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Distribution characteristics and environmental risk assessment following metal(loid)s pollution incidents at Southwest China mining site

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Abstract: Limited information is available on long-term effects of metal(loid)s pollution incidents. Here, we analyze the distribution characteristics and quantification of elements in the Southwest China Keda mining site, which is one of the most populated sites and enables human health and ecological risk assessments of elemental pollution. The results on modified degree of contamination indicated that the soil and sediment were highly contaminated near Dahu Lake. The health risk of children was almost 2.5 times that of adults in surface water, and 7.1 times in soil, respectively. Moreover, Tl and As were the main health risk contributors in surface water and soil, respectively, and As posed the highest ecological risk both in soil and sediment. These results indicated the potential impact of toxic metal(loid)s on the health of residents and environment. Hence, more scientific attention and proper management need to be paid to this environmental challenge in the future.

Key words: thallium; arsenic; mining site; ecological risk; health risk

1 Introduction

Over the past 40 years, China has experienced rapid industrialization and economic development, which has caused adverse effects on the environment [1−4], especially the pollution incidents caused by five poisonous elements [5−8]. Despite these many investigations, thallium (Tl) pollution should not be ignored. KICIŃSKA [9,10] found that arsenic (As), cadmium (Cd) and Tl in the soil layer and grasses still have high pollution and high ecological risk in the vicinity of Zinc Works after 20 years. The abundance of Tl is very low being only 0.75 mg/kg in upper crust, 0.001−0.25 μg/L in groundwater and less than 1 mg/kg in soil [11]. Moreover, Tl is a radioactive and toxic element [12]. The adults' minimum lethal dose was 12 mg/kg and children minimum lethal dose was 8.8−15 mg/kg [13]. During the 1960s and 1970s, there are 189 cases of Tl poisoning patients with related chronic diseases, such as muscle weakness, joint pain, visual impairment and hair loss [14].

Because of critical hazard of Tl, its environmental pollution attracts attention to regulatory authorities and researchers. In recent years, the number of Tl pollution reports has increased. For example, Tl pollution reported on Beijiang River of Guangdong Province in 2008, the Tl pollution reported on Hejiang River in 2013, and the Tl pollution reported on Jialing River in 2016, caused severe damage to the natural resources and

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ecological environment [15,16].

Aside from this, the serious Tl pollution incident occurred in the Keda mining site in Southwest China. This incident was caused by the accumulation of large amounts of waste rocks and tailings [17]. The results showed that surface water of the Keda mining area was polluted [18], and the maximum concentration of Tl tremendously exceeded the permissible limit for Chinese surface water. Furthermore, the drinking water source for people in the urban area was located downstream of the mining site.

In this work, we analyzed the distribution characteristics of metal(loid)s at a mining site in Southwest China to evaluate health and ecological risks following Tl pollution incidents. We aimed to raise awareness of human exposure to Tl in such sites. These results are expected to help policy makers and researchers in assessment and strategic development of alleviating the health risk in mining sites.

2 Experimental

2.1 Study area

Baisha Town is located in the northeast of Fuchuan Yao Autonomous County, Guangxi, China, with the Baisha drainage basin running through the north−south and east−west parts of Baisha Town (Fig. 1). The county is mainly mountainous, and the subtropical monsoon characterizes the climate. The selected site for study is the Keda mining site, located in the east of Baisha Town, and the total area is about 30 km^2. Due to historical contamination and the sudden Tl pollution incident, sampling survey was carried out around the main water bodies containing Shalongchong Reservoir, Zhongnan Reservoir, and Dahu Lake.

2.2 Sample collection

Nitric acid-soaked and cleaned polyethylene plastic bottles were used to collect 1000 mL of water samples from selected sites, and immediately added 1 mL of concentrated nitric acid to adjust the pH to 1−2. Then, the water sample was brought back to the lab using dry ice and stored in refrigerator (4 °C) prior to chemical analyses. The samples of soil and sediment (5 cm in depth) were collected with an Ekman grab sampler. About 500 g samples were collected at each sampling station and stored in a clean, dense bag, and then brought back to the lab at air temperature and stored in the

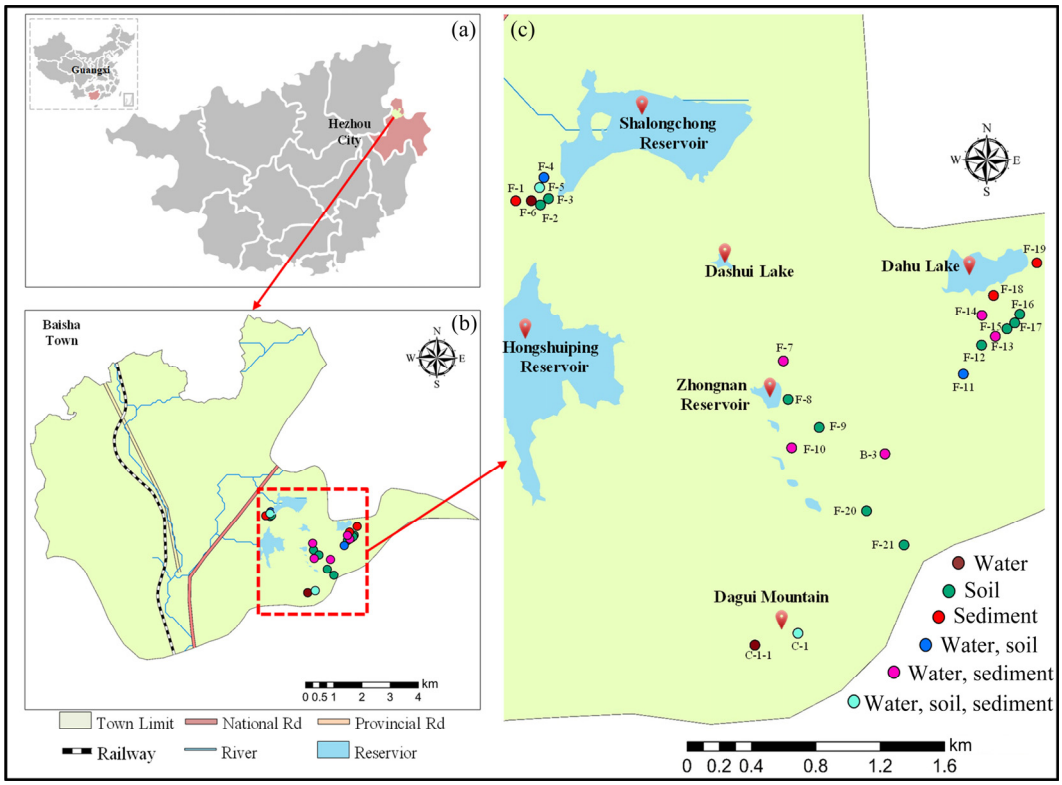

Fig. 1 Distribution map of samples included in present study: (a) Location map of Hezhou City, Guangxi, China; (b) Location map of Baisha Town; (c) Distribution map of sampling points and sampling types)

refrigerator (4 °C). Finally, stones, invertebrate and plant residues were removed from the sample, then air-dried and passed the 150 μm polyethylene sieve [19,20].

2.3 Sample sites

For areas with more human activity, high-density sampling (about one sample every 0.03 km^2) of the soil, river sediment, and surface water was taken. A total of 24 sites were sampled in 4 areas at the Dagui Mountain, Shalongchong Reservoir, Zhongnan Reservoir, and Dahu Lake, where the pollution was most acute, including 11 surface water samples, 14 soil samples, and 10 sediment samples, as shown in Table S1 (in Supplement Materials) and Fig. 1.

2.4 Sample analysis

Soil and sediment samples were air-dried, crushed, and then passed the 150 μm polyethylene sieve [15]. Samples containing 500 mg of soil or sediment were accurately weighed, stored in a crucible, and mixed with concentrated nitric acid (1.42 g/mL, 5 mL, guaranteed reagent) and hydrofluoric acid (1.49 g/mL, 2 mL, guaranteed reagent). The mixture in the crucible was heated (150 °C) until dry [21], and perchloric acid was added (1.76 g/mL, 1 mL, guaranteed reagent) to the solution and heated to digest the residue. The residue in the crucible was dissolved using 5 mL dilute nitric acid (1:1), and transferred the digestion solution into the volumetric flask (25 mL). Finally, adding deionized water to the required volume. Sample solutions were stored in the refrigerator at 4 °C prior to chemical analysis.

The glass electrode probe was used to measure the soils pH (HJ 962−2018) [22]. Briefly, soil samples were dried, sieved and then mixed with distilled water (2.5:1 v/w, solution/soil). The pH of the solution was determined with a commercial test probe (PHS−3C) after stirring and standing.

The concentrations of Cr, Cu, Cd, and Pb in surface water samples were determined by ICP-AES method (PerkinElmer, Optima 5100DV) according to Refs. [23,24]. The method of flame atomic absorption spectrometry was used to determine the concentrations of Cr and Cu, and the method of graphite furnace atomic absorption spectrometry was used to determine the concentrations of Cd and Pb in soil and sediment samples [23−25]. The measurement limits of ICP-AES for Cr, Cu, Cd, and Pb in surface water samples were 0.01, 0.01, 0.003, and 0.05 mg/L, respectively; while those in soil and sediment samples were 1.5, 1.0, 0.01, and 0.05 mg/kg, respectively. The contents of Tl and As in water, soil, and sediment were determined by ICP-MS [25], and the measurement limits of ICP-MS for Tl and As in surface water samples were 0.01 and 0.02 μg/L, respectively; while those in soil and sediment samples were 0.05 and 0.01 mg/kg, respectively.

The quality control of analytical accuracy was carried out by reagent blank and reference soil. The soil standard reference material (GBW-07401) obtained from the China National Center for Standard Reference Materials was digested with the sample and used in Quality Assurance/Quality Control procedures [26].

2.5 Geoaccumulation index (I_{geo})

The geoaccumulation index I_{geo} was proposed to quantify metal(loid)s contamination [27], according to Eq. (1):

$$I_{geo} = \log_2\left(\frac{M^{E}}{1.5M^{E}_{baseline}}\right) \quad (1)$$

where M^E is metal(loid)s concentration. The value 1.5 represents the correction factor. $M^{E}_{baseline}$ is geochemical background value for the same element. The geochemical background value greatly affects the I_{geo} results. The average concentrations of elements in sediments of five provinces, which are all in the Pearl River Basin, are used as the geochemical background values [28]. The geochemical background values for As, Cd, Tl, Cr, Cu, and Pb are 15, 0.26, 0.62, 15, 30, and 30 mg/kg, respectively. Table S2 shows the I_{geo} classification.

2.6 Enrichment factor (E_f)

The enrichment factor E_f was firstly proposed by ZOLLER et al in 1974 [29]. Nowadays, it has widely been used to assess the degree of heavy metal contamination [30], and can be calculated using Eq. (2):

$$E_f = \frac{(C_{Metal}/C_{Fe})_{sample}}{(C_{Metal}/C_{Fe})_{background}} \quad (2)$$

where $(C_{Metal}/C_{Fe})_{sample}$ is the concentration ratio of a heavy metal to Fe in the tested sample and $(C_{Metal}/C_{Fe})_{background}$ is the concentration ratio of the same element to Fe in the geochemical background. Manganese (Mn), aluminum (Al) and iron (Fe) generally serve as the normalizing element to calculate the E_f of heavy metal [31]. In this work, Fe is determined as a reference element because of its high concentration in the earth's crust, and it is not easy to be dissolved in neutral and alkaline environments. In addition, the migration ability is weak and stable. The degree of metal contamination is determined according to seven enrichment factor class, which is shown in Table S3.

2.7 Modified degree of contamination (mC_d)

The mC_d was used to evaluate the pollution of multiple metal(loid)s in environmental matrices [32]. The modified equation for calculating pollution levels is given below

$$C_f^i = \frac{Me^i_{sample}}{Me^i_{baseline}} \tag{3}$$

$$mC_d = \sum_{i=n}^{i=1} C_f^i \Big/ n \tag{4}$$

where C_f^i represents the contamination degree of element i, Me^i_{sample} and $Me^i_{baseline}$ are contamination indexes of the examined sample i and the background value for the identical element, respectively, and n represents the number of analyzed elements. Table S4 shows the classification of mC_d.

2.8 Health risk assessments

Exposure to heavy metals can occur in many ways, for example, via ingesting water and food [33], dermal contact [34] and inhalation [21]. In this study, the critical receptors are the residents around the mining site who may drink the surface water and groundwater polluted by Tl, and Tl pollution in the farmland soil is also at risk [35,36]. A human non-carcinogenic health risk assessment for water and soil is presented below.

2.8.1 Risk assessment of drinking water

The drinking water in the mining site comes from surface water, which is easy to cause oral exposure. The hazard index (H_I) represents the sum of hazard quotient (H^Q) of each metal(loid)s [37,38]. The equation for calculating H^Q value is given below

$$H_I = H^Q_{Tl} + H^Q_{As} + H^Q_{Cd} + H^Q_{Cu} + H^Q_{Pb} + H^Q_{Cr} \tag{5}$$

$$H^Q = \frac{C^D_I}{R^f_D} = \frac{CI_R E_F E_D}{B_W A_T R^f_D} \tag{6}$$

where H^Q refers to the ratio of daily intake (C^D_I, mg/(kg·d) of chemicals to the reference dose (R^f_D), which is the maximum allowable daily intake that will not cause harmful effects on health [38]. C represents the concentration of metal(loid)s in drinking water (mg/L). I_R represents the daily ingestion rate of drinking water (L/(person·d)). E_F represents the exposure frequency (d/a). E_D represents the average duration of exposure (a). B_W represents the average mass of humans (kg). A_T represents average exposure time (d). In addition, the R^f_D values of Tl, As, Cd, Cu, Pb, and Cr are 3×10^{-6}, 3×10^{-4}, 1×10^{-3}, 4×10^{-2}, 0.035, and 1.5 mg/(kg·d), respectively [39]. The risk assessment parameters used for drinking water intake for Eq. (6) are shown in Table S5.

This risk assessment method is not suitable for Pb, as its health criterion is based on uptake rather than intake [40].

2.8.2 Risk assessment of dermal exposure

The primary exposure pathways for metal(loid)s in soil are via oral, respiratory, and dermal. However, only the R^f_D for oral uptake is provided in the Chinese Standard (HJ/T25—1999) in Ref. [39]. Hence, direct soil exposure risk (mainly dermal exposure) value was calculated as follows:

$$H^Q = \frac{C^D_{I\ ingestion}}{R^f_D} = C_S I_{sp} C_F \frac{E_F E_D}{B_W A_{T_1} R^f_D} \tag{7}$$

where C^D_I represents the chronic daily intake (mg/(kg·d)), C_S represents the content of metal(loid)s in soil (mg/kg), I_{sp} represents ingestion rate of soil particle (mg/d), C_F represents the conversion factor (kg/mg), and A_{T_1} represents average exposure time ($365\times E_D$, d/a). Table S6 shows the risk assessment parameters used for soil intake for Eq. (7).

2.9 Ecological risk assessment

Several methods were used to evaluate the ecological risk in soil, such as the potential ecological risk index method [41,42]. This

method was developed from the viewpoint of sedimentology according to the chemical characteristics and environmental behaviors of heavy metal elements, which is applied to the study of heavy metal pollution in soil or river sediment. At present, it is a widely used method in soil research [43]. In addition, the soil cumulative index method [44] is easy to calculate the pollution index according to the actual measurement value and the evaluation standard, the National Soil Environmental Quality Standard, and local soil background values are usually considered as evaluation criteria [45]. The pollution load index method [46] was proposed on the classification of heavy metal pollution. This method is simple and convenient, but it cannot reflect the chemical activity and bioavailability of heavy metals, and does not take into account the background differences of different pollutants [47].

In this study, the most widely used potential ecological risk index method was selected to evaluate the ecological risk in the soil:

$$C_{\mathrm{f}}^{i}=\frac{C_{\mathrm{D}}^{i}}{C_{\mathrm{R}}^{i}},\ E_{\mathrm{r}}^{i}=T_{\mathrm{r}}^{i}\times C_{\mathrm{f}}^{i},\ R_{i}=\sum_{i=1}^{n}E_{\mathrm{r}}^{i}=\sum_{i=1}^{n}T_{\mathrm{r}}^{i}\times\frac{C_{\mathrm{D}}^{i}}{C_{\mathrm{R}}^{i}} \tag{8}$$

where C_{f}^{i} represents the pollution factor for a single metal; C_{D}^{i} represents the concentration of metal(loid)s; C_{R}^{i} represents the reference value for the corresponding metal(loid)s; E_{r}^{i} represents the potential ecological risk; T_{r}^{i} represents the toxic response factor for a single pollutant, as shown in Table S7 [48]; R_i is the sum of all risk indexes for metal(loid)s. The range of pollution degree corresponding to the R_i values and the potential ecological risk indexes are shown in Table S8 [49].

3 Results and discussion

3.1 Distribution of contamination in Keda mining site

Table 1 shows pH values and concentrations of metal(loid)s in surface water. The results demonstrated that the mean concentrations of the six metal(loid)s in surface water followed the order of Cu > Pb > Cr > Cd > As > Tl. Geographically, the lowest pH value and the highest concentrations of As, Cd, Cu, and Pb were found in Dahu Lake (F-13), and the highest concentrations of Tl and Cr are Samples F-5 and F-4 in the Shalongchong Reservoir, respectively. The concentration of metal(loid)s generally exceeded the Environmental Quality Standard for Surface Water (GB 3838—2002), especially for the mean concentrations of Tl and Cu, which were 8.67 and 11970 μg/L being more than 85.70 and 10.97 times the surface water standard limit, respectively. The research area is dominated by pyrite. Mining activities lead to long-term exposure of abandoned sulfur bearing ore. In humid environment, sulfide in waste ore reacts with oxygen in air and water to form ferrous sulfate and sulfuric acid. Therefore, the surface water in some areas is very acidic. The formation of sulfuric acid promotes the dissolution of other heavy metals in minerals into surface water.

The pH values and concentrations of metal (loid)s in soil are summarized in Table 2. The results revealed that the mean concentration of metal(loid)s in soil followed the order of As > Cu > Pb > Cr > Cd > Tl. The lowest pH value was found in Sample F-12 in the northeast direction of the study site, and the highest concentrations of As, Cd, Tl, Cr, Cu, and Pb were clustered in Samples F-17 in the northeast direction, F-8 in the center of the

Table 1 pH values and concentrations of metal(loid)s in surface water

Parameter	pH	Concentration/(μg·L^{-1})					
		As	Cd	Tl	Cr	Cu	Pb
Environmental quality standard for surface water (GB 3838—2002)	6−9	50	5	0.1	50	1000	50
Min	2.45 (F-13)	0.40 (F-11)	4.00 (F-10)	1.23 (F-10)	20.00 (C-1-1)	210.00 (F-11)	60.00 (F-4)
Max	6.34 (B-3)	55.70 (F-13)	43.00 (F-13)	16.10 (F-5)	40.00 (F-4)	72100.00 (F-13)	180.00 (F-13)
Mean	4.62	8.79	20.00	8.67	30.00	11970.00	118.00

study area, F-3 in the northwest direction and F-4 in the northwest direction, F-17 in the northeast direction and F-8 in the middle, respectively (Fig. 1). The mean concentration of As was more than 60.75 times the soil standard limit, indicating the highest level of pollution.

The pH values and concentrations of metal (loid)s in the sediment are shown in Table 3. The results indicated that the average concentrations of metal(loid)s in the sediment followed the order of Tl > As > Cu > Cr > Pb > Cd. The lowest pH value was found in Sample F-1 in the northwest direction, and the highest concentrations of As, Cd, Tl, Cr, Cu, and Pb were clustered in the samples F-13, F-14, F-18 in the northeast direction and F-10 at the center of the study area, respectively. In addition, the mean concentrations of As, Cd, and Cu was more than 34.33, 3.35, and 5.19 times the soil standard limit, respectively.

Overall, the result showed that the concentration of As in sediment was lower than that in soil. The concentration of Tl in surface water exceeded 85.70 times of surface water standard limit. In addition, the concentration of Cu exceeded 10.97 times of surface water standard limit. The pollution status of Cd in three matrices was similar to that of Cu, but the most serious pollution occurred in the sediment, which exceeded 3.35 times of soil standard limit. The concentration of Pb slightly exceeded the surface water standard limit. However, the concentration of Cr did not exceed soil standard limit. LIU et al [50] reported the contamination of Tl and associated metal(loid)s in river sediments from the steel-making industry, and results demonstrated that Tl contamination was accompanied by Cd, Zn, Cu and Pb contamination. In addition, LIU et al [51] investigated Tl dispersal and contamination in river channel surface sediments of a mining, and results indicated that mining and roasting of the pyrite ore constituted the source of 6%−88% of sediment-associated Tl and Pb. These results were consistent with our study, indicating that the pollution around the mining site was mainly caused by multiple metal(loid)s.

3.2 I_{geo} results

Figure 2 shows that the I_{geo} values of As and Cu in the soil are basically higher than 3, and I_{geo} values of Cd, Cr, and Pb are lower than 2. According the standard of contamination levels (Table S2), it

Table 2 pH values and concentrations of metal(loid)s in soil

Parameter	pH	Concentration/(mg·kg^{-1})					
		As	Cd	Tl	Cr	Cu	Pb
Environmental quality standard for soil (GB15618—2018)	>7.5	25	0.6		250	100	170
Min	2.60 (F-12)	53.70 (F-3)	0.07 (C-1)	0.30 (F-17)	25.00 (C-1)	24.00 (C-1)	2.00 (C-1)
Max	6.90 (F-21)	4120.00 (F-17)	6.87 (F-8)	1.81 (F-3)	144.00 (F-4)	2210.00 (F-17)	223.00 (F-8)
Mean	5.22	1543.76	1.44	0.87	63.71	820.57	94.48

Table 3 pH values and concentrations of metal(loid)s in sediment

Parameter	pH	Concentration/(mg·kg^{-1})					
		As	Cd	Tl	Cr	Cu	Pb
Environmental quality standard for soil (GB15618—2018)	>7.5	25	0.6		250	100	170
Min	2.27 (F-1)	10.80 (F-1)	0.39 (F10)	0.40 (F10)	20.00 (C-1)	105.00 (B-3)	4.44 (F-19)
Max	6.78 (F-18)	4370.00 (F-13)	7.55 (F-14)	73000.00 (F-18)	335.00 (F-18)	1690.00 (F-13)	183.00 (F10)
Mean	5.64	883.16	2.61	12470.57	102.50	618.90	100.37
BV (Background value)		15	0.26	0.62	15	30	30

is demonstrated that As and Cu basically reflected severe pollution (Class 4), while Cd, Cr, and Pb basically reflected moderate pollution (Class 2) in all soil samples. There were significant differences between the contamination conditions at different geographical samples. The high As and Cu pollution was found for Samples F-11, F-12, F-15, F-16, and F-17 in the Dahu Lake. Meanwhile, the contamination by Cr and Pb in the northwest direction (F-2, F-3, F-4, and F-5) and in the center of the research area (F-8 and F-9) is higher than that in the northeast direction (F-11, F-12, F-15, F-16, and F-17) (Fig. 1). Moreover, Samples F-21, C-1, and F-20 in the south direction were not significantly polluted by Tl, Cr, Cu, and Pb (I_{geo}<1).

The I_{geo} in the sediment is shown in Fig. 2(b). Cu reflected moderate or severe pollution (Class 3−5) in all sediment samples while Pb reflected low or moderate pollution (Class 1 and 2). The contamination status of the metal(loid)s varied substantially at different samples. The extreme contamination of As was clustered at the stations in Dagui Mountain, the water outlet of the Zhongnan Reservoir and Dahu Lake. Although the mean I_{geo} value of Tl was far higher than 5, which reflected extreme pollution, the Tl pollution was mainly concentrated at Samples F-18 and F-19 in Dahu Lake, while other samples were less polluted. The distribution characteristics of Cr contamination showed the same trend.

3.3 E_f results

Figure 3(a) shows that the E_f of As was basically higher than 1, and E_f of others were lower than 1. According to the standard of contamination categories (Table S3), it is mainly demonstrated that As reflected minor or moderate, or moderately severe enrichment (Class 1, 2 or 3). There were significant differences between the contamination conditions for different geographical samples.

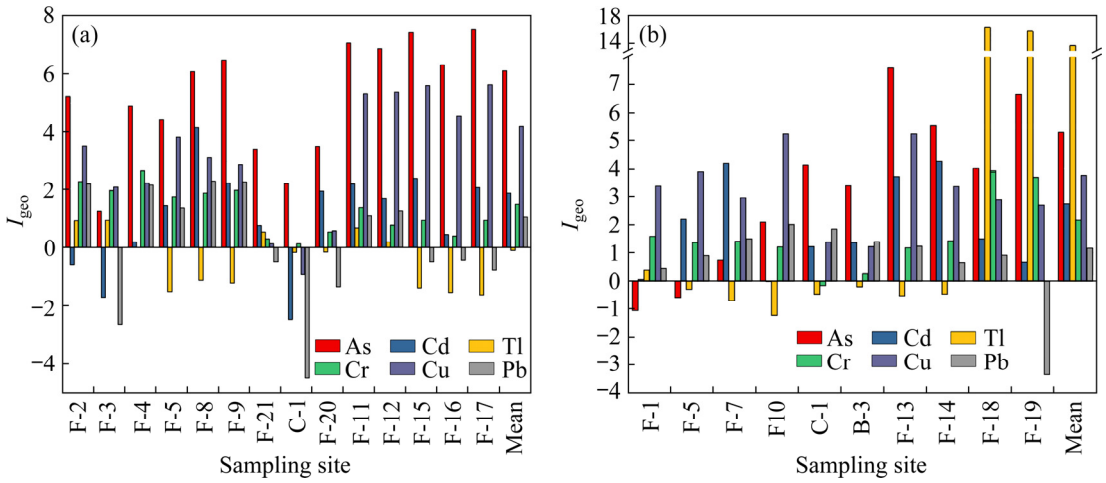

Fig. 2 Geoaccumulation indexes (I_{geo}) of metal(loid)s (As, Cd, Tl, Cr, Cu, Pb) in soil (a) and sediment (b)

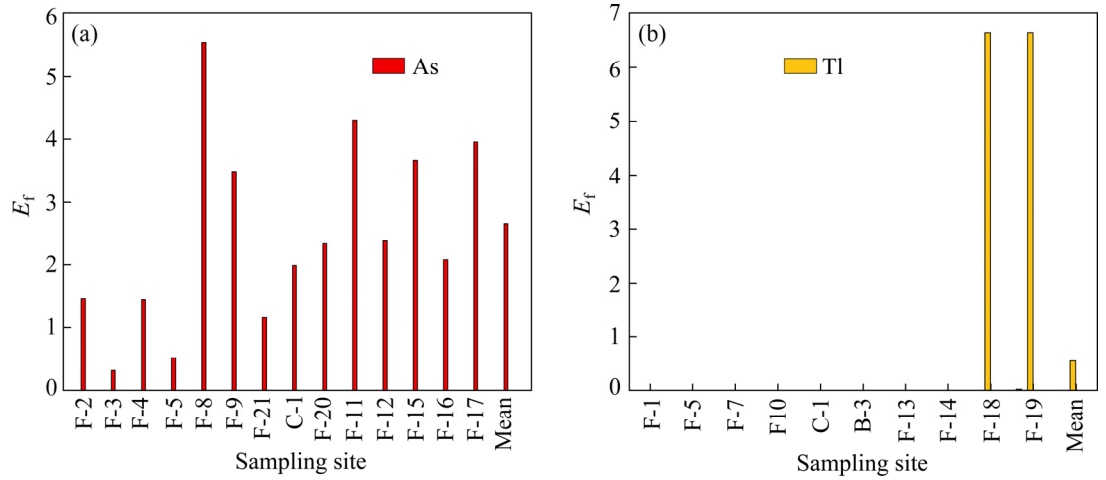

Fig. 3 Enrichment factor (E_f) of metal(loid)s (As and Tl) in soil (a) and sediment (b)

Specifically, the moderately severe enrichment of As was found for Sample F8 in the Zhongnan Reservoir, then the moderate enrichment of As was found for Sample F9 in the Zhongnan Reservoir, and for Samples F-11, F-15, and F-17 in the Dahu Lake. In addition, the E_f in the sediment is shown in Fig. 3(b). Tl reflected moderately severe enrichment (Class 3) in all sediment samples. The moderately severe enrichment of Tl was clustered at the stations in Samples F-18 and F-19 in Dahu Lake, while other samples were less polluted. In a word, the E_F of metal(loid)s showed the same trend as the I_{geo}.

3.4 mC_d results

The determination of I_{geo} indicated the contamination status of individual element, and the mC_d was also determined and used to calculate the mean contamination value of all metal(loid)s. According to the mC_d of soil shown in Fig. 4(a), Dahu Lake (F-11, F-12, F-15, and F-17) in the northeast direction indicated ultra-high contamination (Class 6, Table S4) while extreme high contamination (Class 5) was found in the center of the study area (F-8 and F-9) and in the northwest direction (F-16). Very high contamination (Class 4) occurred in the northwest direction (F-2, F-4, and F-5). In addition, Fig. 4(b) showed that the mC_d contamination status in sediment was similar to the I_{geo} results, and the sediment was ultra-highly contaminated (Class 6) in the northeast direction (F-13, F-18, and F-19), and extremely-high contamination (Class 5) occurred in the northeast direction (F-14), and very highly contamination (Class 4) occurred in the center of the study area (F-7 and F-10).

3.5 Assessment of health risks

The adult health risk assessment of surface

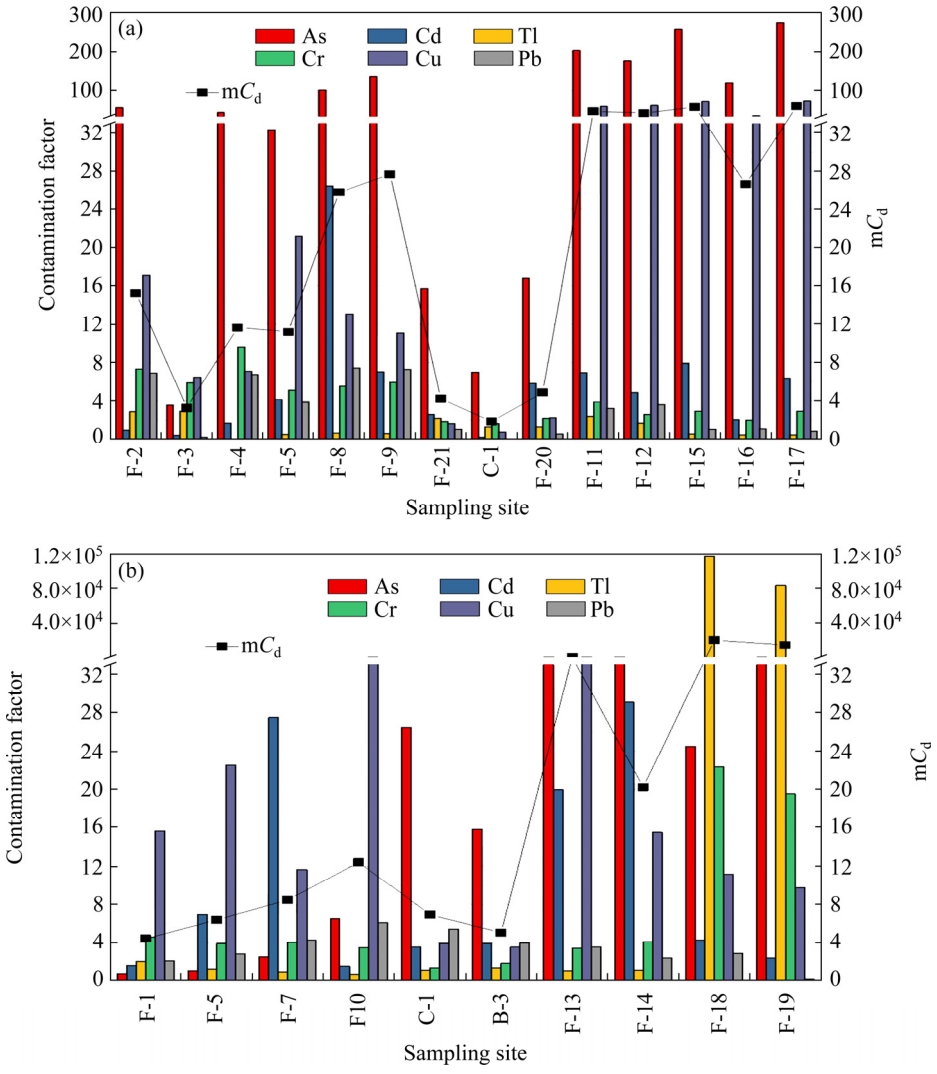

Fig. 4 Contamination status of individual element (As, Cd, Tl, Cr, Cu, Pb) and mean contamination value of all metal(loid)s in soil (a) and sediment (b)

water was calculated by Eqs. (5) and (6), and the results (Table 4) showed that Tl was the main pollution factor, the mean hazard quotient (H^Q) was 48.7893, followed by that of Cu, reaching 5.0520. The risk values (H^Q or H_I)>1.0 demonstrated possible adverse effect. The higher risk values presented the greater threat to human health, indicating the need for stricter control on the use of surface water. In addition, the health risk of children was about 2.5-fold that of adults. This research confirmed Ryan's study [52] and agreed that young children were highly exposed individuals. The risk assessment of metal(loid)s in soils was calculated by Eqs. (4) and (6), and the results (Table 4) showed that the mean H^Q of As reached 8.6873, which was the highest for adult. In addition, the H^Q of As and Tl for children reached 62.0678 and 3.4979, respectively, and the health risk for children was about 7.1-fold that of adults.

3.6 Ecological risk assessment

The potential ecological risk in soil calculated by Eq. (8) showed that As posed an extremely high ecological risk (E_r^i) in 71.4% of the sampling sites (Table 5). In contrast, the potential ecological risks of Pb, Cu, Cr and Tl were relatively low, with only few being moderate ecological risk. The highest risk index (R_i) for sample F-17 indicated a severe risk associated with multiple metal(loid)s in soil near Dahu Lake (Fig. 5).

Table 4 Results of adult and children health risks from exposed metal(loid)s in surface water and soil

Environmental matrix	Parameter	H^Q for adult						H_I for adult
		Cd	As	Tl	Pb	Cu	Cr	
Surface water	Min	0.0675	0.0225	6.9217	0.0289	0.0886	0.0002	7.1295
	Max	0.7259	3.1344	90.6007	0.0868	30.4300	0.0005	124.9783
	Mean	0.3376	0.4946	48.7893	0.0569	5.0520	0.0003	54.7308
Soil	Min	0.0001	0.3022	0.1688	0.0001	0.0010	0.0000	0.4723
	Max	0.0116	23.1848	1.0186	0.0108	0.0933	0.0002	24.3191
	Mean	0.0024	8.6873	0.4896	0.0046	0.0346	0.0001	9.2186
Surface water	Min	0.1689	0.0563	17.3085	0.0724	0.2216	0.0006	17.8282
	Max	1.8153	7.8381	226.5587	0.2171	76.0942	0.0011	312.5245
	Mean	0.8443	1.2369	122.0040	0.1423	12.6331	0.0008	136.8615
Soil	Min	0.0008	2.1590	1.2062	0.0007	0.0072	0.0002	3.3742
	Max	0.0829	165.6472	7.2772	0.0769	0.6664	0.0012	173.7517
	Mean	0.0174	62.0678	3.4979	0.0326	0.2474	0.0005	65.8636

Table 5 Potential ecological risk (E_r^i) and risk index (R_i) for metal(loid)s in soil

Sample	Location	E_r^i						R_i
		Cd	Tl	As	Pb	Cu	Cr	
F-2	Shalongchong Reservoir	13.4	71.3	990.6	51.8	6.4	0.7	1134.2
F-3	Shalongchong Reservoir	6.2	71.7	63.2	1.8	2.4	0.6	145.9
F-4	Shalongchong Reservoir	23.3	0	787.1	50.5	2.7	1.0	864.6
F-5	Shalongchong Reservoir	55.3	13.0	570.6	29.0	7.9	0.5	676.3
F-8	Zhongnan Reservoir	355.3	17.0	1788.2	55.8	4.9	0.6	2221.8
F-9	Zhongnan Reservoir	94.7	15.9	2364.7	54.5	4.2	0.6	2534.6
F-20	Zhongnan Reservoir	78.1	33.1	296.5	4.5	0.9	0.2	413.3
F-21	Zhongnan Reservoir	34.7	53.9	277.6	8.0	0.6	0.2	375.0
C-1	Dagui Mountain	3.6	32.8	123.5	0.5	0.3	0.2	160.9
F-11	Dahu Lake	93.1	59.4	3576.5	24.3	0.0	0.4	3753.7
F-12	Dahu Lake	65.2	42.4	3105.9	27.3	0.0	0.3	3241.1
F-15	Dahu Lake	106.6	14.2	4541.2	8.0	0.0	0.3	4670.3
F-16	Dahu Lake	27.9	12.8	2094.1	8.3	0.0	0.2	2143.3
F-17	Dahu Lake	85.3	12.0	4847.1	6.6	0.0	0.3	4951.3

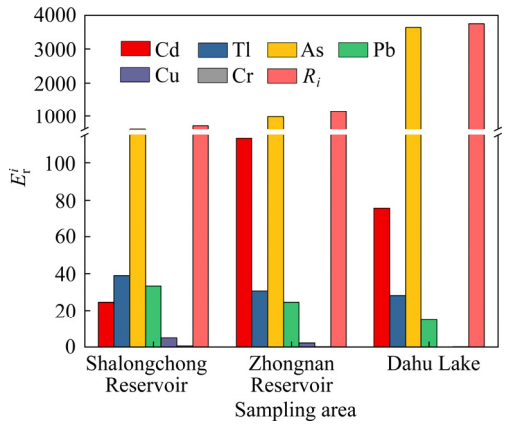

Fig. 5 Potential ecological risk (E_r^i) and risk index (R_i) of metal(loid)s in soils from different sampling areas

The ecological risk of metal(loid)s in sediment was similar to that in soil (Table 6). As remained the highest potential risk, and the ratio of the minor ecological risk among the sediment samples was 20%, and that of extremely high ecological risk was 40%. Cd posed a high potential ecological risk, of which the minor ecological risk among the sediment samples accounted for 20%, and extremely high ecological risk accounted for 50%. The composite risk value (R_i) of the sediment of Dahu Lake was higher than that of soil.

3.7 Considerations

The results of this sudden Tl pollution incident showed that water, soils, and sediments were polluted in the Keda mining site, and the ecological environment and human health were also threatened. Improper management of pollution sources maybe caused more serious secondary pollution. It was reported that Tl pollution in the environment matrices would gradually be migrated to plants [53,54]. LIU et al [55] collected soil and vegetable samples from a site 2 km away from the Tl-containing wastewater plant. Results showed that the mean concentration of Tl in soil was 1.34 mg/kg, and bioconcentration factors and transfer factors of Tl indicated the over-accumulation of Tl in plants. Compared with our present study, higher thallium concentrations were more likely to cause the overaccumulation of Tl in surrounding plants. Numerous studies showed that Tl contamination will be directly or indirectly transferred to the farmland and vegetables around the mining site [56,57], which would seriously affect the ecological environment and aggravate the harm to health of residents. DUAN et al [58] conducted the clinical characteristics examination on six children aged 1−9 years infected with Tl from 2015 to 2019, and results showed that the Tl pollution at concentrations between 13.4 and 60.1 μg/L caused heart damage on six children. Although the urinary Tl concentration dropped below 5 μg/L, these children did not fully recover their liver and kidney function, what's worse, their myocardial function deteriorated after 4 years. The mean concentration of Tl was 8.67 μg/L in our study. Such severe thallium pollution could seriously damage the health if they were exposed to children for a long period of time.

Table 6 Potential ecological risk (E_r^i) and risk index (R_i) for metal(loid)s in sediments

Sample	Location	E_r^i						R_i
		Cd	Tl	As	Pb	Cu	Cr	
F-1	Shalongchong Reservoir	21	48	12.7	15.3	5.9	0.5	103.4
F-5	Shalongchong Reservoir	93.6	30.1	17.5	20.9	8.5	0.4	171
F-7	Zhongnan Reservoir	369.8	22.5	44.1	32	4.4	0.4	473.2
F-10	Zhongnan Reservoir	20	15.7	114.6	45.75	0.0	0.3	196.35
C-1	Dagui Mountain	47.2	26.7	467.1	40.8	1.5	0.1	583.4
B-3	Water outlet of Zhongnan Reservoir	52.2	32	278.8	29.8	1.3	0.2	394.3
F-13	Dahu Lake	268.4	25.5	5141.2	26.5	0.0	0.3	5461.9
F-14	Dahu Lake	390.5	26.5	1223.5	17.7	5.8	0.4	1664.4
F-18	Dahu Lake	303.6	44	431.8	21.2	4.2	0.3	805.1
F-19	Dahu Lake	51.7	0	47.1	125	3.7	0.3	227.8

In addition, relevant research had reported that the highest Tl concentration was 0.0206 mg/L in the Tl pollution incidents of Hejiang River, with a maximum concentration higher than the permissible level by 2000 times. Moreover, more than 2×10^5 people were threatened by pollution incidents downstream of the Keda mining site [17,18]. Although the health risk and adverse effects of Tl were ambiguous, research reported that the low concentration of Tl (0.0134 mg/L) impaired children's liver, renal, and myocardial function [58]. Therefore, in order to minimize the secondary environment pollution, the following scientific management of sources based on the results was more worthy of attention.

Based on the assessment results in the mining site, it was critical to establish a long-term mechanism for environmental risk control, such as strengthening environmental supervision and pollution control. All the enterprises needed to properly dispose abandoned mines, wastewater, surface sewage, waste rock and residue, and centrally cleaned up the polluted areas, or carried out special renovation. This would gradually solve the problems left over by history, and formulated strict monitoring plans and detailed emergency plans. In order to prevent residents from mistakenly drinking contaminated water bodies in the mining site or downstream, and to avoid planting crops on the contaminated soil, daily activities and diet hygiene should be focused on, especially for local children, to control regional health risks. Moreover, according to the seasonal rainfall variation, the reasonable regulation of reservoir discharge could control the environmental risk in the Baisha River Basin and its downstream.

The next step would work out the overall plan for environmental risk system control and ecological environment reconstruction in Keda mining site. The purpose was to clarify the development layout, optimize the industrial structure, carry out special renovation, strengthen source control, and promote cleaner production. The survey found that rivers occasionally exceeded the standard, so it was necessary to continuously monitor the water quality of transboundary rivers and paid close attention to their water quality changes. We could explore the supervision mode of transboundary rivers, clarify their respective responsibilities, and gradually establish an ecological compensation mechanism. When trans-boundary water quality exceeds the standard, the relevant responsibilities should be scientifically defined and the environmental damage should be assessed.

4 Conclusions

(1) The results of metal(loid)s pollution demonstrated that the concentration of As in soil was higher than that in sediment, exceeding the soil standard limit by 60.75 times. The concentrations of Tl and Cu in surface water exceeded 85.70 and 10.97 times of surface water standard limit, respectively. In addition, the most serious pollution of Cd occurred in sediment, which exceeded 3.35 times of soil standard limit. The concentration of Pb slightly exceeded the surface water standard limit. However, the concentration of Cr did not exceed soil standard limit.

(2) The results of I_{geo} showed that As and Cu basically reflected severe pollution in soil, while Tl and As reflected extreme pollution in all sediment samples. Moreover, As reflected different enrichments in soil, and Tl reflected moderately severe enrichment in sediment. In a word, the E_f of metal(loid)s showed the same trend as the I_{geo}. In addition, the mC_d results in soil and sediment demonstrated that Dahu Lake in the northeast direction showed ultra-high contamination.

(3) The results of adults health risk assessment in surface water showed that Tl was the main pollution factor, the mean H^Q was 48.7893, and the health risk of children was 2.5-fold that of adults. In addition, results of adults health risk assessment in soil showed that the mean H^Q of As was 8.6873, which was the highest for adult, and the health risk for children was 7.1-fold that of adults.

(4) The results of potential ecological risk in soil showed that As posed an extremely high ecological risk in 71.4% of the sampling sites. In addition, As and Cd posed a high potential ecological risk in sediment.

Supplementary Materials

Supplement Materials in this paper can be found at: http://tnmsc.csu.edu.cn/download/17-p4062-2021-0927-Supplementary_Materials.pdf.

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西南某矿区金属/类金属污染事件的分布特征及环境风险评价

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摘 要：关于金属/类金属污染事件长期影响的信息有限。本文分析中国西南方可达矿区的元素分布特征和定量化，该矿区是世界上人口较多的矿区之一，能够对元素污染进行人类健康和生态风险评估。污染程度结果表明，大湖塘附近的土壤和沉积物污染程度较高。儿童在地表水和土壤中的健康风险分别是成年人的 2.5 倍和 7.1 倍。此外，铊和砷分别是地表水和土壤的主要健康风险贡献者，而砷在土壤和沉积物中构成的生态风险最高。这些结果表明，有毒金属/类金属对居民健康和环境具有潜在影响。因此，未来需要对该环境挑战给予更多的科学关注和适当的管理。

关键词：铊；砷；采矿场；生态风险；健康风险

(Edited by Wei-ping CHEN)