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Atmospheric mercury in Changbai Mountain area, northeastern China I. The seasonal distribution pattern of total gaseous mercury and its potential sources ☆

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ABSTRACT

An intensive field campaign for the measurement of total gaseous mercury (TGM) concentrations in ambient air was conducted in Changbai Mountain area from 5 August 2005 to 5 July 2006 using an automatic atmospheric mercury analyzer (Tekran 2537A), which was the first time TGM was monitored at a remote area in northeastern China. 99% of the hourly TGM concentrations fell between 1.28 and 9.49 ng m^{-3} with an annual arithmetic mean of 3.58 ± 1.78 ng m^{-3}, which was significantly elevated compared to values obtained in remote areas of Europe and North America. Seasonal mean TGM concentrations displayed a descending trend as follows: winter, spring, fall, and summer. Compared to spring/winter, TGM concentrations were lower in the summer/fall but the standard deviation (SD) of TGM levels was higher and indicated a correlation with anthropogenic emissions. TGM concentrations showed seasonal differences with respect to meteorological parameters: TGM levels in spring/winter were most correlated with wind speed, and correlated with solar radiation only in the winter, while TGM levels in summer/fall periods were most correlated with air temperature. There was a strong diurnal variation of seasonal TGM with significantly higher concentrations in daytime/nighttime compared to the early morning. The seasonal diel TGM pattern indicated regional biofuel and coal combustion were the primary mercury sources.

1. Introduction

Mercury (Hg) is a highly toxic heavy metal and is considered a global pollutant due to its ability to undergo long-distance transport in the atmosphere. It is emitted into the atmosphere by various natural and anthropogenic sources. The majority of Hg in atmosphere is in the form of gaseous elementary Hg vapor, which is relatively inert and has a low depositional velocity compared to ionic forms of Hg (Schroeder and Munthe, 1998). Due to its high volatility and chemical stability, the atmospheric lifetime of gaseous phase elemental Hg is estimated to be 0.5–2 years (Schroeder and Munthe, 1998). Although adverse environmental and human health effects of Hg are predominately associated with the aquatic environment, the atmosphere is a major route for the transport and transformation of Hg from sources to receptor sites (Mason et al., 1994; Fitzgerald et al., 1998).

Monitoring the distribution of airborne Hg is of significant importance to understanding the fate of this element in the environment (Fitzgerald, 1995). Measurements of airborne Hg concentrations have been made worldwide at different locations in Asia, Europe, and North America at urban, rural, and remote sites, and even in the Atlantic ocean regions (Iverfeldt et al., 1995; Lee et al., 1998; Pecheyran et al., 2000; Ebinghaus and Slemr, 2000; Ebinghaus et al., 2002; Poissant, 2000; Kim and Kim, 2001; Liu et al., 2002; Sakata and Marumoto, 2002; Kellerhals et al., 2003; Temme et al., 2003; Cohen et al., 2004; Fang et al., 2004; Feng et al., 2004; Lynam and Keeler, 2005). Aircraft measurements were also conducted to understand the vertical distribution of airborne Hg in the troposphere (i.e. Ebinghaus and Slemr, 2000; Banic et al., 2003). The Panel of Source Attribution of Atmospheric Hg Deposition of the 8th International Conference on Mercury as a Global Pollutant (ICMGP) indicated that a global Hg monitoring

Abbreviations: TGM, total gaseous mercury; WD, wind direction; WS, wind speed; SR, solar radiation; RH, relative humidity

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network is needed to assess long-term changes in atmospheric Hg concentrations (Lindberg et al., 2007). However, current measurement sites are not evenly distributed around the globe. Much less information concerning the distribution of airborne Hg in remote areas in Asia is available from the open literature, even though Asia is now regarded as the largest source of anthropogenic Hg emissions due to the rapid economic development of China and India (Pacyna et al., 2003).

China is now regarded as one of the largest Hg emission countries in the world, and in 1999 it was estimated that total man-made Hg emissions were about 536 ton (Streets et al., 2005). Elevated airborne Hg concentrations in urban areas in China were reported in several recent studies. Liu et al. (2002) observed TGM concentrations in the range of 6–12 ng m^{-3} during the winter in Beijing. TGM concentrations in Guiyang ranged from 5 to 15 ng m^{-3} (Feng et al., 2004). Wang et al. (2006) measured TGM in Chongqing with values between 7.4 and 29.6 ng m^{-3}. Elevated TGM levels in large cities in China were attributed to coal combustion from industrial and residential sectors and zinc smelting (Feng et al., 2004; Streets et al., 2005). However, the distribution of airborne Hg at remote areas of China and potential sources of Hg in ambient air are still unknown. Examples of regional and global models currently used to study the fate of Hg in the atmosphere include the Community Multiscale Air Quality (CMAQ)-Hg model (Bullock and Brehme, 2002), Acid Deposition and Oxidant Model (ADOM) (Petersen et al., 2001), and Sulfur Transport and Deposition Model (STEM)-Hg, which are for evaluating the fate and transport of Hg in East Asia (Pan et al., 2008). However, the calculated values of TGM concentrations using these models were somewhat lower than field measurement data (Fu et al., 2008). In this paper, for the first time a one-year comprehensive TGM measurement campaign in ambient air in Changbai Mountain area was undertaken to characterize the TGM distribution pattern and potential sources of Hg in northeastern China.

2. Materials and methods

2.1. Study area and sampling site

Changbai Mountain (42°24′N, 128°28′E) is in the eastern Jilin Province, northeastern China (Fig. 1), which is near the boundary between China and North Korea. It has a hilly terrain with altitudes ranging from 500 to 2700 m above sea level. The regional topography is characterized by forests and mountains. It has a subtropical continental monsoon climate with abundant precipitation and low temperatures. There is a long cold winter period, which lasts nearly 5 months from late fall to late spring, and its annual average temperature is 2.1 °C. The highest recorded temperature is 30.8 °C, which occurred in mid-August, and the lowest is −33.3 °C, which occurred in early February. Changbai Mountain area is surrounded by a forest ecosystem (a land area of 1964 km^2), and there are no large industrial utilities nearby.

The sampling site is located at the Open Research Station of Changbai Mountain Forest Ecosystems, Chinese Academy of Sciences (CAS), which is situated about 5 km away from the nearest town, Baihe, near Yanji City in Jilin Province. The population of the town is about 49,000, and their main sources of energy are biofuel and coal combustion. The station is about 37 km north of the summit of Changbai Mountain. An air inlet of Teflon sampling tubing was set up 2 m above the roof of a building, about 5 m total above the ground. All tubings were heated with a constant temperature 50 °C to prevent the air from condensation. The measurement campaign was performed from 5 August 2005 to 5 July 2006.

2.2. Analytical methods

TGM monitoring in ambient air was achieved using an automatic Hg vapor analyzer (Tekran 2537A). Briefly, the analytical train of this instrument is based on the amalgamation of Hg onto a pure gold surface followed by thermo-desorption and then analysis is carried out by cold vapor atomic fluorescence spectrophotometry (CVAFS) ($\lambda = 253.7$ nm) providing measurements of TGM in ambient air at sub-ng m^{-3} levels at standard temperature and pressure. A dual-cartridge design allows for alternate sampling and desorption, resulting in continuous measurement of Hg in the air stream. The analyzer was programmed to sample air at a flow rate of 1.5 L min^{-1} for 5 min sampling intervals. Particulate matter was removed by a 47-mm-diameter Teflon filter (0.45 μm). The instrument allows two methods of calibration: manual and automatic permeation source injection. The permeation source rate is determined by calibration with multiple manual injections. The manual vapor injection calibration protocol has been provided by Tekran Model 2505. Accuracy and precision of this experiment have been assessed to be less than 5% (Tekran, 1999). The TGM data sets discussed below were averaged in one-hour time intervals to match the time scale of the meteorological parameters.

2.3. Meteorological parameters

Meteorological parameters including solar radiation (SR), wind direction (WD), wind speed (WS), air temperature (T), air pressure (P), relative humidity (RH), and cumulative precipitation were collected synchronously with TGM. All meteorological data were acquired using an automated Vaisala Milos 520 weather station, with a 1 h observation interval (Vaisala, 2001). Meteorological data were provided by the local weather station from the Open Research Station of Changbai Mountain Forest Ecosystems, CAS. The weather station was about 10 m south, a nd experienced nearly the same meteorological conditions as the sampling air inlet site.

3. Results and discussion

3.1. Overall TGM concentration

The whole sampling period was divided into four seasons based on routine meteorological classification. Fall was considered from 22 August to 21 November 2005; winter was from 22 November 2005 to 21 February 2006; spring was from 22 February to 21 May 2006, and summer included two periods: 5 to 21 August 2005 and 22 May to 5 July 2006, respectively. One sampling interruption occurred from 23 November to 7 December 2005 due to failure of the local power supply.

Fig. 2 depicts the hourly averaged TGM concentrations measured during the sampling campaign. Large variability in the temporal distribution patterns among seasons as well as within seasons was detected. Frequent and high spikes of Hg concentrations

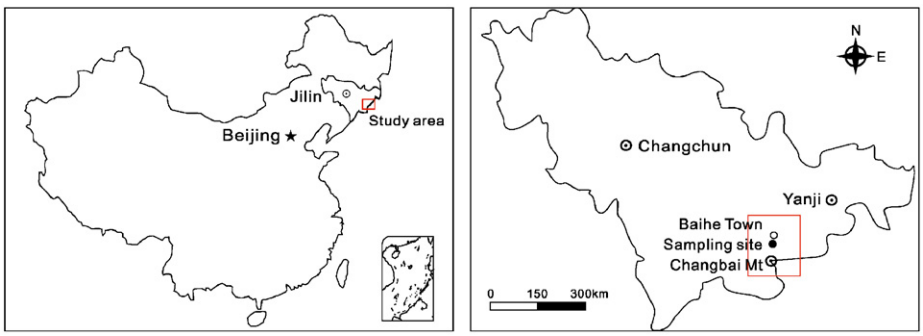

Fig. 1. Location of sampling site in Jilin Province in Northeastern China.

were observed during the study period, which was to some extent related to the effects of anthropogenic sources at the sampling site. This phenomenon was previously observed in other studies (Carpi and Chen, 2002; Dommergue et al., 2002). As a result, the mean TGM concentration in Changbai Mountain area was $3.58 \pm 1.78\,ng\,m^{-3}$, which was significantly elevated compared to values reported using similar instrumentation at a variety of locations in Europe and North America (e.g. Lee et al., 1998; Schroeder and Munthe, 1998; Slemr and Scheel, 1998; Ebinghaus and Slemr, 2000; Poissant, 2000; Urba et al., 2000), but was comparable with those measured at the southern-end district of Seoul, Korea (Kim and Kim, 2001), suburban-industrial and rural locations in Beijing (Liu et al., 2002) and at a remote site in Mt. Gongga in Sichuan Province, southwestern China (Fu et al., 2008). However, TGM concentrations in the study area were much lower than those measured in urban sites in Seoul, Korea (Kim and Kim, 2001), northern Taiwan area (Kuo et al., 2006), and Guiyang city in China (Feng et al., 2004).

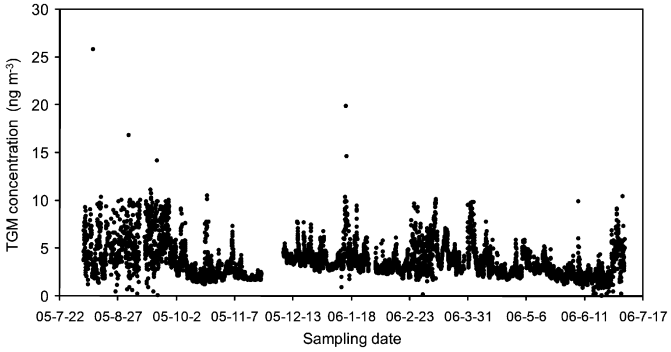

Fig. 2. TGM concentrations in the air of Changbai Mountain area, Northeastern China. Hourly averaged TGM concentrations during the whole study period from 5 August 2005 to 5 July 2006.

Table 1
Statistical summary of TGM concentration ($ng\,m^{-3}$) in Chanbai Mountain area.

	Spring	Summer[a]	Fall	Winter	Whole period
N	1882	1241	1857	1768	6748
Min	0.19	0.08	0.10	0.94	0.08
10%[b]	2.16	1.56	1.77	2.61	1.89
25%[b]	2.55	1.88	1.98	2.99	2.32
Median	3.27	2.32	2.91	3.57	3.15
75%[b]	4.39	3.29	4.63	4.22	4.25
90%[b]	5.92	5.59	7.23	4.93	5.91
Max	10.17	25.84	16.90	19.93	25.84
Arith. mean	3.72	2.97	3.67	3.75	3.58
SD	1.59	1.87	2.20	1.22	1.78
Geo. mean	3.44	2.56	3.15	3.61	3.22

[a] Summer: data in summer contain two periods: August 5 to 21, 2005 and May 22 to July 5, 2006.
[b] 10%: 10%, 25%, 75%, and 90% indicate the TGM values in the 10th percentile, the 25th percentile, the 75th percentile, and the 90th percentiles in every seasonal or all the hourly data sets, respectively.

3.2. Seasonal variation of TGM

Table 1 lists a statistical summary of TGM concentrations for four seasons with significant percentiles including 10, 25, 50, 75, and 90. Generally, 99% of the hourly averaged TGM concentrations fell between 1.28 and $9.49\,ng\,m^{-3}$. Seasonal arithmetic mean of TGM concentrations varied from 2.97 to $3.75\,ng\,m^{-3}$ while geometric mean varied a little lower from 2.56 to $3.61\,ng\,m^{-3}$. Both minimum ($0.08\,ng\,m^{-3}$) and maximum ($25.84\,ng\,m^{-3}$) were observed in summer. Arithmetic SD in fall was highest although average TGM values were not the highest. The lowest SD value along with the highest averaged TGM was observed in winter.

A lognormal frequency distribution pattern was determined for hourly averaged TGM data as shown in Fig. 3. TGM concentrations in ambient air from all seasons were significantly higher than values observed at remote areas in Europe and North America, undoubtedly implying that regional background TGM concentrations were significantly elevated in Changbai Mountain. Although mean TGM in spring/winter were at the same level, TGM frequency distribution patterns were somewhat different. Most frequent TGM values (42.3% of total measurement data in winter) occurred in the range from 3.0 to $4.0\,ng\,m^{-3}$ in winter, while most values were between 2.0 and $3.0\,ng\,m^{-3}$ during the other three seasons. In spring and winter, relative frequencies of TGM concentrations, which ranged from 1.0 to $2.0\,ng\,m^{-3}$, were quite low (less than 5%), but this range constituted a relative frequency of 25.6% and 29.7% in fall and summer, respectively. TGM data for the four seasons showed significant differences ($F = 60.552$, $p < 0.001$) according to one-way ANOVA analysis. This seasonal difference of TGM between spring/winter periods and summer/fall periods was also reported by Slemr and Scheel (1998). These data indicating seasonal differences are a characteristic of TGM in Changbai area.

In addition to seasonal variability in TGM levels, different impact factors were associated with each season. Correlations between seasonal TGM levels and principal meteorological parameters including *T*, WS, SR and RH were significant. TGM levels in spring/winter periods were most significantly correlated with wind speed, while a significant correlation between TGM concentrations and solar radiation was only observed in winter.

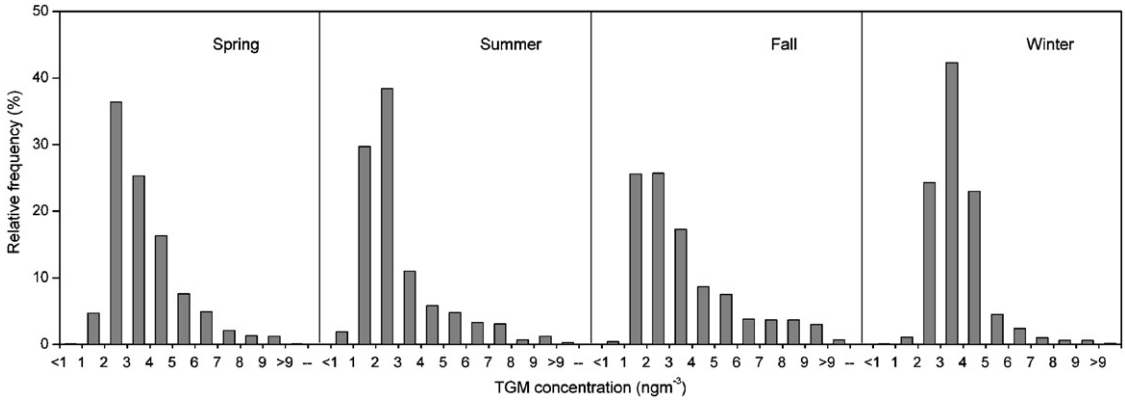

Fig. 3. Relative frequency plot of TGM distribution data measured from sampling site in four seasons.

TGM concentrations in summer/fall periods were most significantly correlated with air temperature. Similar results were also obtained from a PCA model study, which revealed three underlying factors (with eigenvalues >1, Table 2): diurnal mixing, source contributions, and seasonal meteorological conditions. Factor 1 had a moderate positive loading for SR and WS and a moderate negative loading for RH. The loadings for meteorological variables may indicate daytime Hg emissions from the soil surface and influence by wind (air mass). Factor 2 had a particularly strong negative loading for air temperature, and also had moderate positive correlations with air pressure; Factor 3 had heavy positive loading for wind directions, plus moderate positive loadings for air pressure, which suggested effects of climatological seasonality on TGM distributions. Other studies (e.g. Schroeder et al., 1989; Poissant and Casimir, 1998) have demonstrated that an increase in surface temperature significantly promotes the Hg emission rate from surface soil. Therefore, Hg deposited through dry and wet deposition processes and accumulated in surface soil during the cold season may reemit rapidly back to the air during the spring season, which probably results in relatively higher TGM in spring. Hg concentrations in soil were not obtained in this study; however, Hg in soil is likely due to anthropogenically deposited Hg, as Hg mineralization activities were not observed in this region.

Seasonal trends may reflect residential energy consumption differences between cold and warm seasons, as coal combustion is considered as a primary source of atmospheric Hg in most areas of China (Feng et al., 2004; Streets et al., 2005). Domestic coal consumption, which comprised approximately 25% of the total coal consumption in the study area, is utilized during cold seasons from late fall to early spring, which could partly explain the differences of TGM distribution among seasons. TGM concentrations in spring/winter were relatively stable compared with summer/fall and SD in fall was the highest. This strongly suggested that human activities have a significant impact on seasonal TGM distribution, which was observed in other areas in China, e.g., in Guiyang, Changchun, and Beijing (Feng et al., 2004; Fang et al., 2004; Liu et al., 2002).

Table 2
Results from the principal component analysis (PCA) model.

Variables	Factor 1	Factor 2	Factor 3
TGM	−0.07	−0.14	0.29
T	0.30	−0.64	−0.01
WD	0.17	0.32	−0.66
WS	0.49	0.30	−0.21
SR	0.55	−0.13	0.34
P	−0.16	0.55	0.52
RH	−0.59	−0.26	−0.24
Eigenvalues	2.12	1.45	1.12
Variance explained (%)	30.2	21.3	16.1

3.3. Potential sources of TGM

3.3.1. Regional biofuel and coal combustion

Fig. 4 included hourly TGM concentrations in four seasons. In general, hourly TGM concentrations showed a diurnal variation, with highest TGM values around 18:00 in the evening and the lowest occurring at 5:00 in early morning. Peak TGM concentrations at 18:00 were probably due to combustion of coal and biofuel, including wood and leaves for cooking by regional residents. From Fig. 4 the exact peak time for TGM levels varied between seasons: it appeared at 18:00 in spring, 19:00 in summer, 12:00 in fall, and 17:00 in winter. This was consistent with regional cooking habits during four seasons. 60–70% of the regional residents are farmers and they burn biofuel for cooking. Friedli et al. (2003) reported that all the Hg will be liberated from biomass burning, and 87–99.9% is released as elementary Hg. Additionally, around the evening, the predominant wind direction was from the northwest (constituting a relative frequency of 57.3%), which is from a densely populated residential area. Average wind speed around 18:00 was under the median (1.5 m/s); therefore, Hg-contaminated air was not likely diluted by winds. In the late morning till noon, TGM levels increased in each season. Studies show that the continued increase of TGM concentration through morning and into the late afternoon is likely caused by surface emission with increasing solar radiation (Feng et al., 2005; Wang et al., 2006). Although the measurement of Hg exchange fluxes between air and soil surface were not performed in our study,

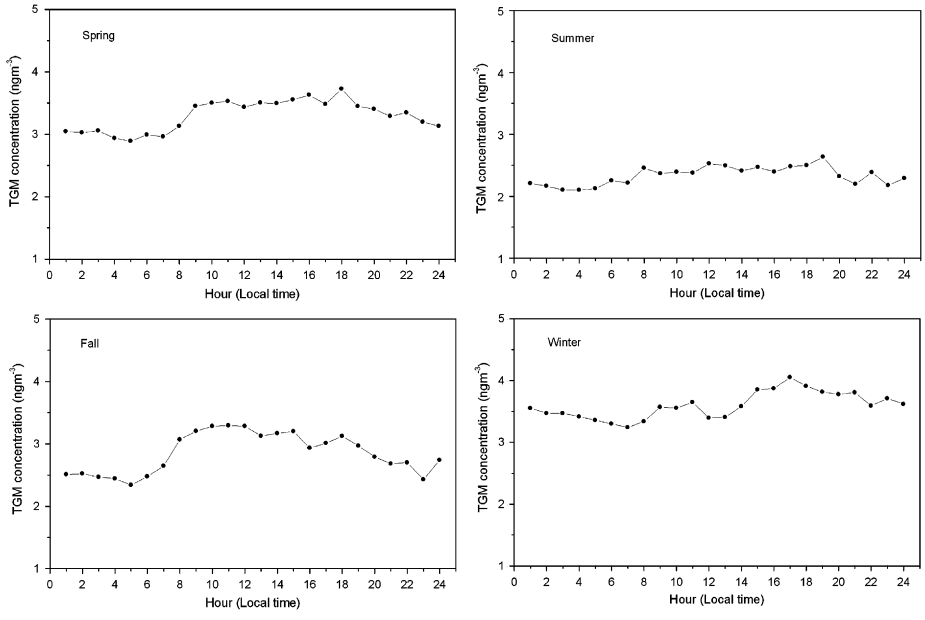

Fig. 4. Hourly TGM concentrations at four seasons based on China central time (CCT) in Changbai Mountain area.

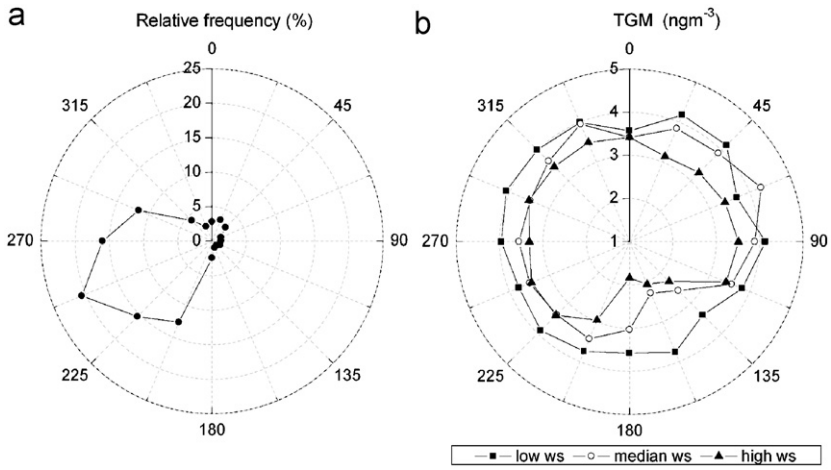

Fig. 5. (a) Frequency increases of wind directions and (b) mean TGM concentrations at wind directions under low wind speed ($N = 1617$), median wind speed ($N = 3421$) and high wind speed ($N = 1453$) modes in Changbai Mountain area.

mercury emission from soil or snow may partly explain the increasing TGM pattern in the morning after sunrise. It was estimated that total annual man-made Hg emissions was about 536 ton in 1999 in China, including 62.1% from coal combustion (Streets et al., 2005). Impacts from intensive domestic coal and biofuel use for house heating and/or living consumption were obvious. Hence, regional anthropogenic Hg sources from energy consumption (e.g. biofuel and coal combustion) contributed significantly to the elevated TGM, similar to most areas of China. It is generally believed shallow nocturnal boundary layers form during most nights, which trap TGM near the surface, while thermal mixing during the day increases the boundary-layer depth, thus diluting the concentrations (Lee et al., 1998); this may also explain the increase of TGM at nighttime. But this fluctuation pattern of hourly TGM is a complex physical and chemical process, and it may represent the effects of multiple processes such as surface deposition, volatilization from surrounding surfaces, together with occasional transport of Hg-enriched air from source regions (Kellerhals et al., 2003).

3.3.2. *Wind directional dependence of TGM*

Fig. 5 shows the frequency distribution of wind along with the mean TGM at different wind directions based on low wind speed ($<1.0\,m\,s^{-1}$), median wind speed ($1.0–3.0\,m\,s^{-1}$) and high wind speed ($>3.0\,m\,s^{-1}$) modes, which was constructed by sorting all the measurements into 22.5° sectors, such that each sector represented a large number of observations. Predominant wind was from the West and SWW sectors, with a relative frequency of 20.6% and 16%, respectively. In general, TGM concentrations were enhanced with winds from any direction at low, median, and high wind speed modes. Fig. 5b shows that the high TGM concentrations in low wind speed mode with less diluting capability can be depicted as a regional mercury source. Frequent southwesterly wind was associated with relatively median TGM, while the northeasterly winds, which were not the predominant wind directions, carried significantly higher Hg concentrations. As mentioned above, densely populated residential areas (e.g., Yanji City) were situated northeast of the sampling site. Wind in low and median wind speed modes from this sector carried high TGM concentrations that exceeded the mean TGM value (Fig. 5b), indicating that TGM levels in the vicinity of the sampling site were generally elevated due to regional Hg emissions. Besides, an opposite circumstance, namely high TGM occurs in low wind speed mode while the lowest TGM occurs in median and high wind speed modes in SE sector, is another evidence for the regional mercury source.

Another potential Hg source may be attributed to gold mining in Jilin Province. Consumption of Hg^0 in Au mining activities throughout China was estimated to be about 400 ton and the amount of Hg^0 lost to surrounding environments was estimated to be 107 ton in 1995 (Feng et al., 2006). Jiapigou gold mining company, which is situated about 150 km northwest, is the largest gold mining plant in Jilin Province using Hg to extract gold. There was no distinct evidence that linked high Hg levels in Changbai Mountain area with Jiapigou gold mining activities, but certainly the gold mining activities released a significant amount of Hg to the air (Wang et al., 2006). Further study is needed to confirm Hg sources in this region, and to offer better insight into the distribution and fate of atmospheric Hg in the Changbai area.

4. Conclusions

Annual mean TGM concentration in Changbai Mountain area based on a one-year study was $3.58 \pm 1.78\,ng\,m^{-3}$, which is significantly elevated compared to the values obtained in remote locations of Europe and North America ($1.5–2.0\,ng\,m^{-3}$) (Schroeder et al., 2001; Ebinghaus et al., 2002). The obtained values were much lower than those reported in urban and rural areas of China. Regional TGM concentrations are elevated mainly due to Hg emissions from coal and biofuel burning from domestic uses. Seasonal mean TGM concentrations displayed a descending trend: winter, spring, fall, and summer. The seasonal diel TGM pattern indicated that regional biofuel and coal combustions were the primary Hg sources. Obviously, a detailed Hg emission inventory in this area is needed to further elucidate TGM distribution characteristics, and to evaluate objectively the ecological consequences.

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