Source apportionment and distribution of atmospheric mercury in urban Beijing, China

ZHOU Xiaocheng[1,2], DU Jianguo[2*], WANG Chuanyuan [3], and LIU Sujian[4]

[1] School of Earth and Space Sciences, University of Science and Technology of China, Hefei 230026, China

[2] Institute of Earthquake Science, China Earthquake Administration, Beijing 100036, China

[3] Yantai Institute of Coastal Zone Research for Sustainable Development, Chinese Academy of Sciences, Yantai 264003, China

[4] Seismological Press, China Earthquake Administration, Beijing 100081, China

* Corresponding author, E-mail: jianguodu@hotmail.com

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Abstract The concentration of gaseous elemental mercury (GEM) in the atmosphere of urban Beijing was measured from October 2003 to September 2004 to investigate the origins and spatial-temporal variations of atmospheric Hg. The mean value of Hg concentration is 17.1 ng·m^{-3} (n=653). The atmospheric Hg data showed spatial-temporal variations throughout the duration of our observation. The maximum GEM concentration (53.7 ng·m^{-3}) was observed at Gucheng in the western area of urban Beijing. The GEM concentrations increased from the north to the south of the studied area, and were higher in winter than in summer. The highest and lowest monthly averages of GEM concentrations were measured to be 23.3 and 4.1 ng·m^{-3} in January and July, respectively. In addition, GEM concentrations are higher in the daytime than at night in Autumn and from 14 March to 15 April, but daily GEM variation showed an inverse pattern from 22 April to 22 May. In winter, two peak values of GEM concentrations occurred at 13:30 and 21:30. Daily variation of GEM concentrations in summer was the lowest in the four seasons. Mercury from coal combustion was estimated to be the main source of anthropogenic emissions in Beijing from October 2003 to September 2004. Additionally, Hg emission from natural gas burning was estimated to be another dominant source of atmospheric Hg in Beijing.

Key words atmospheric mercury; source; environment; Beijing, China

1 Introduction

Mercury (Hg) in the atmosphere exists predominantly in its elemental form (Hg^0). Usually, it has a residence time of about 0.5–2 years in the air and can be transported over long distance (Schroeder and Munthe, 1998; Qiao Shengying et al., 2006). Under a certain condition, Hg deposited in the atmosphere can readily be converted to highly toxic Hg compounds, such as methylmercury (MeHg), which has been found being accumulated in aquatic biota, especially fish (Scheider et al., 1998; Jiang Hongmei et al., 2007; He Tianrong et al., 2008).

Atmospheric Hg originates from both natural and anthropogenic sources. Natural sources include Hg emissions from the Earth's crust, especially from volcanoes, mercury-bearing deposits, and volatilization from the oceans. 50% to 75% of Hg in the atmosphere is related to human activities (Xu Xiaohong et al., 1999). Global and regional distributions and emissions of Hg have been quantified (Pacyna et al., 2001, 2002, 2006; Pirrone et al., 2001). Pacyna et al. (2003) gave a detailed account of global and continental atmospheric Hg emissions from fossil fuel combustion, non-ferrous metal, steel, cement, and waste incineration, a total of 1912.8 t of Hg of the world in 1995, of which 75% was attributed to the stationary combustion of fossil fuels.

Currently, much attention has been paid to the monitoring and reduction of atmospheric Hg in China. The total atmospheric contribution of Hg from coal combustion in China was estimated to be about 302.9 t in 1995 (Wang Qichao et al., 1999). The average value of total gaseous mercury (TGM) in the atmosphere over Guiyang City, China, was reported to be 7.39 ng·m^{-3} (Feng Xinbin et al., 2003). The atmospheric Hg in Beijing monitored for short durations in January, February, and September in 1998 indicated high concentrations of TGM in the residential and central areas of the city (Liu Shili et al., 2002). At the site of the Research Center for Eco-environmental Sciences in northwestern urban Beijing, Wang Qichao

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and Ma Rulong (1997) investigated GEM concentrations in January, April, July, and October in 2005. One-hour continuous measurements were conducted daily at 2:00, 8:00, 14:00, and 20:00, respectively. The highest Hg^0 concentrations appeared in winter, and the lowest in summer.

No comprehensive measurements of GEM in ambient air in urban Beijing have been reported up to date. The goals of this paper are to report the results of continuous Hg monitoring in the atmosphere of urban Beijing for a year, and to discuss the sources of Hg.

2 Methods

2.1 Sampling sites and time

Sixteen sites in urban Beijing were selected for monitoring GEM, with the goal of characterizing the spatial distribution of atmospheric Hg and analyzing the Hg contribution of anthropogenic sources (Fig. 1). Five sites were selected along the West Third Ring Road, and one was at the North Fourth Ring Road to understand the variations of GEM concentrations from north to south. Ten sites (including Site 7 located at the West Third Ring Road) were selected along Fuxing Road, and one was at the Beijing Second Chemical Plant (BSCP) to understand the variations of GEM concentrations from west to east. The sampling height was about 1.1 m above ground and far from any obstacles. The sampling sites were about 200 m away from the road side.
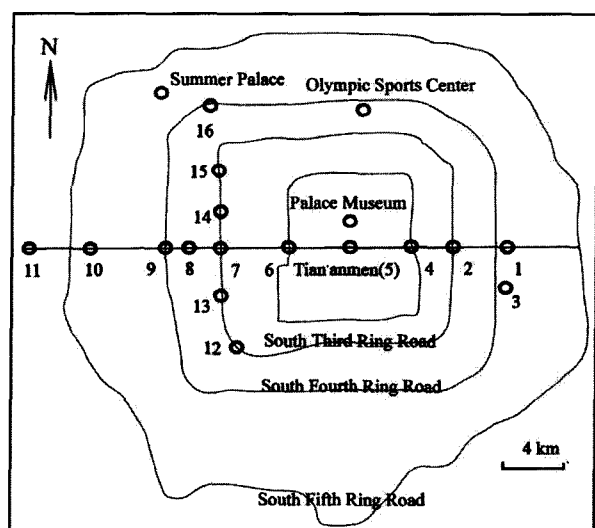

Fig. 1. Locations of sampling sites. 1. Sihui; 2. Guomao; 3. Beijing Second Chemical Plant (BSCP); 4. Jianguomen; 5. Tiananmen; 6. Fuxingmen; 7. Gongzhufeng; 8. Wanshou Road; 9. Wukesong; 10. Laoshan; 11. Gucheng; 12. Fengyi Bridge; 13. Liuli Bridge; 14. Hangtian Bridge; 15. Zizhu Bridge; 16. Suzhou Bridge.

The temporal variations of GEM concentrations

were investigated by conducting measurement daily or every 2 hours from October 5, 2003 to September 30, 2004 at six sampling sites (Fig. 1). The The Wanshou Road (N:39°54'23.17″ , E:116°17'4.9″; Site 8), between the West Third Ring Road and the West Fourth Ring Road, is a heavily traffic area surrounded by public buildings without industrial sources. The Wanshou Road sampling site includes a flat, grassy area and some wooded areas, mainly populated by deciduous poplar trees. The site was equipped with the standard meteorological instrumentation for measuring temperature and relative humidity. The SE and NW winds in Beijing are prevailing in summer and in winter, respectively.

2.2 Sampling and analysis

A QM201G Hg analyzer was employed to measure the concentrations of GEM in the air. The instrument had a detection limit of 1 $ng·m^{-3}$. The air samples were collected at 0.8 m above the ground with a quartz tube filled with a gold filament, through which air was pumped at a flow rate of 1.2 L/min for 15 minutes. The GEM was trapped into gold adsorbents in the form of an amalgam. Then, the sample tube was heated to 850°C to decompose the amalgam. Subsequently, the released Hg was analyzed using the cold atomic fluorescence method (Zhou Xiaocheng et al., 2007a, b; Wang Chuanyuan et al., 2004). The atmospheric temperature and humidity were concurrently recorded. Rainfall data were collected from local weather stations.

2.3 QA/QC for GEM

The Hg-saturated air, stored in a 400-mL sealed flask containing about 2–3 mL of metallic Hg, was used as the experimental standard for calibration at least once per month. No significant differences between the theoretical and measured values of Hg concentrations were found for Hg-saturated air. The margin of error for the measurements was 5%.

3 Results

The data measured at the Wanshou Road sampling site are shown in Figs. 2, 3, 4, and 5. The mean and standard deviation (SD) values of GEM during the entire study periods were calculated to be 17.1 and 11.9 $ng·m^{-3}$ (n=653), respectively. The mean value is much higher than the global background of 2.0 $ng·m^{-3}$ (Lindqvist et al., 1991), and higher than that (7.39 $ng·m^{-3}$) measured in Guiyang City, China (Feng Xinbin et al., 2003).

The concentrations of GEM at 15:30 every day range from 1.98 $ng·m^{-3}$ in summer to 51.4 $ng·m^{-3}$ in

autumn. The distinct relationship between GEM and humidity was not observed, but a weak correlation (R=-0.43) between temperature and GEM was observed. The GEM concentrations showed a decreasing trend in the rainy season (Fig. 2).
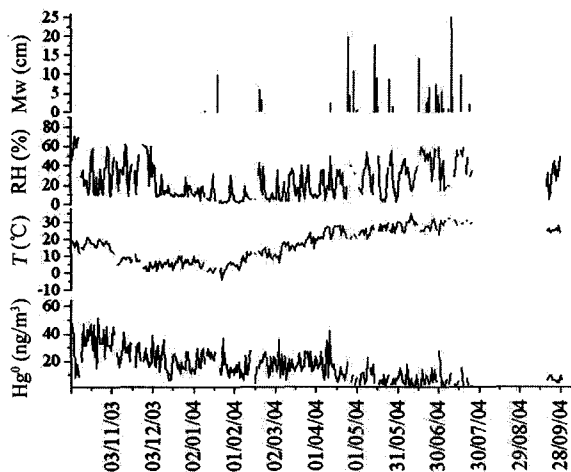

Fig. 2. The variations of Hg concentrations (Hg^0), rainfall (Mw), relative humidity (RH), and temperature (T) at the Wanshou Road sampling site at 15:30 every day from October 2003 to September 2004.
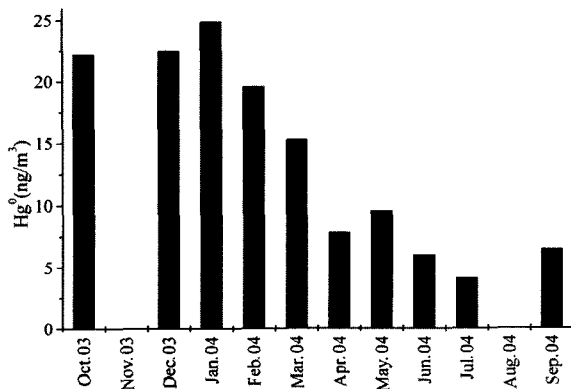

Fig. 3. The monthly mean values of GEM concentrations between October 2003 and September 2004, showing monthly and seasonal variations of GEM in urban Beijing.

The GEM concentrations vary monthly and seasonally (Fig. 3). The highest value of GEM concentrations was 23.3 ng·m^{-3} in January and the lowest was 4.1 ng·m^{-3} in July. The monthly mean values of GEM concentrations decreased following the following order: winter (Dec.–Feb.)>fall (Sep.–Nov.)>spring (Mar.–May)>summer (Jun.–Aug.), which was in accordance with the results of previous investigations (Wang Zhangwei et al., 2007).

The data measured at the Wanshou Road sampling site clearly showed a pattern of diurnal distribution (Figs. 4 and 5). The diurnal distribution pattern of GEM concentrations in autumn distinctly contrasted

with that in summer. The average values of GEM concentrations at 10:30, 15:30, and 16:30 in the period from 5 October to 5 November, 2003, were 16.4, 37.8, and 12.1 ng·m^{-3}, respectively. The values of GEM concentrations during the daytime were notably higher than those at night. The peaks occurred during 13:00–16:00 (Figs. 4a and 5a). Diurnal GEM concentrations were significantly correlated with air temperature (R= 0.91).
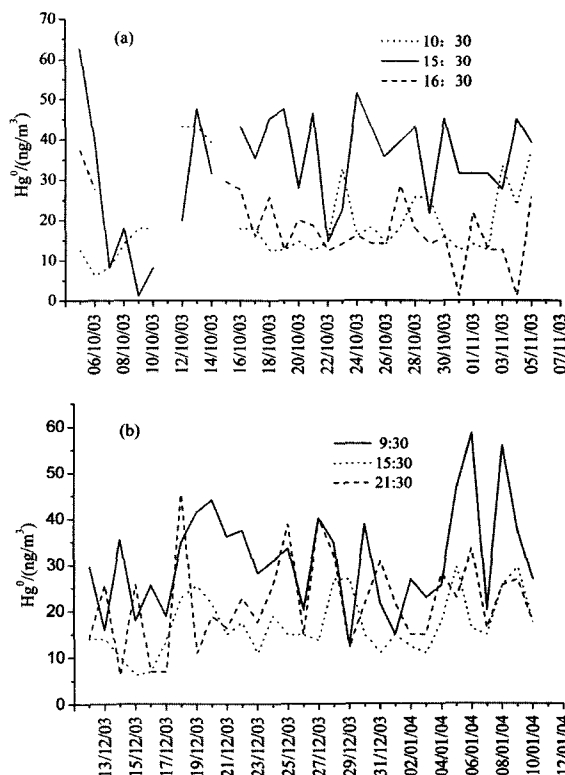

Fig. 4. The variations of GEM concentrations at 10:30, 15:30, and 16:30 during the period from 5 October to 5 November, 2003 (a) and at 9:30, 15:30, and 21:30 during the period from 12 December 2003 to 10 January 2004 (b).

Two peaks of GEM concentrations occurred in the afternoon and early evening in winter. The peak values were 46.7 and 42.7 ng·m^{-3}, as observed at 13:30 and 21:30 from 21 December, 2003, to 24 February, 2004 (Figs. 4b and 5b, c). The average values of GEM concentrations observed at 9:30, 15:30, and 21:30 during the period from 12 December, 2003 to 10 January, 2004 were 31.1, 17.0, and 21.9 ng·m^{-3}, respectively.

The GEM concentrations usually exhibited higher values during the period of 11:30–13:30, and then dropped after 15:30 during the period from March 14 to April 15, 2004 (Fig. 5c). GEM concentrations during the daytime were clearly lower than those at night during the period from April 22 to May 22, 2004 (Fig. 5c, d). The highest value of GEM con-

centrations we recorded in the daytime was 16 ng·m^{-3}.

Diurnal variation of GEM was small in summer (Fig. 5d). The variation magnitude of GEM concen-

trations was smaller during the period from May 28 to July 21, 2004 than that in other seasons in Beijing due to the instability of meteorological conditions.
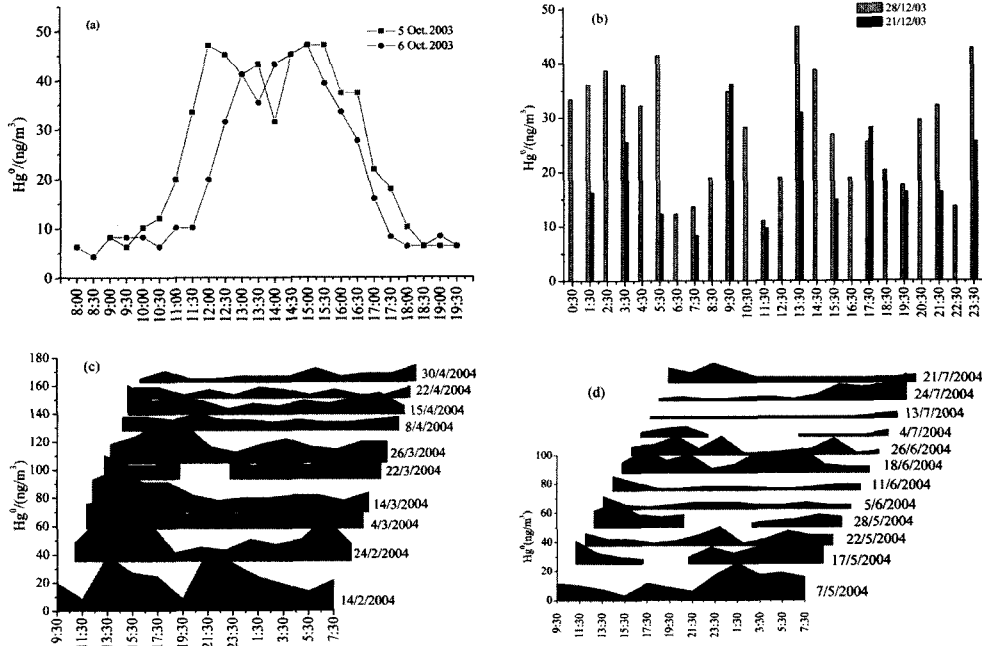

Fig. 5. The diurnal variations of GEM concentrations. (a) 5–6 October, 2003; (b) 21, 28 December, 2003; (c) from 14 February to 30 April, 2004; (d) from 7 May to 21 July, 2004.
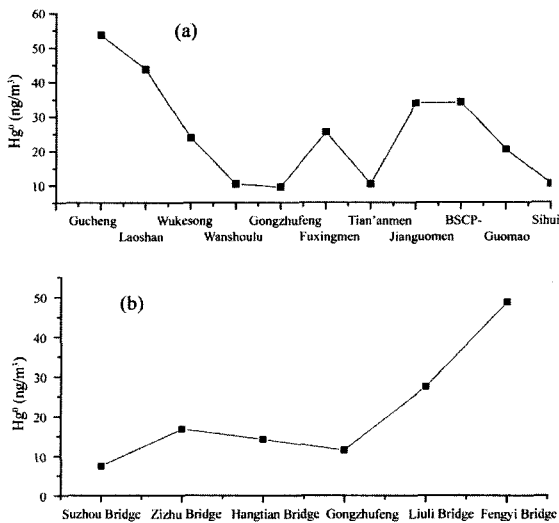

Fig. 6. (a) Variations of GEM concentrations from west to east; (b) variations of GEM concentrations from north to south in the study area.

The spatial distributions of GEM concentrations are presented in Fig. 6. The maximum (53.7 ng·m^{-3}) GEM concentrations were observed at the Gucheng site in western Beijing. The minimum GEM concentrations were 10.4 ng·m^{-3} at the Tiananmen site in the center of Beijing City (Figs. 1 and 6a). Additionally, the increasing tendency of GEM concentrations from

north to south was obvious. The GEM concentrations (48.6 ng·m^{-3}) at Fengyi Bridge in southwestern Beijing were much higher than those (7.5 ng·m^{-3}) at Suzhou Bridge, which is located in northwestern Beijing (Figs. 1 and 6b).

4 Discussion

4.1 Factors controlling GEM variations in urban atmosphere

The temporal patterns of GEM concentrations might be related to many factors, such as the source of GEM, chemicals in the atmosphere, and weather. The concentrations of GEM measured at the Wanshou Road site, where the maximum GEM concentrations were usually observed in winter, clearly showed seasonal variations (Figs. 2–5) similar to those of GEM in Europe and North America (Blanchard et al., 2002). The minimum GEM concentrations were observed in summer at the Wank Summit (Slemr and Scheel, 1998) and in Sweden (Brosset, 1982). Measurements of GEM in the Scandinavian atmosphere demonstrated that the median value of Hg concentrations was 33%, higher in winter than in summer (Iverfeldt, 1991). The data observed by the Canadian Atmospheric Mercury Measurement Network showed a slight seasonal variation pattern, with higher median GEM concentra-

tions in late winter and lower concentrations in late summer at mid-latitude sites in Canada (Kellerhals et al., 2003).

In December and January, the coldest months in Beijing, the high values of Hg concentrations in the observed sites may result from coal consumption (Liu Shili et al., 2002). Total energy consumption reached 45 million tons of coal in Beijing in 2003, of which coal and coke were up to 65% (BMBS, 2004; Xing Fangfang et al., 2007). Primary domestic usage of coal in Beijing is to heat houses during the period from November 15 to March 15 (Wen Zuoxiang and Ma Jingtao, 2003), which can heavily affect the seasonal variation of GEM concentrations in the atmosphere. We have attributed the diurnal distribution pattern of GEM concentrations in winter at Wanshou Road to domestic coal burning. This indicates that Hg emission from domestic coal burning is a significant atmospheric Hg source.

Comparisons between the hourly temperature and the corresponding GEM concentrations suggested that ground emissions of elemental Hg play a role in every season (i.e., higher Hg^0 concentrations in daytime). Hg pollution in Beijing's topsoil is serious, and is the most serious at the center of the city, and gradually decreases to the suburbs (Zhang Xinmin et al., 2006). Wang Dingyong et al. (2003) reported that there was a positive correlation (r=0.741) between the atmospheric Hg concentrations and the Hg contents in soils. The higher GEM concentrations during the daytime could be attributed to solar radiation. Solar radiation can lead to or enhance the photochemical reduction of Hg^{2+} species and photo-induced biological processes (e.g. photosynthesis). Solar radiation can also lead to an increase in surface temperature. These factors, alone or in combination, could lead to the release of elemental Hg into the atmosphere (Kim et al., 1995; Feng Xinbin et al., 2003; Poissant, 2000). In autumn, GEM concentrations show obvious enhancement, which is primarily attributed to the re-emission of Hg from the surface soil. Such processes are enhanced during the daytime at the temperatures above 8 °C, making soil re-emission one of the most significant sources of atmospheric Hg in autumn (Lindberg et al., 1991; Kim and Kim, 2001; Poissant et al., 2004; Bash et al., 2004). In addition, Hg predominantly originating in the atmosphere can accumulate in foliar tissues, which results in lower concentrations of atmospheric GEM in summer. Conversely, deciduous trees begin their dormancy and drop their leaves in autumn, indicating a new Hg input to surface soil (Ericksen et al., 2003). The estimated annual deposition of Hg via litterfall in the Hadlock Brook watershed was greater than Hg deposition from precipitation, and was similar to or greater than Hg deposition via throughfall (Sheehan et al., 2006). In early spring, the concentra-

tions of GEM in the study area are obviously higher in the daytime than at night. Hg is usually deposited on the ground surface during winter and may re-emit back into the air due to increasing surface temperatures in the daytime during early spring (Feng Xinbin et al., 1996; Kim and Kim, 2000).

Wind can affect GEM concentrations in urban atmosphere. Low wind speeds and decreasing air temperatures depress atmospheric convection, which is favoring Hg accumulation in the urban atmosphere from burning fossil fuels. The nocturnal boundary layers formed in the urban area can trap GEM near the surface (Lee et al., 1998), resulting in high Hg concentrations in the atmosphere in winter. Consequently, Hg concentrations were usually higher in Beijing during daytime with high pollution. However, the wind was stronger in late spring, especially in the period of 12:00–14:00 (Yang Xianwei et al., 2002), when Hg concentrations were lower. As reported by Lee et al. (1998) , stronger wind could reduce the concentrations of GEM in the atmosphere.

Rainfall is also a contributing factor to GEM concentrations in the atmosphere. The minimum GEM concentrations were observed in summer at Wanshou Road and obviously lower in July than in May. This correlates with the fact that rainfall was heavier in July than in May. Wallschlager et al. (2000) reported a similar seasonal variation trend of GEM. In addition, atmospheric oxidation of Hg^0 to Hg^{2+} and Hg^+ is the key step in the processing of Hg. The ozone and OH radicals are of potential importance in removal of atmospheric Hg atoms in summer (Lee et al., 1998; Lin and Pehkonen, 1999; Ren Xinrong et al., 2002; Wang Shulan and Chai Fahe, 2002; Jensen and Iverfeldt, 1994). Lindberg et al. (1992) observed a different seasonal variation in TGM concentrations, with the maximum occurring in summer, at a rural site in Tennessee. That was probably due to the influence of local Hg sources and their dependence on air and soil temperatures, with higher summer temperatures increasing Hg fluxes (Lindberg et al., 1992).
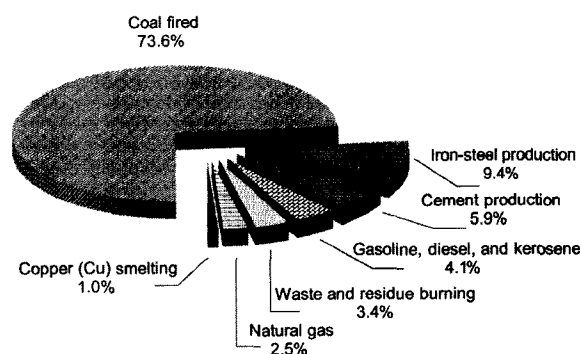

Fig. 7. Mercury emissions from different anthropogenic sources in Beijing in 2003.

The spatial variation of GEM concentrations might be related to the sources of GEM in urban Beijing. The decrease in GEM concentrations from west to east (Fig. 6a) could be attributed to local anthropogenic emissions, especially in the Shijingshan district, an industrial area housing a big electric power plant, cement factories, and an iron and steel plant. The increasing GEM concentrations from Suzhou Bridge to Fengyi Bridge (Figs. 1 and 6b) might be partly explained by the northwest wind moving polluted air from the Shijingshan district to farther south (Zhang Xinmin et al., 2006).

4.2 Source apportionment

Determination of source apportionment for GEM is very important for controlling atmospheric Hg pollution. Detailed source apportionments were conducted by indirect approaches as used by Streets et al. (2005), Wu Ye et al. (2006), and Wang Shuxiao et al. (2006). The emission factors and the estimated results for GEM in Beijing are listed in Table 1 and shown in Fig. 7. The annual Hg contribution to the atmosphere from anthropogenic sources in Beijing was estimated to be 6.79 t.

4.2.1 Hg emission from fossil-fuel combustion

Table 1. Emissions of total mercury from anthropogenic sources in Beijing in 2003, China

Source	Emission factor		Production/ Consumption	Emission of Hg (t)
	Unit	Amount		
Coal burning	g·t^{-1}	0.16[1]	3.11×10^7 t	5.00
Fuel oil (gasoline, diesel, and kerosene)	g·t^{-1}	0.058[3]	4.80×10^6 t	0.28
Natural gas	μg·m^{-3}	7.78[5]	2.38×10^{10} m^3	0.17
Iron-steel production	g·t^{-1}	0.04[2]	1.60×10^7 t	0.64
Copper (Cu)	g·t^{-1}	9.6[6]	7.3×10^3 t	0.07
Cement production	g·t^{-1}	0.04[1]	9.99×10^6 t	0.40
Waste and residue burning	g·t^{-1}	2.8[4]	8.1×10^4 t	0.23
Total				6.79

Note: The data used for estimation were after ITSDNBSC (2004), NBSC (2004), and BMBS (2004); the emission factors were after [1] Streets et al. (2005), [2] Pacyna and Pacyna (2002), [3] US EPA (1995), [4] UN-ECE/EMEP (2004), [5] Chen Jianfa et al. (2001), Liu Quanyou et al. (2006), and [6] Jiang Jingkun (2004).

Hg emission from the consumption of fossil fuels is the predominant source of atmospheric GEM in Beijing (Fig. 7). The Hg emissions from burning coal in Beijing in 2003 were calculated to be about 5 t, i.e., the total Hg emission from non-coal sources (1.2 t, Wu Ye et al., 2006) subtracted from that from anthropogenic sources (6.2 t, Wang Shuxiao et al., 2006). Coal burning was the largest source of anthropogenic Hg emissions in Beijing. Total coal consumption in

Beijing in 2003 was about 31.06 Mt, including 21.58 Mt raw coal, 5.1 Mt cleaned coal, and 4.38 Mt coke. Approximately 13% of total coal consumption was used for heating (BMBS, 2004). The consumed coal in Beijing was mostly derived from Shanxi Province and the Inner Mongolia Autonomous Region, with a small part from local mines. By taking the mean value of Hg in coal as 0.22 μg·g^{-1} (Wang Qichao et al., 1999; Streets et al., 2005), the annual assessment of total mercury from coal combustion is at least 6.8 t. In consideration of the fact that about 17% of Hg in the burned coal remains as bottom ash and a small part remains in particulate matter, the value of Hg emissions from coal burning to the atmosphere in Beijing is also estimated to be about 5 t (Table 1).

The combustion of fuel oil and natural gas has become an important source of Hg emissions, estimated to be 0.28 t and 0.17 t in 2003, respectively (Table 1, Fig. 7). From 1995 to 2005, total final energy-carbon consumption had increased by 71.5% in Beijing (Xing Fangfang et al., 2007). The percentages of coal and oil consumption in the energy structure of Beijing have changed since 1995. Natural gas has replaced coal for heating on a large scale since 2000. The consumption of natural gas had increased by 364.9% in Beijing from 1999 to 2005 (BMBS, 2006), indicating combustion of natural gas as one of the dominant sources of GEM. In 2002, oil and gas began to replace coal (Xing Fangfang et al., 2007). From 2003 to 2005, the percentage of coal consumption had continuously decreased, but oil consumption had increased. Therefore, the total Hg emissions from fuel oil and natural gas increased with increasing consumption of fuel oil and gas, even though Hg concentrations in fuel oil and gas were much lower than those in coal (Won et al., 2007).

4.2.2 Hg emissions from metallurgical industry

The metallurgical industry is an anthropogenic source of atmospheric GEM in Beijing. Hg emissions into the atmosphere (386 t) from the metal-smelting industry (mainly Fe, Zn, Pb, and Cu) were higher than Hg emissions from coal combustion (213.8 t) in China in 1998 (CNEPA, 2000). The steel output from the iron-steel plant located in the Shijingshan area of West Beijing had increased from 15.27 Mt to 16.05 Mt from 1995 to 2003 (NBSC, 1995, 2004). The total Hg released from iron and steel production had mounted up to 0.67 t, and Hg emissions from Cu smelting were at 0.07 t in Beijing in 2003 (Table 1, Fig. 7). Coal-related emissions were excluded from these numbers to avoid double counting with industrial coal use.

4.2.3 Hg emissions from other sources

Another major anthropogenic source of GEM in the atmosphere is cement production in Beijing. Most cement factories are located in the western and northern mountainous regions of Beijing (Chen Xiaoqiu et al., 2005). From 1995 to 2003, cement production had increased from 5.74 Mt to 8.82 Mt (NBSC, 1995, 2004). Total Hg emissions from cement production were 0.4 t in 2003 (Table 1, Fig. 7). Fukuzaki et al. (1986) measured the Hg concentrations in ingredients, fuels, cement, and exhausted gases in a cement factory, and discovered that daily Hg emission from the factory was about 1.5 kg, in which most of Hg originated from limestone.

The incinerated solid wastes were measured at approximately 8.1×10^4 t in Beijing in 2003. If the input of Hg into the furnace is taken at 100%, quantities of transferred Hg from waste to slag, electrostatic precipitator dust, and flue gas in a municipal solid waste incinerator were estimated to be 4%, 24%, and 72%, respectively (Brunner and Moench, 1986). Therefore, the total Hg emission to the atmosphere from incinerating solid wastes was estimated to be 0.23 t in Beijing in 2003 (Table 1, Fig. 7).

5 Conclusions

The concentrations of atmospheric GEM in urban Beijing in 2003 are significantly higher than global levels of approximately 1.5–2.0 ng·m^{-3}. The concentrations of atmospheric GEM in the study area showed daily and seasonal variations and an increasing trend from north to south. The atmospheric GEM concentrations were higher in winter than in summer. The values of GEM concentrations were larger in the daytime than at night in autumn and from 14 March to 15 April, but they were larger at night than during the daytime from April 22 to May 22. These phenomena were controlled by Hg sources and meteorological activities. The source apportionment indicates that coal combustion is the largest anthropogenic source of Hg in Beijing; other anthropogenic sources are the metallurgic industry, cement production, and the usage of oil and gas. Hg emissions from the combustion of natural gas have become a dominant source of atmospheric Hg in Beijing since 2000, when natural gas began to replace coal.

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