion processes and secondary aerosols generated from atmospheric oxidation and gas–to–particle conversion. Several previous studies (Feng et al., 2009; Wang et al., 2016a) have demonstrated that the dominant part of carbonaceous species was secondary organic carbon (SOC) in China, which was formed through photooxidation reactions of volatile organic compounds (VOCs) and  $\text{NO}_x$ . The characteristics of absorbability and hygroscopicity of carbonaceous compounds can enhance the transformation of atmospheric pollutants by aqueous and heterogeneous reactions, which may affect mercury species transformation (Lin and Pehkonen, 1999; Subir et al., 2011; Duan et al., 2015). Additionally, GOM and PBM are thought to be independent of each other, and their concentrations are not dependent on PM components and ambient temperature. However, current researches (Rutter and Schauer, 2007a; Amos et al., 2014) has found that the major RM species are semi-volatile organic compounds, which can partition between gas and particle phases in the atmosphere. Several factors govern their gas–particle phase partitioning process: temperature, relatively humidity, chemical species of divalent mercury, aerosol abundance and interactive surface area. Rutter and Schauer (2007a) measured gas–particle partitioning coefficients of divalent mercury in dry urban areas and in the laboratory, and found that the partitioning process was strongly

dependent on ambient temperature and aerosol constituents. Amos et al. (2014) incorporated the classical gas–particle partition model into a GEOS-Chem global 3-D Hg model and found that mercury deposition at high latitudes increases, with more efficient scavenging of PBM caused by precipitating snow. The partitioning process has considerable influences on the transport, transformation, deposition, and removal of atmospheric mercury (Mason and Pirrone, 2009; Amos et al., 2014).

In this study, the size-specific PBM was measured at three different functional sites in Shanghai, including industrial, urban, and suburban areas. The major aims of this study were to: (1) measure the size-specific distributions of mercury and carbonaceous compounds attached to size-staged particles (0.01–18.0  $\mu\text{m}$ ); (2) evaluate the correlation between size-specific distributed PBM and carbonaceous compounds; (3) investigate the gas–particle partition and dry deposition behaviors of PBM. To the best of our knowledge, this is the first study to analyze both size-specific distributions of PBM, and gas–particle partitioning of RM at multiple functional areas. These research results can be used to enhance our understanding of how different origins affect the size-specific distributions of PBM and OC/EC, and further influences on the gas-particle transformation and atmospheric fates of PBM.

## 2. Materials and methods

### 2.1. Description of sampling site

Particulate matter samples were simultaneously collected in different functional areas in Shanghai (Fig. 1 and Table 1): Gaoqiao Petrochemical Industry (GQPI), Shanghai Academy of Environmental Science (SAES), and Shanghai Jiao Tong University (SJTU). GQPI is one of the largest petrochemical plants in East China, and has the ability to process ~5 million tons of crude oil per year, producing over 3.7 million tons of oil and ~7000 tons of chemical fiber products. SAES is located in urban downtown Shanghai, which is characterized by a main transport artery and viaduct with high density traffic flow every day. SJTU is located in a suburban area with low density residential zones, educational, and high-tech institutions, and has no apparent mercury point source within 1.5 km. The sampling period in this study ranged from March to May 2017, and lasted ~3 months.

### 2.2. Atmospheric mercury sampling

Prior to sampling at each site, three 13-staged size-specific particle samplers, Micro-orifice Uniform Deposition Impactor sampler (Moudi, Model 122R, MSP Corp, Shoreview, USA), were calibrated by placing on the rooftop of the School of Environmental Science and Engineering (SESE) building in SJTU with a distance of <3 m between each sampler, and samples were collected over 12 h, and then weighed. These size-grading airborne particle samplers were implemented synchronously and continuously in the three sampling sites for 2–4 days (47–93 h) each time at an air flow of 30 L/min.

Detailed instrument operation, quality assurance, and control methods can be found in our previous work (Chen et al., 2016). Briefly, the Moudi sampler can collect and separate particles into 13 size fractions: 0.010–0.018, 0.018–0.032, 0.032–0.056, 0.056–0.10, 0.10–0.18, 0.18–0.32, 0.32–0.56, 0.56–1.0, 1.0–1.8, 1.8–3.2, 3.2–5.6, 5.6–10.0, and 10.0–18.0  $\mu\text{m}$ . Prior to sampling, each filter tray was washed with distilled water and ethanol. Two kinds of quartz fiber filters (47 mm diameter for 0.056–18  $\mu\text{m}$  particles, 90 mm diameter for 0.01–0.056  $\mu\text{m}$ ) were pre-baked at 550 °C for 4 h, wrapped in aluminum foil, and then sealed in clean polyethylene bags. Leak and flow tests were strictly conducted

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