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Measurement and Scaling of Mercury on Soil and Air in a Historical Artisanal Gold Mining Area in Northeastern China

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Abstract: The Jiapigou gold mine area, located in the upper reaches of the Songhua River, was the first and largest artisanal gold mine once in China, and it used to be prominent in two marking years (1870 and 1974). Jiapigou area had a gold mining history of more than 190 years, which was first opened in 1820. Gold extraction with algamation was applied as the dominant method of excavation from 1940 to 2008, and a total of more than 100 t of gold were extracted from the mine using this method and it was estimated that 100–200 t Hg were released, thus causing severe mercury environmental pollution in the mining area. In the experimental campaigns of this study, in situ air and soil Hg concentrations and air-soil Hg fluxes were measured from April 2009 to December 2011. The results showed that in the study area the total gaseous mercury (TGM) concentration exhibited remarkable spatial and temporal distribution patterns, i.e. the TGM gradually decreased following the increase in distance to gold mining sites in space, and the values in spring, summer and autumn were elevated by 1–2 orders of magnitude in comparison with those in winter. Furthermore, at other sampling sites the total soil mercury (TSM) concentration in spring was higher than that in autumn, except for the contrary laws demonstrated at the Erdaogou mining site. However, in spring and winter the Hg flux between air and soil was under the control of different environmental factors, and the characteristics were clear and distinct. In spring the Hg flux between air and soil was directly under the control of solar irradiation, and the releasing process was predominant with a remarkable positive correlation to solar irradiation. Nevertheless, in winter the Hg fluxes were indirectly under the control of solar irradiation, which caused thermal inversion due to the thick snow cover. The depositing process was predominant and the correlations between Hg flux and air temperature was remarkably negative, and there was a positive correlation between Hg flux and solar irradiation.

Keywords: artisanal gold mine; mercury (Hg); algamation; flux; terrain; soil

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1 Introduction

Mercury (Hg) is considered to be a global pollutant, as Hg^0 is the predominant form of atmospheric Hg, which has a long residence time in the atmosphere, lasting from 0.5 to 2 years (Lindqvist et al, 1991; Schroeder and Munthe, 1998). It can be transported and deposited to remote places as far as 1000 km from the source (Ci et

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al., 2016; Han et al., 2017; Nie et al., 2017). Hg is released to the atmosphere from both natural and anthropogenic sources (Li et al., 2009; Fu et al., 2016) and the global anthropogenic Hg emission to the atmosphere in 2000 was estimated to be 2190 t (Pacyna et al., 2006). The annual global natural Hg emissions from ocean and terra firma were estimated to be 1800–5800 t (Li et al., 2009). However, Hg emission flux from natural sources is still not well understood, due to the complexity of the emission processes (Wang et al., 2007; Guédron et al., 2013; Arnold et al., 2018).

The bi-directional flux of mercury (Hg) between terrestrial surfaces and the atmosphere is an important component of the global mercury budget (Eckley et al., 2016). The magnitude of soil Hg emissions is related to the soil Hg concentration (Zehner and Gustin, 2002; Gustin et al., 2003; Nacht et al, 2004; Eckley et al., 2011b; Wang et al. 2016), as well as other soil properties such as moisture content, temperature, mineralogy, disturbance and grain size (Nacht et al., 2004; Lin et al., 2010; Eckley et al., 2011a). Meteorological variables such as solar radiation, air temperature, relative humidity (RH), wind speed/turbulence, and air Hg and oxidant concentrations can also affect soil-air Hg fluxes (Carpi and Lindberg, 1997; Xin and Gustin, 2007; Bargagli, 2016). Several studies have used soil-air Hg chamber flux data to scale-up emission estimates spatially and temporally. Some have focused on scaling over relatively small areas exhibiting anthropogenic impacts, like mining sites (Engle and Gustin, 2002; Eckley et al., 2011b; Kocman and Horvat, 2011; Miller et al., 2011; Eckley et al., 2015).

Environmental problems related to the use of elemental Hg for the recovery of Au and Ag has been known since the ancient Roman times (Strode et al., 2009). Once used all over the world, algamation is a popular artisanal method in gold extraction, as the processes it involves are quite simple. Although artisanal Au mining in many countries has led to Hg pollution of terrestrial and aquatic ecosystems since the 1980s, such mining is still prevalent throughout South America (Brazil, Bolivia, Columbia, Venezuela, Peru, Ecuador, French Guiana, Guyana and Suriname), China, Russia, the Philippines, Indonesia, Thailand, Tanzania and Mexico (Strode et al., 2009). It is estimated that in China about 1/3 of the annual Au production involves the Hg amalgamation techniques used during the 1980s and 1990s (Gunson and Veiga, 2004; Zhou et al., 2017). In 1995, the consumption of elemental Hg in Au mining activities throughout China was estimated to be about 400 t, and the amount of elemental Hg lost to surrounding environments was estimated to be 107 t (García-Sánchez et al., 2006). Although artisanal Au mining activities have been officially prohibited in China since 1996, a number of illegal mines still continued the production in some remote areas (Feng et al., 2006).

Many studies have been conducted to investigate the physical health of Au miners from various provinces of China (Wang et al., 2005, 2010; Feng et al., 2006; Li et al., 2009; Wang et al., 2018), and many researches on the sources of the associated Hg contamination have been reported (Lin et al., 1997; Dai et al., 2003; Gunson and Veiga, 2004; Gabriel and Williamson, 2008; Fu et al., 2008; Wang et al., 2016). However, it has been acknowledged that Hg contamination still exists in the ecosystems of the areas surrounding Au mining sites after the traditional artisanal mining method was forbidden, which requires further study (Eckley et al., 2011b; Miller et al., 2011). It was reported that in downstream waters of a gold field in Northeast China, the greatest contents of Hg, Cu, and Pb were found in aquatic samples collected from Weisha River for aquatic animals (Zhu et al., 2016). Another study showed that there was no health risk from exposure to methylmercury by consuming fish from the Songhua River, demonstrating that mercury contamination of the Songhua River had been effectively controlled by nearly 30 years of environmental governance and natural purification (Zhu et al., 2012). This paper mainly focuses on the analysis of the Hg measurement and scaling characteristics on soil and air in the Jiapigou gold mine area, located in Northeastern China, due to its long-term artisanal gold mining history, and reveals the laws on the migration and distribution of Hg emitted previously into the ambient environmental media in the historical artisanal gold mining activities.

2 Study Area

The Jiapigou gold mine area, the first gold mine opened in China with the largest output and almost ten thousand workers in both 1870 and 1974, is located in the mountainous area of southeastern Jilin Province (127°15′E–127°30′E, 42°50′N–43°00′N) of Northeastern China (Fig. 1). The

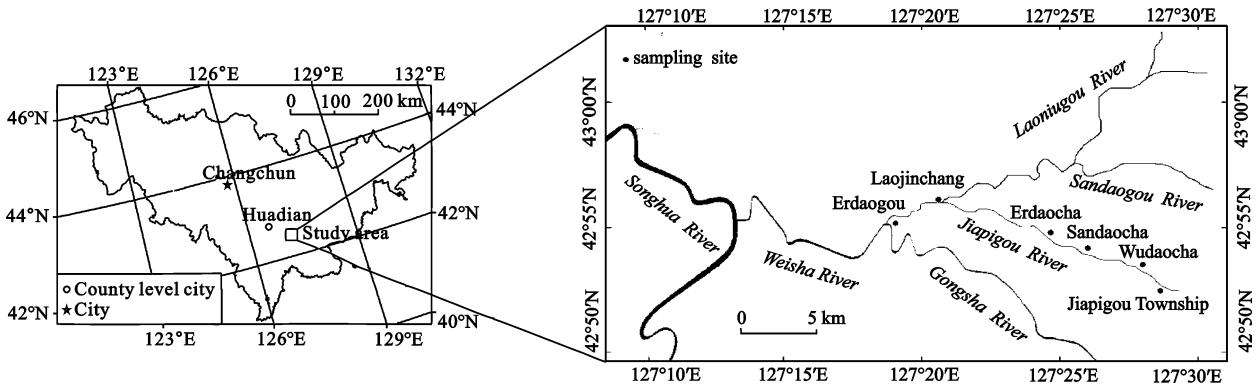

Fig. 1 Sketch map of Jiapigou mining area located on the right bank of Songhua River in Northeast China and the sampling sites

mine is situated on the right bank of the upper Songhua River to the Northwest of the Changbai Mountain, and has an average elevation of 600 m. With a typical temperate zone terrestrial monsoon arid climate, this location is characterized by cold and dry winters and moist and rainy summers. The area's average annual rainfall and annual temperature range from 650 mm to 850 mm and 1.9℃ to 4.4℃, respectively.

The historical documents of Au mining activities at Jiapigou mining area date back to over 1000 years ago. In modern times, placer gold, a branch of various mining categories, was first initiated in Jiapigou area in 1820. Since 1845, primary gold has been discovered and exploited there. As a primary method in the area, gold extraction with algamation was first implemented in 1940, and was not replaced by cyanidation until 2008. Around 2010, seven medium-scale and five small-size gold mine deposits scattered throughout the area, where more than 4000 workers were employed. According to statistics, the annual Au production in the Jiapigou area continuously exceeded two tons. Historically, the Au mills and concentration plants were distributed among the mountains and along several tributary streams of the Songhua River (Fig. 1). The waste water drained from these mills flowed directly into those streams. Artisanal Au mining activities were mainly performed in the valleys and along the streams in the mountainous areas in Jiapigou gold mining area. Around 1870 and 1974, artisanal Au mining activities were quite prominent in the region, in which more than 10 000 people were involved (Wang et al., 2005), and for which elemental Hg was used widely for Au extraction. In China Hg loss is estimated to be 0.68 g of elemental Hg per gram of Au extracted from large-scale Au mining, and in artisanal Au mining about 15 g (Leiva and Morales, 2013). The research results indicate that before the 1980s the grade of gold ores ranged from 2.26–6.80 g/t contained in the superficial layer ores. However, thereafter it is shown that the figure of the deeper layer may range from 6.2–40.0 g/t. It is estimated that the Hg loss can reach almost 10 g of elemental Hg extracted from per 1000 kg of ore. It was recorded that much more than 100 t of Au had been extracted by the method of gold extraction with algamation (Feng et al., 2006). Therefore, according to the Hg releasing factor 1.0–2.0 in artisanal Au mining (Leiva and Morales, 2013), it was estimated that 100–200 t elemental Hg were released into the environment medium through the local gold mining activities.

3 Materials and Methods

3.1 Sampling sites

From April 2009 to December 2011, in the mining area along and down the Jiapigou River, six locations, namely Jiapigou Township, Wudaocha, Sandaocha, Erdaocha, Laojinchang and Erdaogou, were selected as sampling sites (Fig. 1). The six sampling sites were confirmed on the basis that in history along the Jiapigou River among the mountains and valleys there located a large number of gold mines and gold extraction workshops and the sixes were the famous and representative sites, reflecting the applications of algamation and the present Hg contamination situation. Total gaseous mercury (TGM) concentrations and total soil mercury (TSM) concentrations were determined seasonally. Among those, four sample sites deserve mentioning. Erdaocha, featured with woodland, is the place where

Au mining activities have been conducted. Jiapigou Township used to be seated by the old town government and Benqu Au mine is located. Laojinchang is where the new town government located. Erdaogou is mainly used for farmlands. Furthermore, air/soil mercury fluxes were measured at the four sampling sites, during spring and winter, ruling out vegetation influence.

3.2 Air samples and analyses

The TGM were monitored with an automated Hg vapor detector produced by LUMEX (RA-915$^+$, Russia). As a real-time Hg detector, the RA-915$^+$ was based on the Zeeman cold vapor absorption spectrometry technique, with a time resolution of 1 s. It was calibrated with an internal Hg vapor source. Its real-time detection limit and dynamic detection extent were 2 ng/m^3 and 5 ng/m^3 – 2×10^4 ng/m^3, respectively. The average TGM concentrations were recorded every 3 mins in each sampling group, and at each site the measurements were carried out for at least 20 groups. The average values of TGM in the ambient air at each site were calculated based on the total measurements.

3.3 Soil samples and analyses

At the sampling sites, surface soil samples were composite from three to five sub-samples collected from several localities within an area of 1 m^2 using a pre-cleaned plastic shovel. All of the soil samples were collected and stored in sealed polyethylene bags to avoid cross contamination. In the laboratory, all samples were air-dried, ground in a ceramic disc mill, and sieved (mesh = 0.149 mm). The soil samples were digested with an acid mixture of H_2SO_4/HNO_3/$KMnO_4$ for total Hg analysis and determined by atomic fluorescence spectrophotometry (BTI, CHN, AFS-920). The method described in *Soil Quality-Determination of Total Mercury-Cold Atomic Absorption Spectrophotometry* with a resolution of 0.005 mg/kg was used to determine the TSM of the samples, and the mean value of three parallel figures measured in every sample was calculated to represent TSM (Feng et al., 2006). The organic matter contents and pH of soil samples were determined by the potassium dichromate method (H_2SO_4-$K_2Cr_2O_4$) (Feng et al., 2006) and electrode method (CHN, PHSJ-4A), respectively.

Quality control for the Hg determinations was addressed with method blanks, blank spikes, certified reference materials (GBW07405; CRM580), and blind duplicates. The limit of determination was 0.01 ng/g for total Hg. The average total Hg concentration of the geological standard GBW07405 was (0.30 ± 0.01) mg/kg (n = 5), with a range from 0.29 mg/kg to 0.31mg/kg, which was comparable with the certified value of (0.29 ± 0.04) mg/kg. The relative percentage difference was <8.5% for the total Hg obtained from the analysis of the duplicate samples.

3.4 Hg flux measurement

In this experiment, the dynamic flux chamber (DFC) method, which has previously been described in many documents (Lindberg and Zhang, 2000; Feng et al., 2003; Gustin, 2003; Wang et al., 2007), was used to measure the Hg fluxes. A semi-cylinder, open-bottom chamber made of polyfluortetraethylene (PTFE) (Ø = 20×60 cm^2) was used during the sampling process. The Hg flux from the soil exposed in the chamber was calculated using Equ. (1):

$$F = (C_o - C_i) \times Q \div A \quad (1)$$

where F represents the flux of gaseous Hg, consisting mostly of Hg^0 (Schroeder and Munthe, 1998) (ng/(m^2·h)); C_o and C_i are concentrations of Hg in the air of the outlet and inlet of the chamber (ng/m^3), respectively; in this experiment, A refers to the bottom surface area of the chamber ,which is equal to 0.12 m^2 specifically and Q is the flushing flow rate through the chamber, equal to 0.9 m^3/h. The TGM was measured twice in the ambient air entering the inlet of the chamber, and twice in the air exiting through the outlet of the chamber, using an automated air Hg analyzer (RA-915$^+$) with a 3-min sampling time. Switching from the inlet to the outlet of the chamber every 6 minutes was realized by manual control. A high flushing flow rate of 15 L /min (0.9 m^3/h) was adopted to prevent the possibility of underestimating the Hg flux at low flushing flow rates (Lindberg, 1998; Gustin et al., 2003; Feng et al., 2004). The mean values of two C_o and four C_i before and after the C_o were used to sequentially calculate the Hg flux between soil and air. One Hg flux datum was obtained every 12 minutes. The positive and negative results calculated from Eq. (1) represented the Hg emission from soil and air Hg dry deposition, respectively (Feng et al., 2003; Wang et al., 2007).

The meteorological parameters, including air and soil

temperature, solar irradiation and relative humidity, were monitored through a portable meteorology comprehensive observation instrument (DZM2-1, China), with a time resolution of 3 minutes.

The measurements of the DFC blanks were performed before and after the measurement campaigns, by covering the chamber on a PTFE plate. The DFC blanks ((2.0 ± 1.6) ng/(m^2·h), n = 12) were not subtracted from the results of the fluxes, as they were negligible compared to the in situ fluxes. A standard soil sample GBW-07405 (GSS-5) was used to perform QA/QC, and the average total Hg concentration of the geological standard of GBW-07405 was ((0.30 ± 0.01) μg/g, n = 8), which was comparable with certified value of (0.290 ± 0.040) μg/g. The precision of the method obtained from replicate analysis was less than 5%. The results demonstrated that the pretreatment procedures were capable of quantitatively recovering the Hg from the soil samples.

4 Results

4.1 Determination of total gaseous mercury

The summary of TGM measured seasonally from all sampling sites is shown in Table 1. The data indicate that the mean TGM was (55 ± 21) ng/m^3 in the research area during the sampling period, and those in Jiapigou Township, Wudaocha, Sandaocha, Erdaocha, Laojinchang and Erdaogou were (56 ± 33) ng/m^3, (33 ± 13) ng/m^3, (116 ± 28) ng/m^3, (94 ± 33) ng/m^3, (22 ± 8) ng/m^3 and (24 ± 10) ng/m^3, respectively. These values were obviously higher than those shown in the previous study (Wan et al., 2009), in which it was reported that the annual mean value of TGM was (3.22 ± 1.78) ng/m^3 on Changbai Mountain, the highest mountain in Northeast China. The monitoring values in this campaign were also much higher than the TGM background value (1.5–2.0 ng/m^3) of the Northern Hemisphere (Ebinghaus et al., 2002; Lamborg et al., 2002; Steffen et al., 2002).

Table 1 Summary of total gaseous mecury in Jiapigou gold mining area of Northeast China from Apr.2009 to Dec.2011

Sampling site	Season	n	range (ng/m^3)	Mean (ng/m^3)	SD (ng/m^3)	Coefficient of variance
Jiapigou Township 42°52′00″N; 127°29′17″E; 580.0 m	Spring	148	28–1004	77	59	0.76
	Summer	45	25–794	129	55	0.43
	Autumn	45	3–119	15	18	1.20
	Winter	36	1–6	2	1	0.57
Wudaocha 42°52′11″N; 127°27′53″E; 493.9 m	Spring	36	26–132	65	32	0.49
	Summer	25	8–46	28	13	0.45
	Autumn	20	20–41	37	6	0.17
	Winter	36	1–6	3	2	0.58
Sandaocha 42°52′36″N; 127°26′12″E; 441.2 m	Spring	36	38–512	234	38	0.16
	Summer	—	—	—	—	—
	Autumn	25	48–307	109	45	0.41
	Winter	36	2–7	4	1	0.34
Erdaocha 42°52′59″N; 127°25′32″E; 425.0 m	Spring	39	19–160	72	42	0.58
	Summer	30	15–131	49	23	0.48
	Autumn	30	71–324	248	62	0.25
	Winter	36	1–15	5	4	0.78
Laojinchang 42°53′23″N; 127°21′14″E; 340.5 m	Spring	114	5–83	32	20	0.63
	Summer	25	4–70	25	8	0.33
	Autumn	20	23–25	24	0	0.02
	Winter	36	4–11	8	2	0.25
Erdaogou 42°53′11″N; 127°18′33″E; 316.3 m	Spring	57	5–137	39	26	0.66
	Summer	20	16–176	37	12	0.34
	Autumn	45	4–29	12	3	0.22
	Winter	36	7–11	9	1	0.12

4.2 Determination of total soil mercury

In Table 2, the data of the soil Hg concentrations in spring and autumn is shown as follows. With the exception that the TSM of Erdaogou in autumn was higher than that in spring, the values of TSM from all the other sites were higher in spring than those of autumn. The mean value of TSM in the research area was (0.913 ± 0.041) mg/kg, which was higher than the value of 0.25 mg/kg observed in Changbai Mountain, China, as well as the average background value of 0.041mg/kg in Jilin Province, where the research area was located (Steffen et al., 2002). But it was lower than the mean value (4.7 ± 1.8) mg/kg in the Tongguan Au mining area, Shaanxi Province, which was another well-known large-scale artisanal Au mine area in China and the soil was significantly contaminated due to Au mining activities similarly (Feng et al., 2006).

4.3 Determination of Hg flux between soil and air

Among the six sites, the statistical summary of Hg flux between soil and air in winter and spring at the four representative ones, that is Jiapigou Township, Erdaocha, Laojinchang and Erdaogou is shown in Table 3. The mean flux values were (7 ± 46) ng/(m^2·h), (23 ± 171) ng/(m^2·h), (129 ± 496) ng/(m^2·h) and (99 ± 44) ng/(m^2·h) in spring, and (–13 ± 7) ng/(m^2·h), (–24 ± 18) ng/(m^2·h), (–21 ± 16) ng/(m^2·h) and (–4 ± 9) ng/(m^2·h) in autumn, respectively. In spring the Hg flux was mainly released from the soil to air, but in winter the reverse was true, deposited primarily from the air to soil. According to flux data illustrated in Table 3 and the area 50–150 km^2 of Jiapigou gold mine, it was estimated that around 8–24 t Hg were released from the contaminated soil by Au mining activities (for 7 months) and 1–3 t Hg were deposited to the snow cover in winter (for 5 months), and more details on net Hg output from the earth surface to the air in artisanal Au mines need further study.

Table 2 Statistical summary of total soil mecury in Jiapigou gold mining area of Northeast China in Apr.2009–Dec.2011 and soil properties

Sampling sites	Spring			Autumn		
	Mean value of TSMs (μg/g)	pH	Organic matter content (g/kg)	Mean value of TSMs (μg/g)	pH	Organic matter content (g/kg)
Jiapigou Township	1.474 ± 0.000	5.88	33.9 ± 1.3	0.696 ± 0.025	5.89	33.8 ± 1.4
Wudaocha	0.945 ± 0.000	5.99	46.9 ± 1.7	0.163 ± 0.018	6.01	45.6 ± 1.6
Sandaocha	0.507 ± 0.050	6.05	50.4 ± 1.9	0.274 ± 0.030	6.03	48.7 ± 1.9
Erdaocha	1.863 ± 0.098	6.07	78.9 ± 2.3	1.713 ± 0.056	6.07	80.4 ± 2.2
Laojinchang	0.726 ± 0.007	5.98	80.7 ± 2.3	0.102 ± 0.014	6.05	82.3 ± 2.4
Erdaogou	0.687 ± 0.022	6.41	104.7 ± 2.9	0.826 ± 0.053	6.44	105.1 ± 2.7

Note: n=32.

Table 3 Statistical summary of Hg flux between soil and air (ng/(m^2·h))

Sampling site	Sampling time	Mean	Max	Min	SD	Release/balance/deposit
Jiapigou Township	2009-04-08 T7:00–19:00	7	128	–139	46	33/3/24
	2011-12-13 T7:00–17:00	–13	0	–38	7	0/4/46
Erdaocha	2009-04-09 T7:00–19:00	23	585	–443	171	30/0/30
	2011-12-14 T7:00–17:00	–24	0	–98	18	0/2/48
Laojinchang	2009-04-10 T7:00–19:00	129	1400	–1420	496	31/0/29
	2011-12-15 T7:00–17:00	–21	0	–68	16	0/1/49
Erdaogou	2009-04-11 T7:00–19:00	99	173	20	44	60/0/0
	2011-12-16 T7:00–17:00	–4	15	–23	9	0/13/37

Notes: n=60; SD, stand difference

5 Discussions

5.1 Characteristics of total gaseous mercury

The TGM decreased with the distance increasing toward the Benqu gold mine in Jiapigou Township and Sandaocha gold mine. With distances of 20 km to Benqu and 15 km to Sandaocha, Erdaogou is located in the outermost area, where the mean TGM was (24 ± 10) ng/m^3, only half of the mean value in the research area. Compared with TGM 29 ng/m^3 in the point with a distance of 1 km in the down wind direction to the gold mining mill in Tongguan Township of Shanxi Province, as reported by Feng (Feng et al., 2006), in Jiapigou gold mining area it was shown that the air had been severely polluted by Hg.

In terms of temporal distribution, in spring, summer and autumn the values of TGM were elevated by 1–2 orders of magnitude in comparison with those in winter. Jiapigou Township is situated in the center of the gold mining area, and Erdaogou on the outlying edge of the research area. Moreover, from Jiapigou Township to Erdaogou the elevation decreased from 580.0 m to 316.3 m, and alternating valleys in the terrain caused the air currents to undergo irregular motions in all directions near the earth surface to a certain extent. It is shown in Table 1 that in and around the gold mining area the values of TGM did not observe clear change patterns in fluctuation and demonstration. In two of the mining gold deposits of Jiapigou area, Benqu and Sandaocha, TGM presented high values in summer and autumn, which was conjectured to be closely related to the dominant wind direction in seasons and irregular air current motions caused by the special terrain of the area. However, in winter the TGM at all sampling sites ranged around low concentration levels and were similar to each other. The values of TGM at Laojinchang and Erdaogou described the following temporal distribution: spring > summer > autumn > winter, which was estimated to be related to both the snow cover in winter that hindered the Hg flux between the air and soil, and the distance in between since the two points were located in relatively outlying spaces.

Historically, primary gold had been explored in the alternating valleys of the entire research area, but only the gold mine deposits of Jiapigou Township (Benqu mine) and Sandaocha remain today. Although the extraction gold method of algamation and floatation used since 1940 was replaced by all-slime cyanidation in 2008, according to the information presented in Jiapigou-gold-mine's official homepage, gold extraction with algamation application in almost 7 decades have led to a great amount of Hg being released into the environmental medium. After 2008, the soil previously contaminated by gold extraction has been the source of Hg release into the air in this area, and in recent years recycling of gold tailing and exploration of ore spoils have also contributed to the TGM. In winter the seasonal snow cover prevented the Hg flux (migration) between air and soil, which was the main reason for the low TGM concentration level in comparison with those in other seasons, and Hg released from bare contaminated soil in spring and vegetation in summer and autumn was the major contribution to the TGM.

5.2 Characteristics of total soil mercury

During the process of gold extraction with algamation, Hg entered the environment through exhaust gas and waste water, and then gradually was accumulated in the environmental medium. The research data showed that in the gold mining area the TSM decreased with the distance increasing from the center to the outlying edge of the research area. In addition, during early spring the snow from the winter gradually melted and the snow-water carried Hg in the air deposited into the soil, leading to an increase in the soil Hg concentration. Previous researchers found out that in artisanal Au mine the total Hg of the surface soil most likely derived from atmospheric Hg deposition (Dai et al., 2003). However, under the effects of rain washing and eluviating in the rainy season, Hg was carried by rain-water beneath the soil surface layer, after which it flowed down to the streams at the base of the valley, and the continual rainfall process caused the TSM in autumn to decrease remarkably. Furthermore, the process of matter gathering from higher to lower terrains led to the value of TSM of the lowest sampling site Erdaogou to be higher in autumn than that in spring (Table 2).

According to Newton (Newton et al., 1976), the maximum absorption amount of Hg on the surface of Bentonite ranged from pH 4.5 to 5.5, and the similar report showed that the maximum of absorption amount of additive Hg to soil occurred from pH 3.0 to 5.0 (Yin et al., 1996). It was found that the affinity of Hg and humus decreased following the pH increasing and the

higher the pH was, the fewer the absorption amount of Hg presented (Huljuv,1986; Kerndorff and Schnitzer, 1980). In this study through analyzing the types of soil in the six sampling sites, the author found out they were featured with the newly formed alluvial soil and the origin of the soils derived from the dark brown forest soil in the forests spreading over the mountains in Jiapigou gold mining area. Furthermore, the TSM concentrations and the pH and organic matter contents did not correlate with each other. In addition, the accumulated tendency of alluvial deposits from high to low elevation performed significantly (Table 2), thus indicating the TSM transferred with the processes of soils erosion and deposition in the mountainous Au mining area.

5.3 Characteristics of Hg flux between soil and air

In the research area, under the impact of the terrain of the low hills and foothills and crossbar ravines, the solar irradiation intensity reaches the Earth's surface unevenly and the air warming process occurs in the afternoon, at which time the Hg flux was released from the soil in Jiapigou Township, Erdaocha, Laojinchang and Erdaogou and the values reached the maximum of the whole day, i.e. 128 ng/(m^2·h), 585 ng/(m^2·h), 1400 ng/(m^2·h) and 173 ng/(m^2·h)[1], respectively (Fig. 2 and Table 3). In spring there were three diurnal fluctuation patterns of the Hg flux at the interface between the soil and air at all of the sampling sites. The respective patterns were as follows: 1) Jiapigou Township pattern. The Hg flux level continuously ranged within a broad wave motion and the two processes of release and deposit remained in approximate balance, which was due to the fact that sampling site was located at the base of the basin surrounded by mountains, and the closed terrain conditions limited the diffusion of the atmospheric Hg. 2) Erdaocha and Laojinchang pattern. In this pattern there was a distinct inflexion in time, before which the Hg fluxes were in narrow wave motions, and once they surpassed a certain point they changed into a broad wave motion. Due to the fact that Laojinchang and Erdaocha were situated at the base of the narrow valley, under the impact of the surrounding vegetation and particular terrain, only several hours of direct solar irradiation could reach the surface per day. The inflexion in time appears to follow the direct solar irradiation intensity change. 3) Erdaogou pattern. There was a single

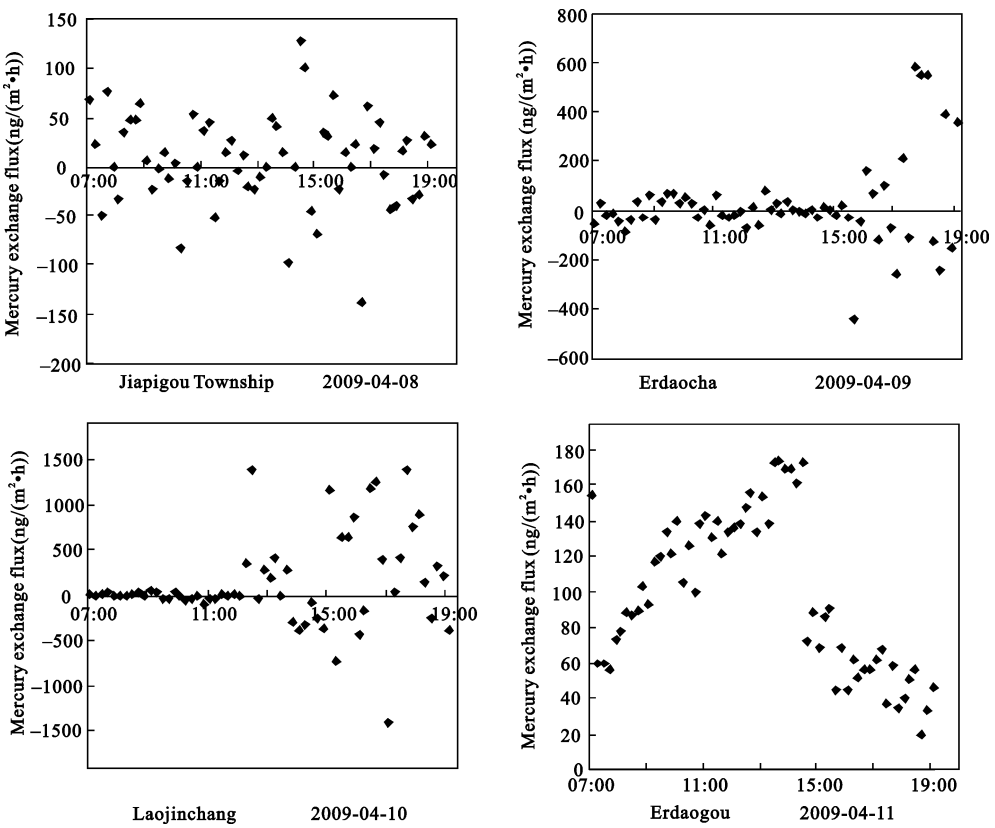

Fig. 2 Hg flux between soil and air in spring

peak of Hg flux between the soil and air and the typical diurnal Hg flux was released from the soil to air. The peak value appeared in the afternoon after the highest air temperature occurred, and the Hg flux level showed a clear change from the fresh to moderate. Located in an open washland area, the soil after sunrise in Erdaogou was exposed to intensive direct solar irradiation, and thus the site presented a pattern that was in accordance with previous reports (Gustin, 2003; Gunson and Veiga, 2004; Feng et al., 2006; García-Sánchez et al., 2006; Wang et al., 2010)

The Hg fluxes deposited from the air to the soil (snow surface) at all sampling sites during the period of research are shown in Table 3. In the gold mining area during winter the Earth's surface was covered by homogeneous seasonal snow cover for 4–5 months. Generally, the snow surface reflects 85%–95% of the solar short-wave radiation and only the infrared radiation can be absorbed, with a heat radiant emissivity of 0.98–0.99, which is similar to black body radiation. Therefore this snow cover led to the remarkable descent of temperature near the Earth through the cold underlying surface inducing temperature inversion. This temperature inversion resulting from the snow cover caused the air Hg to accumulate and a clear Hg deposit process from air to the Earth's surface (snow surface) occurred.

In Fig. 3, it can be seen that there was a remarkable low value distribution on the Hg flux in the daytime which appeared in accordance with the maximum of solar irradiation intensity. The stronger the solar irradiation was, the thicker the temperature inversion became, resulting in the Hg flux continuing to intensively deposit from the air to the Earth's surface.

5.4 Relationships between Hg flux and impact factors

Field and laboratory studies clearly demonstrated that meteorological parameters, such as solar irradiation, soil temperature and precipitation play important roles in Hg emission from contaminated soil (Kerndorff and Schnitzer, 1980; Carpi and Lindberg, 1997; Gillis and Miller, 2000; Gustin et al., 2000; Feng et al., 2005). It is generally believed that solar irradiation can accelerate

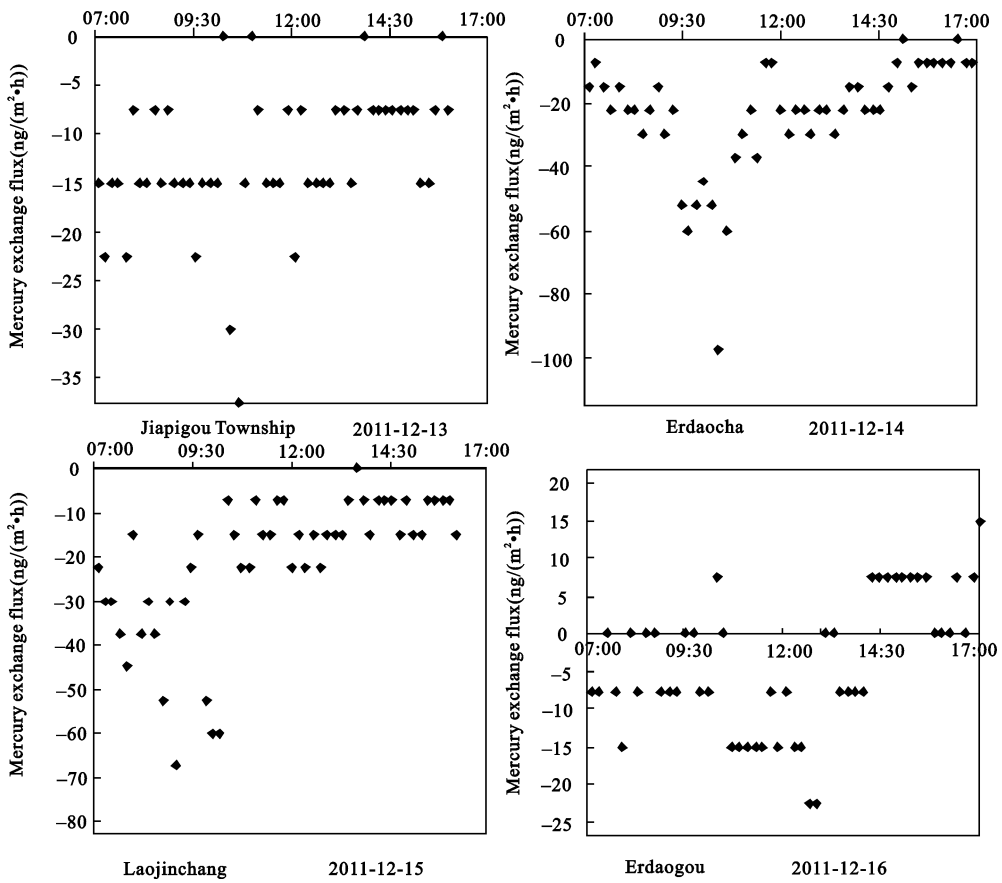

Fig. 3 Hg flux between soil and air in winter

Hg emission from soil significantly, because reactive Hg can be photo-reduced to Hg 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 by 10℃ (Moore and Carpi, 2005). In addition, 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 (Gustin, 2003; Nacht et al., 2004). Therefore, in the study of non snow-covering in spring (Table 4) and snow-covering in winter (Table 5), the correlations between air/soil Hg fluxes and impact parameters were investigated in spring and winter as shown in Table 4 and Table 5.

In spring, it was shown in Table 4 that there was a remarkable positive correlation between the Hg flux and solar irradiation at all four sites and no correlation can be observed between the Hg flux and TGM, the Hg flux and air temperature or the Hg flux and soil temperature in Jiapigou Township, Erdaocha and Laojinchang. In addition, in Erdaogou the Hg flux had a remarkable positive correlation with the air temperature and soil temperature. In spring when the solar irradiation strengthened the Hg photo-reduction effect in the naked soil of the Earth's surface, the Hg flux showed release tendency from the soil to the air, which was reinforced at the same time. However, due to the impact of the fluctuations of the local terrain, the air current near to the Earth's surface appeared to show irregular movements, thus leading to the three special patterns of Hg flux between the soil and air, as shown in Fig. 3.

In winter, at the four sites a clear negative linear correlativity between the Hg flux and TGM and air temperature can be seen, and a positive linear correlation between Hg flux and solar irradiation appeared (Table 5). In winter homogeneous snow covered the Earth's surface and caused temperature inversion to occur near the surface, due to the coldness underlying snow, and with the solar irradiation intensity strengthening, the air Hg was accumulated near the Earth's surface and then deposited there gradually. Following the increase of solar irradiation, the air Hg under the temperature inversion received more gaseous Hg supplements from its upper air layer, thus aggravating the deposit process, which gradually became weaker until the temperature inversion disappeared, as shown in Fig. 3.

6 Conclusions

The Jiapigou gold mining area is located in a mountainous area of Northeast China, where clear patterns of spatial and temporal distribution on TGM can be seen. In terms of spatial distribution, the TGM decreased following the increase in distance to gold mining sites centered in Jiapigou Township (the Benqu mine) and the Sandaocha gold mine, and in Erdaocha, with a distance of 20 km to Jiapigou Township, the TGM decreased to half of the mean value of the research area. In terms of temporal distribution, the values of TGM of all the sampling sites in spring, summer and autumn were elevated by 1–2 orders of magnitude compared to those in winter. In the other sampling sites, the values of TSM in spring were higher than those in autumn, with the exception of Erdaogou, where the reverse was true. However, in spring and winter the Hg fluxes between the air and soil were under different controls of environmental factors, and the characteristics were clear and distinct. In spring the Hg fluxes were directly under the control of solar

Table 4 Pearson's correlation coefficient and *P* value between Hg flux and impact factors in spring

Factors	Jiapigou Township	Erdaocha	Laojinchang	Erdaogou
Air Hg concentration	–0.403, $P = 0.021$	–0.213, $P = 0.010$	0.415, P=0.001	0.244, $P = 0.007$
Solar irradiation	0.799, $P = 0.003$	0.802, $P = 0.001$	0.881, P=0.002	0.723, $P = 0.000$
Air temperature	–0.062, $P = 0.064$	–0.162, $P = 0.890$	–0.162, P=0.081	0.854, $P = 0.000$
Soil temperature	–0.032, $P = 0.061$	–0.133, $P = 0.608$	0.063, P=0.073	0.539, $P = 0.000$

Table 5 Pearson's correlation coefficient and *P* values between Hg flux and impact factors in winter

Factors	Jiapigou Township	Erdaocha	Laojinchang	Erdaogou
Air Hg concentration	–0.770, $P = 0.011$	–0.832, $P = 0.006$	–0.865, $P = 0.014$	–0.661, $P = 0.008$
Solar irradiation	0.771, $P = 0.009$	0.714, $P = 0.003$	0.742, $P = 0.001$	0.681, $P = 0.003$
Air temperature	–0.698, $P = 0.002$	–0.521, $P = 0.000$	–0.673, $P = 0.08$	–0.627, $P = 0.014$

irradiation, and the release process was predominant and had remarkable positive correlation with solar irradiation. However, in winter the Hg fluxes were indirectly under the control of solar irradiation, which caused thermal inversion near the Earth's surface due to the thick snow cover. The depositing process was predominant and the correlations between Hg flux and TGM and air temperature were significantly negative, and a clear positive correlation between Hg flux and solar irradiation can be found. In conclusion, long-term and large-scale Au mining activities using elemental Hg amalgamation techniques in Jiapigou mining area of Northeast China have resulted in local environmental Hg contamination. Residents in these Au mill areas were exposed to concentrations of Hg vapor from the soil contaminated by mining, which thus exerted potential adverse impact on human health.

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