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ORIGINAL ARTICLE

Heavy metal(loid) pollution in mine wastes of a Carlin-type gold mine in southwestern Guizhou, China and its environmental impacts

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Abstract Mercury pollution resulting from artisanal gold mining is a serious environmental problem in many developing countries. In this study, we analyzed heavy metal(loid) contamination in mine wastes from a Carlin-type gold mine in southwestern Guizhou, China. Highly elevated levels of As, Hg, Tl, Sb, and Cd—up to 5844, 28, 29, 581 and 3.0 mg/kg, respectively—were observed in the mine wastes, but no significant accumulation of Cr, Ni, Cu, Zn, and Pb was found. The smelting process of gold ores had significant impacts on the enrichment of As, Tl, and Hg in the mine wastes. Due to the significant metal(loid) pollution in this gold mine, the ecological risks are classified as "very high." Hg and As are the major pollutants with a relative contribution of 55.9 % and 24.2 % to the risk index, respectively. Phytoremediation could be used to reduce heavy metal contamination and recycle the gold simultaneously. Hg–As–Tl pollution should be considered in gold extraction at Carlin-type gold mines.

Keywords Heavy metals · Metalloid · Potential ecological risk assessment · Enrichment factor · Mine waste · Carlin-type gold mine

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1 Introduction

China produced the most gold in the world in 2007, and kept this record at least through the following 6 years. Gold output in China increased 11.7 % in 2012 with a yield of 403 tonnes (CGA 2013). Gold resources are unevenly distributed in China. Many types of gold ore have been found in northern and eastern China. Primary gold ore occurs mainly in Shandong, Henan, Hebei, Shaanxi, Jilin, and Hubei Provinces; while gold placers are found mainly along the margins of northeastern China, such as Heilongjiang, Shaanxi, Gansu, Sichuan, and Inner Mongolia. Recently, western provinces such as Guizhou and Yunnan have gained a sharp increase in gold production.

In Guizhou Province, gold production has increased since the advent of bacterial pretreatment to extract invisible gold in sulfide minerals a few years ago. Carlin-type gold ore is a typical refractory ore, characterized by low gold grade and high sulfur and arsenic concentrations. Gold usually occurs in these ores as discrete particulates (<0.1 μm in diameter) within sulfide minerals. For Carlin-type gold ore, bioleaching is advantageous over other traditional methods (Qiu et al. 2012).

Carlin-type gold deposits (also known as sediment-hosted gold deposits) are the largest hydrothermal gold deposits in the world and are widely distributed in the United States and China (Tretbar et al. 2000; Hu et al. 2002). The region of southwestern Guizhou, is an important component of the Yunnan–Guizhou–Guangxi "gold triangle" area. Carlin-type gold deposits in southwestern Guizhou, China, are hosted in late Paleozoic and early Mesozoic sedimentary rocks along the southwest margin of the Precambrian Yangtze craton. The deposits can be classified as two types, namely the fault type and the strata-bound type, based on their occurrence, shape and structural

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controls (Zhang et al. 2003; Xia et al. 2012). They have similar characteristics to Carlin-type gold deposits in Nevada, including notable enrichment in As, Sb, Hg, and Tl (Hu et al. 2002; Xia et al. 2012).

Unfortunately, many modern gold mining practices have led to elevated levels of pollutants in air, soil, and water. Artisanal and small-scale gold mining (ASGM) was the largest anthropogenic source of Hg emission to the air in the world in 2010 at 727 tonnes or 37 % of the global anthropogenic emissions (UNEP 2013). ASGM has been recognized as one of the Top 10 Toxic Pollution Problems in 2012 by Blacksmith Institute. The technique is popular in developing countries because of its simple processes and low costs; it accounts for about 20 % of global gold production. The Hg pollution resulting from ASGM is a serious environmental problem in South America, China, Russia, Southeast Asia, and Africa (Lacerda and Salomons 1998; Feng et al. 2006). In 1996, ASGM was banned in China and dissolution with cyanide (CN) has been widely developed. In this technique, finely ground high-grade ore is mixed with CN; low-grade ores are stacked into heaps and sprayed with a CN solution.

Carlin-type gold deposits show enrichment in Au, Sb, Hg, Tl, and Ba. This enrichment results from hydrothermal circulation with a temperature of up to 300 °C. The associated trace elements are released into the environment during mining and refining processes. This paper evaluates heavy metal and metalloid pollution and their possible sources in Laowanchang gold mine in Guizhou Province, China. The results provide scientific input to pollution control and remediation efforts in Carlin-type gold mines.

2 Materials and methods

2.1 Study area

The Laowanchang gold mine is located in the south of Qinglong County (Fig. 1). The ore is a Carlin-type gold deposit with the characteristic Au–As–Sb–Hg–Tl trace element assemblage. It is considered to be the product of weathering evolution of a primary Carlin-type gold deposit. It formed through the accumulation of ore-forming material by karst collapse, followed by lateralization (Yang and Liu 2003; Yang et al. 2004). The granulating-dump-leaching technique was used to extract gold in this mine. The processing techniques included grinding, concentration, dehydration, and pressure oxidation before cyanidation (Fig. 2; Ren 2003). The total gold reserve of this mine was 2278 kg, with a grade of 0.5–1.25 g/t. During the period of 1993–2008, significant quantities of mine wastes were produced and piled in the mining area.

2.2 Sample collection

In October 2010, 49 mine waste and surface soil samples were collected in the study area (Fig. 1). The sampling sites can be categorized into five types: mine wastes in the settling tank (A), mine wastes (B), crude ores (C), corn field soil (D), and background soil (E). The settling tank was a separation unit separating solids from the waste water. Suspended particles were settled to the bottom of the tank and the waste water was discharged. The corn fields are very close to mine waste heaps. The control site selected for comparison with the polluted sites was located ∼5 km from the gold mine sites.

Crude ores were collected from different areas and smelted at Laowanchang sites. The mine wastes were heaped in Laowanchang and Hongyan sites. The crude ores were extracted from the land surface and the grasses and trees removed, causing soil erosion in the mining area.

Every sample consisted of a composite of four to five subsamples within an area of 1 m^2. A profile of the mine waste was collected at site QL5, and it was divided into ten depths with an interval of 30 cm. We collected mine ore waste samples at different depths. In the laboratory, the samples were air-dried, ground, and passed through a 100-mesh nylon sieve prior to geochemical analysis.

2.3 Analytical methods

The prepared samples were digested in the mixed acid of HNO_3–HF, and Cd, Sb, Tl, Cr, Ni, Cu, Zn, Pb, and Sc concentrations in the digests were determined by Inductively Coupled Plasma Mass Spectroscopy (ICPMS, Element, Finnigan MAT Co) according to the method developed by Qi and Grégoire (2000). Arsenic concentrations in samples were digested by HNO_3–HCl–H_2O_2 and determined by atomic fluorescence spectrometry (AFS-920, Beijing Jitian Instrument Corporation).

Mercury concentrations in the samples were analyzed by the RA-915 + Hg analyzer coupled with the PYRO-915 + attachment (Lumex Co., Russia). Au concentrations in the samples were analyzed by aqua regia digestion and atomic absorption spectrometry detection. CN concentrations in the samples were determined by the Pyridine Pyrazolone Spectrophotometric Method (Xu et al. 1999) with a limit of detection (LOD) of 0.025 mg/kg. Concentrations below the LOD were assumed to be the LOD in the calculation and assessment. In addition, the pH was determined with a pH meter in 2.5:1 (v/m) water/solid suspension.

Quality assurance and quality control consisted of method blanks, duplicates, and certified reference

Fig. 1 Location of the sampling sites in the Laowanchang gold mine, southwestern Guizhou, China
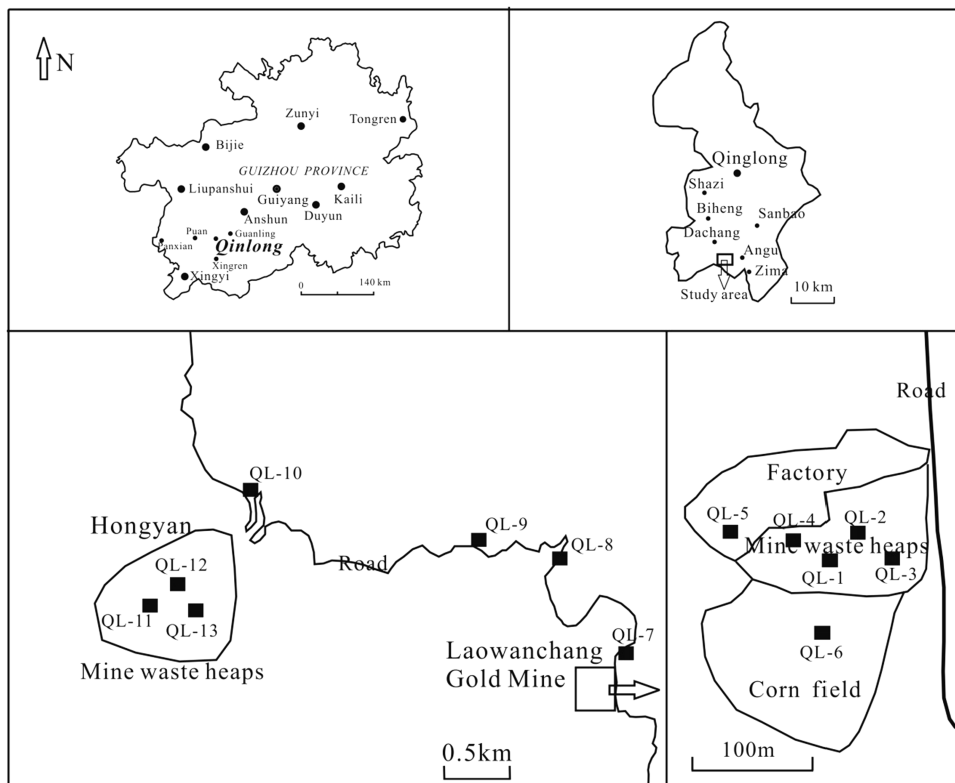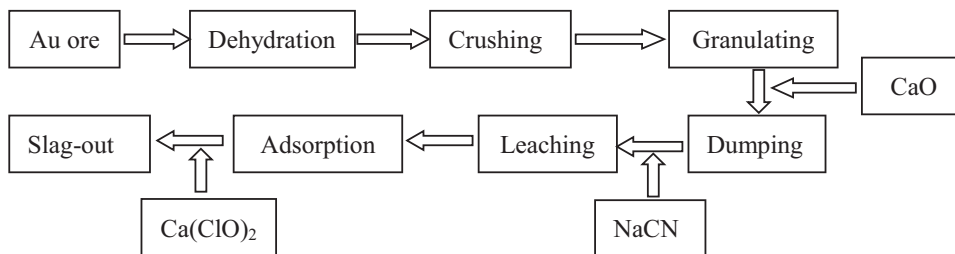

Fig. 2 The sketch map of the process of gold smelting in the Laowanchang mine

materials. Recoveries of certified reference materials (GBW07405 and GBWE070023) for all elements ranged between 90.2 % and 106 % (Table 1). The relative percentage difference was <10 % for heavy metals and metalloids in duplicate samples. All chemical analyses were carried out in the Institute of Geochemistry, Chinese Academy of Sciences.

2.4 Potential ecological risk assessment

The method of potential ecological risk assessment was created by Hakanson (1980) and has been widely applied to the pollution assessment of sediment and soil. It integrates concentration and ecological toxicity of different pollutants, and gives a potential ecological risk index as shown in Eq. (1):

Table 1 List of certified reference materials used in the present study and the results obtained

Element	GBW07405											GBWE070023
	Sc	Cr	Ni	Cu	Zn	Cd	Sb	Tl	Pb	Hg	As	Au
Measured value (mg/kg)	15.3	125	38.0	134	496	0.47	34.1	1.68	534	0.28	420	3.57
Certified value (mg/kg)	17.0	118	40.0	144	494	0.45	35.0	1.60	552	0.29	412	3.60
Recovery (%)	90.2	106	95.1	93.4	100.5	104.4	97.5	105	96.8	97.6	101.9	100.8

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$$RI = \sum_{i=1}^{n} Ei = \sum_{i=1}^{n} Ti \times Ci \qquad (1)$$

where RI is the potential ecological risk index; Ei is the potential ecological risk factor for a given pollutant (i); Ti is the "toxic-response" factor for a given pollutant as calculated by Hakanson (1980), i.e., Hg = 40, Cd = 30, As = Tl = Sb = 10, Pb = Cu = Ni = 5, Cr = 2, Zn = 1; and Ci is the ratio of metal(loid) concentration in the sample to the corresponding background level. RI can be divided into four levels: RI < 150, low ecological risk; 150 < RI < 300, moderate ecological risk; 300 < RI < 600, considerable ecological risk; and RI > 600, very high ecological risk.

2.5 Calculation of enrichment factors

The Enrichment Factor (EF) has been widely used to quantify the extent of human-induced contamination; it is defined as the relative abundance of a chemical element in the environmental media compared with the background (Shotyk et al. 2000). EF is calculated according to Eq. (2):

$$EF = \frac{M[\text{sample}]/X[\text{sample}]}{M[\text{background}]/X[\text{background}]} \qquad (2)$$

where M is the concentration of the element under consideration and X is the concentration of the chosen reference element. The reference element is conservative, and the most commonly used elements are Al, Fe, Sc, and Ti (Hernandez et al. 2003; Abrahim and Parker 2008). In this study, Sc was selected as it is generally considered to be mainly originated from natural sources (rock weathering).

3 Results

3.1 Major elements and pH

The gold ore is composed of SiO_2, Fe_2O_3, Al_2O_3, and TiO_2 (Chen 1999) as shown in Table 2. Compared with Guizhou red soil, the gold ore is rich in SiO_2 and Fe, and depleted in Ca and Mg. This indicates the gold mine originated from a primary Carlin-type gold deposit through strong weathering evolution. The deposit formed through the accumulation of ore-forming material by karst collapse followed by lateralization (Chen 1999; Yang et al. 2004).

The pH in the mine wastes and soil samples are listed in Table 3. The pH in the mine waste was higher than that in the crude or soil due to addition of quicklime during the smelting process (Fig. 2).

Table 2 Composition of major elements of the gold ore and Guizhou red soil (Data from Chen 1999) (%)

Major elements	Ore 1	Ore 2	Guizhou red soil
SiO_2	56.03	61.84	49.91
Al_2O_3	15.55	9.17	17.80
TiO_2	1.99	2.14	0.48
Fe_2O_3	13.08	13.98	10.41
FeO	0.07	0.19	0.13
CaO	0.10	0.17	3.75
MgO	0.26	0.15	1.46
K_2O	2.03	1.43	0.29
Na_2O	0.13	0.26	0.27
P_2O_5	0.93	1.80	
MnO	0.16	0.01	
LOI	7.40	5.99	

3.2 Heavy metal(loid) concentrations

The background concentrations of heavy metal(loid)s in Guizhou soil and heavy metal(loid) concentrations at the reference site are listed in Table 4. The background levels of As, Hg, Tl, and Cd at the reference site of the study area were comparable with the background levels of these elements in soil in Guizhou. However, the concentration of Sb and Au in soil at the reference site in this study was significantly higher than the background concentrations of these elements in soil of Guizhou Province. This might be related to Sb and Au mineralization in this region.

No significant accumulation of Cr, Ni, Cu, Zn, and Pb was observed in the mine wastes (Table 3). The maximum concentrations of As, Hg, Tl, Sb, and Cd were 5844, 28, 29, 581, and 3.0 mg/kg, respectively. The As and Hg concentrations were elevated by three orders of magnitude compared with the background values at the reference site. The maximum concentrations of CN and Au were 4.0 and 4.8 mg/kg, respectively.

Concentrations of selected trace elements differed significantly between different types of samples (Fig. 3). Among these types, the highest concentrations for As, Hg, Tl, Au, and CN were observed in types A and B. However, for Sb and Cd, the highest concentrations were observed in type C samples, indicating high metal levels in the crude ores related to mineralization.

3.3 Mine waste profile

The concentrations of elements, pH, and organic matter in the profile of mine waste are shown in Fig. 4. Sb and Cd concentrations were nearly constant at different depths. Hg,

Table 3 Statistics of studied elements and parameters in different types of mine waste (Mean ± SD)

	A (n = 5)	B (n = 35)	C (n = 4)	D (n = 4)	E (n = 1)	Total
pH	8.27 ± 0.11	8.11 ± 0.87	6.48 ± 1.28	6.84 ± 0.39	5.29	7.83 ± 1.05
Hg (mg/kg)	18.8 ± 6.52	8.68 ± 2.53	4.31 ± 0.70	2.81 ± 2.34	0.12	8.71 ± 5.02
As (mg/kg)	3847 ± 223	2879 ± 1443	1145 ± 608	990 ± 872	19.7	2624 ± 1516
Cd (mg/kg)	1.06 ± 0.47	0.95 ± 0.41	1.82 ± 1.33	0.47 ± 0.097	0.07	0.98 ± 0.58
Sb (mg/kg)	256 ± 29.6	289 ± 104	345 ± 121	171 ± 16.4	29	730 ± 604
Tl (mg/kg)	17.7 ± 2.52	11.3 ± 7.15	3.83 ± 1.02	3.30 ± 2.16	0.44	10.5 ± 7.29
Cr (mg/kg)	127 ± 5.75	144 ± 18.8	234 ± 44.6	102 ± 6.02	130	146 ± 35.0
Ni (mg/kg)	79.9 ± 6.18	75.3 ± 22.6	137 ± 48.5	46.7 ± 3.46	30.6	77.5 ± 30.6
Cu (mg/kg)	104 ± 11.0	100 ± 14.2	119 ± 27.6	49.1 ± 7.78	126	98.3 ± 21.5
Zn (mg/kg)	198 ± 11.8	196 ± 54.0	244 ± 54.6	131 ± 8.15	123	193 ± 54.0
Pb (mg/kg)	30.9 ± 7.31	18.6 ± 2.66	46.7 ± 36.0	27.5 ± 0.71	16.9	21.4 ± 5.87
Au (mg/kg)	3.21 ± 1.06	0.59 ± 0.17	0.54 ± 0.28	0.95 ± 0.62	0.35	0.88 ± 0.89
CN (mg/kg)	1.43 ± 1.57	0.31 ± 0.18	0.039 ± 0.01	0.11 ± 0.019	0.025	0.38 ± 0.60
Sc (mg/kg)	12.7 ± 0.70	13.4 ± 2.28	22.1 ± 7.51	8.42 ± 0.53	29.2	14.0 ± 4.53

Table 4 The heavy metal(loid) concentrations in background soil of Guizhou, soil from control site in the study area, and the national limit levels (mg/kg)

Element	Guizhou soil background (a)	Soil sample from the control site in this study	Class III level of soil environmental quality standard in China (b)
As	14.9	20	40
Hg	0.1	0.12	1.5
Tl	0.71	0.44	–
Cd	0.31	0.07	1
Sb	1.35	29	10
Cr	79.4	130	400
Ni	32.8	30.6	200
Cu	29.4	126	400
Zn	89.9	123	500
Pb	29.4	16.9	500
Au	1.06×10^{-3}	0.35	–
Sc	16.3	29.2	–

a, (He 1998); b, (SEPAC 1995)

Tl, Au, and CN concentrations varied greatly at different depths, but the overall trend was consistent.

Arsenic concentrations in the profile decreased with depth. Redox potential (Eh) and pH are the most important factors controlling As speciation and mobility. Arsenic is relatively mobile under a reducing environment in contrast to other heavy metals (Smedley and Kinniburgh 2002). Reducing conditions and high pH (8.36–8.70) at the bottom of mine wastes can lead to the desorption of As from mineral oxides and to the reductive dissolution of Fe and Mn oxides. Thus, As can migrate to underground water from mine wastes and its concentrations were notably less in the bottom layer.

4 Discussion

4.1 Risk index

The ecological impacts of heavy metals and metalloids in mine wastes and soils are listed in Table 5. RI for all sample types reached "very high" levels, indicating serious heavy metal and metalloid pollution in the study area. The average relative contribution from each element, as calculated as the percentage of Ei compared to RI was: Hg 55.9 %, As 24.2 %, Cd 12.4 %, Tl 4.1 %, and Sb 2.5 %. Average relative contributions from Cr, Ni, Cu, Zn, and Pb were very small (<0.5 %). Hg and As are the major
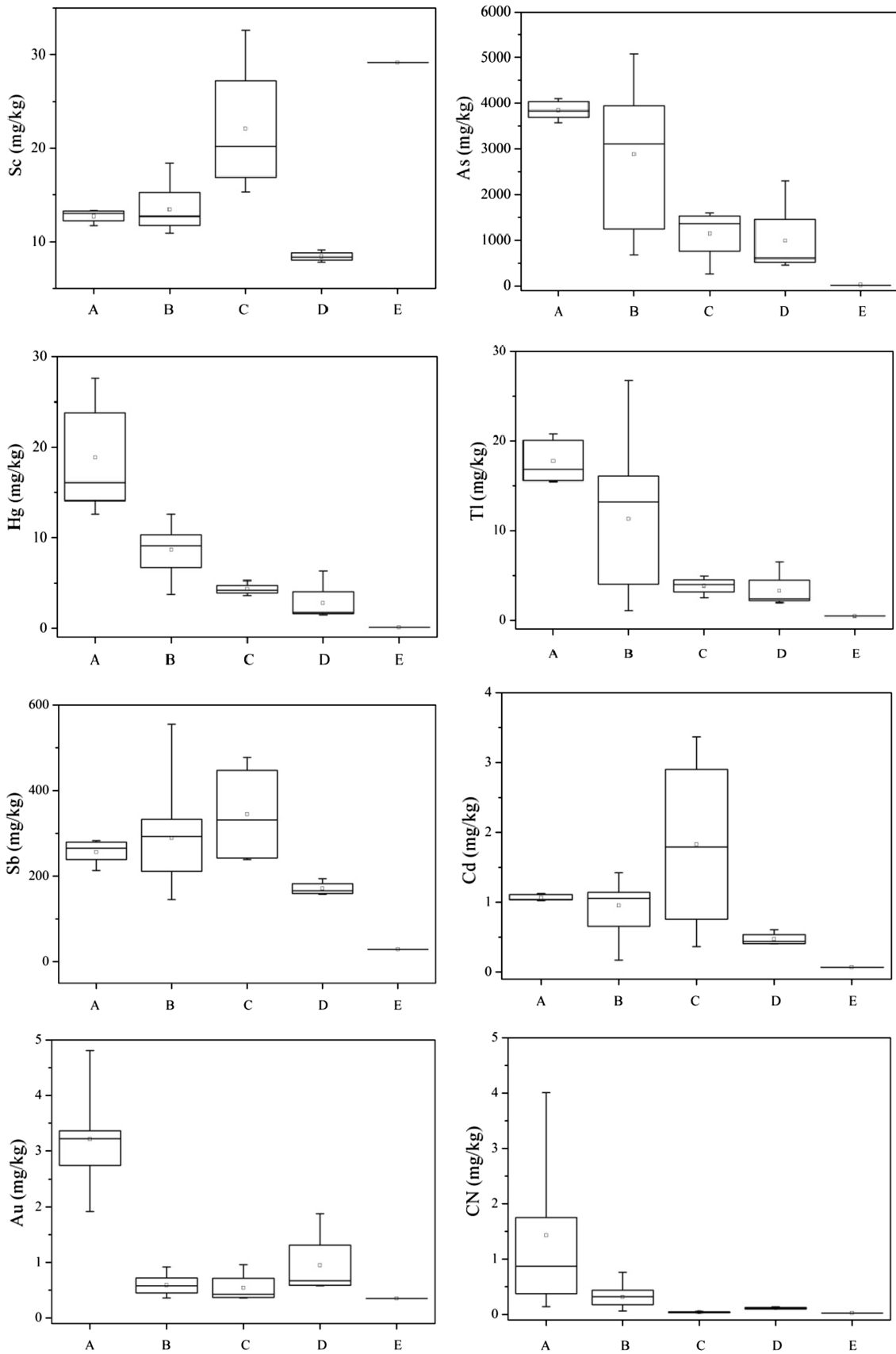
◀ **Fig. 3** *Box-plot* of trace element concentrations in different types of samples. Each *box* represents inter-quartile range (25th and 75th percentiles), the band near the *middle* of the *box* is the 50th (*median*), and the whisker represents the 5th and 95th percentiles

pollutants in this gold mine, and remedial techniques are urgently needed to mitigate the historical pollution before land reclamation.

A comparison of heavy metal pollution in different gold mines in China is shown in Table 6. Due to the widespread use of Hg for gold extraction before 1996 in China, Hg is the major pollutant in most gold mines of China. Other heavy metals (Cd and Cu) were also observed at elevated levels in some gold mines. The ecological risks in the study mine were much higher than other gold mines. Even though amalgamation was not used in this mine, Hg is still the major pollutant in this Carlin-type gold mining area.

4.2 Pollution source analysis

In order to evaluate the impact of the smelting process on the accumulation of heavy metals and metalloids in the mine wastes, trace element concentrations in the crude ore were considered as the background values to calculate EFs. The results are listed in Table 7. The values of EF, using Sc as the reference element, were in the range of 0.9–8.0. The EFs were 0.9 and 1.0 for Cd and 1.4 and 1.3 for Sb, indicating no significant enrichment. High EF values were observed for Hg, As, and Tl, with a range of 3.3–8.0. These three elements (Hg, As, and Tl) had accumulated

Table 5 Result of potential ecological risk assessment in the study area

	A	B	C	D
Ei (Hg)	6266.7	2893.3	1436.7	936.7
Ei (As)	1952.8	1461.4	581.2	502.5
Ei (Cd)	454.3	407.1	780.0	201.4
Ei (Sb)	88.3	99.7	119.0	59.0
Ei (Tl)	402.3	256.8	87.0	75.0
Ei (Cr)	2.0	2.2	3.6	1.6
Ei (Ni)	13.1	12.3	22.4	7.6
Ei (Cu)	4.1	4.0	4.7	1.9
Ei (Zn)	1.6	1.6	2.0	1.1
Ei (Pb)	9.1	5.5	13.8	8.1
RI	9194.2	5144.0	3050.4	1794.9
Description	Very high	Very high	Very high	Very high

significantly in the mine wastes of the settling tank through the smelting process, but there was nearly no impact on Cd and Sb from the smelting process. The geochemical behavior of heavy metals and metalloids varied in the smelting process. According to the EF values, the trace elements can be divided into two groups: Hg–As–Tl and Cd–Sb.

The correlation analysis between all trace elements is shown in Table 8. The As, Hg, and Tl concentrations significantly correlated with each other ($p < 0.001$); and Cd and Sb significantly correlated ($p < 0.01$). According to the correlation factors, the elements can be divided into two

Fig. 4 Distribution patterns of trace element concentrations and other parameters in the mine waste profile
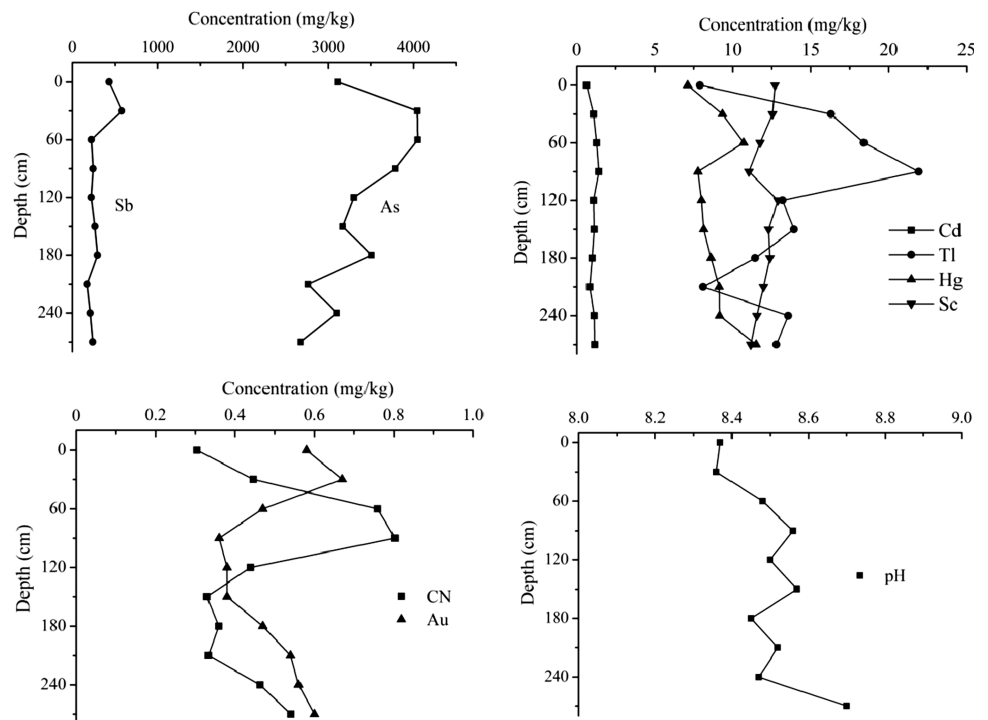
Table 6 Comparison of heavy metal pollution in gold mines of China

Area	Gold mine	Type	Process	Media	Elements (mg/kg)	RI	Reference
Zhaoyuan, Shandong	Linglong	Quartz reef	Amalgamation	Surface water	Hg (61 µg/L)		Liang et al. (2011)
					Cd (30 µg/L)		
					Cu (1640 µg/L)		
Tongguan, Shaanxi	Xiaoqinling	Quartz reef	Amalgamation	Soil	Hg (2.75)	Hg (845)	Wu et al. (2010)
					Cd (0.55)	Cd (97)	
Huaye, Jilin	Jiapigou	Quartz reef	Amalgamation	Soil	Hg (0.56)	Hg (465)	Yang and Wang (2013)
					Cd (0.56)	Cd (141)	
Miyun, Beijing	Detiangou, Qifengcha	Quartz reef	Amalgamation	Soil	Hg (0.14)	Hg (187)	Chen et al. (2012)
Qinglong, Guizhou	Laowanchang	Carlin-type	Cyanidation	Mine waste, soil	Hg (8.71)	Hg (6267)	This study
					As (2624)	As (1953)	
					Cd (0.98)	Cd (454)	
					Sb (730)	Sb (88)	
					Tl (10.5)	Tl (402)	

Table 7 Enrichment factors of mine wastes through smelting process

	Hg	As	Tl	Cd	Sb
$EF_{A/C}$	7.6	5.8	8.0	1.0	1.3
$EF_{B/C}$	3.3	4.1	4.9	0.9	1.4

Table 8 Pearson correlations between different trace elements in the samples

	As	Cd	Sb	Tl	Hg
As	1				
Cd	0.22	1			
Sb	−0.05	0.37**	1		
Tl	0.91***	0.31*	−0.13	1	
Hg	0.67***	0.22	−0.13	0.72***	1

*** p < 0.001; ** p < 0.01; * p < 0.05

groups: As–Hg–Tl and Cd–Sb. In other words, the results were identical with those of the EF analysis and indicated that the crude ores were the main pollution source and the smelting process caused significant enrichment of Hg, As, and Tl in the mine wastes.

4.3 Implications for phytoremediation

This study revealed heavy metal and metalloid (Hg, As, Tl, Cd, and Sb) contamination and gold enrichment in mine wastes. In 1998, Anderson et al. (1998) first reported harvesting a crop of gold in plants—gold phytomining. The increase in gold prices during recent decades combined with higher gold concentrations in plant tissues and higher biomass yield has made gold phytomining economically feasible (Wilson-Corral et al. 2012). Gold phytomining trials in the field have been reported in the Fazenda Brasileiro gold mine, Brazil (Anderson et al. 2005), and in the Magistral mine, Mexico (Wilson-Corral et al. 2011). An economic assessment in Mexico indicated the gross profit was US$14,537/ha after subtracting the cost of producing biomass (US$8,330/ha) (Wilson-Corral et al. 2012). In addition to gold recovery, hyperaccumulator plants can be used to extract heavy metals. Balancing the cost of phytoremediation and the economic benefit is an important issue. The Laowanchang mine is an ideal site to carry out phytomining and phytoremediation simultaneously.

5 Conclusions

Highly elevated levels of As, Hg, Tl, Sb, and Cd were observed in the mine wastes from a Carlin-type gold mine in southwestern Guizhou, China. The ecological risks all reached the "very high" level, indicating heavy metal(loid) pollution associated with this gold mine. Hg and As were the major pollutants with relative contributions of 55.9 and 24.2 % to RI, respectively. The smelting process of gold ores had significant impacts on the enrichment of As, Tl, and Hg in the mine wastes.

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