

Distribution and assessment of residual mercury from gold mining in Changbai Mountain Range Northeastern China

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Abstract. Gold mining was first initiated in Jiapigou area, Huadian city of Northeastern China about 200 years ago. Before 2006, the mercury amalgamation technique was used in the gold mining process, which led to severe mercury contamination. The aim of this paper is to explore the influences of residual mercury on the environment media after eliminating the amalgamation process to extract gold. The mercury concentrations of the atmosphere and the soil were determined in autumn of 2011 and spring of 2012. The soil environmental quality was assessed by the index of geoaccumulation. The results indicated that the maximum value of gaseous mercury was $25\text{ng}\cdot\text{m}^{-3}$ in autumn and $19.5\text{ng}\cdot\text{m}^{-3}$ in spring; the maximum value of mercury in the soil was $2.06\text{mg}\cdot\text{kg}^{-1}$ in autumn and $2.51\text{mg}\cdot\text{kg}^{-1}$ in spring. It can be seen that the peak concentrations of the gaseous mercury happened at the gold mine area and tailings, while the peak mercury concentrations in the soil were located at the places near the mining sites and the residential area in the valley. Furthermore, the regression analysis of the total mercury contents between the atmosphere and the soil showed a significant correlation, which indicated that there was certain circulation of the mercury between the regional atmosphere and soil. In general, after the elimination of the amalgamation technique in gold extraction, the distance to the mercury source, the special conditions of hilly weather and landforms and the mercury exchange flux are the main factors of mercury contamination.

1. Introduction

The accumulation, migration and circulation of mercury in the environment could contaminate the air, water and soil, and cause problems for human health. After the Minamata disease event of Japan, poison accidents of mercury and methyl mercury to organisms including humans have recurred frequently, which has become the focus of research [1-3]. The mercury released in the gold extraction process by the amalgamation technique was the man-made source of mercury pollution in the gold mining area [4]. The mercury amalgamation technique is relatively inexpensive and readily available, so the overwhelming majority of artisanal and small-scale gold miners utilize mercury to separate gold from the unwanted gangue materials [5, 6].

Miners separate the gold from the amalgam by “burning” the amalgam, volatilizing the elemental mercury and dispersing it throughout mining camps. Then, the mercury continually enters into the soil, water and other environmental media near the mines, which becomes a threat to the local animal, plant



and human communities. Some countries ban mercury use in artisanal and small-scale gold mining (ASGM) outright [7, 8], but some less developed areas still keep using this technology. One good example is that the privately owned Clean Tech Mine in the Manica Province in Mozambique has eliminated the use of mercury at their mine. Instead, gold is isolated by centrifugation and magnetic removal of gangue materials [9]. There are still many small-scale/artisanal gold mines that use the mercury amalgamation technique. However, the mercury contamination in the ASGM area and its surrounding area caused by long-term gold mining activity should not be neglected [10, 11]. As a result, the remaining mercury pollution of the gold mine area has gradually aroused great attention in recent years [12-14]. The gold mining has caused severe pollution in the local environment and a big threat to the human health of the local mine area in South America [15, 16], Africa [17] and Asia [18].

Mercury releases from Artisanal and small scale gold mining (ASGM) were estimated to be about 1400 tonnes/year, making it the largest global demand sector for mercury. This sector produces about 12-15% of the world's gold. An estimated 10-15 million miners, including 4-5 million women and children, are involved in the sector [7]. The study of Lecce et al. [10] on historic gold mining in North Carolina, USA showed that the mercury concentrations in the channel and floodplain sediment downstream from the Gold Hill mining district ranged from 0.01 to 2.2mg•kg⁻¹, with a maximum concentration more than 35 times background levels. Balzino et al. [19] suggested that contrary to common belief, Hg was recovered from the bottom of the river instead of being released into it. The total Hg-mass balance is actually closed with a positive recovery and with an emission factor. Despite this result, Hg continues to be volatilized into the atmosphere in the form of vapors. The investigation by Saiki et al. [20] showed that the mercury and methyl mercury concentration of the samples from water, soil and sediment in the downstream area of the Sierra Nevada foothill Reservoir are much higher than they are upstream [20], so gold mine areas often became important pollution sources in these areas. Egler et al. [21] studied the total mercury concentration in soil and vegetables and the mercury concentration relationship between soil and vegetables in two small gold mine areas, São Chico and Creporizinho of Amazon, Brazil. They found that the total mercury concentrations of the soil samples in the two areas were all above the local background value, and the total mercury of the edible parts of the vegetables in São Chico was close to the standard of Food and Agricultural Organization/World Health Organization/JECFA [21].

Lacerda [22] argued that most mercury was discharged into the atmosphere due to the uncontrolled burning of the amalgam and bullion smelting in small-scale gold mining. China is rich in gold resources, and gold mining has a long history, but in the past, the mercury amalgamation technique was widely used in the extraction of the primary gold. The reports on gold mine mercury contamination were mainly concentrated on the Shanxi areas. The study of Feng et al. [23] on the gold mine area in Tongguan, Shanxi, showed that the total gaseous mercury (TGM) in the environmental atmosphere was severely higher than the background concentration, and its average TGM concentration reached 18,000 ng•m⁻³. The largest mercury concentration in the soil was 19.50mg•kg⁻¹, but the mercury concentration in the background area was only 0.25mg•kg⁻¹. The total mercury concentration range in vegetables and wheat was from 42mg•kg⁻¹ to 640mg•kg⁻¹, which were all beyond the upper limit of the standard value of Chinese vegetables (0.01mg•kg⁻¹) and food fish (0.02mg•kg⁻¹) [23].

There has been a long history of gold mining in Northeastern China, and most gold mines were located in the mountainous areas, such as the Jiapigou gold mining area, which is located in the upper reaches of the Songhua River, Changbai Mountain Range. In this area, mercury contamination was serious in ambient air, water and sediment. Some research of the mercury pollution investigations had already been done in this area [24] before the ending of amalgamation use, but there is no deeper discussion of the contamination at the present situation and the control measures after the elimination of the mercury amalgamation technique in the gold mine area. Based on the regional gold mining features and the terrain and weather conditions, this article studied the current distribution features of mercury concentrations in the atmosphere and soil in this region. Furthermore, the reasons of the formation of the mercury contamination in the air/soil of the area and the mechanism of mercury

migration and circulation in the environment were discussed. Finally, the geoaccumulation index was used to assess the local soil environmental quality. The study provides the scientific basis to understand more about the characteristics of gold mine mercury contamination, as well as methods to control this type of regional mercury contamination.

2. Study area and methods

2.1. Study area

The Jiapigou gold mine area in Jiapigou Town, Huadian City, Jilin Province, Northeastern China was selected as the study area, which is located in the northwest of Changbai Mountain (42°41'–43°00'N and 127°15'–127°30'E). As a typical hilly area, the topography of this area is high in the southeast and low in the northwest. There are many valleys and channels in the area, and most of the landforms are two mountains with one valley in the middle. There are more than 20 mountains that are above 450 m in the region, and the highest mountain is 557 m. It has a typical mid-latitude zone continental monsoon climate, which is cold and dry in winter and wet and rainy in summer. The average annual temperature is 1.9–4.4°C, and the average temperature is 9.5–12°C in spring and 13–20°C in autumn. The average annual rainfall is 650–850 mm, the predominant wind direction is southwest in summer and northwest in winter, and the average wind speed is 2.2 m/s. It is rainy in summer and snowy in winter, so the weather conditions are not conducive to sampling and research. The average temperature in autumn is higher than that in spring, so the authors sampled in both spring and autumn.

Jiapigou gold mine area is situated in the northeast of the Zhongchao ancient land plate and the southeast of the Huifa River deep fracture. The northwest west Jiapigou-Dashilizi tectonic zone controls the distribution of the goldfields and deposits. This gold mine belt is 50 km long and 1–3 km wide, and mainly includes the Jiapigou gold mine, the Laoniugou gold mine, the Erdaodianzi gold mine, the Daxiangou gold mine, the Banmiaozhi gold mine, the Sandaogou gold mine, the Huanan gold mine and the Laojinchang gold mine, as well as other small-scale gold mines. Placer gold was exploited in this gold mine area in 1821, and rock gold was exploited in 1845. The Jiapigou gold mine has been exploited for nearly 190 years. First, indigenous alchemy was adopted for gold extraction after 1821, and then the mercury amalgamation has been used for the gold extraction since 1940 until 2006. Considering the amalgamation of serious pollution to the environment, the international community does not advocate the use of amalgamation, and China's government officially prohibited the mercury amalgamation technique in 1996 [8], so the miners in this area have used cyanidation instead of amalgamation for gold extraction since 2006. According to investigations, 2000 t ore can annually be exploited in Jiapigou alone, approximately 20 kg·yr⁻¹ of mercury were used in the amalgamation, and 50%–60% of the mercury could enter into the water, soil and atmospheric environment through point and non-point source ways [24].

In order to further understand the effects of the past and current gold mining activities on the environment, three operating gold mines (Laoniugou gold mine, Jiapigou gold mine and Sandaocha little gold mine) and one abandoned gold mine (Laojinchang gold mine) as well as 240 km² of its surrounding area in Jiapigou gold mine area were chosen as the study area.

2.2. Sampling and analytical methods

In order to explore more about the effects of mercury contamination on environmental quality in the gold mine and the surrounding areas, the sampling sites should contain all of the current types of mercury discharge (gold mine site, mineral processing plant and tailings) and residential area; in addition, resident coal-fired cooking time and busy traffic times of the study area should be avoided when sampling.

In October 2011 and April 2012, considering the landform characteristics of Jiapigou Town and the position of the gold mines, the sampling sites of the atmosphere and soil were set along the main roads and country roads that led to the gold mines. Laojinchang village, the center of Jiapigou gold mine area, was selected as the starting site, and one sample site every 3 km was set along the roads that led

in the Wudaogou (Laoniugou gold mine) direction, Jiapigou gold mine direction and Erdaogou direction respectively. There were 23 sampling sites in total. The locations and description of the natural conditions of the sampling sites are shown in figure 1 and table 1.

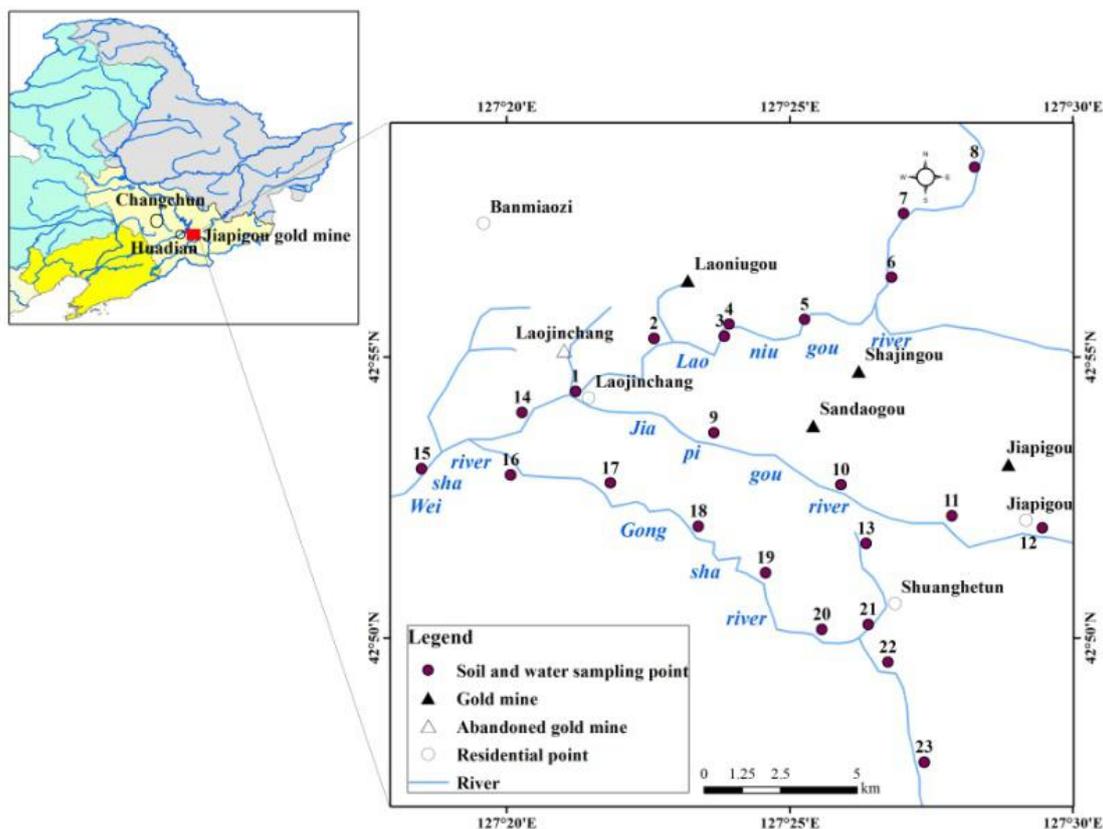


Figure 1. Locations of sampling sites in study area.

Table 1. Natural conditions of sampling sites.

Sample No.(#)	Name	Natural conditions	Sample No.(#)	Name	Natural conditions
1	Laojinchang	Abandoned gold mine	13	Xingan village	Residential area, country road
2	Laoniugou	Residential area	14	Erdaogouku	Road, river confluence
3	Laoniugou gold mine	Gold mine	15	Banchang	Road
4	Gaolitun	Mineral processing plant	16	Xinshengtun	Country road
5	Shajingou	Tributary converge site, Gold mine	17	Erdaogou	Country road
6	Sidaogou	Country road	18	Lingqian	Country road
7	Wudaogou	Country road	19	Daheyang	Country road
8	Hamagou	<i>Rana</i>	20	Xiaoyinggo	Country road

		<i>chensinensis</i> (Chinese brown frog) cultivation farm		u	
9	Toudaocha	Regional road	21	Shuanghetun	Residential area
10	Sandaocha	Small-scale gold mine, tailing pond, mineral processing plant, residential area	22	Majiadian	Wood farm, country road
11	Miaolingtun	Residential area, tailing pond	23	Majiadian'	<i>Rana chensinensis</i> (Chinese brown frog) cultivation farm
12	Jiapigou	Gold mine, residential area, road			

As figure 1 and table 1 show, Laoniugou gold mine (3#), Sandaocha (10#) and Jiapigou (12#) are near the sites of the gold mines that are still currently operating; Laojinchang (1#) is near the site of abandoned gold mine; there are small mineral processing plants around Gaolitun (4#) and Sandaocha (10#); gold mine tailing ponds are in Sandaocha (10#) and Miaolingtun (11#); residential areas are in Laoniugou (2#), Sandaocha (10#), Miaolingtun (11#), Jiapigou (12#), and Shuanghetun (21#); Erdaogoukou (14#) and Banchang (15#) have the roads that lead to Huadian City; Toudaocha (9#) and Jiapigou (12#) have the roads that lead to Jiapigou Town; others are country roads; and Laojinchang (1#), Shajingou (5#) and Erdaogoukou (14#) are the places where rivers converged. The sampling sites have covered all the conditions of study area.

The atmosphere and soil control samples were collected at Longwan Nature Reserve, Huinan County, Jilin Province, which was set as the clean area, 56 km southwest of the study area. The natural conditions of the control site including the topography, hydrometeorological and ecological environment are similar to those in the polluted area, but without large-scale gold mines or mine area.

The concentration of gaseous mercury in the air was determined by automatic mercury vapor analyzer RA-915+ produced by Lumex Company, Russia. Three consecutive values of gaseous mercury were read at each sampling site, and nine instantaneous values were obtained in all. Meanwhile, the latitude and longitude coordinates and altitude of the sampling sites were measured by GPS.

The mixed surface soil samples of multi-sampling sites were collected, and each of the mixed soil samples was about 1 kg and the surplus soil was discarded by the quarter method. The samples were kept in a PVC ziplock bag and brought back to the laboratory. The concentration of mercury in the soil was determined by the national standard method. The soil samples were wet digested by HNO₃-H₂SO₄-V₂O₅ and a constant volume of the solutions was determined by cold atomic fluorescence spectrometry AFS-2202. Parallel samples were taken from each of the samples, and the standard reference sample of the soil was used for validation.

2.3. Calculation method

2.3.1. Kriging interpolation. Interpolation predicts values for cells in a raster from a limited number of sample data points. It can be used to predict unknown values for any geographic point of data. The Kriging interpolation method was used to analyze the spatial distribution of the mercury concentration, and ArcGIS software was used to build the model of Kriging interpolation in the study.

It is not required to have a normal distribution of the original points data for prediction. However, when the data were normally distributed, Kriging interpolation will be the best method of unbiased estimation. In the study, only the gaseous mercury data set in autumn were characterized by normal

distribution, so logarithmic transformation for all of data have been done before the Kriging interpolation.

2.3.2. Geoaccumulation index. The geoaccumulation index (I_{geo}) was used to analyze and evaluate the mercury contamination level in the soil of Jiapigou gold mine area, and this could provide solid information for the management monitoring of the soil environment in this area.

The geoaccumulation index method was proposed by Muller [25] to evaluate the heavy metal contamination level in sediment. Due to the common characteristics of soil and sediment, the method was also widely used in assessing the heavy metal contamination level in soil. The computational formula is:

$$I_{geo} = \text{Log}_2[C_n / (k \times B_n)] \quad (1)$$

where C_n is the measured concentration in the sediment for metal n ; B_n is the geochemical background value for metal n in the local rock; k is the coefficient of the change of the background value of the metal elements caused by the difference of local rock (usually 1.5). Generally, the geoaccumulation index includes seven grades, and the corresponding contamination levels are shown in table 2 [25].

Table 2. Ranks of geoaccumulation index and corresponding contamination level.

Item	Contamination level						
	Uncontaminated	Uncontaminated/moderately contaminated	Moderately contaminated	Moderately/strongly contaminated	Strongly contaminated	Strongly/extremely contaminated	Extremely contaminated
I_{geo}	≤ 0	0-1	1-2	2-3	3-4	4-5	> 5
Level	0	1	2	3	4	5	6

Because of industrial pollution and human activity, the mercury background value in the soil was difficult to determine, so the geochemical background value (B_n) of the mercury in the local rock that the above formula mentioned was usually the mercury value in less polluted areas. Kabata-Pendias [26] estimated the background mercury values of all different types of soils in the world, which were all between 0.05-0.30 mg·kg⁻¹. The study of Wang *et al.* [27] on the soil environmental element chemistry of China indicated that the average mercury background value in soil of China was 0.038 mg·kg⁻¹, and the average mercury background value in soil in Jilin Province (0.027 mg·kg⁻¹) was lower than that of the whole nation, which was related to the geochemical characteristics of mercury being the dispersed element; moreover, the mercury background values were different in different types of soils [27]. The average mercury concentrations for dark brown forest soil and dark brown forest soil were 0.044 -0.086 mg·kg⁻¹, so we selected 0.063 mg·kg⁻¹ as the background value in this research.

3. Results and Discussion

3.1. Results of mercury concentration

3.1.1. Total mercury concentrations in atmosphere and soil. The characteristic values of the total mercury concentrations in the regional atmosphere and soil of the study area are shown in table 3 and Table 4 respectively, which are calculated in Excel. All of the mean values between spring and autumn were significant by the one-way ANOVA test.

Table 3. Characteristic values of total Hg concentrations in the atmosphere of Jiapigou gold mine area in spring and autumn.

Time	Maximum /ng·m ⁻³	Minimum /ng·m ⁻³	Arithmetic mean /ng·m ⁻³	Standard deviation /ng·m ⁻³	Coefficient of variation (C.V.) /%	Number of samples (n)	Arithmetic mean±Standard deviation in Control site /ng·m ⁻³
Spring	19.5	1.33	5.27	6.56	5.81	207	5.0±0.51
Autumn	25.0	1.00	11.45	8.16	7.11	138	8.0±0.23

Table 4. Characteristic values of total Hg concentration in the soil of Jiapigou gold mine area in spring and autumn.

Time	Maximum /mg·kg ⁻¹	Minimum /mg·kg ⁻¹	Arithmetic mean /mg·kg ⁻¹	Standard deviation/mg·kg ⁻¹	Coefficient of variation (C.V.) /%	Number of samples (n)	Arithmetic mean±Standard deviation in Control site /mg·kg ⁻¹
Spring	2.51	0.09	0.58	0.62	109.54	46	0.39±0.16
Autumn	2.06	0.06	0.44	0.52	117.33	46	0.43±0.14

The results from table 3 and table 4 showed that the mean value of the mercury concentration in the atmosphere was 5.27 ng·m⁻³ in spring, and 11.45 ng·m⁻³ in autumn in the study area, and the mean mercury value in the soil was 0.58 mg·kg⁻¹ in spring and 0.44 mg·kg⁻¹ in autumn. All of the mean values were more than those in the control sites.

The Kriging interpolation method in ArcGIS was used to analyze the spatial distribution of the total mercury concentrations of the atmosphere and soil in spring and autumn in Jiapigou gold mine area, and the results are shown in figure 2 and figure 3.

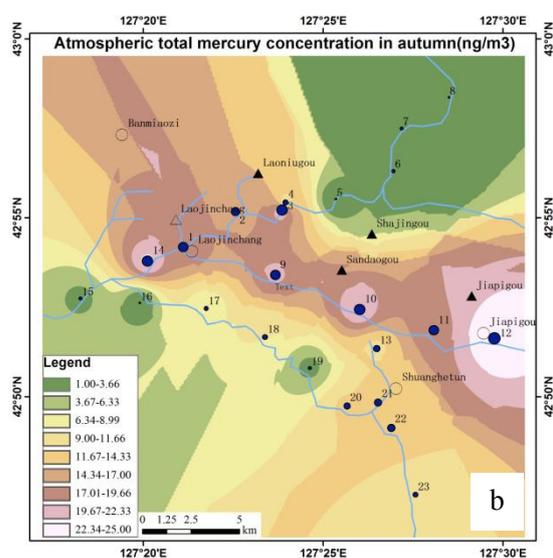
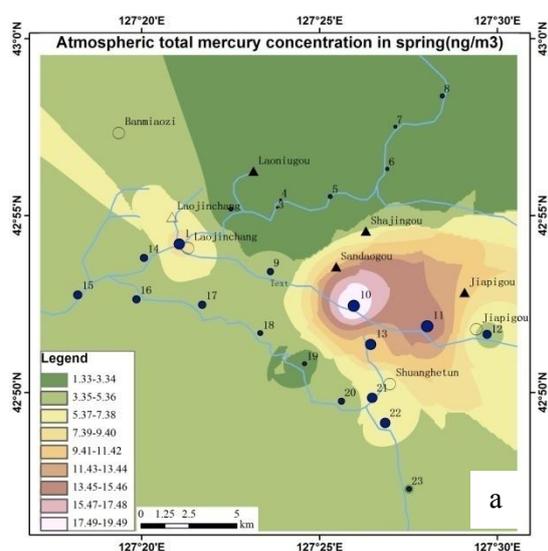


Figure 2. Spatial distribution of the total mercury concentration in the atmosphere in spring (2a) and in autumn (2b).

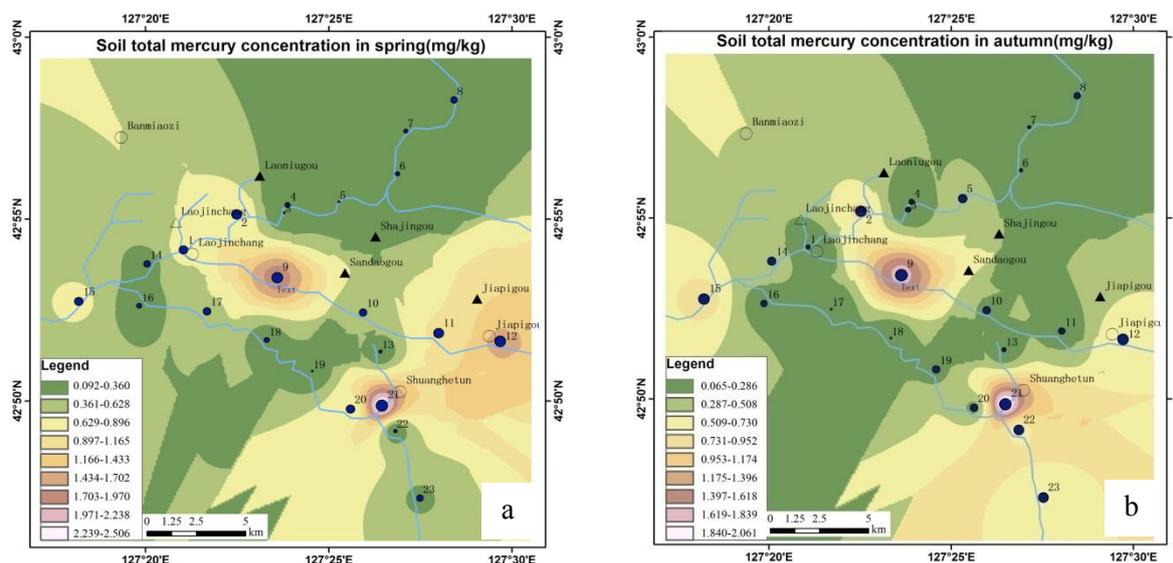


Figure 3. Spatial distribution of the total mercury concentration in the soil in spring (3a) and in autumn (3b).

It was indicated that the concentration of mercury in atmosphere gradually decreased from the centers, Sandaocha (10#), Miaolingtun (11#) and Xingancun (13#), to their surrounding areas in spring; and from the center, Jiapigou (12#), Laoniugou gold mine (3#), Toudaocha (9#) and Sandaocha (10#), to its surrounding areas in autumn. Meanwhile, the mercury concentrations in the soil were similar in spring and autumn, that is, the concentration of mercury gradually decreased from the centers, Shuanghetun (21#), Toudaocha (9#), Jiapigou (12#) and Laoniugou (2#), to their surrounding areas (table 2 and figure 2 to figure 5).

3.1.2. Correlation of mercury concentrations in atmosphere and soil. Correlation analysis was done after the elimination of several extreme outliers of mercury contents in atmosphere and soil, and it should be within the allowable range. The correlation analysis was calculated in Excel. R^2 is the regression determination coefficient and r is the Pearson correlation coefficient.

The regression equation of the total mercury concentrations in atmosphere and soil in spring was $y = 3.0086x + 2.6007$, $R^2 = 0.6505$ and $r = 0.8065$ ($P < 0.001$), and in autumn, $y = 15.191x + 2.3336$, $R^2 = 0.5802$ and $r = 0.7617$ ($P < 0.001$). Therefore, the mercury concentrations in the atmosphere and soil have a significant correlativity.

3.2. Distribution characteristics of mercury concentrations

3.2.1. Distribution characteristics of mercury peak values. The spatial distributions of gaseous mercury concentrations in spring and autumn are shown in figure 2. It can be seen that the concentrations of gaseous mercury substantially decreased from the center (gold mine area) to the surrounding area in general. In autumn, the peak values of gaseous mercury concentration emerged at Jiapigou gold mine, Laoniugou gold mine and Sandaocha gold mine, which showed that the mercury concentration in the atmosphere had a close relationship with gold mining activity; in spring, the peak value of gaseous mercury concentration emerged only at Sandaocha gold mine and the mercury concentrations were not high at the other two gold mines. The reason of the mercury peak values emerging at Sandaocha both in spring and autumn was that there was a small-scale gold mine, a tailing pond and a mineral processing plant near Sandaocha site. The concentration of gaseous mercury tends

to decrease, which was related to the elimination of the mercury amalgamation technique [28, 29]. In the autumn of 2008, the maximum total gaseous mercury concentration was $119.0 \text{ ng}\cdot\text{m}^{-3}$ in Jiapigou [29], while the concentration was $19.5 \text{ ng}\cdot\text{m}^{-3}$ in spring of 2012 and $25.0 \text{ ng}\cdot\text{m}^{-3}$ in autumn of 2011 in our study. Excluding weather-related conditions, it can be mainly related to prohibiting the use of the plate amalgamation technique for a long time.

The spatial distributions of the soil mercury concentration in spring and in autumn are shown in Figure 3. It was indicated that the mercury peak values were not at the gold mining sites at all, but at the places near mining sites and residential areas.

3.2.2. Correlation of mercury concentrations in atmosphere and soil. The characteristics of mercury determine that its contamination scope has regional limitations. The circulation and transportation of mercury in the atmosphere and soil was the main mode of movement for regional mercury contamination, and mercury could migrate far away through the media of water and atmosphere. The regression analysis of the total mercury contents between the atmosphere and the soil showed a significant correlation, which indicated that there was certain circulation of the mercury between the regional atmosphere and soil. The residual mercury in the soil can be released into the atmosphere, and the gaseous mercury attached to the particulate matter can come into the soil through dry and wet deposition in certain weather conditions [19].

3.2.3. Causes for mercury contamination in atmosphere and soil. Mason et al. [30] estimated the contribution of gaseous mercury to terrestrial mercury at different latitudes in the northern hemisphere. At $10^{\circ}\text{N}\sim 30^{\circ}\text{N}$ and $30^{\circ}\text{N}\sim 70^{\circ}\text{N}$, the mercury settlement values were $19.8\mu\text{g}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ and $15.8\mu\text{g}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ respectively. Schlüter [31] summarized the mechanism and related reports about mercury evaporation at present, which showed that in the topsoil, the concentrations of elemental mercury and methyl mercury formed from biotic and abiotic ways were significantly affected by the evaporation rate from the soil to the atmosphere. Furthermore, the evaporation rate of mercury was restricted by climate change and the migration of soil volatile matter in mercury-enriched mine areas. So there are many factors affecting the transfer and circulation of mercury between the atmosphere and soil.

For the study area, the mercury contamination in the atmosphere and soil might be caused by the following factors.

- Gold mine production. It is assumed that the distribution of mercury in the soil and atmosphere was related to long-term production of gold in the Jiapigou gold mine area. Jiapigou gold mine has been exploited since 1921, and the plate amalgamation technique has been used in gold extraction in the area from 1940 to 2006. In the mining process, mercury metal ions were heated into elemental mercury and then released into the atmosphere. The gaseous mercury migrates with the air movement in the atmosphere, and it can enter into soil through the dry and wet deposition around the mining area. Mercury was absorbed by clay particles in the soil, so the mercury concentration of the surface soil increased.

In 2006, the mercury amalgamation technique was replaced by cyanidation for the gold extraction in Jiapigou gold mine, which reduced and even terminated the release of mercury. However, the residual mercury in the soil was constantly released into atmosphere and became the mercury source of the study area.

- The mechanism of soil absorption. The mercury concentration in the soil correlated significantly with the gaseous mercury concentrations in Chongqing region, China, and the correlation coefficient was 0.741 [32]. The authors concluded that the soil organic matter and clay had a strong ability of adsorption to mercury, so the wet and dry mercury deposition into the soil is mostly fixed, which can increase the mercury concentration in the soil [32]. Jiapigou gold mine is located in the northeast of China. The soil type in this area was dark brown forest soil with high organic matter (5.0%~5.72%). The soil texture is mostly loam or clay loam; in addition, the climate is humid for these are mountainous and forest areas, therefore the gaseous mercury showed strong adsorption. As so many similar conditions above,

it was suggested that the soil in this area had a similar absorption mechanism with Wang *et al.*'s [32] study.

- Mercury exchange flux. Zhang *et al.* [28] suggested the mechanism of mercury exchange flux between atmosphere and soil at the gold mining site and its surrounding area. The soil intensely released mercury into the atmosphere in autumn, but in spring, the mercury in the atmosphere could be settled into the soil by wet or dry deposition, so the mercury concentration in the soil in spring was generally slightly higher than that in autumn. The results of the paper were consistent with the finding published by Zhang *et al.* [28].
- Landform characteristics. The landform characteristics of the study area was of two mountains with a valley in the middle, which made the closed valley landforms of Shuanghetun and Toudaocha, and became obstructive areas for the diffusion of atmospheric flow and peak values areas for soil mercury.

In summary, the source of soil mercury contamination is complex, not only through mining release into the atmosphere and affects the surrounding areas, but also due to mercury dry and wet deposition into the soil through the atmosphere released from coal fired by local people. In addition, it also includes the soil's natural mercury release. This research avoided sampling at the time of coal firing and traffic peak hours, and it can be partially excluded from these sources, but it can also be further proof that the mercury in the soil is the cumulative result of a variety of sources for many years, which are sinks and sources of mercury contamination in gold mining district, and the result is similar with other scholars' results [33].

3.3. *Assessment of mercury contamination in soil.* Considering the determined results of mercury concentrations in the study area, the geo-accumulation indexes of 23 sampling sites were calculated (table 5).

Table 5. Geo-accumulation index of mercury contamination in Jiapigou gold mine area.

Sampling site	Spring		Autumn		Sampling site	Spring		Autumn	
	<i>I</i> _{geo}	Lev _{el}	<i>I</i> _{geo}	Lev _{el}		<i>I</i> _{geo}	Lev _{el}	<i>I</i> _{geo}	Lev _{el}
1# Laojinchang	2.942	3	0.112	1	13# Xingancun	0.596	1	0.099	1
2# Laoniugou	3.266	4	3.057	4	14# Erdaogoukou	1.560	2	2.337	3
3# Laoniugou gold mine	0.268	1	0.532	1	15# Banchang	2.580	3	-0.542	0
4# Gaolitun	1.082	2	0.322	1	16# Xinshengtun	1.214	2	0.752	1
5# Shajingou	0.090	1	2.357	3	17# Erdaogou	2.863	3	3.128	4
6# Sidaogou	0.842	1	-0.189	0	18# Lingqian	1.209	2	-0.400	0
7# Wudaogou	1.004	2	-0.069	0	19# Daheyuan	-0.047	0	1.307	2
8# Hamagou	1.7	2	0.78	1	20#	2.78	3	1.20	2

	95		2		Xiaoyinggou	2		5	
9# Toudaocha	4.3 01	5	4.18 0	5	21# Shuanghetun	4.73 1	5	4.44 9	5
10# Sandaocha	2.4 22	3	1.33 1	2	22# Majiadian	0.49 3	1	2.68 1	3
11# Miaolingtun	3.3 22	4	0.78 5	1	23# Majiadian	1.76 8	2	2.55 6	3
12# Jiapigou	3.9 63	4	2.88 1	3					

Kriging interpolation was used to analyze the spatial differentiation of mercury contamination levels (figure 4). As indicated in the figures, the sampling sites of Shuanghetun (21#) and Toudaocha (9#) in the study area were strongly/extremely contaminated with Hg (in level 5) both in autumn and spring, and the sampling site of Laoniugou (2#) was strongly contaminated with Hg (in level 4) both in autumn and spring; all these places were not far from the gold mining site and in the valley of low topography, where the surrounding mountains blocked the atmospheric flow with mercury and the mercury was deposited in this place. Laoniugou gold mine (3#) and Xingancun (13#) were uncontaminated/moderately contaminated with Hg (in level 1) both in autumn and spring. (table 2) In addition, the contamination levels of the other sampling sites were different in spring and autumn. Details can be found in table 5.

The study indicated that, for soil mercury contamination, strongly/extremely contaminated (level 5) and strongly contaminated (level 4) area accounted for 21.74% of Jiapigou gold mine area, and moderately/strongly contaminated (level 3) area accounted for 21.74% and uncontaminated (level 0) area only accounted for 4.35% in spring; and the corresponding values accounted for 17.4%, 21.74% and 17.35% respectively in autumn. In conclusion, the mercury contamination in soil was severe in Jiapigou gold mine area, especially in the mining area and its surrounding area; they were all beyond the moderately/strongly contaminated level.

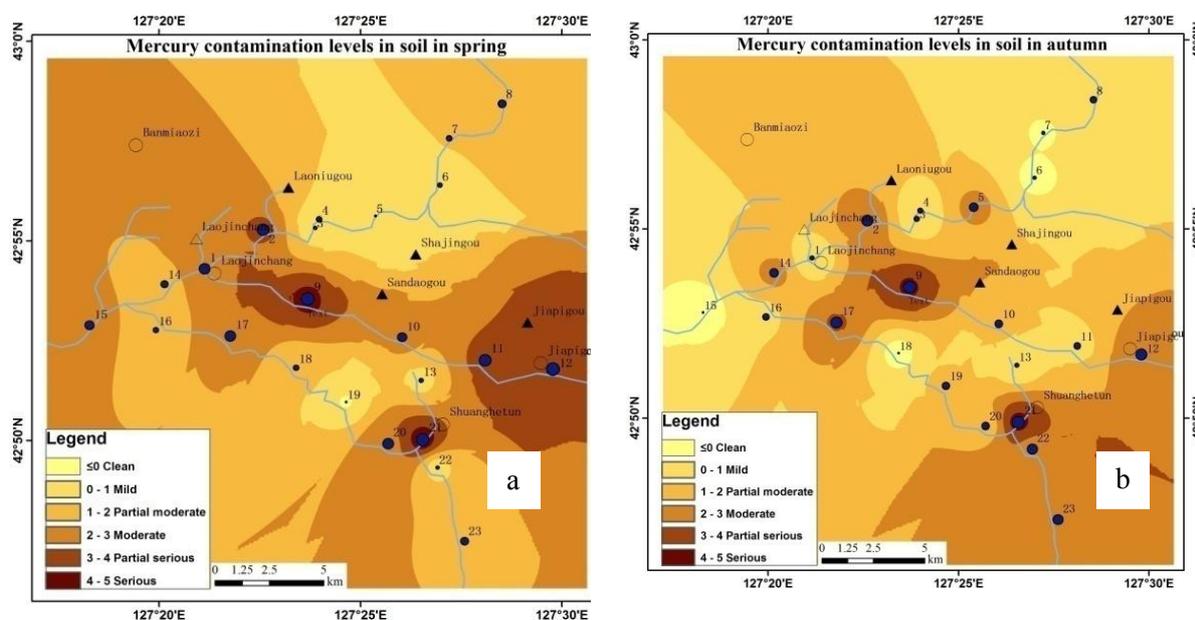


Figure 4. Mercury contamination levels in soil in spring (4a) and in autumn (4b).

4. Conclusions

- The Jiapigou gold mine has been exploited for nearly 190 years; the highest atmosphere mercury concentrations were 3.9 and 3.13 times those at the control site in spring and autumn respectively. The distribution of TGM in the study area was significantly related with the original mercury sources. The peak of TGM is in the existing gold mine area, mineral processing plant and tailings, decreasing gradually from the center to the surrounding area. Overall, the mercury contamination was attributed to long-term use of backward technology for gold extraction, and it gradually reduced after elimination of the technology.
- The highest soil mercury concentrations were 6.44 and 4.79 times those at the control site in spring and autumn respectively. The ranking of mercury concentrations in soil from high to low were the places near mining sites, the gold mining sites and then the others. The mercury concentrations in soil were significantly positively correlated with those in the atmosphere, which indicated that mercury can be transported between the atmosphere and the soil. The data from the study strongly supported the conclusion that the mercury circulation consisted of mercury release from soil into atmosphere and gaseous mercury settlement into the soil through dry and wet deposition in certain weather conditions.
- The mercury peak value of the soil emerged at the valley with low topography that was close to the gold mining site. The combination of strongly/extremely contaminated (level 5) and strongly contaminated (level 4) area accounted for 21.74% and 17.4% in spring and autumn respectively. Based on the local meteorological conditions, it was predicted that the soil mercury contamination was affected not only by long-term amalgamation of gold mine and tailings, but also by the local impact of the closure of the valley topography and climate conditions.

The study presents the mercury contamination situation in the Jiapigou gold mining area after termination of the use of amalgamation technique for gold extraction. These investigations are reasonable for better understanding the potential risk of metal-contaminated areas for human health in these areas.

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