Atmospheric Pollution Research 5 (2014) 769–778

Atmospheric Pollution Research

www.atmospolres.com

Characterization of atmospheric mercury at a suburban site of central China from wintertime to springtime

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ABSTRACT

Atmospheric mercury exits primarily as gaseous mercury and particulate mercury (PHg). Change in the species of atmospheric mercury will pose significant impact on the biogeochemical process of mercury. Here total gaseous mercury (TGM) and total particulate mercury (TPM) were measured from heating season in wintertime to springtime with frequent dust storm during February to May 2009 in the suburban of Hefei, central China, where atmospheric mercury measurements were completely absent. The average concentrations of TGM and TPM were 2.57±1.37 ng/m^3 and 0.32±0.10 ng/m^3, respectively. Variations in the TGM were affected by both emissions and meteorological parameters. In the heating period (February), due to coal combustion TGM concentrations were significantly higher than those in the spring (March, April and May). A clear different diurnal variation in TGM concentration was also observed both in late winter and in spring, accompanying with the advance of sunrise. The percentage of total particulate mercury (TPM) in total atmospheric Hg ranged from 5.8%–19.2%, with relatively high levels appeared in March and April. PHg was mainly derived from direct emissions by coal combustion in February and May, while it was dominated by transformation from gaseous Hg on particles in March and April due to dust storms, which may result in more deposition of mercury to ecosystem.

Keywords: *Total gaseous mercury; particulate mercury; diurnal variation; sources; dust storm*

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Article History:
Received: 19 March 2014
Revised: 29 May 2014
Accepted: 10 June 2014

doi: 10.5094/APR.2014.086

1. Introduction

Mercury (Hg) is a ubiquitous pollutant with high biotoxicity and bioaccumulation through food chains, and its major pathway of transport is through the atmosphere. Both anthropogenic activities and natural processes can emit Hg into the atmosphere (Pirrone et al., 2010). Three important species of atmospheric Hg are known as gaseous elemental Hg (GEM, Hg^0), reactive gaseous Hg (RGM) and particulate Hg (PHg). GEM is the predominant species of atmospheric Hg with long residence time (0.5–2 years) and long–range transport in the atmosphere, and thus is a global pollutant (Schroeder and Munthe, 1998; Weiss–Penzias et al., 2007). Although RGM and PHg account for a small fraction of total atmospheric Hg and are not considered in long–range transport, they are not negligible because they can easily deposit to earth's surface from the atmosphere and impact terrestrial and aquatic ecosystems (Keeler et al., 1995; Lee et al., 2001; Xie et al., 2008). Furthermore, RGM and PHg have been suggested to be critical in Hg removal processes from the atmosphere and the cycling of Hg in the environment (Keeler et al., 1995; Lindberg and Stratton, 1998).

Monitoring the distribution of atmospheric Hg and its species is of significant importance to understand the fate of this element in the environment (Fitzgerald, 1995). Measurements of airborne Hg concentrations have thus been made worldwide at different locations. China is one of the largest mercury emitting countries in the world (Streets et al., 2005). However, atmospheric mercury measurements in China are scarce and in the central China completely absent. Up to date there are some monitoring sites on metropolitan areas such as Beijing, Shanghai, Guangzhou and Guiyang, and some background regions such as Mt. Changbai, Mt. Gongga and Mt. Waliguan (Table 1). However, the knowledge of atmospheric Hg in the vast central and western China where the economy booming is little. In this study, we firstly report atmospheric mercury in central China. The monitoring site was located in the suburban of Hefei, which is a quickly developing city with a population of approximately 8.0 million. The levels, diurnal and monthly variations and the distribution of total gaseous Hg (TGM) and total particulate mercury (TPM) during a period from heating season to springtime with dust storm were reported. The dominant factors influencing TGM and TPM in the ambient air during different months were also discussed. The results will provide special parameterization for model evaluation of the regional Hg pollution in a special climate region from temperate zone to subtropical zone.

2. Materials and Methods

2.1. TGM monitoring

Highly time–resolved TGM monitoring was carried out in situ using an automatic Mercury Vapor Analyzer (Tekran 2537B, Toronto, Canada) during a campaign from February to May 2009 in Hefei (31°52′ N, 117°17′ E), the capital of Anhui Province in central China (Figure 1). Hefei is one of the fastest growing cities in China, located between the Yangtz River and the Huaihe River, and near the geographical dividing line of North and South China. The climate there belongs to a transition region from temperate zone

to subtropical zone. The city is affected by coal combustion for heating in winter and dust storms from North China in spring. During the monitoring period, pollutants in the atmosphere such as NO_X, SO_2, Hg or PM were in high levels compared to the other seasons, and thus air quality was bad. Besides, Hefei is located upwind of Nanjing and Shanghai, megacities in East China. The monitoring there is propitious to comprehend the regional transport of Hg. During the monitoring period, Tekran 2537B was placed on the top of a main building (20 meters from the ground) of Anhui Institute of Optics and Fine Mechanics (AIOFM), located on Science Island. Science Island is in the suburban regions of Hefei, surrounded by farmlands in the northwest side and Dongpu Reservoir in the other sides. It is more than 5 km away from coal–fired power plants, iron and steel works and backbone roads (Figure 1). The monitoring site is not directly affected by obvious pollution sources, and therefore the results from this monitoring could represent the background of TGM in Hefei.

The detailed method for instrumental analysis of TGM was previously described by Xia et al. (2010). Briefly, Tekran 2537B continuously measured TGM with a 5–min integrated sampling frequency at an air flow rate of 1.5 L/min. Automated recalibrations were performed for Hg^0 every 24 h using an internal permeation source. Manual injections were used to evaluate these automated calibrations using a saturated Hg vapor standard before and after the field campaign. The relative percent differences between manual injections and automated calibrations were <2%, and the relative differences for duplicate injections were all <2%. The TGM detection limit in this operation mode is lower than 0.1 ng/m^3. The measurements by the Tekran instrument represent the TGM for this case because both GEM and GRM were collected on the gold collectors and transformed to Hg^0 during the amalgamation and thermal desorption (Temme et al., 2003). There were 6 009, 8 696, 7 298 and 8 674 TGM data for February, March, April and May, respectively.

Table 1. *Average concentrations of TGM and PHg at some typical locations in China and some other countries*

Location	Classification	Time	TGM (ng/m^3)	PHg (ng/m^3)	Reference
Hefei, China	Suburban	Feb–May 2009	2.53±1.36	0.32±0.10 [b]	This study
Beijing, China	Urban	Jan, Feb, and Sep 1998	7.9–34.9		Liu et al. (2002)
	Suburban	Jan, Feb, and Sep 1998	5.3–12.4		Liu et al. (2002)
	Remote	Jan, Feb, and Sep 1998	2.5–5		Liu et al. (2002)
	Urban	Jan–Dec 2003		1.18±0.82 [b]	Wang et al. (2006)
	Suburban	Jan–Dec 2003		0.68±0.62 [b]	Wang et al. (2006)
Shanghai, China	Urban	March 2002–Sep 2003		0.33–0.56 [b]	Xiu et al. (2009)
	Urban	Aug–Sep 2009	2.7±1.7		Friedli et al. (2011)
Changchun, China	Urban	Jul 1999 to Jan 2000	18.4	0.28 [b]	Fang et al. (2004)
	Suburban	Jul 1999 to Jan 2000	11.7	0.11 [b]	Fang et al. (2004)
Guiyang, China	Urban	Nov 2001–Nov 2002	9.7±10.2 [a]	0.37±0.68 [d]	Fu et al. (2011)
Guangzhou, China	Remote	Nov–Dec 2008	2.94±2.02		Li et al. (2011)
Taiwan, China				2.63±1.57 [c]	Tsai et al. (2003)
				1.86±1.01 [d]	Tsai et al. (2003)
Mt. Gongga, China	Remote	May 2005–July 2007	3.98±1.62	0.031±0.032 [b]	Fu et al. (2008a, 2008b)
Mt. Changbai, China	Remote	Aug 2005–Jul 2006	3.58±1.78	0.077±0.136 [d]	Wan et al. (2009a, 2009b)
Mt. Waliguan	Remote	Sep 2007–Aug 2008	1.98±0.98 [a]	0.019±0.018 [d]	Fu et al. (2012b)
Detroit, US	Urban	Jan–Dec 2003	2.2±1.3 [a]	0.021±0.03 [d]	Liu et al. (2007)
Alert, Canada	Remote	February–June 2005	1.0±0.4 [a]	0.1±0.12 [d]	Cobbett et al. (2007)
Rocky Mountains, US	Remote	October 2006–May 2007	1.51±0.12 [a]		Obrist et al. (2008)
Mace Head, Ireland	Remote	1998–2004	1.72		Kock et al. (2005)
Zingst, Germany	Remote	1998–2004	1.66		Kock et al. (2005)
Tokyo, Japan	Urban	April 2000–March 2001		0.098±0.051 [b]	Sakata and Marumoto (2002)
Soul, Korea	Urban	January 1999–August 2000	5.26±3.27		Kim and Kim (2001)
Great Lakes Region, US	Remote			0.011–0.022 [b]	Keeler et al. (1995)
	Urban			0.094 [b]	Keeler et al. (1995)
Southeast coastal cities of China	Urban	Nov. 2010, Jan., Apr., and Aug. 2011		0.14±0.13 [d,e]	Xu et al. (2013)
	Suburban	Nov. 2010, Jan., Apr., and Aug. 2011		0.037±0.019 [d,e]	Xu et al. (2013)
	Remote	Nov. 2010, Jan., Apr., and Aug. 2011		0.024±0.015 [d,e]	Xu et al. (2013)
Background value in the Northern Hemisphere			1.5–1.7		Lindberg et al. (2007)

[a] *GEM*
[b] *TPM*
[c] *Hg in PM_{10}*
[d] *Hg in $PM_{2.5}$*
[e] *Accounting for 46.8–71.9% of the total particulate mercury*