

Soil heavy metal pollution and risk assessment associated with the Zn-Pb mining region in Yunnan, Southwest China

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Received: 13 September 2017 / Accepted: 22 February 2018 / Published online: 7 March 2018
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Abstract The environmental assessment and identification of sources of heavy metals in Zn-Pb ore deposits are important steps for the effective prevention of subsequent contamination and for the development of corrective measures. The concentrations of eight heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) in soils from 40 sampling points around the Jinding Zn-Pb mine in Yunnan, China, were analyzed. An environmental quality assessment of the obtained data was performed using five different contamination and pollution indexes. Statistical analyses were performed to identify the relations among the heavy metals and the pH in soils and possible sources of pollution. The concentrations of As, Cd, Pb, and Zn were extremely high, and 23, 95, 25, and 35% of the samples, respectively, exceeded the

heavy metal limits set in the Chinese Environmental Quality Standard for Soils (GB15618-1995, grade III). According to the contamination and pollution indexes, environmental risks in the area are high or extremely high. The highest risk is represented by Cd contamination, the median concentration of which exceeds the GB15618-1995 limit. Based on the combination of statistical analyses and geostatistical mapping, we identified three groups of heavy metals that originate from different sources. The main sources of As, Cd, Pb, Zn, and Cu are mining activities, airborne particulates from smelters, and the weathering of tailings. The main sources of Hg are dust fallout and gaseous emissions from smelters and tailing dams. Cr and Ni originate from lithogenic sources.

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Keywords Heavy metals · Soil contamination · Pollution indexes · Environmental risk assessment · Multivariate analysis · Spatial distribution

Introduction

Heavy metal contamination associated with industrial activities, such as the mining, processing, and smelting of metal ores, is a global concern, as these activities are responsible for the release of heavy metals into soil, water, and air, which poses potential health risks to local inhabitants (Zheng et al. 2007; Lim et al. 2008; Li et al. 2014a; Cai et al. 2015). In particular, the mining of Zn-Pb ores is one of the primary sources of heavy metal pollution in the environment and poses a risk due to the toxicity of lead and the combined pollution of Zn, Cd, As, Cu, etc. (Jung and Thornton 1996; Rodríguez et al. 2009; Zhang et al. 2012). Heavy metal pollution in soil has attracted much attention because metals are difficult to decompose and pollution can accumulate in the food chain (Liu et al. 2013; Li et al. 2014b).

The Jinding Zn-Pb mine is a world-class zinc-lead deposit with reserves of over 10 million tons and is located in Lanping County, Yunnan Province, Southwest China. Large-scale mining activities in the Jinding Zn-Pb deposit began in the 1980s and have caused serious environmental problems in and around the area (Wen et al. 2015). Recent environmental studies have mainly focused on the heavy metal contamination of soils in the mining area (Li et al. 2009a, b) and the Bijiang River, which flows through the western edge of the mining area (Li et al. 2008a, b; Wang et al. 2009; Li et al. 2010; Yi et al. 2012). However, systematic research on the distribution of heavy metals in soils and the determination of their sources is still rare. This article presents a more complex view on the issue of heavy metal contamination and includes information relating to heavy metal polluted soils, such as their spatial distributions, the background and threshold values, the pollution indexes, and statistical characteristics.

A wide range of different assessment methods are commonly used to assess contamination and ecological risk in the areas influenced by ore mining (Yang et al., 2010; Wang et al., 2015; Bai et al., 2009; Li et al., 2014a). The contamination factor, modified contamination degree, comprehensive potential ecological risk index, and Nemerow comprehensive index were used to assess the level and distribution of the heavy metal pollution and ecological risk in the soils in and around the Jinding mine.

Multivariate analyses, such as cluster analysis (CA) and principal component analysis (PCA), are the most common statistical methods to process multivariate data in environmental studies (Li et al. 2009c; Sekabira et al. 2010; Ma et al. 2015; Hlavsová et al. 2016). They were used, altogether with correlation analysis and spatial distribution, to find the relationships between investigated heavy metals, pollution possible sources, and the main principles of heavy metal migration.

The objectives of this study are (1) to examine the degree and extent of Pb, Zn, Cd, As, Cr, Cu, Hg, and Ni contamination in soils associated with Jinding mining activities; (2) to assess the potential environmental risk; and (3) to determine the major sources of heavy metal contamination in soils.

Materials and methods

Study area

In Jinding (26° 25.5', 99° 24.5'), which has a subtropical monsoon mountain climate, the average annual temperature is 11.7 °C, and the annual average precipitation is 1015.5 mm. The wet season runs from June to October, accounting for 80% of the total precipitation, while the dry season runs from November to May.

As the largest Zn-Pb deposit in China, Jinding Zn-Pb mine (Fig. 1) covers a surface area of approximately 8 km², and the deposit has reserves of ~200 Mt. of ore, grading 6.08% Zn and 1.29% Pb, i.e., a metal reserve of ~15 Mt. (Xue et al. 2007). Jinding is also rich in Tl, Cd, Ag, and Sr, the contents of which are higher than those in the standard large-scale deposits in China. There are over 100 ore bodies in the Jinding ore district, which are divided into seven ore blocks. The host rocks are sandstones for the Upper Ore Zone and breccia-bearing sandstones and breccia for the Lower Ore Zone. The main minerals in the primary ores are sphalerite (ZnS), galena (PbS), and pyrite (FeS₂). Minor ores of chalcopyrite (CuFeS₂), wurtzite (ZnS), argentite (Ag₂S), and tetrahedrite (Cu₁₂Sb₄S₁₃) are also identified (Xue et al. 2007).

The mining history dates back to the Ming and Qing Dynasty, with evidence of slags for silver left by ancient workers in Paomaping. The Jinding Zn-Pb deposit was discovered in 1960 and subsequently explored in the 1960s–1980s. Large-scale mining activities in the Jinding Zn-Pb deposit began in the 1980s and have continued since. In the 1980s, there were over 100 underground pits

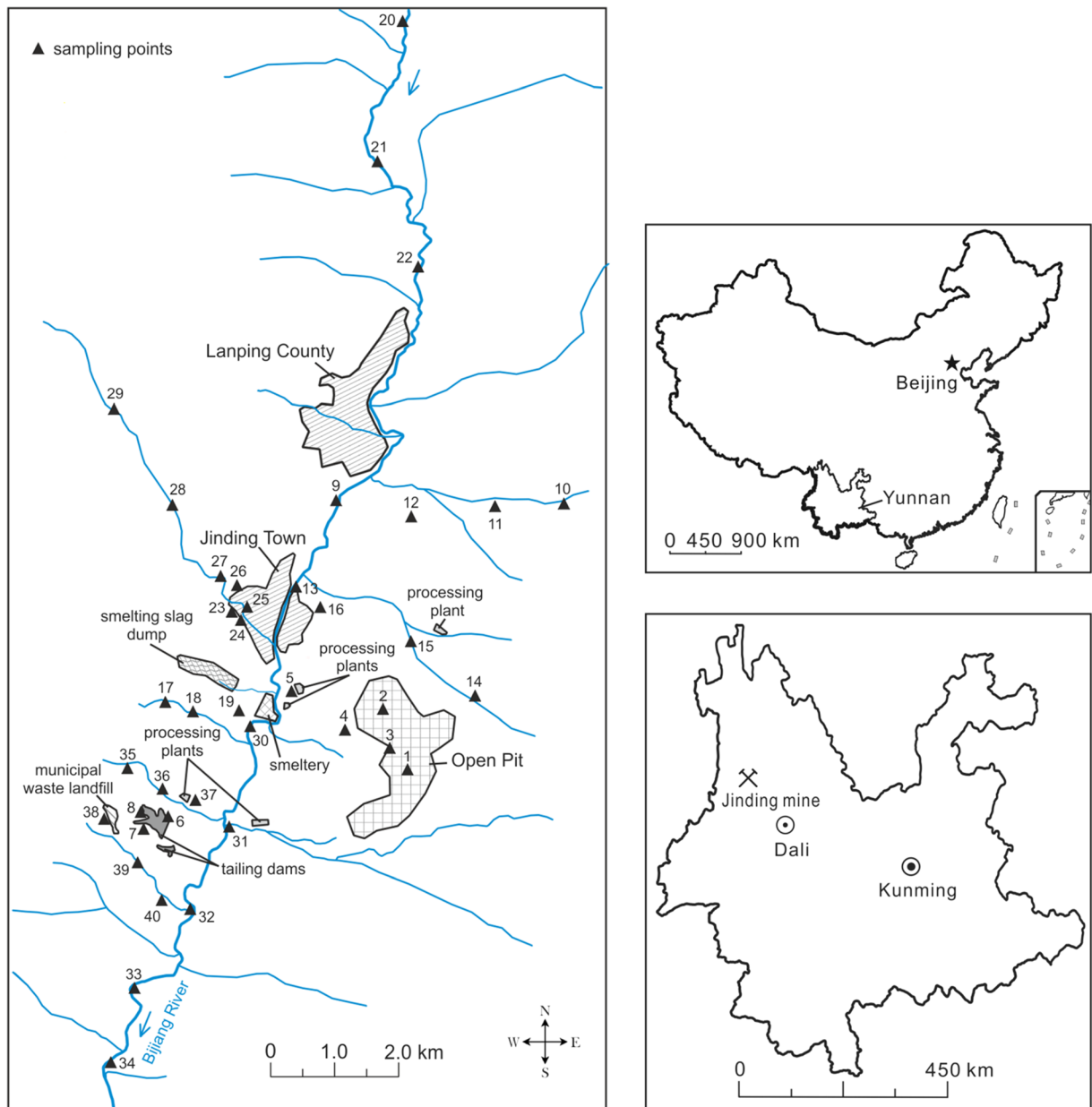


Fig. 1 Locations of sampling points in the Jinding mining area

due to unauthorized and unsystematic mining. In the 1990s, the local government reorganized the Jinding mining order, and opencast mining has become the major development method. In 2016, the open pit covered approximately 0.7 km². At present, approximately 80% of the ores come from surface mining, including the primary (sulfide) and secondary (oxidized) ores. The remaining 20% of the ores come from underground mining, mainly including primary (sulfide) ores (Wen et al. 2015). Waste

rock is piled in two dumps near the open pit. There are several processing plants around the open pit and two large tailing ponds. Large-scale smelting has been carried out in the vicinity of Jinding mine since 2005. To address the past and current environmental problems, many countermeasures in mining, processing, and smelting have been implemented. There are several villages and much arable land in the vicinity of the Jinding mining area and along the Bijiang River and its tributaries.

Sampling

A total of 40 soil samples were collected from the study area in January 2016. The locations of the sampling points are shown in Fig. 1. Three samples were collected directly in the open pit, and 37 samples were drawn from the areas surrounding the open pit, i.e., the premises of the processing plant, the smelter, and the tailing ponds. The majority of the sampling points were along the Bijiang River and its tributaries, where numerous agricultural fields are located. Soil samples were collected from the soil horizon. The maximum sampling depth was 20 cm. Soil samples were stored in polyethylene zip bags and transferred to the laboratory.

Sample preparation and chemical analysis

All samples were processed and analyzed at the Ministry of Land and Mineral Resources Supervision and Test Center Kunming, ISO 9001 and ISO 17025 Accredited Lab in China. The samples were oven-dried at a temperature of 40 °C for 72 h, disaggregated, sieved through a 0.15-mm plastic sieve, and milled for chemical analysis. The total concentrations of Pb, Cd, Cr, Cu, Zn, and Ni were determined by inductively coupled plasma mass spectroscopy (ICP-MS). The total As content was determined by atomic fluorescence spectroscopy (AFS), while the total Hg content was determined by cold vapor atomic fluorescence spectrometry (CV-AFS). All analytical data were subjected to the strict quality assurance and control. Only Guaranteed Reagent from Sinopharm Chemical Reagent Co. Ltd. was used for analytical tests in this study. The ultrapure deionized water was used for the test. The duplicate sample method was used with relative deviation less than 10%. Detection limits for analyzed elements and used analytical methods are shown in the Table 1. The pH of soil samples was determined by a pH meter INESA PHS-25. Soil sample was mixed with ultrapure water in polyethylene centrifuge tube. The mixture was centrifuged and measured in the supernatant solution after 1 h shaking.

Data analysis and assessment method

Statistical methods and geostatistical mapping

Exploratory data analysis (EDA), statistical analysis for descriptive statistics, and correlation analysis were performed using the statistical software Statgraphics Plus

5.0 and OriginPro 8.5. The Shapiro-Wilk test was performed for normality testing of the datasets. The significance levels of $p < 0.05$ and 0.01 were used for all statistical analyses and tests.

Information about the geochemical background values and threshold values in Yunnan province is very limited, and we found only two studies detailing the values (Bai et al. 2009; Zhong et al. 2016). For this reason, to determine the geochemical background values and threshold values, we used the calculated distribution function method described by Matschullat et al. (2000). This is a non-parametric method that eliminates outliers to obtain a normal distribution of the dataset. As reported by Matschullat et al. (2000) and Nakić et al. (2007), the calculated distribution function method is suitable to determine the geochemical background in connection with the anthropogenic activities responsible for an increase (positive anomalies) in the observed parameter. Geochemical background concentrations and threshold concentrations were calculated from soil samples taken from peripheral regions of the study area that were unaffected by mining or ore processing.

To identify the relations among the heavy metals and the pH in soils and possible sources of pollution, correlation analysis and multivariate analysis (cluster analysis and principal component analysis) were performed. Currently, cluster analysis (CA) and principal component analysis (PCA) are the most common statistical methods to process multivariate data in environmental studies (Li et al. 2009c; Sekabira et al. 2010; Ma et al. 2015; Hlavsová et al. 2016). Because most of the data were not normally distributed, the Box-Cox transformation was performed to ensure the assumption of normality. The transformed data were subsequently used in the multivariate analysis.

The essence of CA is to group objects (samples) and variables into clusters based on the similarity (or dissimilarity) of their chemical properties. The main advantage of CA is its ability to display multi-dimensional datasets in n -dimensional space, where n is the number of monitored variables. CA was performed using Ward's hierarchical clustering method with the Euclidean distance. The results of CA are shown in a dendrogram. The essence of PCA is a linear transformation of original variables into a smaller number of uncorrelated latent variables (principal components, PCs). The results of PCA are shown in a biplot. The multivariate analyses (CA and PCA) were performed using the OriginPro 9.1 software (trial version).

Table 1 Analytical methods used for soils

Elements	Methods	Instrument model	Detection limit	Instrumental error
Cr	ICP-MS	iCAP Q (Thermo Fisher Scientific)	0.82 mg/kg	ΔlgC < 0.05
Cd	ICP-MS	iCAP Q (Thermo Fisher Scientific)	0.015 mg/kg	ΔlgC < 0.05
Pb	ICP-MS	iCAP Q (Thermo Fisher Scientific)	0.96 mg/kg	ΔlgC < 0.05
Total As	AFS	AFS 3100(Beijing Haiguang Instrument Co., Ltd)	0.27 mg/kg	ΔlgC < 0.05
Total Hg	CV-AFS	XGY-1011 (Institute of geophysical and geochemical exploration CAGS)	0.0004 mg/kg	ΔlgC < 0.05
Cu	ICP-MS	iCAP Q (Thermo Fisher Scientific)	0.89 mg/kg	ΔlgC < 0.05
Zn	ICP-MS	iCAP Q (Thermo Fisher Scientific)	2.15 mg/kg	ΔlgC < 0.05
Ni	ICP-MS	iCAP Q (Thermo Fisher Scientific)	0.44 mg/kg	ΔlgC < 0.05

Geostatistical mapping was performed using Surfer 13 (Golden Software, USA). The method of ordinary kriging with an omnidirectional variogram was used for grid calculation. Based on the cross-validation results, the linear interpolation method was applied to construct spatial distribution maps of the heavy metals and pH in the soil (0–20 cm). As in the multivariate analysis, the transformed data were used for geostatistical analysis. Finally, the transformed data were converted using back transformation into the normal (original) values that appear on the maps.

Assessment methods

To assess heavy metal pollution in the soils, the contamination factor, modified contamination degree, comprehensive potential ecological risk index, and Nemerow comprehensive index were examined.

The contamination factor C_f^i was proposed by Håkanson (1980) and is calculated as follows:

$$C_f^i = \frac{C_i}{C_n^i} \tag{1}$$

where C_i is the concentration of heavy metal i in the soil sample (mg/kg) and C_n^i is the geochemical background value of metal i . The geochemical background values for As, Cd, Cr, Cu, Ni, Pb, Zn (Bai et al. 2009), and Hg (Zhong et al. 2016) in the whole Yunnan Province were used in the calculation. These values are summarized in Table 2. Håkanson (1980) divided the C_f value into four categories: $C_f < 1$, low contamination; $1 \leq C_f < 3$, moderate contamination; $3 \leq C_f < 6$, considerable contamination; and $C_f \geq 6$, very high contamination.

The degree of contamination mC_d was calculated based on Abraham’s modification of the Håkanson

contamination degree C_d . mC_d represents a generalized form of the overall degree of contamination at a sampling point (Håkanson 1980; Abraham 2005). Eq. (2) can be used to express the modified contamination degree.

$$mC_d = \frac{\sum_{i=1}^N C_f^i}{N} \tag{2}$$

where N is the number of elements analyzed and C_f is the contamination factor. Brady et al. (2015) classified mC_d into seven grades: $mC_d < 1.5$, unpolluted; $1.5 \leq mC_d < 2$, slightly polluted; $2 \leq mC_d < 4$, moderately polluted; $4 \leq mC_d < 8$, considerably polluted; $8 \leq mC_d < 16$, highly polluted; $16 \leq mC_d < 32$, strongly polluted; and $mC_d \geq 32$, extremely polluted.

The Nemerow comprehensive index P_i was calculated using the following Eq. (3):

$$P_i = \sqrt{\frac{(mC_d)^2 + (C_{fmax}^i)^2}{2}} \tag{3}$$

where mC_d is the modified contamination degree calculated from Eq. (2) and C_{fmax} is the maximum value of the contamination factor calculated from Eq. (1) (Li et al. 2008c). Soil pollution according to Nemerow index can be classified into five groups: $P_i \leq 0.7$, clean; $0.7 < P_i \leq 1$, warning limit; $1 < P_i \leq 2$, slight pollution; $2 < P_i \leq 3$, moderate pollution; and $P_i > 3$, heavy pollution (Hong-Gui et al. 2012). Recently, the Nemerow index has been widely used to assess soils (Li et al. 2015), stream sediments (Singovszka et al. 2016), and waters (Cheng et al. 2016).

The potential ecological risk index comprehensively evaluates the risk behavior of a metal in the environment (Håkanson 1980). The potential ecological risk index for a discrete metal factor E_r

Table 2 Concentrations of heavy metals ($\mu\text{g/g}$) and the pH in soil samples from and around the Jinding Zn-Pb mining area

Soil	pH	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Open pit ($n = 3$)									
Range (min–max)	7.69–7.93	249–282	419–808	19.2–29.1	21.4–89.3	0.12–0.90	9.12–25.30	3800–5200	32,400–64,300
Mean \pm S.D.	7.93 \pm 0.14	275 \pm 20	452 \pm 230	27.9 \pm 5.8	34.9 \pm 40.1	0.15 \pm 0.46	12.88 \pm 9.57	3800 \pm 827	36,350 \pm 18,842
Over-limit ratio (%)	0	100	100	0	0	0	0	100	100
Around open pit ($n = 37$)									
Range (min–max)	5.67–7.89	9.21–111	0.30–83.5	34.5–81.8	24.3–49.3	0.045–0.53	5.50–35.00	23.7–6700	57.7–8100
Median	6.94	29.4	3.52	57.3	31.8	0.18	22.3	115.3	233.1
IQR	0.82	15.8	6.54	13.7	9.1	0.14	7.3	353	376
Over-limit ratio (%)	13.5	16.2	94.6	0	0	0	0	21.6	29.7
Background value	6.69	22.19	2.07	55.74	27.91	0.17	20.94	58.97	134.10
Threshold value	< 5.78; > 7.60	39.78	4.22	73.61	31.98	0.35	34.59	96.17	243.27
Background value in Yunnan Province ^a	–	18.40	0.22	65.20	46.30	0.12 ^b	42.50	40.60	89.70
Chinese standard for grade III soil ^c	> 6.5	40	1.0	300	400	1.5	200	500	500

^aThe background value of the soil environment in Yunnan Province—Bai et al. (2009)

^bZhong et al. (2016)

^cNational Environmental Protection Agency of China, 1995 (GB15618-1995). Grade III soil refers to soil from woodlands, arable land in the vicinity of mines, and districts with high background values

and potential ecological risk index for combined factors RI are defined as follows:

$$E_r^i = C_f^i * T_r^i \tag{4}$$

$$RI = \sum_{i=1}^n E_r^i \tag{5}$$

where E_r is the potential ecological risk associated with metal i , C_f is the contamination factor of heavy metal i , and T_r is the toxicity coefficient of metal i . The toxicity coefficients for Zn, Cr, Cu, Pb, As, Cd, and Hg were determined by Håkanson (1980) to be 1, 2, 5, 5, 10, 30, and 40, respectively. The degree of E_r is classified as follows: $E_r < 40$, low risk; $40 \leq E_r < 80$, moderate risk; $80 \leq E_r < 160$, considerable risk; $160 \leq E_r < 320$, high risk; and $E_r \geq 320$, very high risk. Håkanson (1980) evaluated the RI of eight parameters (As, Cd, Cr, Cu, Hg, Pb, Zn, and PCBs). Because PCBs are not included in this study, it is necessary to modify the original grading for the RI using the weighted average method (Zhang et al. 2013; Wu et al. 2015; Lin et al. 2016). The modified grades for the RI of seven heavy metals are as follows: $RI < 105$, low risk; $105 \leq RI < 210$, moderate risk; $210 \leq RI < 420$, considerable risk; and $RI \geq 420$, very high risk (Lin et al. 2016).

Results and discussion

Heavy metal concentrations

Based on EDA, we found that most of the variables were not normally distributed and had many outliers. The outliers can be very important for the identification of pollution sources and therefore were not excluded. Thus, the median and interquartile range (IQR) was used to estimate the measure of location and variability. For soil samples from the open pit ($n = 3$), we used the arithmetic mean from the two nearest concentrations to estimate the location. As stated by Meloun and Militký (2011), this approach is better than the calculation of the median from all three values. To estimate the variability (the standard deviation), we proceeded according to Eq. (6), as defined by Potts (1992):

$$s = \frac{w}{k} \tag{6}$$

where s is the standard deviation, w is the difference between the highest and lowest value, and k is a factor dependent on

the number of values (observations). The value of k for $n = 3$ is 1.693.

The concentrations of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn ($\mu\text{g/g}$ dry weight) and the pH in the surface soils are given in Table 2. Because of the extremely high concentrations of As, Cd, Pb, and Zn in the soil samples from the open pit compared to the other samples, the samples from the open pit were treated separately.

The concentrations of heavy metals were compared with the Chinese standard (GB15618-1995) for grade III soils (soil from woodlands, arable land in the vicinity of mines and districts with high background values) (Bai et al. 2009; Zhong et al. 2016). Although the medians of the metal concentration are relatively low and do not exceed the limits of the GB15618-1995 standard (except for Cd), the concentrations of As, Cd, Hg, Pb, and Zn exceed the standard limits in many sampling points. Moreover, the median concentrations of these metals are high above the background values for soils from Yunnan province (Bai et al. 2009; Zhong et al. 2016). On the other hand, the median concentrations of Cu, Cr, and Ni are below both the standard limits (GB15618-1995) and the Yunnan background value (Bai et al. 2009; Zhong et al. 2016).

The pH of the soils ranged from 5.67 to 7.93 with a mean of 7.08, indicating the neutral nature of most of the soil samples. The pH of five of the samples was below 6.5, i.e., the limit of the Chinese standard (GB15618-1995) for grade III soil. The soil sample with the lowest pH of 5.67, indicating an acidic nature, was collected from the forest around the largest tailing pond. The soil pH of most of the samples was not significantly modified from the neutral value, which may be attributed to the high content of calcite. Calcite is an important gangue mineral and can neutralize acidic water.

The concentration of As in the soils ranged from 9.21 to 282 $\mu\text{g/g}$; 22.5% of the samples exceeded the safe limit of 40 $\mu\text{g/g}$ set by GB15618-1995, and 75% of the samples were above the natural background values of Yunnan Province soils (Bai et al. 2009; Zhong et al. 2016). The concentration of As in the soils from the open pit ranged from 249 to 282 $\mu\text{g/g}$, and outside the open pit they were between 9.21 and 111 $\mu\text{g/g}$. The highest concentration of As in the analyzed soils was 15 times higher than the natural background values and seven times higher than the limit set in GB15618-1995.

The concentration of Cd in the soils was very high and ranged from 419 to 808 $\mu\text{g/g}$ in the open pit and between 0.30–83.5 $\mu\text{g/g}$ outside the open pit; 95% of

the samples exceeded the safe limit of 1.0 $\mu\text{g/g}$ set in GB15618-1995, and all the samples showed Cd values that were far above the natural background of soils from Yunnan Province. The high concentration of Cd in the soils may be caused by the presence of Cd in the isomorphic admixture of sphalerite and smithsonite (Li et al. 2008b; Li et al. 2009a). Moreover, the calculated background value of Cd in soils from the Jinding mining area, 2.07 $\mu\text{g/g}$, exceeds the safe limit of 1.0 $\mu\text{g/g}$ set in GB15618-1995. This value is close to the background value around Jinding calculated by Yan (1.70 $\mu\text{g/g}$) (Yan 1998). The highest analyzed values of Cd are extreme and are over 800 times higher than that in GB15618-1995 and over 3700 times higher than the natural Yunnan soil background.

The concentration of Pb in soils ranged from 23.7 to 6700 $\mu\text{g/g}$; 25% of the samples exceeded the safe limit of 500 $\mu\text{g/g}$ set in GB15618-1995, and more than 92% of the samples exceeded the natural Yunnan soil background. The concentration of Pb in the soils from the open pit ranged from 3800 to 5200 $\mu\text{g/g}$ and from 23.7 to 6800 $\mu\text{g/g}$ outside the open pit. The highest concentration of Pb in the analyzed soils was 128 times higher than the natural background value and 13 times higher than the limit set in GB15618-1995. The highest concentration of Pb was not found in the open pit but in the soil collected near the border of the mine premises. This may be attributed to the prevailing south winds in the Jinding mining area, which spreads Pb in the form of aerosol.

The concentration of Zn in the soils ranged from 57.7 to 64,300 $\mu\text{g/g}$. Approximately 35% of all samples exceeded the safe limit of 500 $\mu\text{g/g}$ set in GB15618-1995, and more than 92% of the samples exceeded the natural Yunnan soil background. The concentration of Zn in the soils from the open pit ranged from 32,400 to 64,300 $\mu\text{g/g}$, which is much higher than the concentration of Zn outside the open pit (58–8100 $\mu\text{g/g}$). The highest concentration of Zn analyzed in the open pit soils was 716 times higher than the natural background value and 128 times higher than the limit set in GB15618-1995.

As mentioned above, the concentration of Cr, Cu, Hg and Ni were below the limits set in the national standard GB15618-1995, and only the concentration of Hg was significantly over the natural background of the Yunnan soils (by more than seven times).

The peak values of the main heavy metal pollutants (Pb, Zn, and Cd) were compared with the concentrations in the soils from other Zn-Pb mining and smelting areas.

The results of Li et al. (2005) from Shaoxing, Zhejiang Province, China, show a higher concentration of Pb (16,397 $\mu\text{g/g}$) but significantly lower concentrations of Cd (9.3 $\mu\text{g/g}$) and Zn (1221 $\mu\text{g/g}$). According to Rieuwerts et al. (2000), the concentrations of all major heavy metals in soil from the mining area in Přebram, Czech Republic, were much lower than the results in this study. Rieuwerts et al. (2000) observed the peak concentrations of 33 $\mu\text{g/g}$ for Cd, 2070 $\mu\text{g/g}$ for Pb, and 3890 $\mu\text{g/g}$ for Zn. Some of the highest concentrations were described by Yang et al. (2010). The peak concentration of Pb in their study from Guizhou Province, China, was extremely high (37,700 $\mu\text{g/g}$). On the other hand, the peak concentrations of Cd (131 $\mu\text{g/g}$) and Zn (31,625 $\mu\text{g/g}$) were lower than our results. In addition, Yang et al. (2010) summarized the results from nine studies of soils affected by Zn-Pb smelting, and all the peak concentrations of Cd and Zn were lower than those from the Jinding mining area.

Figure 2 shows the geostatistical maps with spatial distributions of the analyzed heavy metals and the pH. The highest concentrations of As, Cd, Pb, and Zn are located at the open pit, and they spread to the surroundings predominantly southwards. The concentrations of Hg have two peaks, one in the open pit and the other close to the smelting factory. In contrast, the concentrations of Cr, Cu, and Ni have minimal values in the open pit, and their peak concentrations are outside its borders.

Contamination and environmental risk assessment

The contamination factor C_f was calculated from Eq. (1) using the background values of Yunnan Province (as shown in Table 2). The average contamination factors of soils from the open pit area decrease in the order of $\text{Cd} > \text{Zn} > \text{Pb} > \text{As} > \text{Hg} > \text{Cu} > \text{Ni} > \text{Cr}$ and outside the open pit, $\text{Cd} > \text{Pb} > \text{Zn} > \text{As} = \text{Hg} > \text{Cr} > \text{Cu} > \text{Ni}$. The contamination factors of soils from the Jinding area, based on Håkanson's classification categories, indicate that all soil samples from the open pit are highly contaminated by Pb, Cd, Zn, and As and one sample is highly contaminated by Hg (Fig. 3). The soils outside the open pit are highly contaminated by Cd (95% of samples), Pb (32% of samples), Zn (24% of samples), and As (3% of samples). More than 10% of the samples show considerable contamination by Hg. The contamination by Cr, Cu, and Ni is moderate to low in all samples.

Fig. 2 Spatial distribution maps of heavy metals and the pH in soils from the Jinding mining area

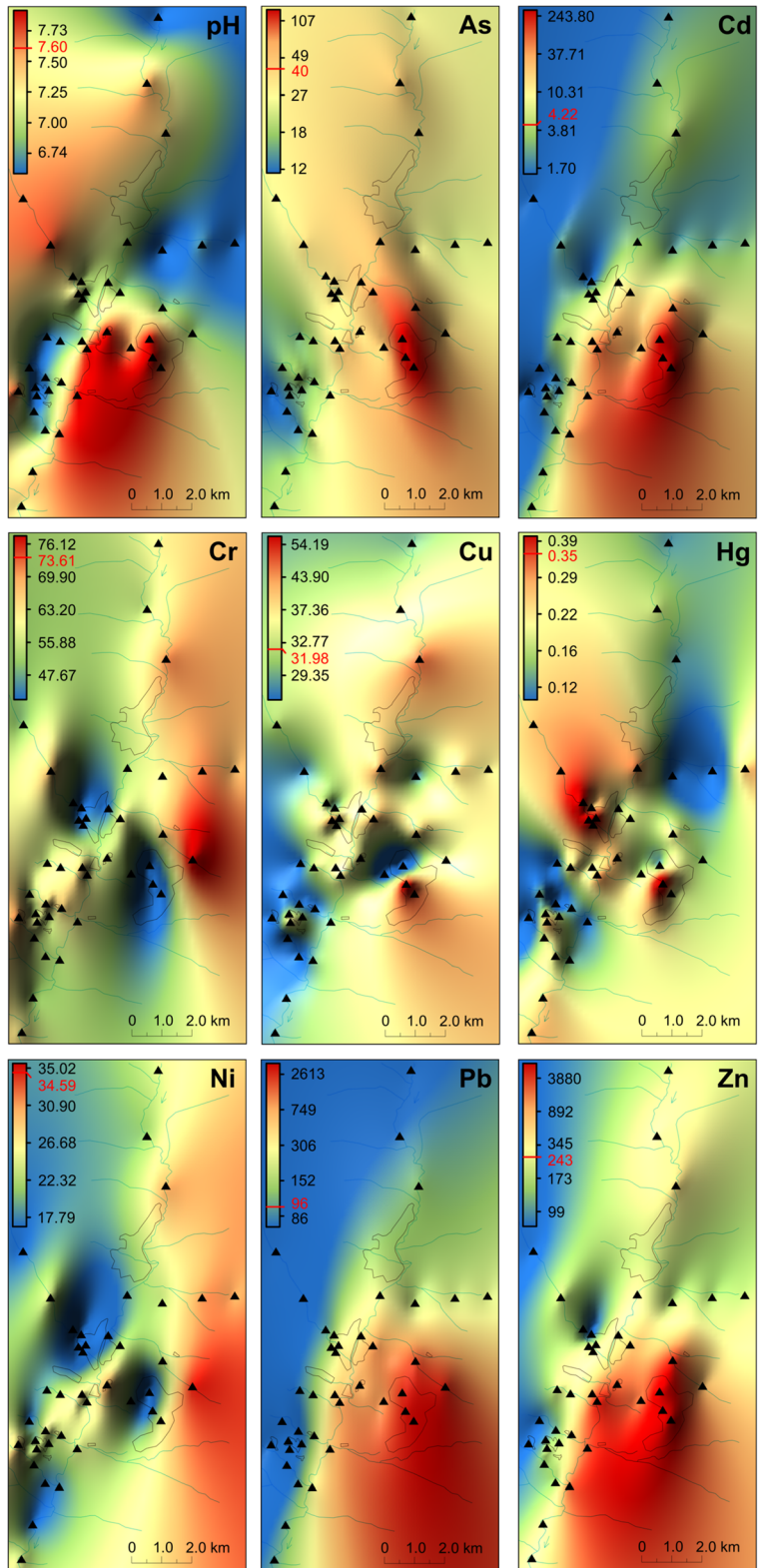
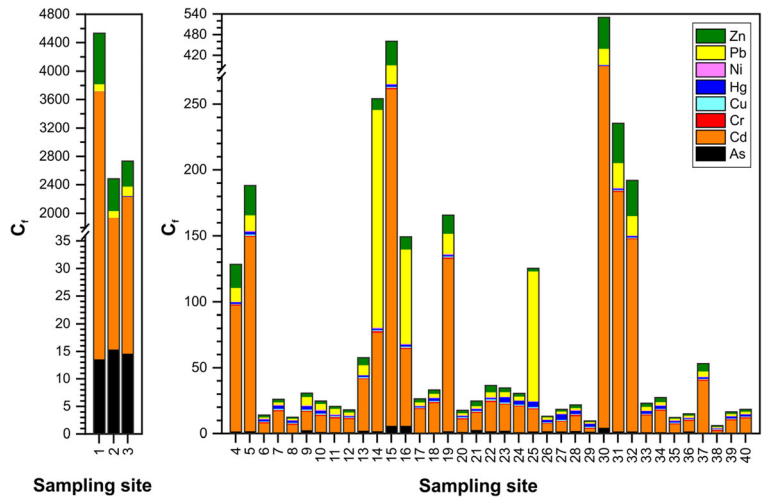


Fig. 3 Charts of the contamination factor



The mC_d values for soils from the open pit sampling points range from 310.28 to 566.75 and indicate ultra-high contamination in all three sampling points (Fig. 4). The mC_d values for the sampling points outside the open pit range from 0.74 to 66.21. According to the mC_d category, the pollution degree outside the open pit shows extreme pollution in 5% of the sampling points, strong pollution in 19%, high pollution in 3%, considerable pollution in 14%, moderate pollution in 40%, slight pollution in 14%, and no pollution in 5% of the sampling points. The average modified contamination degree indicates that the study area around Jinding mine is highly polluted ($mC_d = 10.37$).

The pollution in and around the Jinding mine is shown to be more significant when using the Nemerow comprehensive pollution index P_i . The values of P_i in the open pit

sampling points are between 1376.67 and 2651.30, indicating heavy pollution ($P_i \geq 3$). The P_i values of soils from the sampling points outside the open pit range from 1.11 to 274.86 (Fig. 5). The Nemerow index indicates heavy pollution for most soils (95%).

The average E_r for single metals in soil samples from the open pit decrease in the order of $Cd > Pb > Zn > Hg > As > Cu > Cr$ and suggest a very high risk for Cd, Pb, and Zn; very high risk for Hg in one sample; and considerable risk for As (Fig. 6). The average E_r for single metals in soil samples outside the open pit decrease in the order of $Cd > Pb > Hg > As > Zn > Cu > Cr$. A very high risk in soils outside the open pit is indicated for Cd (68% of samples) and Pb (11% of samples); high risk for Cd (27% of samples), Hg (5% of samples), and Pb (3% of samples); and considerable

Fig. 4 Charts of the modified contamination degree

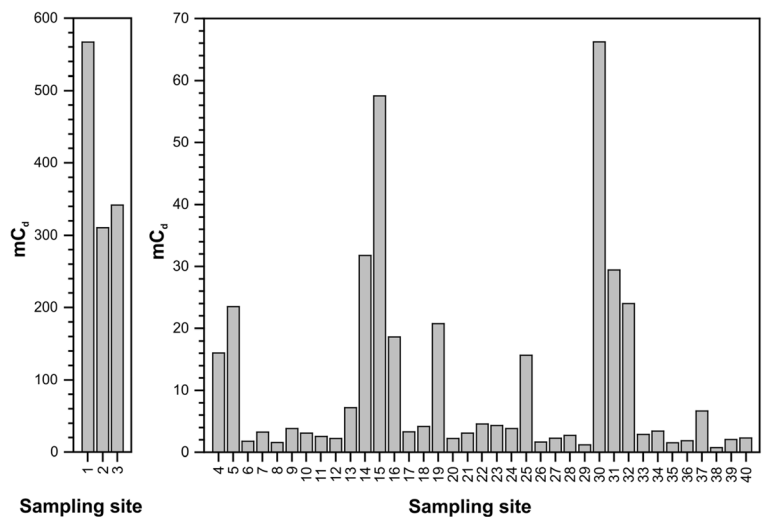
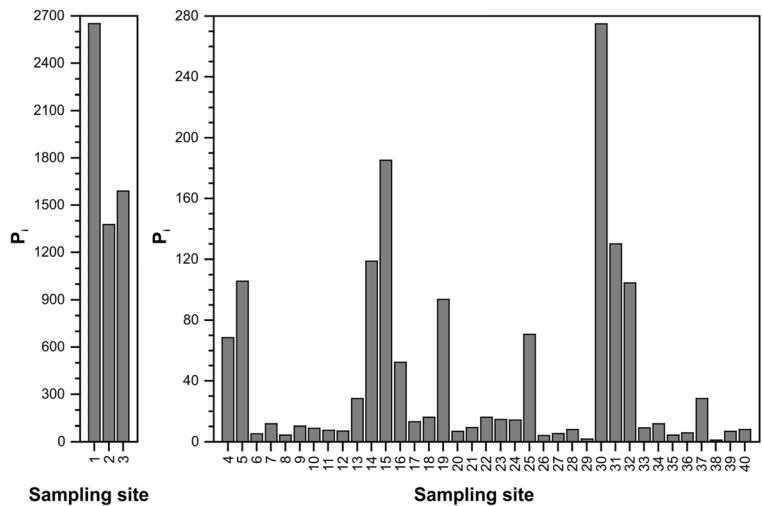


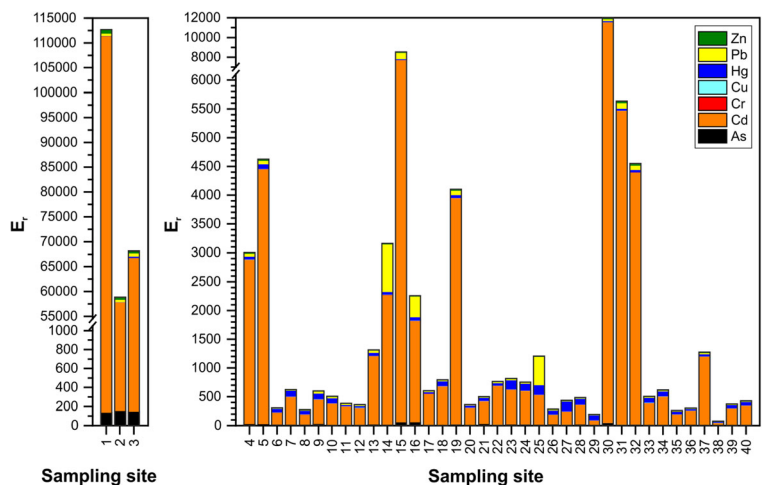
Fig. 5 Charts of the Nemerow comprehensive pollution index



risk for As (8% samples), Hg (32% samples), and Pb (3% samples). In contrast, all samples (including these from the open pit) indicate low risk for Cu and Cr.

The RI of almost all samples is extremely high. The values for soils from the open pit sampling points range between 58,774 and 112,580. According to the categories described in the subsection “Assessment methods,” these values indicate a very high risk at all three sampling points. The RI values for sampling points outside the open pit range from 71 to 11,975. According to the RI category, the ecological risk outside the open pit shows a high risk in more than 75% of the sampling points, a considerable risk in 19%, a moderate risk in 3%, and a low risk 3% of the sampling points. The average RI indicates that the study area around the Jinding mine has high ecological risk (RI = 1700).

Fig. 6 Charts of the potential ecological risk index



The extremely high concentration of Cd together with the very high ecological risk expressed by E_p and mC_d agrees with the study of Wang et al. (2015), which showed high values of Cd in the blood in humans living in the Lanping area. Their results also indicate a negative dependence of the Cd concentration in human blood on the distance from the mining district.

Correlation analysis results

Correlation analysis of the concentrations of heavy metals and the pH in soil samples in the Jinding Zn-Pb mining area ($n = 40$) was performed. Since most variables were not normally distributed and had many outliers, Spearman’s correlation coefficient (r_s) was used. The results are shown in Table 3.

Table 3 Matrix of Spearman’s correlation coefficients for heavy metals and the pH in the soil samples

	pH	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
pH	1								
As	0.541 **	1							
Cd	0.473 **	0.595 **	1						
Cr	-0.270	-0.265	-0.111	1					
Cu	0.081	0.515 **	0.370 *	-0.063	1				
Hg	0.158	0.247	0.075	-0.418 **	0.498 **	1			
Ni	-0.089	-0.110	0.139	0.843 **	0.125	-0.383 *	1		
Pb	0.376 *	0.649 **	0.888 **	-0.091	0.489 **	0.120	0.100	1	
Zn	0.539 **	0.634 **	0.974 **	-0.095	0.357 *	0.034	0.156	0.847 **	1

* Significant at $p < 0.05$; ** Significant at $p < 0.01$

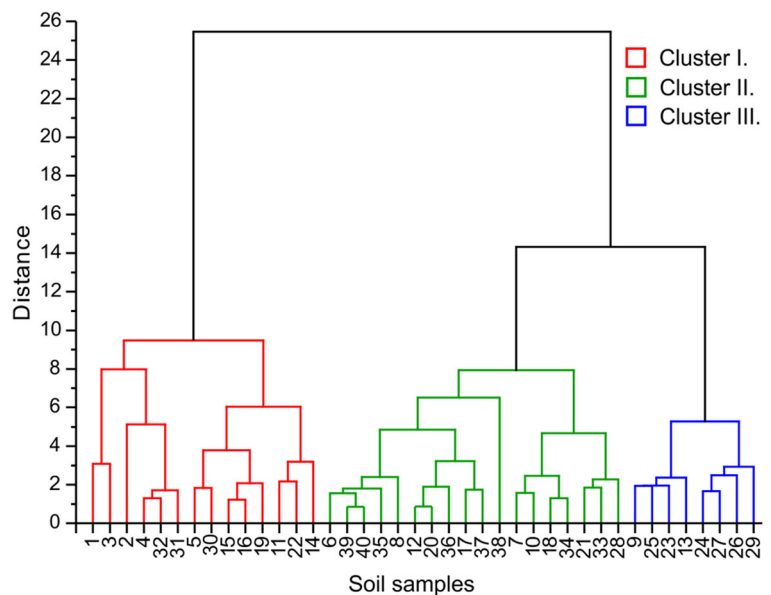
Significant positive correlations ($r_s = 0.489$ to 0.974 at a level of significance of 0.01) existed among As, Cd, Pb, Zn, As, and Cu and between Pb and Cu. In addition, a slightly weaker positive linear relationship (at a level of significance of 0.05) was found between Cd and Cu ($r_s = 0.370$) and between Zn and Cu ($r_s = 0.357$). The positive correlations indicate that the main source for this group of inorganic pollutants is the mining of Zn-Pb ores and other related processes. In addition, a significant positive dependence existed between Cr and Ni ($r_s = 0.843$) and between Cu and Hg ($r_s = 0.498$). On the other hand, a significant negative dependence between Cr and Hg ($r_s = -0.418$) and weakly negative dependence between Ni and Hg ($r_s = -0.383$) were found. These dependencies suggest the

existence of two groups of inorganic pollutants (Cr, Ni and Cu, Hg) originating from different sources. The significant linear relationship between the metals and pH was found only in As, Cd, Zn ($r_s = 0.473$ to 0.541 at a level of significance of 0.01) and Pb ($r_s = 0.376$ at a level of significance of 0.05).

Multivariate analysis results

Three groups of soil samples were obtained from the cluster analysis (CA). The CA results are shown as a dendrogram in Fig. 7. The first cluster included soil samples with the highest concentrations of As, Cd, Pb, and Zn, higher Cu concentrations, and highest pH values. This group of soil samples was collected near

Fig. 7 Dendrogram obtained by CA of the soil samples



the smelters, in mining areas, and in their close proximity. Their high concentrations are caused by intensive mining and smelting activities in this part of the study area. The results are consistent with the correlation analysis and spatial distribution of As, Cd, Pb, Zn, Cu, and the pH (Fig. 2). This is in agreement with the results of Wei et al. (2009), Yang et al. (2010) and Ma et al. (2015), who stated that the main sources of As, Cd, Pb, Zn, and Cu are mining activities of Zn-Pb ores, airborne particulates, and wastewater from smelters, the weathering and leaching of tailing and waste minerals, and acidic mine drainage.

The second cluster included soil samples taken primarily in the southwest part, as well as a smaller portion from the northeast part of the study area. This group was characterized by moderately elevated concentrations of Cr and Ni. The concentrations of Cr and Ni slightly varied, and in most soil samples, they were lower or comparable with their background values in Yunnan Province. This suggests that Cr and Ni originated mainly from lithogenic sources. This assumption is confirmed by the results of Spearman’s correlation analysis.

The third cluster consisted of soil samples with the highest concentrations of Hg. Tang et al. (2017) stated that Hg largely occurs as an isomorphous substitution for Zn in sphalerite in the Jinding Zn-Pb deposit. Mercury readily vaporizes during ore smelting and processing, especially during the roasting of Zn-Pb ores (Zhang et al. 2008). In this case, the significant source of this inorganic pollutant is the smelters and ore processing plants.

CA results for heavy metals and the pH are shown as a dendrogram in Fig. 8. The dendrogram of variables

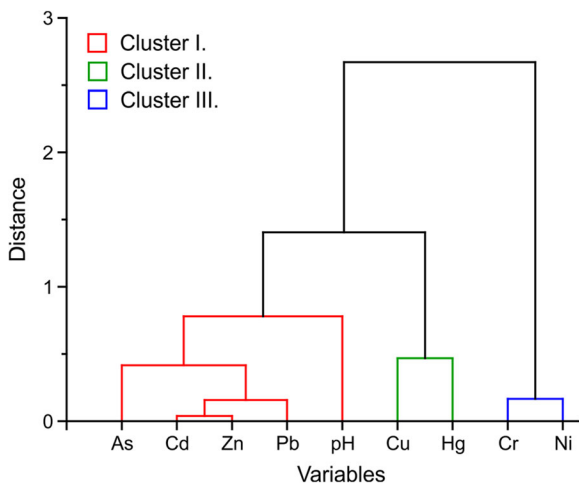


Fig. 8 Dendrogram obtained by CA for heavy metals and the pH

displays three clusters: (1) including As, Cd, Zn, Pb, and pH; (2) including Cu and Hg; and (3) including Cr and Ni. Based on the abovementioned results, the main sources of As, Cd, Zn, and Pb are mining and smelting activities, while Cr and Ni come from natural sources. The most likely sources of Cu and Hg are dust fallout and gaseous emissions from smelters and dust fallout from the dried-out parts of tailing dams (Wei et al. 2009; Křibek et al. 2010).

Similar results were obtained from principal component analysis (PCA). After performing PCA, a total of two significant principal components were extracted. The first principal component (PC1) and second principal component (PC2) accounted for 45.37 and 23.53% of the total variance, respectively, and the two components could explain 68.89% of the total variance. The PCA results are shown as a biplot representing PC1 vs. PC2 (Fig. 9).

In the biplot, three groups of soil samples can be easily identified, as in the dendrogram (Fig. 7). PC1 included Zn, Pb, Cd, Cu, As, pH, and Hg and can be defined as the mining factor. PC2 in its positive part is primarily comprised by the elemental association of Cr and Ni. The negative part includes only Hg. This is consistent with the correlation analysis, spatial distribution, and CA. The main sources of Cr and Ni are probably lateritic soils within the study area. As stated by Schwertmann and Pfab (1996) and de Oliveira et al. (2001), Cr and Ni in laterites are associated with goethite. As mentioned above, there are two main sources of Hg. The first is the processing of Zn-Pb ores, and the second is the dust fallout and gaseous emissions from smelters and tailing dams. It is clear that PC2 can

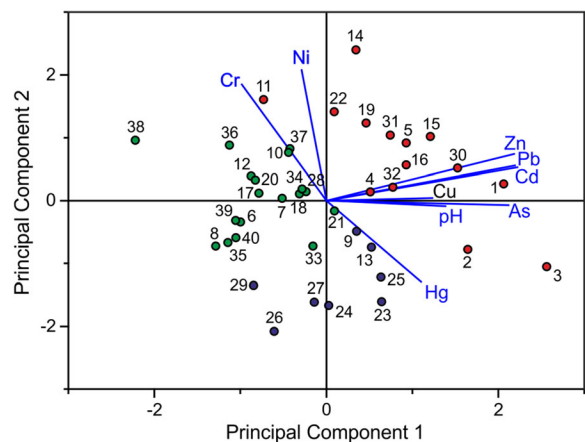


Fig. 9 Biplot of PC1 vs. PC2

be labeled as a combination of lithogenic factors and ore processing factors.

Conclusions

Based on the obtained results, the following can be concluded:

- Zinc and lead show the highest median total concentrations.
- The median total concentrations of arsenic, cadmium, lead, and zinc are significantly higher than the natural background for soils from the Yunnan province.
- The maximal obtained concentrations of As, Cd, Pb, and Zn are 7, 800, 13, and 128 times higher, respectively, than the limits for grade III soils set by the Chinese standard GB15618-1995.
- The concentration of cadmium in almost all of the samples (95%) exceeded the safe limit of 1.0 $\mu\text{g/g}$ set in GB15618-1995.
- The mean contamination factors indicate high contamination by Cd, Pb and Zn.
- The mean single metal potential ecological risk indexes indicate a very high risk for Cd and a high risk for Pb.
- Based on the mean mC_d , P_i and RI, it can be stated that the study area is extremely contaminated and heavily polluted, representing a very high ecological risk.
- The study area around the Jinding Pb-Zn mine can be regarded as a heavily polluted site, where the highest ecological risk is represented by an extreme contamination of cadmium.
- New geochemical background and threshold values were calculated for the entire Jinding mining area, namely, for As, Cd, Cr, Cu, Hg, Ni, Pb, Zn, and the pH value.
- Correlation analysis, spatial distribution, CA and PCA gave similar conclusions. All studied heavy metals can be divided into three main groups originating from three different sources. The main group of heavy metals with the highest concentrations consists of As, Cd, Pb, Zn, and Cu, and their sources are mainly mining and smelting activities. The second group consists only of Hg which originates mainly from the processing of ores, especially from the roasting of Zn-Pb ores and smelting activities.

The third group contains Cr and Ni, the concentrations of which are comparable with the Yunnan background values, and their origin is mainly lithogenic.

- Finally, the sources of a minor proportion of the Hg and Cu pollution are dust fallout and gaseous emission from smelters and tailing dams.

Acknowledgements All sources of funding of the study should be disclosed. Please clearly indicate grants that you have received in support of your research work. Clearly state if you received funds for covering the costs to publish in open access. Financial support from the Department of Land Resources of Yunnan Province of China (Grant No. Yun Land Resources Scientific 2013-1) is gratefully acknowledged. This publication has been financially supported by the Moravian-Silesian Region within the project Support of VŠB-TUO activities with China. We are particularly grateful to Lanping County Land Resources Bureau and Yunnan Jinding Zinc Industry Limited Company for help with field work. This paper was prepared within the framework of the project LO1404: Sustainable development of ENET Centre.

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