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Total gaseous mercury in the atmosphere of Guiyang, PR China

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Abstract

Four measurement campaigns were carried out to monitor total gaseous mercury (TGM) at one site in Guiyang City, PR China in the following periods: April 19–30, 2000; February 26–March 14, 2001; June 26–July 20, 2001; and October 9–November 22, 2001, respectively. High temporal resolved data were obtained by using automated mercury analyzers Gardis 1A and Tekran 2537A. TGM data from all measurement periods followed the typical log normal distribution pattern. The geometric mean of TGM from different seasons were 8.56, 7.45, 5.20 and 8.33 ng m^{-3} in spring 2000, winter 2001, summer 2001 and autumn 2001, respectively. The overall average TGM covering the sampling periods was 7.39 ng m^{-3}, which is significantly elevated comparing to global background of approximately 1.5–2.0 ng m^{-3}. The major anthropogenic atmospheric mercury emission sources differed significantly among seasons, which caused the seasonal variability of TGM level. Distinct daily variability of TGM was observed among seasons. The daytime TGM concentrations were larger than that of nighttime in spring and winter seasons, while in summer and autumn the opposite daily TGM distribution pattern was observed.

Keywords: Total gaseous mercury; Atmosphere; Guiyang; Source; Mercury measurement; Trends

1. Introduction

Both human activities and natural processes emit gaseous phase mercury into the atmosphere (Schroeder and Munthe, 1998; Lindqvist and Rodhe, 1985; Mason et al., 1994). Man-made emissions of mercury arise from fossil-fuel combustion, metal smelting, refining and manufacturing, chlor-alkali plants, waste incineration and gold mining (Schroeder and Munthe, 1998; Sloss, 1994). Natural source include outgassing of the earth's mantle/crustal material, evasion from surfacial soils, water bodies (both fresh- and saltwater), vegetation surfaces, wild fires, volcanoes, and geothermal sources (Schroeder and Munthe, 1998). Re-emission involving gaseous evasion of previously deposited mercury is usually categorized into natural source because it is difficult to differentiate between them.

The amount of mercury mobilized and released into the atmosphere has increased since the begin-

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ning of the industrial age (Mason et al., 1994). Measurements of atmospheric mercury over the Atlantic Ocean suggest a global increase between 1977 and 1990 (Slemr and Langer, 1992). Long term monitoring of total gaseous mercury (TGM) at two European background sites revealed a deceasing trend in atmospheric mercury since 1990 (Slemr and Scheel, 1998; Iverfeldt et al., 1995; Slemr et al., 1995). On the contrary, long term monitoring of TGM at a Canadian Artic site has not shown any significant changes of annual average concentration since 1995 (Schroeder, 2001). In industrialized counties from Europe and northern American, anthropogenic mercury emissions are been reducing since considerable efforts are devoted to curbing the use of mercury and strict regulations are applied to major anthropogenic mercury emission sources (Hylander, 2001). On the other hand, man-made mercury emissions are however still increasing in developing countries in contrast to industrialized countries (Hylander, 2001; Wang et al., 2000; Feng et al., in press). In countries such as China and India this is because of installation of chlor-alkali plants with Hg electrodes and increased burning of coal (United Nations, 1994), while in Brazil, China, Colombia, Ecuador, French Guyana, Guyana, Ghana, India, Peru, Siberia, Tanzania, Venezuela, the Philippines and other countries in southeast Asia it is due to gold extraction with Hg (Ebinghaus et al., 1999a; Ghose, 1994; Ikingura and Akagi, 1996; Malm, 1998).

Whereas data are relatively abundant to characterize the temporal distributions of airborne Hg in many regions of Europe and Northern America (e.g. Slemr and Scheel, 1998; Iverfeldt et al., 1995; Lee et al., 1998; Schroeder and Markes, 1994; Ames et al., 1998; Urba et al., 2000; Poissant, 2000), it is not the case for other regions including Asia. Recently, measurements of atmospheric mercury performed in Seoul, Korea (Kim and Kim, 2000, 2001) showed the TGM concentration in urban air is elevated comparing to the global background values which is believed to be from 1.5 to 2.0 ng m^{-3} (Lindqvist et al., 1991). China is believed to be an increasing atmospheric mercury emission source (Hylander, 2001), however, no comprehensive measurements of TGM in ambient air of China, to our knowledge, have been done yet. In this paper, we present results of the first comprehensive measurements of TGM at one site in Guiyang City, China.

2. Materials and methods

The provincial capital of Guizhou, Guiyang is classified as one of the most seriously polluted cities in China. Its climate represents a typical subtropical humid monsoon with an average annual temperature of 15 °C and a precipitation of 1100–1400 mm (The Government of Guizhou Province, 1995). Acid rain caused by coal combustion emissions is regarded as the main environmental burden. According to Statistical Bureau of Guizhou Province (1995), the daily average concentrations of SO_2 and total suspended particulate matter (TSP) in the air of Guiyang in 1992 were 468 and 395 $\mu g\, m^{-3}$, respectively, exceeding the national air quality standards, which are 100 and 300 $\mu g\, m^{-3}$ for SO_2 and TSP, respectively. Preliminary studies demonstrated that approximately 2.2 t Hg was emitted to the air from coal burning in Guiyang 1998 (Feng et al., in press). It is well known that coal produced in Guizhou has relatively larger mercury and sulfur concentration than that produced in other provinces in China (Feng and Hong, 1999; Feng et al., in press). The amount of coal burnt in Guiyang is predicted to increase due to the regional economic development, and therefore the impact of coal burning on atmospheric mercury pollution in this region needs scrutinizing.

A measurement site in Guiyang, where the Institute of Geochemistry, Chinese Academy of Sciences is located, was chosen for measurement of TGM in the air (Fig. 1). The study site is a dense residential area, and a number of industries (such as Guiyang Coal Fired Power Plant and Guizhou Cement Production Plant which are the largest single mercury emission point sources) are located southwest of the study site within 20 km. Two small boilers supplying hot water to public bathrooms are situated North and Northeast of the sample site within 300 m. The inlet air of the Teflon sampling tubing was located at 2 m above

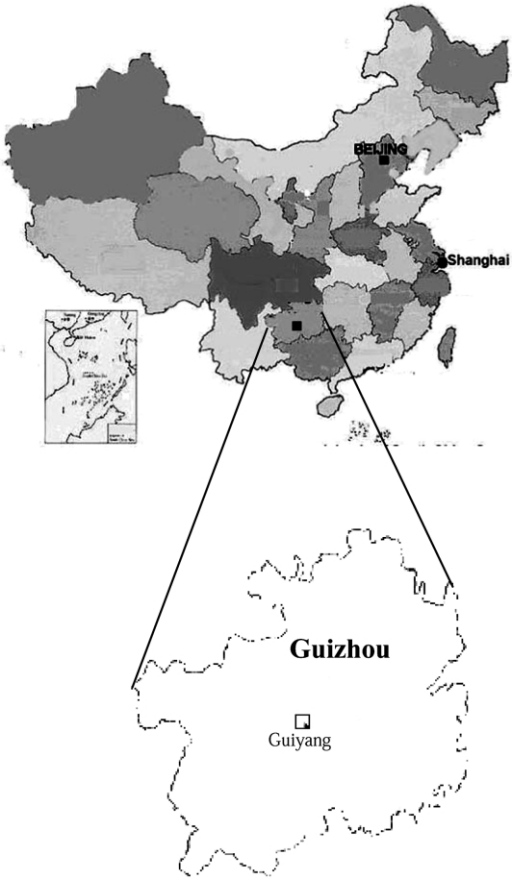

Fig. 1. The location of sampling site in Guiyang (■).

the roof of the State Key Laboratory of Environmental Geochemistry building.

Four measurement campaigns were performed April 19–30, 2000, February 26–March 14, 2001, June 26–July 20, 2001, October 9–November 22, 2001, respectively. In the first campaign carried out April 19–30, 2000, an automated mercury analyzer Gardis 1A (Urba et al., 1995) that used dual amalgamation and cold vapor atomic absorption spectrometry for the analysis of TGM after 10 min of sample collection was employed. For the rest of the three campaigns, measurements of the TGM concentration in the air was performed with an automatic Mercury Vapor Analyzer Tekran® 2537A. The Tekran® has two channels which trap vapor-phase mercury onto gold adsorbents. The two adsorbent cartridges thermally desorb alternately, after which Hg is detected using cold vapor atomic fluorescence spectroscopy, enabling a time resolution down to 5 min. Both instruments were calibrated by injecting a known amount of mercury vapor. These two instruments were recently inter-compared at two workshops, one at Mace Head, Ireland (Ebinghaus et al., 1999b) and another at Sasetta, Italy (Munthe et al., 2001), and good agreement was reported for the TGM concentrations measured by these two instruments. TGM consists of both Hg^0 and reactive gaseous mercury (RGM), and a previous study (Feng et al., in press) showed that RGM only constituted approximately 3.9% of TGM in air of Guiyang.

For the first and second measurement campaigns, meteorological data of hourly averaged air temperature, relative humidity, wind direction and speed were collected from a nearby weather station. For the last two campaigns, the meteorological data were measured using a portable weather station (GL300B Weather station-Global Logger IIIB from Global water, USA) at the sampling site with the same time resolution (5 min) with Tekran 2537A.

3. Results and discussion

3.1. Seasonal distribution patterns of TGM

Table 1 summarizes the TGM concentrations and the meteorological parameters determined during four comparative study periods. Four campaigns represented different seasons in Guiyang as we can see from the average air temperatures monitored during the sampling periods (Table 1). April 2000 campaign represented the spring season, whereas February to March, June to July and October to November 2001 campaigns represented winter, summer and autumn, respectively. TGM concentrations measured in the four measurement campaigns were significantly elevated compared to those reported using the similar instrumentation at a variety of locations in Europe and North America (e.g. Schroeder and Schneeberger, 1996; Ebinghaus et al., 1999a; Schroeder et al., 1998; Urba et al., 2000; Poissant, 2000; Lee et al., 1998;

Table 1
A statistic summary of the concentrations of gaseous elemental mercury (TGM) and the meteorological parameters measured during four campaigns

	TGM ($ng\ m^{-3}$)	Wind speed ($m\ s^{-1}$)	Temperature (°C)	RH (%)	Insolation ($W\ m^{-2}$)
April 19–30, 2000					
Average	8.85	3.78	15.3	85.5	
S.D.	5.50	1.64	3.2	11.4	
Median	8.15	4.00	14.8	87.0	
Min	3.57	0	10.7	47.0	
Max	146.75	9.00	27.2	99.0	
N	1933	237	255	251	
Geometric mean	8.56				
February 26–March 14, 2001					
Average	8.18	3.52	9.5	76.2	
S.D.	4.15	1.69	4.9	17.6	
Median	6.99	3.00	9.1	81.0	
Min	3.53	0	−0.1	24.0	
Max	112.34	9.00	24.8	99.0	
N	3926	504	504	503	
Geometric mean	7.46				
June 26–July 20, 2001 Summer, 2001					
Average	5.71	0.14	24.8	82.1	7.03
S.D.	2.26	0.42	4.6	16.0	10.30
Median	4.93	1.40	23.8	85.8	1.26
Min	1.70	0	16.9	36.3	0.32
Max	70.00	5.02	41.0	100.0	44.56
N	6501	8850	8850	8850	8850
Geometric mean	5.20				
October 9–November 22, 2001					
Average	9.14	0.13	15.3	82.8	4.07
S.D.	4.64	0.39	5.06	17.9	7.22
Median	7.97	0.14	15.5	87.6	0.55
Min	2.73	0	3.1	13.9	0.29
Max	129.80	3.74	27.7	100.0	41.33
N	8900	10 064	10 064	10 064	10 064
Geometric mean	8.33				

Slemr and Scheel, 1998), but were comparable with those measured in Kagoshima City, Japan which is influenced by volcano eruptions (Tomiyasu et al., 2000) and Seoul, Korea during the 1980s (Kim and Kim, 2000).

Fig. 2 depicts the high time resolved TGM distribution measured in the four campaigns. A large variability of the temporal TGM distribution pattern was observed as shown in Fig. 2, which is contrary to the observation from background measurements (Schroeder and Schneeberger, 1996; Ebinghaus et al., 1999b; Schroeder et al., 1998). The large temporal variability was also consistently observed at measurement site close to sources by a number of investigators (e.g. Kim and Kim 2000; Schmolke et al., 1999; Burk and Keeler, 1996). Frequency distributions of the high time resolved TGM data from four measurement campaigns are shown in Fig. 3. Clearly, TGM data from all measurement periods followed the typical log normal distribution pattern. For data set that follows log normal distribution pattern, the geometric mean represents objectively the average of the data set (Hu, 1991), and the geometric means of TGM from the four measurement campaigns are therefore listed in Table 1. A distinct seasonal

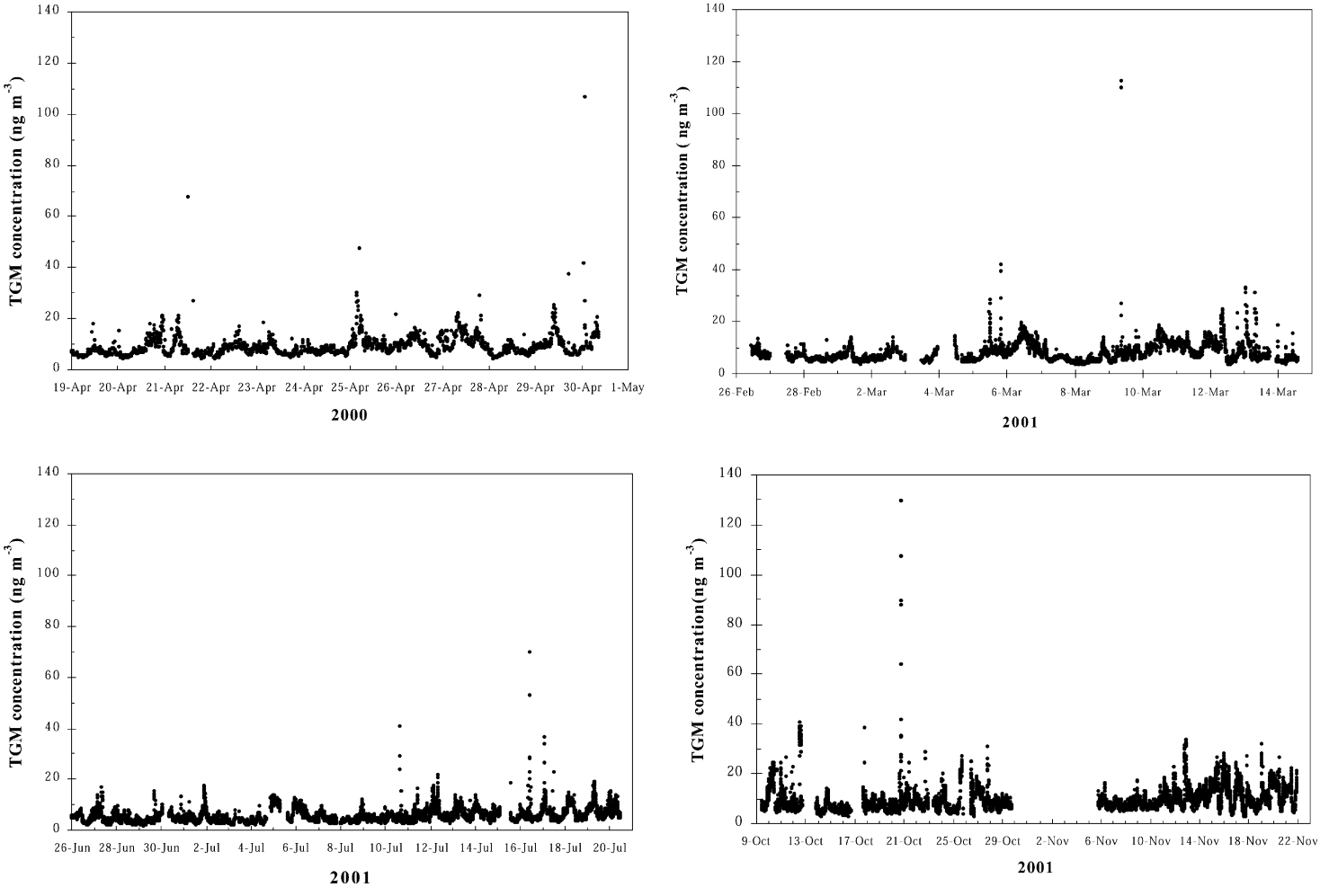

Fig. 2. Temporal variability of TGM in four sampling periods (10 min average values for April 2000 campaign and 5 min average for 2001 campaigns).

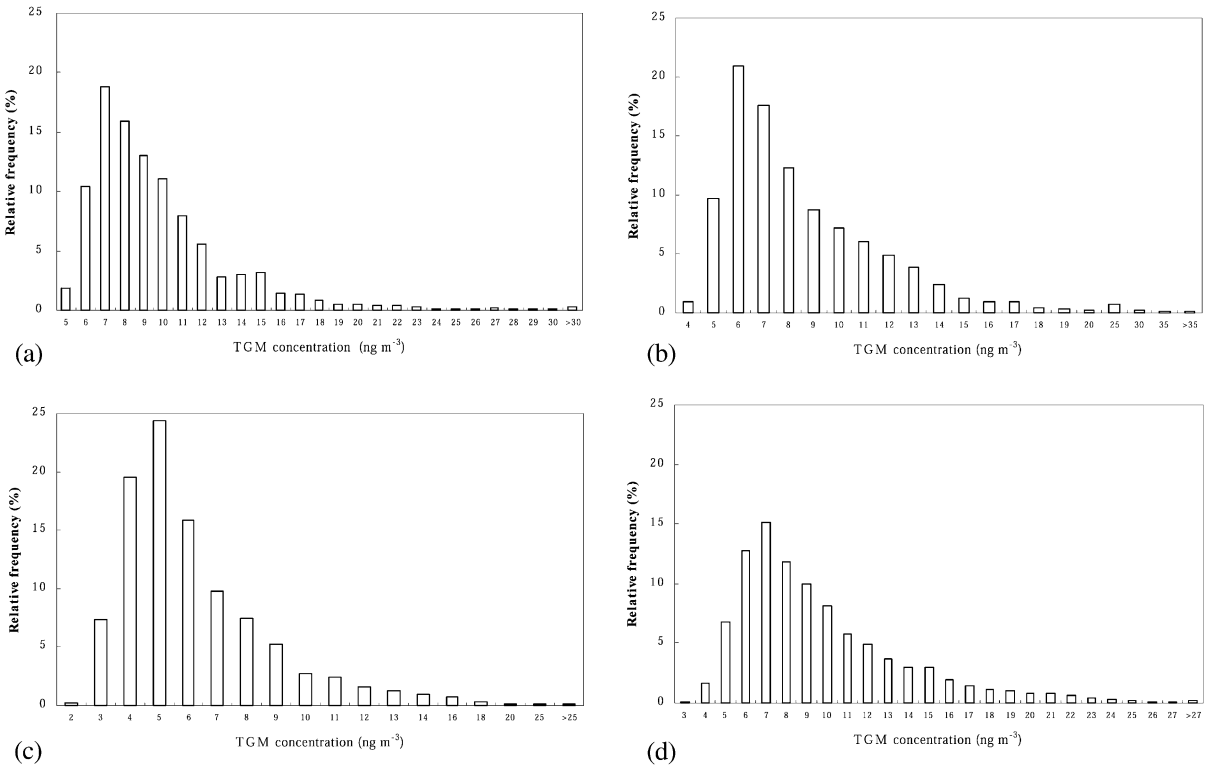

Fig. 3. Relative frequency plot of TGM distribution data measured from the sampling site in Guiyang during 4 campaigns. (a) Spring 2000 campaign; (b) winter 2001 campaign; (c) summer 2001 campaign; (d) autumn 2001 campaign.

distribution pattern of TGM was observed, and the TGM concentration levels are distinctively lower during summer than that during the other seasons (Fig. 4 and Table 1). The annual average TGM concentration based on the four measurement campaigns was 7.39 ng m^{-3}.

The major anthropogenic atmospheric mercury emission source in Guiyang is from coal combustion (Feng et al., in press), and coal is burnt both by industry utilities and domestic uses. The main domestic coal consumption which comprises approximately 25% total coal consumption is diverted to house heating during cold seasons from late autumn through winter to early spring, which could explain partly the differences of TGM among the seasons. Seasonal geometric mean TGM concentrations were: summer < winter < autumn < spring. Surprisingly, the average TGM concentration in winter is slightly lower than that in spring and autumn (the possible reasons will be given later in this section). Although TGM data from all sampling periods follow a log normal distribution implying that the regional transport processes may play a role in the observed pattern, we do strongly believe that local sources contribute more significantly to the observed TGM pattern because Guiyang City is the most polluted and industrialized area in the Province.

Fig. 5 shows the windrose pattern for the study periods, which was constructed by sorting all the hourly (spring 2000 and winter 2001) or the 5 min (summer and autumn 2001) measurements by 22.5° sectors, such that each sector represents a large number of measurements. Northeasterly, easterly and southwesterly winds are predominant in spring, autumn and winter measurement periods, and whereas southerly and southeasterly winds are dominant in summer measurement period. Wind

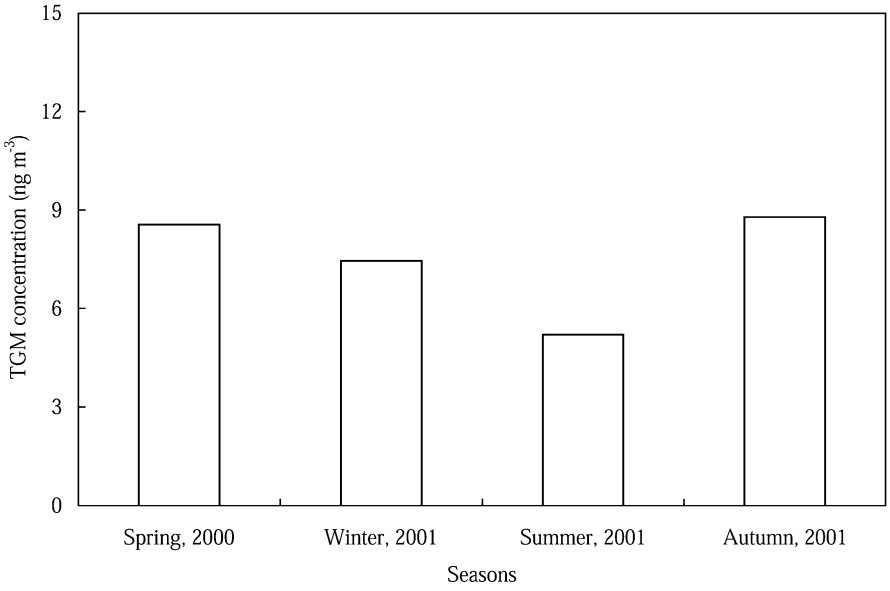

Fig. 4. Seasonal variation of the geometric mean of TGM values observed at the sampling site in Guiyang.

directional dependence of TGM is shown in Fig. 6. In summer 2001 campaign, northeasterly, easterly, southerly and southwesterly winds that are not the predominant wind directions carry significantly higher concentrations, which may decipher partly the low average TGM obtained. In summer, the major atmospheric mercury sources are from industries since generally no domestic coal consumption occurs during this period of time. From the wind directional dependence of TGM shown in Fig. 6c, it is distinctly shown that northerly, northeasterly and southwesterly winds carry significantly higher concentrations. A dense industrial area is located to the southwest of the sampling site within 20 km and two small boilers are situated at 300 m north and northeast of the research site. It is obvious that the industry area and the small boilers are atmospheric mercury sources. In winter 2001 campaign, TGM concentrations in the air were enhanced due to intensive domestic coal use for house heating and the impacts from the industry area and the small boilers were not obviously seen (Fig. 6b). Hence emissions from domestic coal burning were the primary atmospheric mercury in winter. In spring 2000 campaign, northeasterly, easterly and southwesterly winds that are from emission source areas are the dominant wind directions, which could partly explain the high average TGM concentration measured. It is evident that mercury concentrations were generally elevated comparing to summer campaign. Apart from the fact as mentioned above that still some domestic coal burning activities occurred during this season, rapid re-emissions of mercury that deposited in the surface in winter time could be also responsible for the high TGM measured. Earlier studies (Feng, 1997; Feng et al., in press) showed that RGM and particulate mercury concentrations in the air of Guiyang are generally elevated up to several hundreds pg m^{-3}, which could result in tremendous mercury deposition to the surface by both dry and wet processes. Numerous studies (e.g. Feng et al., 1996; Xiao et al., 1991; Poissant and Casimir, 1998; Schroeder et al., 1989; Kim et al., 1995) have demonstrated that surface temperature increase will significantly promote mercury emission rate from the surface soil. Therefore, mercury deposited and accumulated in the surface soil during winter season may re-emit rapidly back to the air during spring season, which results in much higher TGM concentrations in spring season than in winter. In autumn 2001 campaign, winds

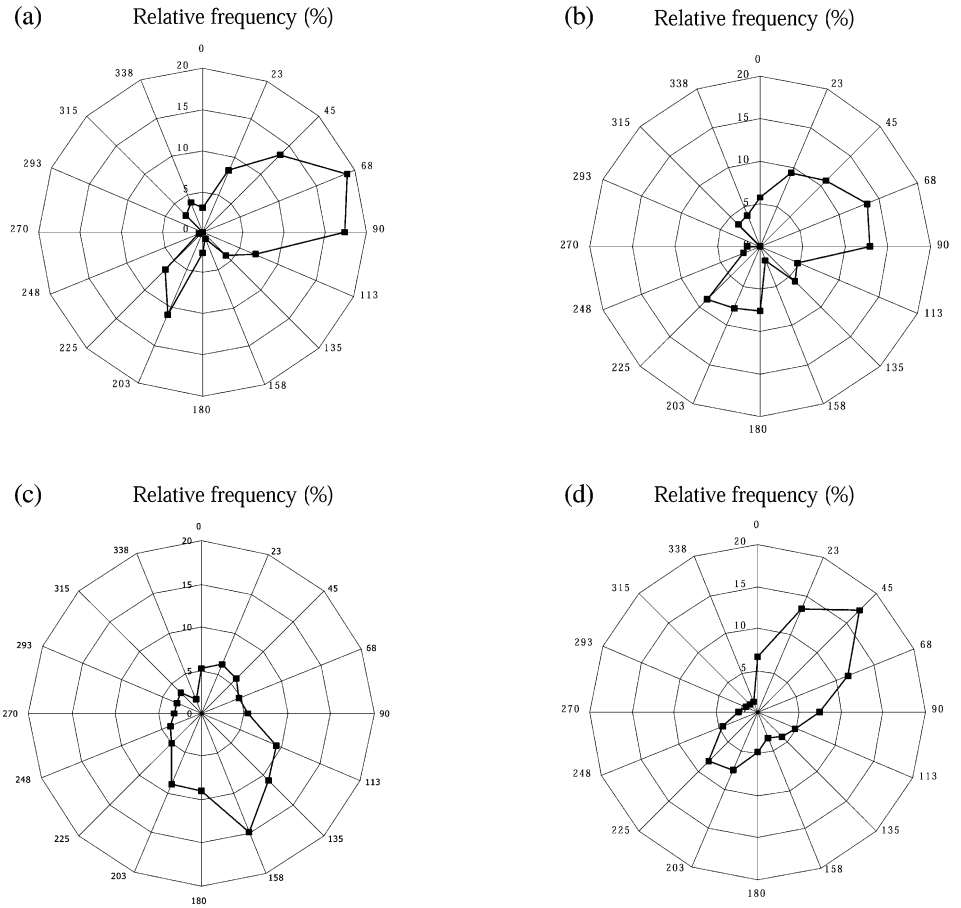

Fig. 5. Wind frequency roses during the four measurement campaigns. (a) Spring 2000 campaign; (b) winter 2001 campaign; (c) summer 2001 campaign; (d) autumn 2001 campaign.

from the small boilers which were the dominant wind directions carried significantly high TGM concentrations, but no clear influence from the industry area could be noticed. In autumn, burning of leaves is one of the main means to dispose of the falling leaves in Guiyang, which obviously is an important atmospheric mercury emission source. Meanwhile, some families started to heat their houses by burning coals during the sampling period. All these sources could definitely contribute to the high average TGM concentration observed in the sampling period.

3.2. Diurnal distribution patterns of TGM

The diurnal variability of TGM in different seasons is depicted in Fig. 7. In spring, it is apparently that TGM concentrations in daytime are higher than those in nighttime (Fig. 7a). If the spring 2000 results are compared on day/night basis, the daytime average TGM value of 9.53 $ng\,m^{-3}$ is approximately 17% larger than the nighttime average value of 8.17 $ng\,m^{-3}$. The distinct daily TGM distribution pattern further supports the hypothesis that re-emission of mercury from the surfacial soils of which the process is enhanced during daytime due to relative high temperatures may be one of the most important atmospheric sources in spring (Feng et al., 1996; Xiao et al., 1991; Kim et al., 1995). On the other hand, nocturnal deposition (Feng et al., 1996; Xiao et al., 1991) may also occur and explain the observed diurnal pattern.

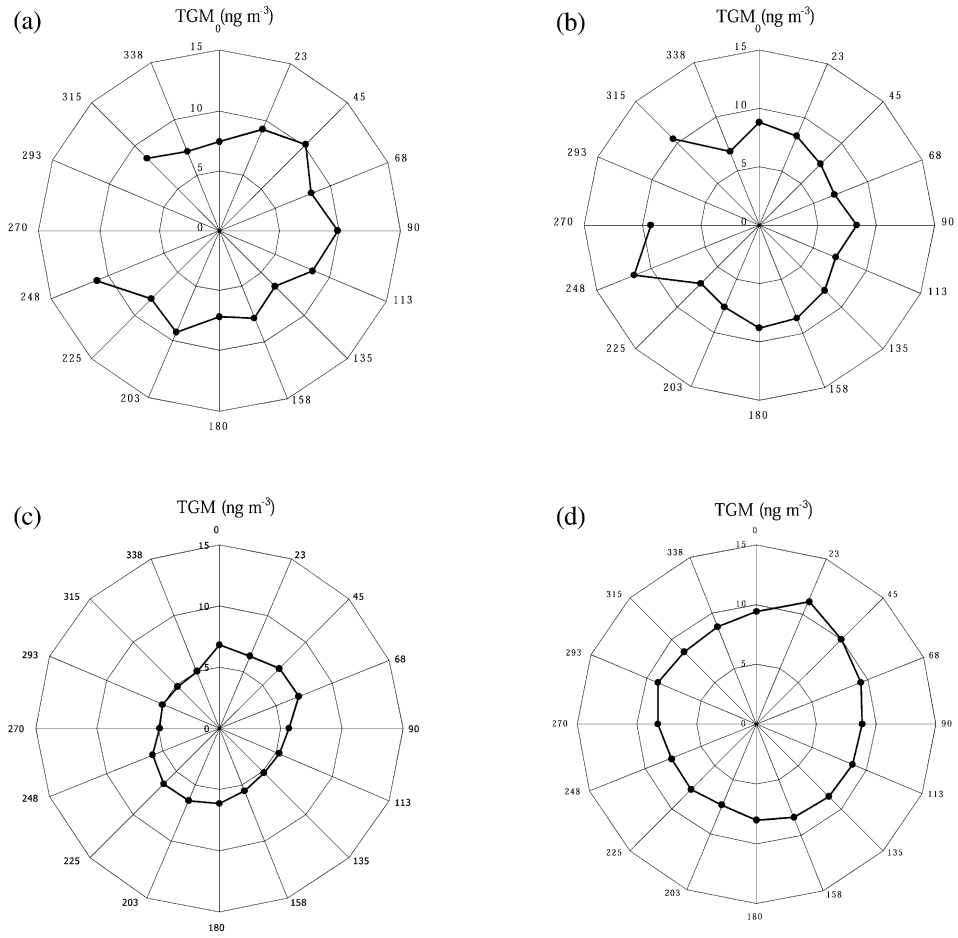

Fig. 6. Wind directional dependence of TGM data for four measurement campaigns. (a) Spring 2000 campaign; (b) winter 2001 campaign; (c) summer 2001 campaign; (d) autumn 2001 campaign.

In winter, two TGM peaks occurred in early morning and early evening, respectively (Fig. 7b), which corresponded to the two periods when people usually start their furnace to acquire heating for the houses. The notable feature of diurnal distribution pattern of TGM in winter again highlights that domestic coal burning is an important atmospheric mercury source. The average TGM concentration of daytime is 8.31 ng m^{-3}, which is slightly larger than that in nighttime which is 8.08 ng m^{-3}.

The diurnal distribution pattern of TGM in summer and autumn contrasted significantly to that in spring and winter, as the average day TGM concentration is notably smaller than that in nighttime (Fig. 7c and d). The same diurnal distribution pattern was also reported from a rural measurement in England through seasons (Lee et al., 1998). It is generally believed that on most nights shallow nocturnal boundary layers form, trapping TGM near the surface, while thermal mixing during the day increases the boundary-layer depth thus diluting the concentrations (Lee et al., 1998). The major atmospheric mercury source in summer is from industry emissions, and the emission rate is relatively steady. The above mentioned mechanism therefore could be the only explanation to the fact that the average nighttime TGM 6.29 ng m^{-3} was elevated in comparison with that of daytime 5.28 ng m^{-3} in summer campaign. While emission from

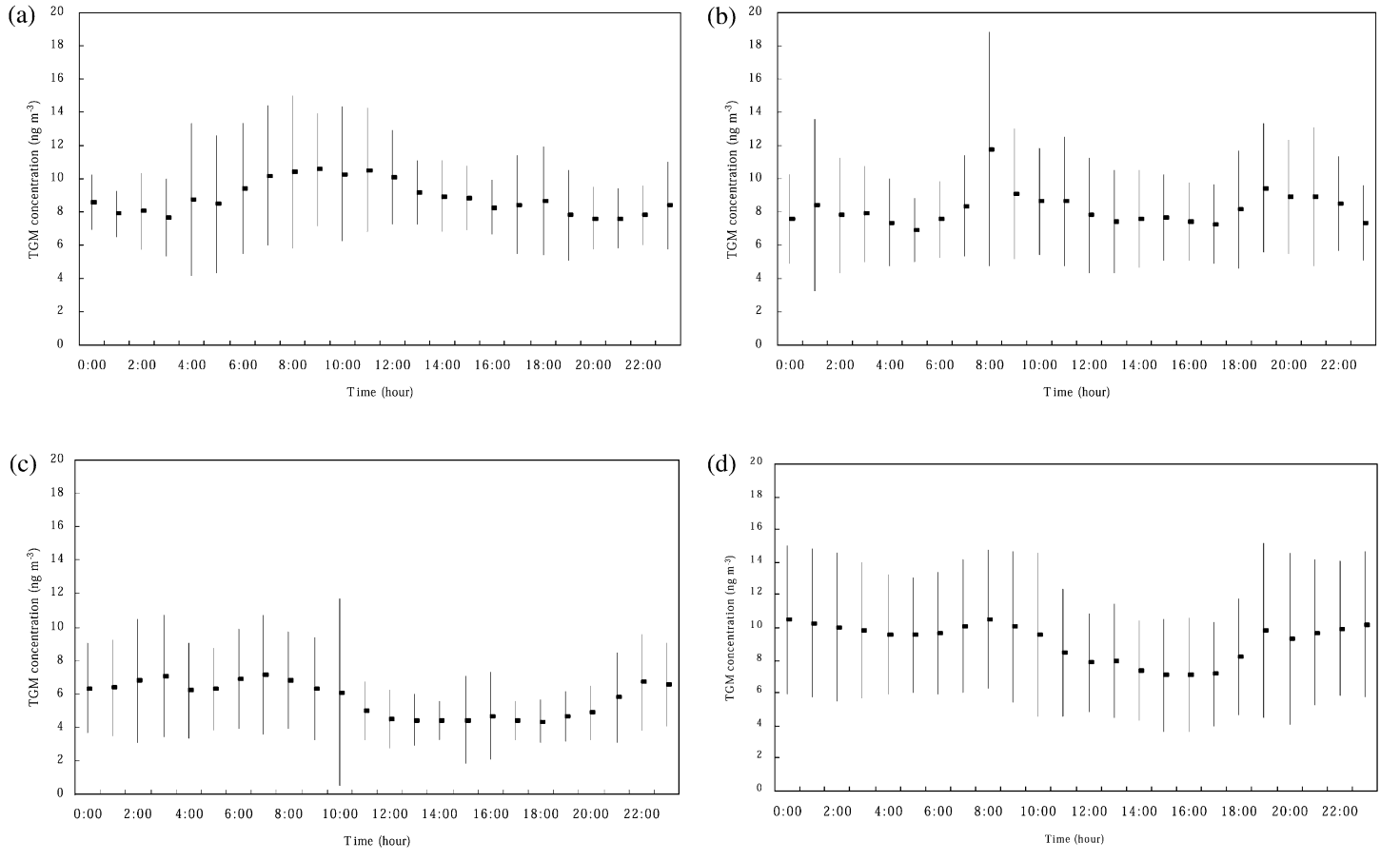

Fig. 7. Diurnal variability of TGM at four measurement campaigns. (a) Spring 2000 campaign; (b) winter 2001 campaign; (c) summer 2001 campaign; (d) autumn 2001 campaign.

burning leaves is one of the most significant atmospheric sources in autumn, and the process usually starts after 18:00 and lasts for a few hours. Obviously, burning leaves could be another cause to the elevated TGM concentration in nighttime (9.88 ng m^{-3}) compared to that in daytime (8.46 ng m^{-3}) during autumn season.

4. Conclusions

The average TGM concentration in Guiyang based on the four measurement campaigns representing different seasons is 7.39 ng m^{-3}, which is significantly elevated comparing to global background of approximately 1.5–2.0 ng m^{-3}. A distinctly seasonal distribution pattern of TGM was observed, and the seasonal geometric mean TGM concentrations were: summer < winter < autumn < spring. The major anthropogenic atmospheric mercury emission sources differed significantly among seasons, which explained the seasonal variability of TGM level. The daytime TGM concentrations were larger than that of nighttime in spring and winter seasons, while in summer and autumn the opposite daily TGM distribution pattern was observed. Obviously, a detailed mercury emission inventory in this area is needed to further elucidate TGM distribution characteristics, to evaluate objectively the ecological consequences, and to find out an efficient solution to prevent atmospheric mercury emissions.

Acknowledgments

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