Mercury concentrations and air/soil fluxes in Wuchuan mercury mining district, Guizhou province, China

Shaofeng Wang[a,b,c], Xinbin Feng[a,*], Guangle Qiu[a], Lihai Shang[a,c],
Ping Li[a,c], Zhongqing Wei[a,b]

[a]*State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, PR China*
[b]*Key Laboratory of Terrestrial Ecological Processes, Institute of Applied Ecology, Chinese Academy of Sciences,
Shenyang 110016, PR China*
[c]*Graduate School of the Chinese Academy Sciences, Beijing 100039, PR China*

Abstract

Wuchuan Hg mine, located in the Circum-Pacific Global Mercuriferous Belt, is one of the important Hg production centers in Guizhou province, China. Soil Hg concentrations in this area are elevated by 2–4 orders of magnitude compared to the national background value in soil which is $0.038 \, \mu g \, g^{-1}$. In situ air Hg concentrations and air/soil Hg fluxes were measured at five sampling sites in Wuchuan Hg mining area (WMMA) from 19 to 26 December 2003 and from 18 to 25 December 2004. The results showed that air Hg concentrations were 2–4 orders of magnitude higher than those observed in background areas in Europe and North America due to a large amount of Hg emission from artisanal Hg smelting activities. The average in situ Hg fluxes at site Laohugou, Jiaoyan, Luoxi, Sankeng and Huanglong were -5493, 124, -924, -13 and $140 \, ng \, m^{-2} \, h^{-1}$, respectively. Diurnal pattern of Hg flux was not found and a number of negative Hg fluxes were observed in our sampling campaigns. The correlations between Hg fluxes and meteorological parameters such as solar irradiation, air temperature, soil temperature and relative humidity and air Hg concentrations were investigated. The commonly observed significant correlations between Hg fluxes and meteorological parameters observed in many previous studies were not obtained in WMMA. However, significantly negative correlations between Hg flux and air Hg concentration were observed at all sites. Our study demonstrated that highly elevated air Hg concentrations could suppress Hg emission processes even from Hg-enriched soil. At specific conditions in WMMA, air Hg concentrations play a dominant role in controlling Hg emission from soil.
© 2007 Elsevier Ltd. All rights reserved.

Keywords: Air/soil Hg exchange; Air Hg concentration; Source; Sink; Guizhou province

1. Introduction

Once released to air, mercury (Hg⁰) can be transported to remote areas by atmospheric cycles

*Corresponding author. Tel.: +86 851 5891356;
fax: +86 851 5891609.
E-mail address: fengxinbin@vip.skleg.cn (X. Feng).

due to its long residence time in the atmosphere (0.5–2 yr) (Schroeder and Munthe, 1998; Ebinghaus et al., 2002a). Both natural processes and human activities can emit a great amount of gaseous Hg to the atmosphere (Schroeder and Munthe, 1998). It has been estimated that from the industrial revolution, the average Hg concentration in the troposphere has been elevated by a factor of three

(Fitzgerald, 1995). A recent publication has estimated that Hg emission flux from anthropogenic sources was 2190 t in 2000 (Pacyna et al., 2006). However, Hg emission flux from natural sources is still not well known because of the complexity of the emission processes. Geologically, Hg-enriched areas have been regarded as a huge source of atmospheric Hg for a long time (Mason et al., 1994; Gustin, 2003). In the last decade, considerable progresses on understanding Hg emission from Hg-enriched area have been made (e.g. Gustin, 2003; Feng et al., 2005; Wang et al., 2005). The results showed that the Hg emission rates from Hg-enriched area are much greater than the values measured in background area (e.g. Poissant and Casimir, 1998) and the contribution of gaseous Hg to the atmosphere from Hg-enriched soil in mercuriferous belt was largely underestimated (Gustin, 2003; Feng et al., 2005).

Guizhou province (N: 24°30′–29°13′, E: 103°1′–109°30′), the most important Hg production base of China, is situated in the Circum-Pacific Global Mercuriferous Belt. Hg concentrations of soil in this area are much elevated up to 2–4 orders of magnitude compared to the national background value that is 0.038 μg g^{-1} (Feng et al., 2005). Some studies were conducted on Hg emission flux measurements from soils with elevated Hg concentrations in Guizhou province (Feng et al., 1996, 2003, 2005; Wang et al., 2003, 2005). It was found that Hg-enriched soil in Guizhou province is an important Hg source to the atmosphere. Wuchuan Hg mining area (WMMA), located in northeastern Guizhou, is a large Hg mine in China (Fig. 1). It has a long history of Hg production for 400 years and more than 4070 t of metallic Hg has been produced (Qu Liya, 2004). The state Wuchuan Hg mining plant went into operation in 1949 and was shut down in 2003. However, illegally artisanal Hg smelters are still in operation. Long-term Hg refining activities induced serious Hg contaminations to water, soil, sediment, and air compartments in Wuchuan (Qu Liya, 2004; Qiu et al., 2006). Large quantities of gangues, ore tailings, and smelting residues (calcites) have been produced into piles and spoil heaps. Due to heavy pollution, air Hg concentration in WMMA was much elevated (Li et al., 2006a) by 2–4 orders of magnitude compared to the values observed in pristine areas in Europe and North America, that is 1.5–2 ng m^{-3} (Ebinghaus et al., 2002b; Poissant et al., 2005). It has been proven that the health of local residents and
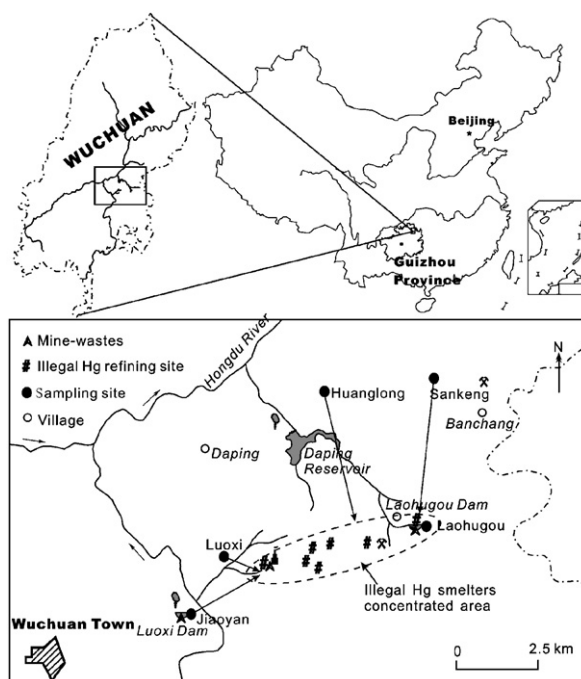

Fig. 1. The sketch map of the location of Wuchuan Hg mining area and sampling sites.

workers for artisanal Hg smelting activities has been harmed seriously by elevated air Hg concentration in WMMA (Li et al., 2006a). Previous field and laboratory experiments indicated that elevated air Hg concentration may decrease Hg emission flux from soil or induce Hg deposition to soil/land surfaces (e.g. Gillis and Miller, 2000; Nacht et al., 2004; Wang et al., 2005). In this human-disturbed area, the objectives of this study is to (1) determine the contribution of gaseous Hg from widely distributed Hg-enriched soil and retorted tailings to the atmosphere and (2) understand the mechanism of air/soil Hg exchange under this anthropogenically Hg-polluted condition.

2. Experimental

2.1. Background of the study area

The WMMA (E: 107°31′–108°31′, N: 28°11′–29°05′) is part of a hilly and karstic terrain, ranging in elevation from 325 to 1743 m above sea level. Its climate presents a typical subtropical humid monsoon with an average temperature of 15.5 °C and an average annual rainfall of 1272 mm.

Similar to the most of Hg mines worldwide, the dominant Hg ore is cinnabar in WMMA. Previous

large-scale Hg-mining activities were primarily conducted at Luoxi and Laohugou (Fig. 1). Approximately 185 t of metallic Hg introduced in exhausts, mine wastes, and drainages were eventually released into the local environment (Qiu et al., 2006).

WMMA also is a center of artisanal Hg smelting activities using an indigenous method. Numerous artisanal Hg smelters were scattered in the valley from Laohugou to Luoxi (Fig. 1). Apparently, environmental impacts related to those illegal Hg-refining activities are even more serious. All smelting residues from the artisanal Hg smelters were discarded in the vicinity of the smelters along the stream channels. Because the simple smelting processes are without any environmental protection measures, the Hg emission factors (the proportion of Hg in ore is released to the ambient air) ranged from 6.9% to 32.1% and the annual Hg emission was up to 3.7–9.6 t (Li et al., 2006b).

2.2. Sampling sites

In situ air Hg concentrations and air/soil Hg fluxes were measured at three sites (Laohugou, Jiaoyan, Luoxi) from December 19 to 26, 2003, and at two sites (Sankeng, Huanglong) from 18 to 25 December, 2004 in WMMA, respectively (Fig. 1). Except that the substrates of site Laohugou and Jiaoyan were roasted cinnabar and gangue, the substrates of other sites were field soil. Site Laohugou was selected nearby a group of artisanal Hg smelters in a valley. Site Jiaoyan was chosen on the dam of ore tailings. Site Luoxi is located in a crop field which is about 0.8 km away from the Wuchuan state Hg-refining plant and several artisanal Hg smelters were in operation during our sampling period. For comparison, site Sankeng and Huanglong were chosen several kilometers away from Hg smelters concentrated area (Fig. 1). The straight distance between the illegal Hg smelting area and the five sampling sites in Laohugou, Jiaoyan, Luoxi, Sankeng and Huanglong are 0, 2.0, 0.8, 4.9 and 4.8 km, respectively (Fig. 1).

2.3. Hg flux measurement

The dynamic flux chamber (DFC) method used to measure Hg fluxes has been described in many documents (e.g. Feng et al., 2005; Wallschläger et al., 1999; Xiao et al., 1991). A semi-cylinder, open-bottom chamber made of

Quartz ($\varnothing 20 \times 30$ cm) was used during our sampling campaigns. Hg flux from the soil exposed in the chamber was calculated using Eq. (1):

$$= (C_o - C_i) \times Q/A, \qquad (1)$$

where F is the flux of gaseous Hg, which consists of mostly Hg^0 (Schroeder and Munthe, 1998) in ng $Hg\,m^{-2}\,h^{-1}$; C_o and C_i are concentrations of Hg in air of the outlet and inlet of the chamber in $ng\,m^{-3}$, respectively; A is the bottom surface area of the chamber in m^2 ($0.06\,m^2$); and Q is the flushing flow rate through the chamber in $m^3\,h^{-1}$ ($0.9\,m^3\,h^{-1}$). Gaseous Hg concentrations were measured twice in the ambient air entering the inlet of the chamber and twice in the air exiting through outlet of the chamber using an automated air Hg analyzer (Tekran 2537A) with a 5 min sampling time. Switching from the inlet to the outlet of the chamber every 10 min was achieved using a magnetic valve (Tekran 1110). Another mass flow controller combined with a second magnetic valve, which is synchronous with the first one, was employed to compensate flow rate decrease when Tekran is sampling air from the inlet of the chamber. A high flushing flow rate of $151\,min^{-1}$ ($0.9\,m^{-3}\,h^{-1}$) was adopted to prevent the possibility of underestimating Hg flux at low flushing flow rates (Gillis and Miller, 2000; Zhang et al., 2002; Lindberg et al., 2002). The mean of two C_o and the mean of four C_i before and after C_o were used to calculate Hg flux between soil and air sequentially. One Hg flux datum was obtained every 20 min. The positive and negative results calculated from Eq. (1) represent Hg emission from soil and atmospheric Hg dry deposition, respectively (Feng et al., 2005).

2.4. Soil Hg concentration and meteorological parameters

Representative surface soil samples from the five sampling sites were collected from the area covered by the flux chamber. All soil samples were air-dried, milled and sieved (<80 mesh). In all, 300–500 mg sample was oxidized with 5 ml concentrated $HNO_3 + HCl$ (1:3 v/v) in a Teflon vial using a microwave oven at a power of 1 kW (MDS2000, from CEM, USA) for 50 min. The digested solution was then transferred to a 100 ml volumetric flask, and the volume was made up to 100 ml by adding Mill-Q water. Total Hg concentration was determined using BrCl oxidation and $SnCl_2$ reduction

coupled with cold-vapor atomic absorption spectrometry (F-732, China).

The meteorological parameters including air and soil temperature, wind speed and direction, solar irradiation and relative humidity were monitored using a mini-weather station (Global Water IIIB, USA) with a time resolution of 5 min. The detailed description on the weather station was given by Feng et al. (2004, 2005).

2.5. QA/QC

The automatic air Hg analyzer (Tekran 2537A) was calibrated manually before and after the sampling campaign by injecting a volume of Hg-saturated air with known concentration. The measurements of the blank of DFC were performed before and after the measurement campaigns by covering the chamber on a quartz glass plate. Blanks of DFC (2.0 ± 1.6 ng m^{-2} h^{-1}, $n = 12$) were not subtracted from results of fluxes because it is negligible compared to in situ fluxes. A standard soil sample GBW-07405 (GSS-5) was used to accomplish QA/QC, and the average total Hg concentration of the geological standard of GBW-07405 was 0.30 ± 0.01 µg g^{-1} ($n = 8$), which is comparable with certified value of 0.290 ± 0.04 µg g^{-1}. The precision of our method obtained from replicate analysis is less than 5%. The results demonstrated that the pretreatment procedures can quantitatively recover Hg from the soil samples.

3. Results and discussion

3.1. Air Hg concentration and air/soil Hg flux

Air Hg concentrations (C_i) measured from the chamber inlet at all sampling sites are shown in Table 1 and Fig. 2. The average air Hg concentrations at site Laohugou, Jiaoyan, Luoxi, Sankeng and Huanglong were 2114.5, 159.9, 528.2, 50.5 and 56.3 ng m^{-3}, respectively. The highest level of air Hg concentration (12,222.0 ng m^{-3}) was found at site Laohugou. It is obviously seen that air Hg concentrations were significantly elevated by 2–4 orders of magnitude compared to the value observed at the global background sites in Europe and North America ($\sim$1.5 ng m^{-3}, Schroeder et al., 1998). Average air Hg concentration decreased exponentially with the distance between sampling sites and the artisanal Hg smelting area as shown in Fig. 3 and a significant correlation ($R^2 = 0.98$) between air Hg concentration and the distance away from the artisanal Hg smelting site were observed. At the same time, it can be seen that the air Hg concentrations changed with the wind direction as shown in Fig. 4 and it is obvious that Hg emission from artisanal Hg smelting area had a strong impact on the air Hg concentrations in WMMA. High temporal resolved air Hg concentration is shown in Fig. 2. Due to the influence of Hg smelting activities, the temporal variations of air Hg concentrations were observed at five sampling sites. Previous study (Feng et al., 2004) demonstrated that DFC method could not give true Hg fluxes

Table 1
Statistical results of Hg flux, TGM concentration in air and total Hg concentration in soil at all sampling sites in Wuchuan Hg mining area

Site	Sampling time	Hg flux (ng m^{-2} h^{-1})			Air Hg concentration (ng m^{-3})		Soil Hg concentration (µg g^{-1})
			Mean	n	Mean	n	
Laohugou	19/12/2003 17:50–	Emission	5143 ± 3738	26	2114.5 ± 1479.6	154	94.7
	20/12/2003 19:45	Deposition	10916 ± 8339	51			
Jiaoyan	21/12/2003 16:30–	Emission	305 ± 199	78	159.9 ± 78.0	229	150.7
	23/12/2003 16:05	Deposition	291 ± 356	34			
Luoxi	24/12/2003 11:35–	Emission	422 ± 444	43	528.2 ± 464.0	166	1.1
	25/12/200316:05	Deposition	2407 ± 2560	39			
Sankeng	19/12/2004-17:05–	Emission	32 ± 47	52	19.5 ± 5.3	304	2.7
	21/122004 20:15	Deposition	36 ± 36	100			
Huanglong	22/12/2004 16:10–	Emission	234 ± 257	175	56.3 ± 40.4	442	3.0
	25/12/2004 17:30	Deposition	219 ± 316	46			

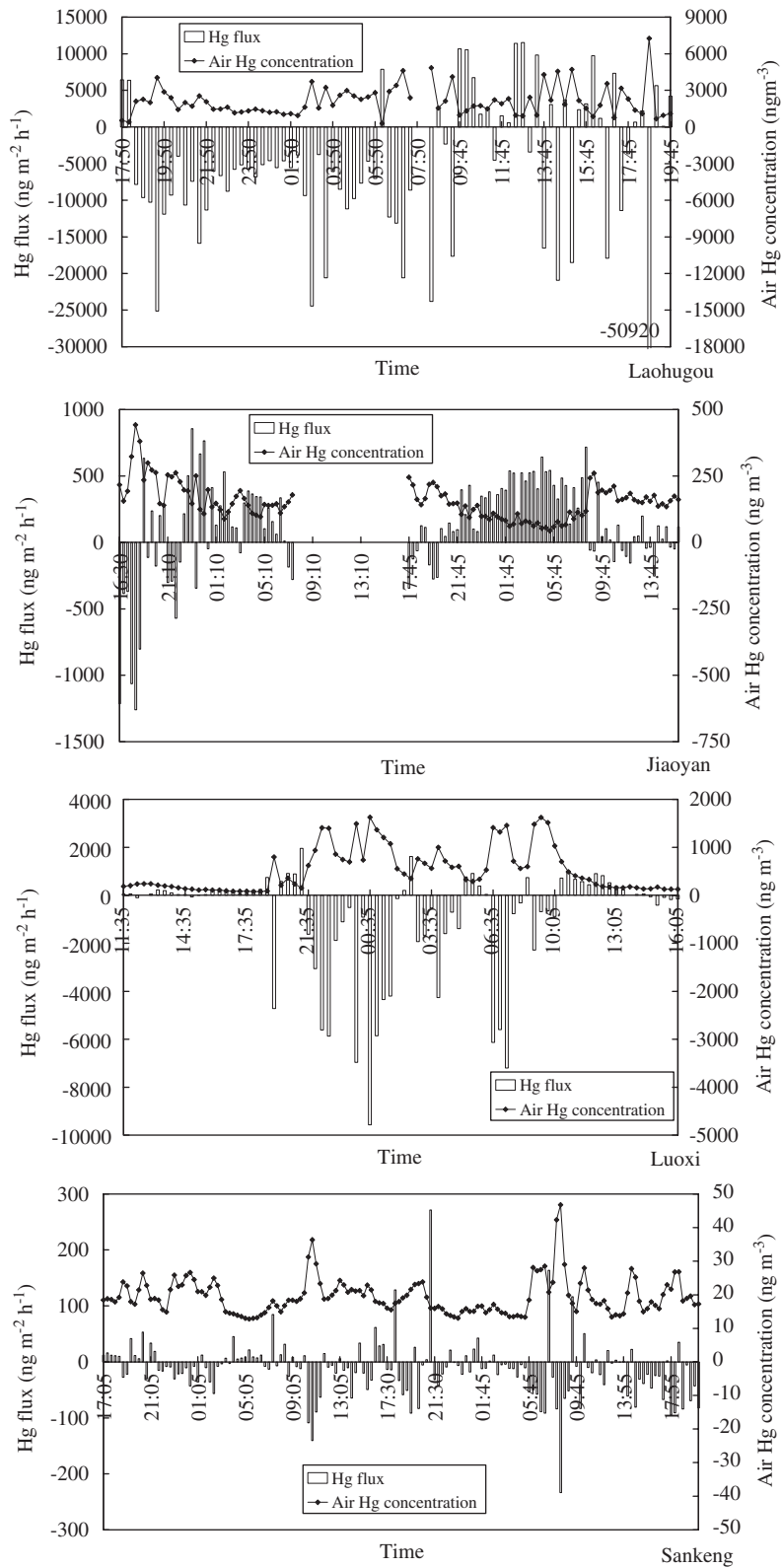Hg flux and air Hg concentration are shown as mean$\pm$standard deviation.

Fig. 2. Temporal variation of air Hg concentrations and Hg flux at all sampling sites. From top to bottom, plots for Laohugou, Jiaoyan, Luoxi, Sankeng and Huanglong. Air Hg concentrations shown in the figure are average value of 20 min data. Meteorological parameters shown in the figure are recorded on resolution of 5 min.
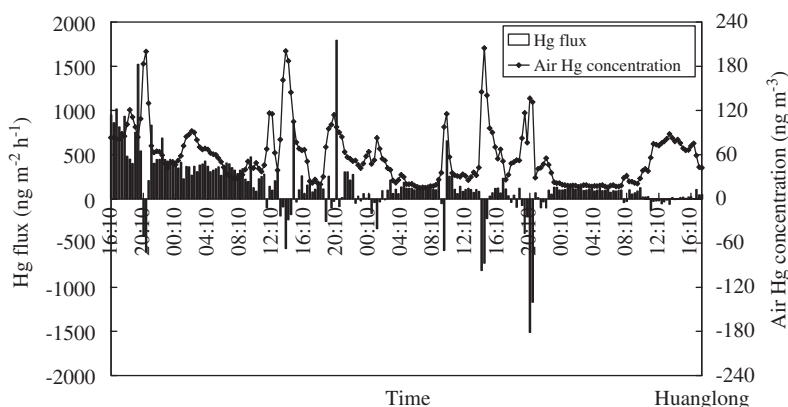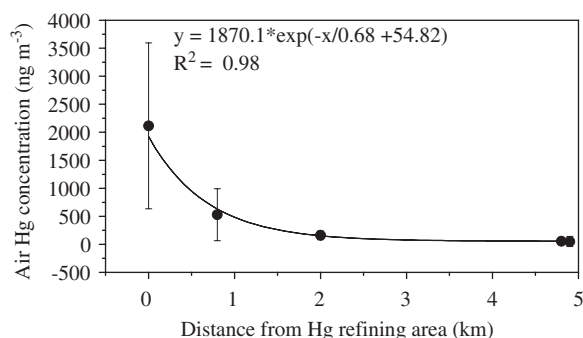
Fig. 2. (*Continued*)

Fig. 3. Plots of air Hg concentration decreasing following increase in distance.

at a condition with short-term large variation of air Hg concentrations. Generally, the air Hg concentrations in our measurement campaigns changed progressively with only few occasional short-term spikes, therefore the Hg fluxes measured using DFC method can represent the true Hg fluxes.

The results of air/soil Hg fluxes are given in Table 1 and Fig. 2. The average Hg exchange fluxes at Laohugou, Jiaoyan, Luoxi, Sankeng and Huanglong were −5493, 124, −924, −13 and 140 ng m^{-2} h^{-1}, respectively. It is obvious that net Hg emission fluxes were obtained from sites Jiaoyan and Huanglong, and negative Hg fluxes were found at sites Laohugou, Luoxi and Sankeng, respectively. Such high-frequency of Hg deposition in WMMA was not reported in other area. These air/soil Hg fluxes were 2–3 orders of magnitude stronger than those in the background area (Zhang et al., 2001). It is clear that Hg-enriched soil or roasted cinnabar could be the Hg sources or sink to

the atmosphere in WMMA. Previous studies reported that Hg emission flux from soil showed clearly diurnal trends in Hg-enriched area and this diurnal model was used to estimate annually Hg emission flux from the whole district (Coolbaugh et al., 2002; Nacht et al., 2004). However, Hg fluxes fluctuated strongly and did not show any diurnal patterns at all sampling sites in WMMA (Fig. 2). Highly elevated air Hg concentrations and variation of Hg concentrations may contribute the fluctuation of Hg fluxes observed in our study as discussed in Section 3.4.

3.2. Relationship between Hg flux and soil Hg concentration

At the sites Laohugou and Jiaoyan, elevated soil Hg concentrations of 94.7 and 150.7 μg g^{-1} were found, respectively. Relatively lower soil Hg concentrations of 1.1, 2.7 and 3.0 μg g^{-1} were found at sampling sites of Luoxi, Sankeng and Huanglong, respectively (Table 1). Compared to the average background soil Hg concentration in China (0.038 μg g^{-1}, Feng et al., 2005), soil Hg concentrations are 2–4 orders of magnitude elevated in WMMA. Previous studies have demonstrated that soil Hg concentration is an important factor controlling Hg emission from soil and a log–log relationship between soil Hg concentration and Hg flux can be constructed (e.g. Gustin, 2003; Nacht et al., 2004). However, in spite of high soil Hg concentration, strong negative Hg flux was observed instead of Hg emission from soil at almost all sites and in most time. The highest negative Hg flux was obtained at site Laohugou with the high soil Hg concentration of 94.7 μg g^{-1} (Table 1). The log–log
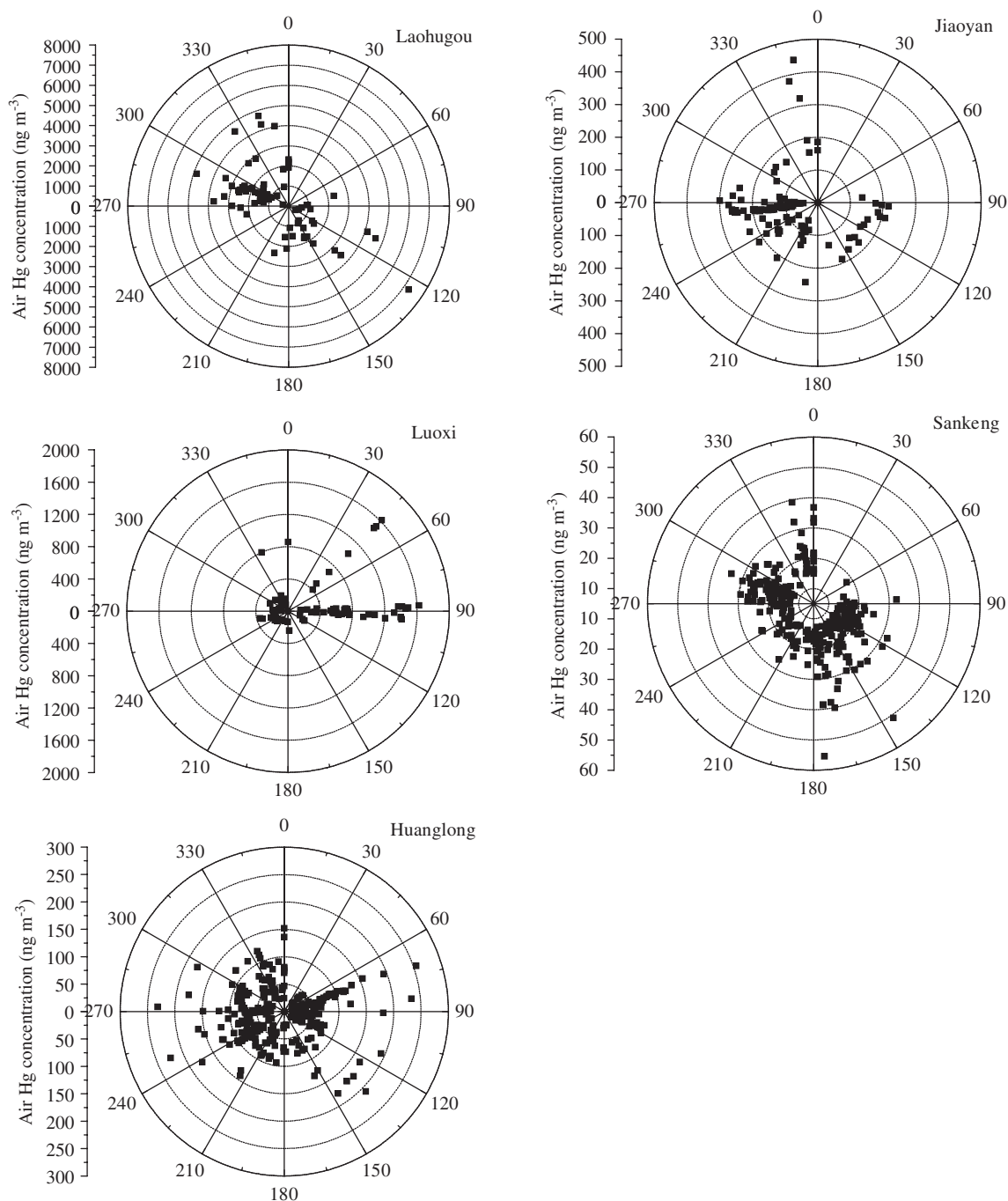
Fig. 4. Plots of air Hg concentration following the change of wind direction at sites Laohugou, Jiaoyan, Luoxi, Sankeng and Huanglong.

relationship between soil Hg concentration and Hg flux was not observed in our study. This intriguing observation implies that highly elevated air Hg concentrations may suppress Hg emission fluxes, which concealed the impact of soil Hg concentration on Hg flux.

3.3. Relationships between Hg flux and meteorological parameters

Field and laboratory studies clearly demonstrated that meteorological parameters, such as solar irradiation, soil temperature and precipitation, play

an important role in Hg emission from soil (e.g. Carpi and Lindberg, 1997; Coolbaugh et al., 2002; Gustin et al., 2000, 2002; Feng et al., 2005; Gillis and Miller, 2000; Lindberg et al., 1999). Through the correlation analysis (SPSS 11.0), the correlations between air/soil Hg fluxes and meteorological parameters were investigated as shown in Table 3.

It is generally believed that solar irradiation could accelerate Hg emission from soil significantly because reactive Hg can be photo-reduced to Hg^0 in soil (Gustin et al., 2002). It was also demonstrated that Hg emission flux could be two times higher when soil and air temperature increased 10 °C (Moore and Carpi, 2005). However, weak correlations between Hg flux and meteorological parameters such as solar radiation, air temperature and soil temperature were observed at site Laohugou and Luoxi, and no significant correlations were found at other three sites, respectively as shown in Table 3. On the one hand, the sampling campaigns were conducted in winter when the solar radiations were weak, air temperatures were low and the relative humidity was high as shown in Table 2. It is obvious that the low intensity of solar radiation and low temperatures will not favor the formation of Hg^0 in soil and subsequently Hg emission from soil. Even though Hg flux from arid soil can be accelerated by watering (Lindberg et al., 1999), Hg emission flux will decrease significantly when the soil

is saturated with water (Gustin and Stamenkovic, 2005). Thus Hg flux cannot be accelerated by high relative humidity because soil was almost saturated in the long-time moisture weather. On the other hand, the highly elevated air Hg concentrations definitely controlled the Hg emission processes from soil enriched with Hg at this specific condition in WMMA as discussed in Section 3.4 (Table 3).

3.4. Relationship between air Hg concentration and Hg flux

From Fig. 2, it could be seen that Hg fluxes varied oppositely corresponding to air Hg concentrations. The relationships between air Hg concentration and Hg flux were investigated (Table 4). The results showed that significantly negative linear

Table 4
Linear relationships between air Hg concentrations and Hg fluxes

Site	Coefficient
Laohugou	$-0.87, p < 0.0001$
Jiaoyan	$-0.78, p < 0.0001$
Luoxi	$-0.81, p < 0.0001$
Sankeng	$-0.79, p < 0.0001$
Huanglong	$-0.36, p < 0.002$

Table 2
Summary of meteorological parameters at all sampling sites in Wuchuan Hg mining area during sampling campaigns

Sampling site	Air temperature (°C)	Soil temperature (°C)	Relative humidity (%)	Solar irradiation (W m^{-2})
Laohugou	1.2 ± 1.3	2.5 ± 2.3	74.1 ± 14.6	20.1 ± 66.9
Jiaoyan	1.7 ± 2.4	4.0 ± 1.9	90.1 ± 10.4	3.0 ± 39.4
Luoxi	7.1 ± 1.4	7.8 ± 1.0	81.8 ± 13.1	30.1 ± 66.6
Sankeng	2.2 ± 2.2	3.3 ± 2.8	63.3 ± 16.6	2.7 ± 52.2
Huanglong	5.3 ± 3.0	6.6 ± 2.2	81.4 ± 14.4	11.0 ± 53.3

All data are shown as mean ± standard deviation.

Table 3
Summary of Pearson correlation coefficients (p is 1-tailed significance) between Hg fluxes and air temperature (AT), soil temperature (ST), relative humidity (RH) and solar irradiation (SI), respectively

Site	Hg flux vs. AT	Hg flux vs. ST	Hg flux vs. RH	Hg flux vs. SI
Laohugou	$0.316, p = 0.003$	$0.278, p = 0.007$	$-0.305, p = 0.004$	$0.322, p = 0.002$
Jiaoyan	$-0.518, p < 0.0001$	$-0.554, p < 0.0001$	$0.590, p < 0.0001$	$-0.191, p = 0.022$
Luoxi	$0.348, p = 0.001$	$0.284, p = 0.005$	$-0.348, p = 0.001$	$0.370, p < 0.0001$
Sankeng	$-0.137, p = 0.083$	$-0.103, p = 0.104$	$0.103, p = 0.046$	$-0.113, p = 0.103$
Huanglong	$-0.326, p = 0.022$	$-0.277, p = 0.020$	$0.138, p < 0.0001$	$-0.136, p < 0.0001$

relationships between air Hg concentrations and Hg fluxes were observed at all sampling sites.

Previous researches found that elevated air Hg concentrations could lead to Hg emission flux decreasing, or negative flux (Engle et al., 2001; Gillis and Miller, 2000; Nacht et al., 2004). Wang et al. (2005) observed totally different correlations between Hg fluxes and air Hg concentrations in daytime and nighttime in Lanmuchang Hg-Tl mining area in Guizhou, China. Hg fluxes significantly positively correlated with air Hg concentrations during daytime, but negatively correlated with air Hg during nighttime. In theory, Hg exchange flux between soil and air is determined by two factors: (i) the rate of Hg diffusion on interface and (ii) the rate of Hg adsorption by soil surface. According to the two-resistance exchange interface model (Zhang et al., 2002), the rate of Hg diffusion is controlled by the gradient of Hg concentration between soil, air and the above air. Thus, when air Hg concentration is elevated, the potential of Hg diffusion from soil to the atmosphere will decrease. This can lead to the rate of Hg diffusion from soil decreasing or Hg diffusing from the ambient air to soil air (negative flux). Synthetically, highly elevated air Hg concentrations can inhibit/decrease Hg emission flux from soil. As described in Section 3.1, air Hg concentrations were elevated by 2–4 orders of magnitude compared to those values observed in pristine areas in Europe and North America because of a huge amount of Hg emission from artisanal Hg smelting activities. Thus, our study further demonstrated that highly elevated air Hg could suppress Hg emission processes even from Hg-enriched soil. At the specific conditions in WMMA, air Hg concentrations play a dominant role in controlling Hg emission from soil.

4. Conclusion

Air Hg concentrations in WMMA were much elevated by 2–4 orders of magnitude compared to the values observed in the background areas in Europe and North America as a result of Hg emission from artisanal Hg smelting activities. WMMA may become an important local and regional Hg emission source to the atmosphere. Massive negative Hg fluxes were measured on Hg-enriched soil surfaces using dynamic flux chamber method and significantly negative correlations between air Hg concentration and Hg flux were observed. Soil may be served as the sink of atmospheric Hg in WMMA under dark, humid and cold weather conditions. Huge fluctuation of Hg fluxes demonstrated that the exchange process of gaseous Hg between soil and air was so dynamic and air Hg concentration was the dominant controlling factor of air/soil Hg exchange in WMMA in cold season.

References

Carpi, A., Lindberg, S., 1997. Sunlight-mediated emission of elemental mercury from soil amended with municipal sewage sludge. Environmental Science & Technology 31, 2085–2091.

Coolbaugh, M.F., Gustin, M.S., Rytuba, J.J., 2002. Annual emissions of mercury to the atmosphere from natural sources in Nevada and California. Environmental Geology 42, 338–349.

Ebinghaus, R., Kock, H.H., Temme, C., Einax, J.W., Löwe, A.G., Richter, A., Burrows, J.P., Schroeder, W.H., 2002a. Antarctic springtime depletion of atmospheric mercury. Environmental Science & Technology 36, 1238–1244.

Ebinghaus, R., Kock, H.H., Coggins, A.M., Spain, T.G., Jennings, S.G., Temme, C., 2002b. Long-term measurements of atmospheric mercury at Mace Head, Irish west coast, between 1995 and 2001. Atmospheric Environment 36, 5267–5276.

Engle, M.A., Gustin, M.S., Zhang, H., 2001. Quantifying natural source mercury emission from the Ivanhoe Mining District, north-central Nevada, USA. Atmospheric Environment 35, 3987–3997.

Feng, X., Chen, Y., Zhu, W., 1996. The fluxes of volatile mercury over soil surface in Guizhou Province. Environmental Sciences 17, 20–25 (in Chinese).

Feng, X.B., Wang, S.F., Qiu, G.G., 2003. Total gaseous mercury emissions from soils in mercury mineralization belt, Southwestern China. Geochimica et Cosmochimica Acta 67, A94.

Feng, X., Yan, H., Wang, S., Qiu, G., Tang, S., Shang, L., Dai, Q., Hou, Y., 2004. Seasonal variation of gaseous mercury exchange rate between air and water surface over Baihua reservoir, Guizhou, China. Atmospheric Environment 38, 4721–4732.

Feng, X., Wang, S., Qiu, G., Hou, Y., Tang, S., 2005. Total gaseous mercury emissions from soil in Guiyang, Guizhou, China. Journal of Geophysical Research, doi:10.1029/2004JD005643.

Fitzgerald, W.F., 1995. Is mercury increasing in the atmosphere? The need for an atmospheric mercury network (AMNET). Water, Air, and Soil Pollution 80, 245–254.

Gillis, A.A., Miller, D.R., 2000. Some local environmental effects on mercury emission and absorption at a soil surface. The Science of the Total Environment 260, 191–200.

Gustin, M.S., 2003. Are mercury emissions from geologic sources significant? A status report. The Science of the Total Environment 304, 153–167.

Gustin, M.S., Stamenkovic, J., 2005. Effect of watering and soil moisture on mercury emission from soils. Biogeochemistry 76, 215–232.

Gustin, M.S., Lindberg, S.E., Austin, K., Coolbaugh, M., Vette, A., Zhang, H., 2000. Assessing the contribution of natural sources to regional atmospheric mercury budgets. The Science of the Total Environment 259, 61–71.

Gustin, M.S., Biester, H., Kim, C.S., 2002. Investigation of the light-enhanced emission of mercury from naturally enriched substrates. Atmospheric Environment 36, 3241–3254.

Li, P., Feng, X., Qiu, G., Jiang, H., Li, Z.G., Fu, X., Bai, W., Sakamoto, M., Liu, X., Yoshida, M., Iwata, T., Wang, D.Y., zhang, C., 2006a. Mercury vapor exposure survey and health impact evaluation of indigenous smelting method workers in Wuchuan district, Guizhou province. Asian Journal of Ecotoxicology 1, 30–34 (in Chinese).

Li, P., Feng, X., Qiu, G., Wang, S., 2006b. Mercury emission from the indigenous method of mercury smelting in Wuchuan mercury mining areas, Guizhou province. Environmental Science 27, 837–840 (in Chinese).

Lindberg, S.E., Zhang, H., Gustin, M., Vette, A., Marsik, F., Owens, J., Casimir, A., Ebinghaus, R., Edwards, G., Fitzgerald, C., Kemp, J., Kock, H.H., London, J., Majewski, M., Poissant, L., Pilote, M., Rasmussen, P., Schaedlich, F., Schneeberger, D., Sommar, J., Turner, R., Wallschläger, D., Xiao, Z., 1999. Increases in mercury emissions from desert soils in response to rainfall and irrigation. Journal of Geophysical Research 104, 21879–21888.

Lindberg, S.E., Zhang, H., Vette, A.F., Gustin, M.S., Barnett, M.O., Kuiken, T., 2002. Dynamic flux chamber measurement of gaseous mercury emission fluxes over soils: Part 2—effect of flushing flow rate and verification of a two-resistance exchange interface simulation model. Atmospheric Environment 36, 847–859.

Mason, R., Fitzgerald, W.F., Morel, M.M., 1994. The biogeochemical cycling of elemental mercury: anthropogenic influences. Geochimica et Cosmochimica Acta 58, 3191–3198.

Moore, C., Carpi, A., 2005. Mechanisms of the emission of mercury from soil: Role of UV radiation. Journal of Geophysical Research, doi:10.1029/2004JD005567.

Nacht, D.M., Gustin, M.S., Engle, M.A., Zehner, R.E., Giglini, A.D., 2004. Atmospheric mercury emissions and speciation at the Sulphur Bank Superfund site, northern California. Environmental Science & Technology 38, 1977–1983.

Pacyna, E.G., Pacyna, J.M., Steenhuisen, F., Wilson, S., 2006. Global anthropogenic mercury emission inventory for 2000. Atmospheric Environment 40, 22, 4048–4063.

Poissant, L., Casimir, A., 1998. Water–air and soil–air exchange rate of total gaseous mercury measured at background sites. Atmospheric Environment 32, 883–893.

Poissant, L., Pilote, M., Beauvais, C., Constant, P., Zhang, H., 2005. A year of continuous measurements of three atmospheric mercury species (GEM,RGM and Hgp) in southern Quebec, Canada. Atmospheric Environment 39, 1275–1287.

Qiu, G.L., Feng, X.B., Wang, S.F., Shang, L.H., 2006. Environmental contamination of mercury from Hg-mining areas in Wuchuan, northeastern Guizhou, China. Environmental Pollution 142, 549–558.

Qu Liya, 2004. A Study on the Prevention and Remedy of Hg-contamination in Guizhou. Guizhou Press, Guiyang.

Schroeder, W.H., Munthe, J., 1998. Atmospheric mercury—an overview. Atmospheric Environment 32, 809–822.

Schroeder, W.H., Anlauf, K.G., Barrie, L.A., Lu, J.Y., Steffen, A., Schneeberger, D.R., Berg, T., 1998. Arctic springtime depletion of mercury. Nature 394, 331–332.

Wallschläger, D., Turner, R.R., London, J., Ebinghaus, R., Kock, H.H., Sommar, J., Xiao, Z., 1999. Factors affecting the measurement of mercury emission from soils with flux chambers. Journal of Geophysical Research 104, 21859–21871.

Wang, S., Feng, X., Qiu, G., 2003. The study of mercury exchange rate between air and soil surface in Hongfeng reservoir region, Guizhou, PR China. Journal of Physics IV France 107, 1357–1360.

Wang, S., Feng, X., Qiu, G., Wei, Z., Xiao, T., 2005. Mercury emission to atmosphere from Lanmuchang Hg-Tl mining area, Southwestern Guizhou, China. Atmospheric Environment 39, 7459–7473.

Xiao, Z., Munthe, J., Schroeder, W.H., Lindqvist, O., 1991. Vertical fluxes of volatile mercury over forest soil and lake surfaces in Sweden. Tellus 43B, 267–279.

Zhang, H., Lindberg, S.E., Marsik, F.J., Keeler, G.J., 2001. Mercury air/surface exchange kinetics of background soils of the Tahquamenon River watershed in the Michigan upper peninsula. Water, Air and Soil Pollution 126, 151–169.

Zhang, H., Lindberg, S.E., Barnett, M.O., Vette, A.F., Gustin, M.S., 2002. Dynamic flux chamber measurement of gaseous mercury emission fluxes over soils. Part 1: simulation of gaseous mercury emissions from soils using a two-resistance exchange interface model. Atmospheric Environment 36, 835–846.