Temporal variation of total gaseous mercury in the air of Guiyang, China

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Received 17 September 2003; revised 17 November 2003; accepted 1 December 2003; published 5 February 2004.

[1] Total gaseous mercury (TGM) concentrations in ambient air were monitored at an urban site in Guiyang from 23 November 2001 to 30 November 2002 using a high temporal resolution 5 min mercury vapor analyzer (Tekran 2537A). TGM concentrations follow lognormal frequency distribution pattern, and the mean TGM concentration at the measurement site is 8.40 ng m^{-3} on the basis of 1 year observation. TGM concentrations in Guiyang are significantly elevated compared to the continental global background values. Coal combustion from both industrial and domestic uses is estimated to be the primary atmospheric source. A seasonal distribution pattern of TGM with a descending order of winter, spring, fall, and summer was observed. The highest TGM concentration in winter is attributed to household heating using coal. A consistent diurnal distribution pattern of TGM was obtained among all seasons, and the nighttime TGM concentration is elevated compared to the daytime values. Meteorological conditions are responsible for the formation of diurnal TGM distribution pattern. Tremendous efforts are needed to reduce mercury emissions from coal combustion to decrease TGM concentrations in Guiyang. *INDEX TERMS:* 0330 Atmospheric Composition and Structure: Geochemical cycles; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0399 Atmospheric Composition and Structure: General or miscellaneous; *KEYWORDS:* mercury, atmosphere, urban, anthropogenic, diurnal pattern

Citation: Feng, X., L. Shang, S. Wang, S. Tang, and W. Zheng (2004), Temporal variation of total gaseous mercury in the air of Guiyang, China, *J. Geophys. Res.*, *109*, D03303, doi:10.1029/2003JD004159.

1. Introduction

[2] Because of its high volatility, low chemical reactivity, and low solubility in water, Hg^0, which makes up more than 95% of total gaseous mercury (TGM), has a residence time in the atmosphere from 0.5 to 2 yr. This means Hg^0 can be transported far beyond the regions where it is emitted and eventually becomes well-mixed hemispherically [*Schroeder and Munthe*, 1998; *Lindqvist and Rodhe*, 1985]. Long-range transport of atmospheric Hg, its deposition, bioaccumulation and the enrichment of highly toxic methylmercury compounds in aquatic food chain pose a serious environmental problem even in remote areas like the Arctic and Antarctic [e.g., *Jewett et al.*, 2003; *Temme et al.*, 2003a]. Numerous efforts have therefore devoted to curbing the use of mercury and consequently to reducing its emissions into the atmosphere in developed countries [e.g., *Organisation for Economic Co-operation and Development*, 1994].

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[3] Measurements of TGM over the Atlantic Ocean in the years 1977–1980, 1990, and 1994 suggest a global increase between 1977 and 1990 and a decrease after 1990 [*Slemr and Langer*, 1992; *Slemr et al.*, 1995; *Slemr*, 1996]. Long-term measurements from two European sites (one from Sweden and the other from Germany) also reveal a decreasing trend in TGM concentration since 1990 and possibly a few years before [*Slemr et al.*, 1995; *Iverfeldt et al.*, 1995; *Slemr and Scheel*, 1998]. More recent measurements started from 1995 at Alert in the Canadian Arctic and at Mace Head in the Irish west coast, however, demonstrate that both sites do not show a specific trend of TGM concentration over time, and that the long-term average TGM concentration is around 1.5 to 1.7 ng m^{-3} [*Schroeder et al.*, 2001; *Ebinghaus et al.*, 2002a]. The latest study [*Slemr et al.*, 2003] showed that atmospheric mercury concentrations increased in the late 1970s to a peak in the 1980s, then decreased to a minimum at about 1996, and have been nearly constant since. The trend of worldwide anthropogenic mercury emission rate is difficult to predict because the reduction of anthropogenic emissions in developed countries that is

Figure 1. The location of the sampling site in Guiyang (solid square).

well estimated and the increase of man-made emissions in developing countries that is poorly studied occur simultaneously [*Hylander*, 2001; *Wang et al.*, 2000; *Feng et al.*, 2002; *Ebinghaus et al.*, 1999a; *Ghose*, 1994; *Ikingura and Akagi*, 1996; *Malm*, 1998; *Pacyna and Pacyna*, 2002].

[4] Concentrations of TGM in the air at remote continental and mid-oceanic locations in both hemispheres have been well documented [*Lindqvist et al.*, 1991; *Fitzgerald et al.*, 1991; *Mason et al.*, 1992; *Slemr et al.*, 1995; *Slemr and Scheel*, 1998; *Schroeder et al.*, 1998; *Ebinghaus et al.*, 2002a, 2002b; *Lindberg et al.*, 2002; *Lamborg et al.*, 1999; *Lee et al.*, 1998; *Poissant.*, 2000]. A small but definite interhemispheric gradient in TGM that reflects global transport from Northern hemispheric sources is observed [*Ames et al.*, 1998; *Urba et al.*, 2000; *Lamborg et al.*, 2002; *Ebinghaus et al.*, 2002a, 2002b; *Baker et al.*, 2002; *Lindberg et al.*, 2002; *Schroeder et al.*, 1998; *Temme et al.*, 2003b]. The background level of TGM in South hemisphere is about 1.1 to 1.4 ng m^{-3} [*Ebinghaus et al.*, 2002b; *Baker et al.*, 2002], while that in North hemisphere is believed to be 1.5 to 2.0 ng m^{-3} [*Ebinghaus et al.*, 2002b; *Schroeder et al.*, 2001; *Lamborg et al.*, 2002]. On the contrary, TGM concentrations in urban air, especially in developing countries are however understudied. Atmospheric mercury monitoring in urban air in developing countries is of crucial importance because man-made mercury emissions are not well controlled yet. The current TGM concentrations in urban air in developing countries might represent TGM concentration in urban air of the developed countries when the Industrial Revolution started. Recent studies conducted in Seoul, Korea, and in Beijing and Guiyang, China depicted much elevated TGM concentrations compared to the global background values [*Kim and Kim*, 2000, 2001a, 2001b, 2002; *Liu et al.*, 2002; *Feng et al.*, 2002]. China is regarded as one of the largest anthropogenic Hg emission countries. Unfortunately however, TGM concentrations in ambient air are only been measured at few locations in China. Here we reported the result of a one-year continuous TGM measurement from a single sampling location during November 2001 to November 2002.

2. Experimental

[5] Guiyang, the Capital of Guizhou Province, is classified as one of the most seriously polluted cities in China. SO_2 and particulate matter emitted mainly from coal combustion are the main airborne pollutants in Guiyang. According to the *Statistical Bureau of Guizhou Province* [1995], the daily average concentrations of SO_2 and total particulate matter (TPM) in the air of Guiyang in 1992 were 468 (163.8 ppb) and 395 $\mu g\ m^{-3}$, respectively, exceeding the national air quality standards, which are 100 (35 ppb) and 300 $\mu g\ m^{-3}$ for SO_2 and TPM, respectively. Preliminary studies demonstrated that about 2.2 t Hg were emitted to the air from coal burning in Guiyang in 1998 [*Feng et al.*, 2002]. It is well known that coal produced in Guizhou has relatively high mercury and sulfur concentration than that produced in other provinces in China [*Feng and Hong*, 1999; *Feng et al.*, 2002]. The amount of coal burnt in Guiyang is expected to increase with the regional economic development, and therefore the impact of coal burning on atmospheric mercury pollution in this region needs scrutinizing.

[6] A measurement site selected for TGM measurement is located at the Institute of Geochemistry, Chinese Academy of Sciences, Guiyang (Figure 1) . The site characteristics and all the relevant information have been described elsewhere [*Feng et al.*, 2002]. The inlet air of the Teflon sampling tubing was located at 2 m above the roof of the State Key Laboratory of Environmental Geochemistry building. Continuous measurements of TGM concentrations with 5 min time resolution have been carried out between 23 November 2001 and 30 November 2002 by using the automated atomic fluorescence (AFS) mercury vapor analyzer, Tekran 2537A [*Schroeder et al.*, 1995a]. The instrument features two cartridges. While one is adsorbing mercury during sampling period, the other is being desorbed thermally and analyzed subsequently for TGM. The functions of each cartridge are then reversed, allowing continuous sampling of the incoming air stream. A 45 mm diameter Teflon prefilter (pore size 0.45 μm) protects the sampling cartridges against contamination of airborne particulate matters. The amalgamated mercury is thermally desorbed into an Argon carrier gas stream and analyzed using an internal AFS detector. The instrument is calibrated on a routine base (twice a week) by injecting a known amount of mercury vapor.

[7] The analytical uncertainty of the Tekran 2537A has been assessed through comparison with other automated

Table 1. Results of TGM Concentrations Measured With Tekran 2537A and Manual Gold Trap Methods[a]

	Mean TGM Concentration, ng m^{-3}		
Sampling Period	Tekran 2537A	Manual Method	Percent Difference
20–21 Feb. 2001	10.25	10.78	−5.04%
21–22 Feb. 2001	15.37	14.95	2.78%
23–24 Feb. 2001	9.78	10.23	−4.50%
26–27 June 2001	5.79	5.50	5.14%
27–28 June 2001	11.25	12.23	−8.35%
23–24 March 2003	13.56	14.12	−4.04%
24–25 March 2003	14.35	14.56	−1.45%

[a]Sampling location in Institute of Geochemistry, Chinese Acedamy of Sciences, Guiyang.

mercury vapor analyzer (i.e., Gardis 1A) and with commonly used manual methods at an urban/industrial site in Windsor, Ontario, Canada [*Schroeder et al.*, 1995b], at a remote site in Tuscany, Italy [*Munthe et al.*, 2001] as well as at Mace Head, Ireland [*Ebinghaus et al.*, 1999b]. The results showed good agreement between the different techniques. We also performed comparison between the Tekran 2537A and manual gold trap methods at the sampling site. The manual gold trap method is based on gold trap amalgamation and subsequent analysis using CVAFS [*Brosset*, 1987]. TGM in ambient air is collected on a gold trap, which consists of a quartz tube (6mm inner diameter and 10 cm in length) filled with gold coated quartz chips. The sampling flow rate is set at 300 ml min^{-1}, and the sampling time is 24h. The 5-min interval samples from Tekran 2537A were averaged over 24h period, and the results were compared with the manual gold trap samples. The comparison results were tabulated in Table 1. The results in Table 1 depict a good agreement between the mean 24h TGM concentrations measured with the two different methods and validate Tekran's field operating procedure. On the basis of 3 times of standard deviation when Tekran 2537A were sampling zero air (air without Hg), a detection limit of the order of 0.3 ng m^{-3} can be achieved under these conditions. The meteorological parameters such as wind speed, wind direction, air temperature, relative humidity and solar irradiation were measured using a portable weather

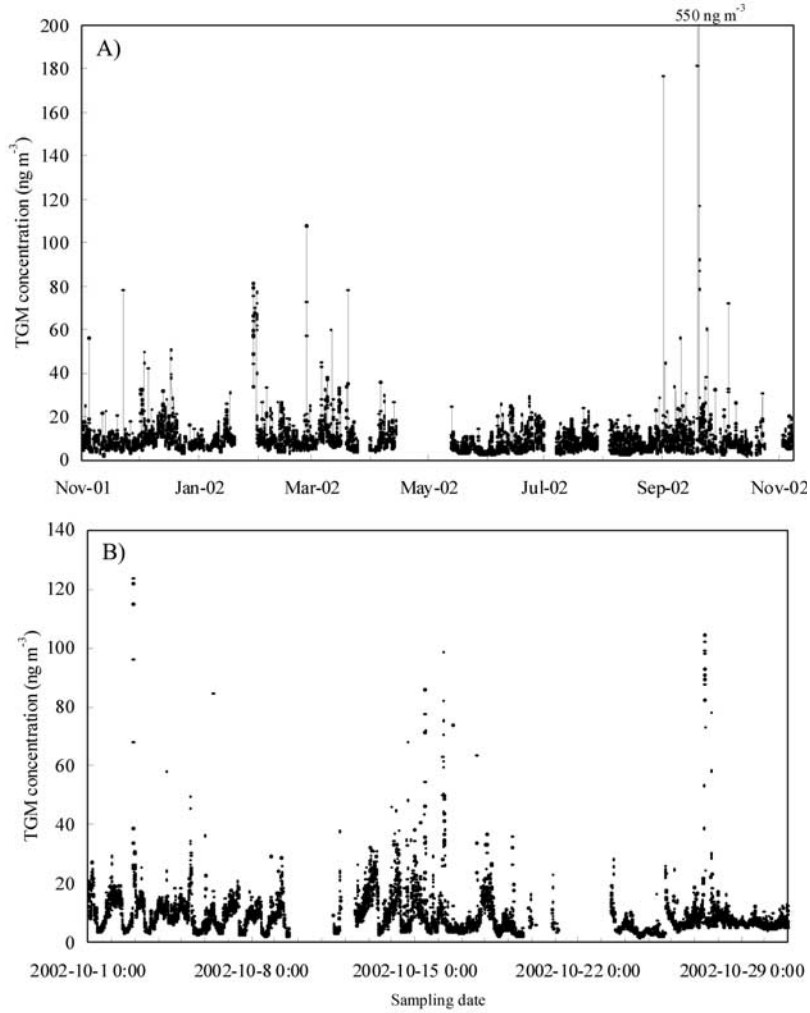

Figure 2. Total gaseous mercury (TGM) concentration in the air of Guiyang, China. (a) Hourly averaged TGM concentrations during the whole study period from 23 November 2001 to 30 November 2002. (b) TGM concentrations in October 2002 with a sampling time of 5 min.

Table 2. The Statistical Summary of High-Time Resolution (5 min) TGM Data

	Original Data					Mean With 3SD Removed			
	Minimum	Maximum	Mean	SD	n	Median	Mean	SD	n
Spring	1.90	405.63	8.50	7.46	14223	6.60	7.97	2.03	14017
Summer	1.53	127.69	7.51	4.80	21155	6.20	7.16	3.89	20796
Fall	1.08	632.07	8.20	9.58	21867	6.76	7.86	4.31	21777
Winter	2.04	407.97	10.54	10.26	20296	8.24	9.54	5.16	19955
All-data	1.08	632.07	8.68	8.44	77541	6.95	8.40	4.87	76545

station (GL300B Weather station, Global water, USA) at the sampling site with the same time resolution (5 min) as Tekran 2537A.

3. Results and Discussion

3.1. Overall TGM Distribution Characteristics in the Study Site

[8] In this study we intended to monitor TGM concentrations on routine basis; however, interruptions were inevitable for certain periods due either to the instrument maintenance or to the employment of the analytical systems for other study purposes. The longest periods which we were unable to retrieve data include the following 6 terms: (1) 11–19 February 2002; (2) 17–21 April 2002; (3) 7 May to 3 June 2002; (4) 23–28 July 2002; (5) 20–25 August 2002; and (6) 15–24 November 2002. Except for those breaking periods, we collected a total of 77,541 individual data points from our Hg measurements. The hourly averaged TGM concentrations over the sampling periods are depicted in Figure2a. The lowest hourly averaged TGM concentration is 1.6 ng m^{-3} whereas the highest one is 550 ng m^{-3}. The hourly averaged concentration values show a short-term variability on the scale of a few hours, which may be caused by local sources. In order to see more clearly the short term variability of TGM, we present TGM concentration measured in October 2002 with a sampling time of 5 min in Figure2b. The data sets were screened initially by such criteria as mean + 3SD (standard deviation) to objectively reduce the bias induced from a few extreme high values. The statistics result for the data collected for the whole study period is summarized in Table 2.

[9] A lognormal frequency distribution was observed for the hourly averaged TGM data as shown in Figure 3. This implied that local anthropogenic mercury emissions had significant impacts on the distribution of TGM in the air at the sampling site. The average TGM concentration over the whole one year sampling period is 8.40 ng m^{-3}. It is obvious that our TGM concentrations are significantly elevated compared to the global continental background values.

[10] Winds during the whole sampling period are from the NE, ENE, and E (Figure 4a). TGM concentrations in air from any directions are significantly higher than the background values (Figure 4b), implying that the regional background TGM concentrations in the atmosphere are elevated. Our preliminary measurements at several rural sampling sites in Guiyang showed elevated TGM concentrations as well [e.g., *Feng et al.*, 2003, unpublished data, 2003]. Obviously anthropogenic sources play a predominant role on the distribution of TGM in the air. Coal combustion is the primary man-made source of Hg in Guiyang and Guizhou Province, and currently of 8.3 t Hg emitted annually to the air from coal combustion in Guizhou, 2.2 t is emitted in Guiyang [*Feng et al.*, 2002].

[11] It is noted that winds from 0 to 180°Carry more TGM than those from 180–360° (Figure 4b). It is certain that Guiyang Steel Plant and an Organic Chemical Plant situated in about 2 km southeast and 2 km east of the sampling site contribute to high TGM concentrations. Meanwhile 3 small boilers supplying hot water to public bathhouses located within 1km northeast and east of the study site may as well be sources of mercury in air from those directions. Guiyang Coal Fired Power Plant (GCFPP)

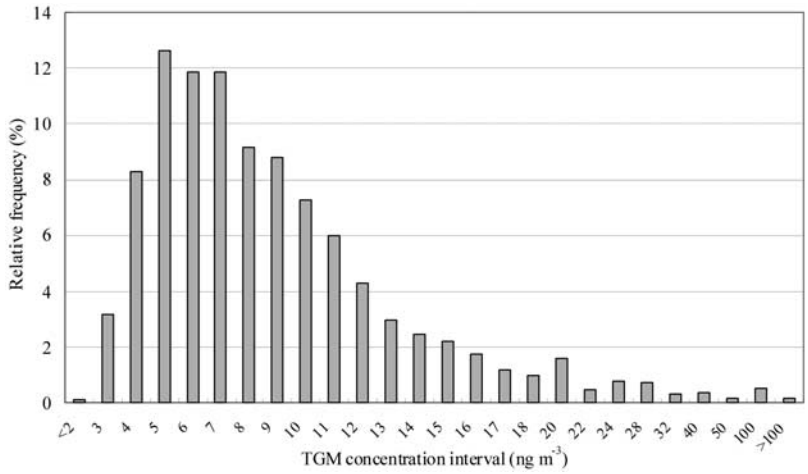

Figure 3. Frequency distribution of TGM measurements covered the whole sampling period. The calculation is based on the original data set without bias reduction.

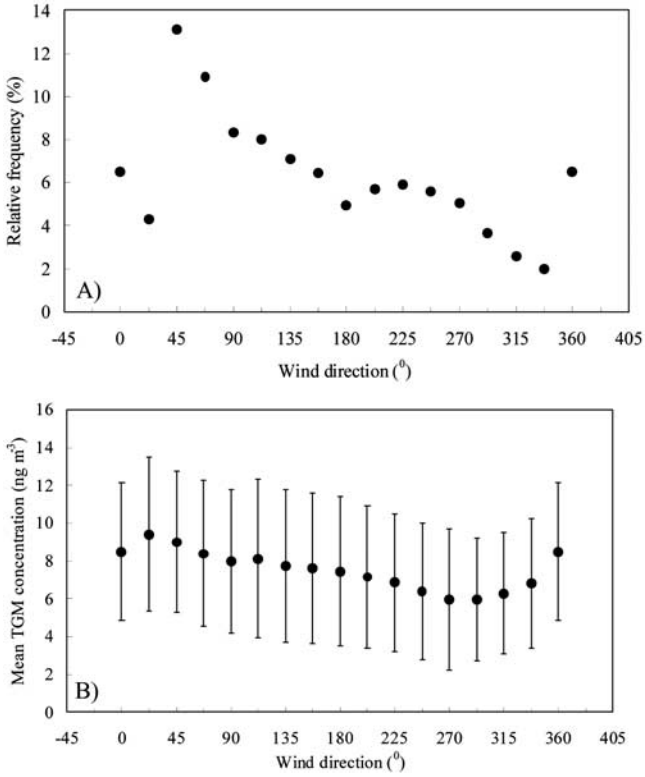

Figure 4. (a) Wind frequency roses covered the whole sampling period and (b) wind directional dependence of TGM data for the whole sampling period.

and Guizhou Cement Plant (GCP) situated within 20 km southwest of the study site are the largest single mercury emission sources, but it seems that they are less important on the distribution of TGM than those small local sources.

3.2. Seasonal Distribution Patterns of TGM

[12] TGM data set was subdivided into their respective seasons defined according to the solstice dates (21 March, 21 June, 21 September, and 21 December) [*Poissant*, 2000], and the seasonal statistical summary of TGM is listed in Table 2 and ambient air temperatures for the different seasons are presented in Table 3. The seasonal mean TGM values decreased on the descending order: winter, spring, fall, and summer. The differences among the seasonal averages are quite substantial and the difference between average for winter and summer is 2.38 ng m^{-3}. This difference corresponds to approximately 28% when referred to the overall mean TGM concentration of 8.40 ng m^{-3}.

[13] The same type of seasonal variation has also been observed by *Slemr* [1996] at the Wank Summit, by *Ebinghaus et al.* [2002a] at Mace Head, Irish west coast, by *Poissant* [2000] in Quebec, and by *Brosset* [1987] in Sweden. This type of seasonal variation is explained by *Slemr* [1996]. The seasonal variation, with summer minimums observed at the Wank Mountain, and in Sweden [*Brosset*, 1982], is characteristic of the majority of trace gases of which almost all are removed from the atmosphere by oxidation processes [*Warneck*, 1988]. The major oxidation species in the troposphere is the OH radical which has a pronounced seasonal cycle at middle and higher latitudes [*Warneck*, 1988]. Higher OH concentrations in summer lead to faster removal by oxidation and to a summer minimum in pollutant concentrations. On the other hand, *Ebinghuas et al.* [2002a] argued that the seasonal trend observed in Mace Head data may be due to the cycle in the burning of fossil fuels. Fossil fuel combustion for domestic heating in the greatest in winter and spring period. *Weiss-Penzias et al.* [2003], however, showed surprising losses of Hg^0 during local pollution event at a marine boundary layer sampling site. The third explanation is that a seasonal cycle in the deposition of mercury has also been observed in many studies [*Jensen and Iverfeldt*, 1994]. Maximum washout occurs in summer, and may result from greater oxidation of Hg^0 to more soluble species. It is possible that the seasonal trend observed in Guiyang data may be primarily due to the cycle in the burning of coal. A different seasonal cycle in TGM concentration measurements with a summer maximum was observed at a rural site in Tennessee by *Lindberg et al.* [1992]. This is probably due to the influence of local mercury sources and their dependence on air and soil temperature, with higher summer temperatures increasing mercury fluxes [*Lindberg et al.*, 1992].

[14] To offer better insights into the seasonal distribution characteristics of TGM, the monthly mean of TGM at the sampling site is computed and shown in Figure 5. It is quite evident that the average TGM concentrations in cold months (December, January, February, and March) are significantly elevated compared to those in the rest months excluding May and October. TGM measurement was only performed 6 days in May 2002, the average TGM concentration may not be representative and hence can be excluded from the discussion. The high average TGM concentration in October could be attributed to burning of tree leaves as discussed below. The mean TGM concentration for the cold months is 9.30 ng m^{-3}, whereas that for the counterpart is 7.37 ng m^{-3}; this shows that mean TGM concentrations in cold seasons are 26% higher than that in the counterpart. From December to March, household heating increase. Generally 90% of coal consumption is related to industrial activities such as power generation and smelting industries, while 10% of them is diverted to residential heating in China [*National Statistic Bureau*, 1999]. In 2001, 5.1 million ton coal was consumed in Guiyang, and 0.51 million ton coal was used for domestic heating [*Guiyang Statistic Agency*, 2002]. Assuming that coal consumption from industrial uses is evenly distributed in each month, and that the rest of coal consumption occurred during cold months, a monthly increasing rate of 33.3% in coal consumption during cold months can be calculated. Anthracite and bituminous coal are two major types of coal used in Guiyang, and the average mercury concentration in these coals is 0.53 mg kg^{-1}. Mercury in coal will be vaporized at high temperature, and mercury emission rate depends strongly on combustion techniques, but not on coal types

Table 3. Statistical Summary of Ambient Temperature Measured Every 5 min

	Minimum, °C	Maximum, °C	Mean, °C	SD	n
Spring	−19.0	40.8	10.2	16.6	15175
Summer	11.1	49.9	23.3	5.9	23488
Fall	−0.3	38.6	9.7	7.0	12017
Winter	−23.0	28.6	7.0	8.5	19598

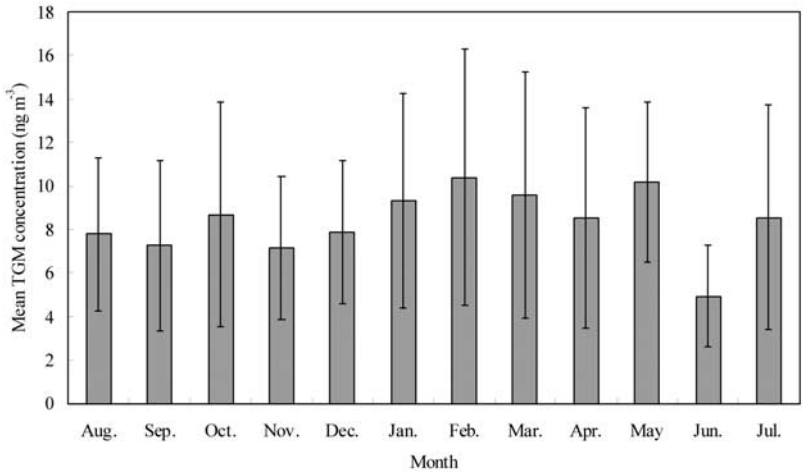

Figure 5. Seasonal variations based on monthly means of TGM concentrations between November 2001 and November 2002 in Guiyang, China.

[*Clark and Sloss*, 1992]. Generally small-scale boilers are utilized for domestic heating in Guiyang. The behavior of mercury in coal combustion boilers is poorly studied in China. Our study showed that mercury emission rates are relatively low from small boilers due to insufficient combustion compared to large-scale boilers [*Tang et al.*, 2003]. About 40% mercury in coal was recovered in bottom ash in this kind of small boilers, while only 1% was distributed in bottom ash in large scale boilers [*Meij et al.*, 2002]. On the other hand, no flue gas cleaning devices are installed in the small boilers in Guiyang, while for those large scale industrial boilers at least wet flue gas precipitators (WFGP) which could remove 80% particulate and part SO_2 are installed. It is demonstrated that the WFGP device could at least remove 25% of mercury entering the boiler in coal [*Tang et al.*, 2003]. In the aggregate, mercury emission rate into the air from small boilers in Guiyang is about 16% less than the large boilers. Assuming that the average mercury concentrations in coals used for both industrial boilers and small scale boiler are the same because these coals are produced from the same coal basins in Guizhou, it can be calculated that 33% more coal consumption due to domestic heating will result in 28% more mercury emissions from coal combustion in Guiyang as a whole. This number is quite consistent with the observed TGM increase rate (26%) in the air of our sampling site. It is therefore quite convincing that coal uses for household heating are the main cause of high TGM concentration observed in winter. Moreover, it highlights that coal combustion is the major anthropogenic mercury emission source in Guiyang. From Figure2a, we can see that more frequent short-term variability occurred in the cold months than in warm months, which again suggests the local mercury emissions from household heating occurred during winter. From middle September to the end of October, short-term variability occurred frequently as well. This is a response to burning of leaves that is the major way to dispose of tree leaves in fall in Guiyang. Though no research is performed on mercury emission from burning of tree leaves in Guiyang, it is already demonstrated that burning of biomass releases a significant atmospheric emission source [*Friedli et al.*, 2003]. More research is needed to quantify mercury emission rate from this source in Guiyang.

3.3. Diurnal Variation of TGM Concentration

[15] Figure 6 depicts diurnal variation of TGM concentration for both all and seasonally divided data sets. Results of this analysis clearly demonstrate the pattern for enhanced TGM concentrations during nighttime relative to daytime. This result is comparable to those of *Lee et al.* [1998] for a rural region of England, and to those of *Kim and Kim* [2001a] for an urban site in Seoul, Korea. TGM concentration differences between daytime and nighttime are more significant in summer and fall than those in spring and winter (Figure 6).

[16] The nighttime dominance of TGM is generally attributed to the fact that on most nights, shallow nocturnal boundary layers form to trap TGM near the surface. On the other hand, thermal mixing during the daytime increases the boundary layer depth, thus diluting TGM concentrations [*Lee et al.*, 1998]. Diurnal variation of wind speed for both all and seasonally divided data sets is also shown in Figure6. The results show the existence of strong negative correlations between TGM concentration and wind speed. The correlation coefficient between TGM concentration and wind speed (r) for all data set is -0.88 ($p < 0.01$), while that for spring, summer, fall and winter data sets are -0.81 ($p < 0.01$), -0.54 ($p < 0.01$), -0.88 ($p < 0.01$), and -0.78 ($p < 0.01$), respectively. Our measurement site is surrounded by small mountains, which results in a certain wind directions (NE, ENE, and E) being predominant. We believe that the consistent diurnal wind speed pattern in all seasons is responsible for the consistent TGM diurnal distribution obtained at our sampling site.

4. Conclusions

[17] Guiyang is well-known as one of the major cities in China where acid depositions occur frequently due to emissions from coal combustion. The concentration level of Hg from Guiyang is significantly elevated compared to the continental global background values. TGM concentrations from our study appear to follow log normal frequency distribution pattern. The mean of TGM concentration at the measurement site is 8.40 ng m^{-3} on the basis of one year

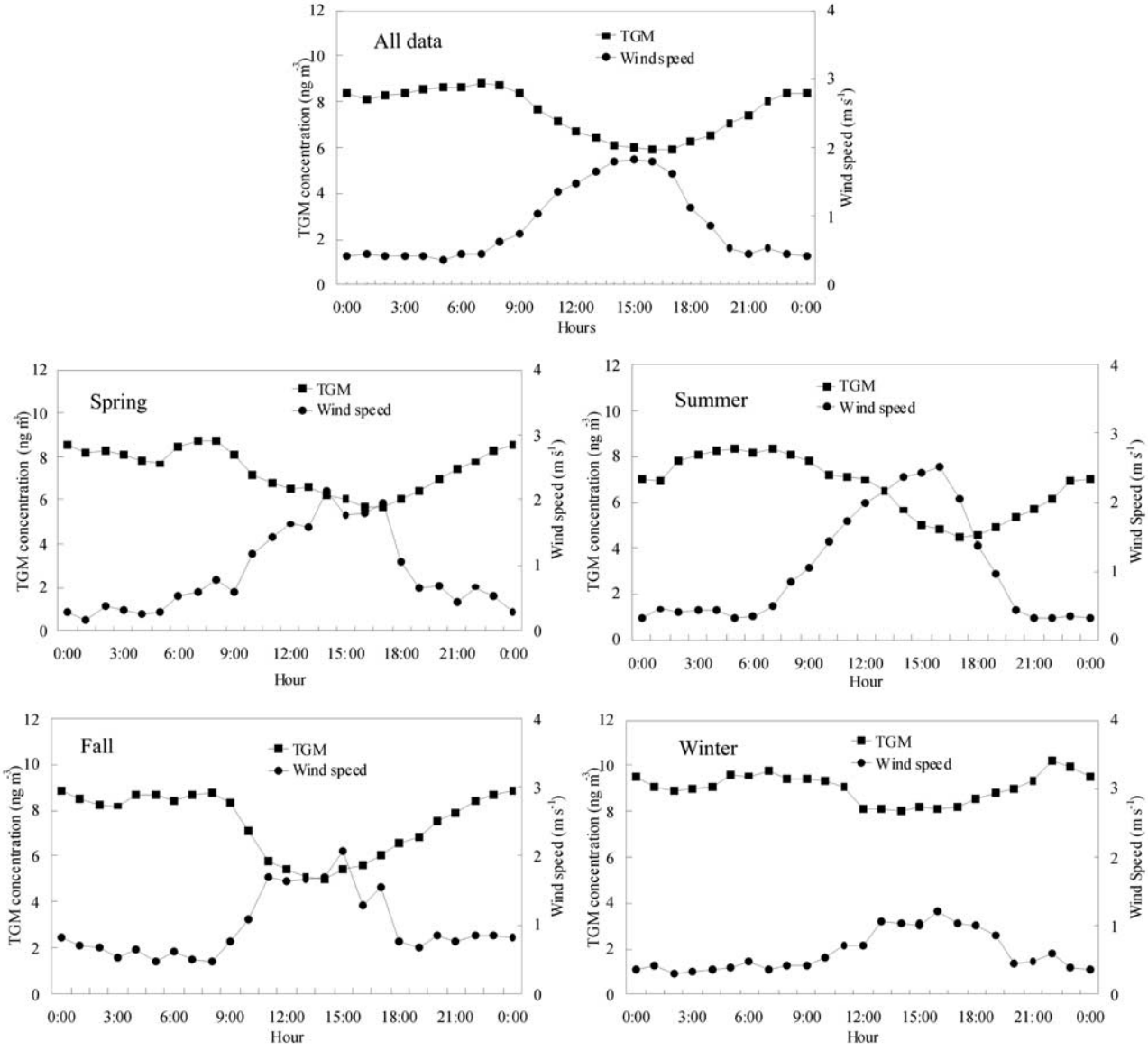

Figure 6. Diurnal variation of TGM and wind speed for both all and seasonally divided data sets. Both TGM and wind speed date are hourly averages.

observation. Small local emission sources pose more significant influences on the TGM concentration at the measurement site than large point sources that are much far away from the sampling site.

[18] We observed a seasonal distribution pattern of TGM with a descending order: winter, spring, fall and summer. The highest TGM concentration in winter can be attributed to household heating. Diurnal distribution pattern of TGM obtained for all seasons seems to be highly consistent, while the nighttime TGM concentration is elevated compared to the daytime values.

[19] Coal combustion from both industrial and domestic uses is the primary atmospheric source of Hg, and the seasonal TGM distribution pattern is attributed to different amount of coal consumption between cold and warm seasons. We believe that the consistent diurnal wind speed pattern in all seasons is responsible for the consistent TGM diurnal distribution obtained at our sampling site.

[20] Mercury emission from soil is not considered in our discussion since soil mercury concentrations (about 0.2 $\mu g\ g^{-1}$) in Guiyang are generally not very high. Measurement of mercury exchange flux between air and soil in Guiyang, however, will definitely help explain our data set better. This work is ongoing in our lab. Moreover, mercury speciation (Hg^0, reactive gaseous mercury and particulate mercury) measurements in air of Guiyang will also be propitious to elucidate our data set better. Unfortunately so far we do not have enough data on mercury speciation yet.

[21] **Acknowledgments.** This research was financially supported by Chinese Academy of Sciences through "A Hundred Talent Plan" and as well by the Chinese Natural Science Foundation (40173037).

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