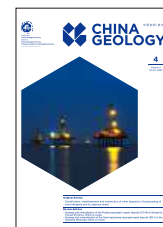




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Source analysis and risk evaluation of heavy metal in the river sediment of polymetallic mining area: Taking the Tonglūshan skarn type Cu-Fe-Au deposit as an example, Hubei section of the Yangtze River Basin, China

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ABSTRACT

In this paper, 25 sampling points of overlying deposits in Tonglūshan mining area, Daye City, Hubei Province, China were tested for heavy metal content to explore pollution characteristics, pollution sources and ecological risks of heavy metals in sediments. A geo-accumulation index method was used to evaluate the degree of heavy metal pollution in the sediment. The mean sediment quality guideline quotient was used for evaluating the ecological risk level of heavy metal in the sediment. And a method of correlation analysis, clustering analysis, and principal component analysis was used for preliminary analysis on the source of heavy metal in the sediment. It was indicated that there was extremely heavy metal pollution in the sediment, among which Cd was extremely polluted, Cu strongly contaminated, Zn, As, and Hg moderately contaminated, and Pb, Cr, and Ni were slightly contaminated. It was also indicated by the mean sediment quality guideline-quotient result that there was a high ecological risk of heavy metals in the sediment, and 64% of the sample sites had extremely high hidden biotoxic effects. For distribution, the contamination of branches was worse than that of the main channel of Daye Dagang, and the deposition of each heavy metal was mainly influenced by the distance from this sample site to the sewage draining exit of a tailings pond. The source analysis showed that the heavy metals in the sediment come from pollution discharging of mining and beneficiation companies, tailings ponds, smelting companies, and transport vehicles. In the study area, due to the influence of heavy metal discharging from these sources, the ecotoxicity of heavy metals in the sediment was extremely high, and Cd was the most toxic pollutant. The research figured out the key restoration area and elements for ecological restoration in the sediment of the Tonglūshan mining area, which could be referenced by monitoring and governance of heavy metal pollution in the sediment of the polymetallic mining area.

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1. Introduction

Sediment is an important constituent part of the water environment. However, it is also a main “source” and “gathering” of environmental pollutants, which significantly impacts the ecosystem structure and the service function of

ivers (Liu K et al., 2019). Heavy metals are typical pollutants characterized by degradation-resistant, strong toxicity, and other features (Zhang JQ et al., 2017; Liu Q et al., 2019; Yang A et al., 2020), 85% of which are deposited and enriched in overlying deposits. After years of accumulation, their concentration is stable, and the environmental effect and the degree of environmental hazards are different, which is liable to influence the biological assimilation of the aquatic organism and generate bioaccumulation of heavy metals, thus threatening the ecosystem and human health (Zhao G et al., 2017; Zhang H et al., 2017). The petroleum industry, mining, combustion of fossil fuel, and other anthropogenic

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discharging sources are regarded as the main sources of heavy metals (Tian HZ et al., 2015; Wang P et al., 2019; Liu RP et al., 2020), and the contamination level and distribution of heavy metals are largely influenced by the type of pollution sources, as well as their spatial distribution (Yin S et al., 2016; Islam MS et al., 2018). Therefore, pollution characteristics, source analysis, and ecological risks of heavy metals in sediment are important contents of the pollution survey (Liu K et al., 2019).

A considerable amount of wastewater, waste gas, and solid waste is generated during the exploitation of mines and beneficiation of minerals, and heavy metals in these wastes are deposited and transferred to surrounding soil and river via atmospheric dust-fall, overland runoff, and other ways (Wang YP et al., 1998; Wang YG et al., 2022).

As a time-honored city, the problem of heavy metal pollution in Daye City is particularly acute. Research showed that the content of seven heavy metals in the sediment of Daye, Hubei Province is relatively high (Chen GC et al., 2020). In the sediment of the Daye Lake basin, five heavy metals, including Cd, Cr, Pb, and Hg, have extremely high hidden ecological risks, and the ecological risk of Cd is the highest (Zhou GQ et al., 2016). The ecological risk of Cd is very high for four heavy metals, Ni, Cd, Cu, and Pb, in the sediment of Daye Lake. Cd and Cu mainly come from agricultural production activities. Ni is closely related to discharging of industrial wastewater, and transportation pollution is the main source of Pb (Zhang JQ et al., 2017). These researchers mainly focused on wastewater, contamination level, and the level of ecological risk of 5–7 heavy metals in the sediment of Daye Lake. And there is seldom research on the existing state, pollution sources, and the level of ecological risk of heavy metals in the sediment of the Tonglūshan mining area.

This study takes eight heavy metals, Cr, Ni, Cu, Pb, Zn, Cd, As, and Hg in the sediment of the Tonglūshan mining area as the research object. Using a geo-accumulation index method to evaluate the contamination level of heavy metal; using the mean sediment quality guideline-quotient to evaluate the level of ecological risk of heavy metal, and using the method of correlation analysis, clustering analysis, and principal component to analyze the source of heavy metal. To provide some scientific and theoretical basis for the ecological restoration of sediments and the ecological risk management of sediments in polymetallic mining areas.

2. Overview of the research area

The research area is located in the Tonglūshan mining area, Daye City, Hubei Province, China, which is a low hilly area with a continental climate, warm and humid, and abundant rainfall. The soil is mainly red earth and laterite. The mining area is a typical Cu-Fe-Au skarn-type polymetallic deposit of the ore-concentrated area in southeast Hubei Province, belonging to the outcome of metallogenic activity related to the intrusive acid rock of Yanshanian. The outcropping stratum is mainly marble and dolomitic marble of

the Daye group of the Lower Triassic (Wang MF et al., 2019). A main useful constituent of Fe, Cu, Au, etc., and associated constituents of Ag, Co, Mo, S, Pb, Zn, as well as Ga, Se, Te, W, etc., are highly concentrated.

The main rivers within the research section include the Jinhu subdistrict section of Daye Dagang, as well as its two southern tributaries. The Dagang River originates in Hongfeng reservoir, Lingxiang Town, Daye City. It flows into Daye Lake at Qiaonan Village of Kangtou, Daye City, with an overall length of 34.5 km and a catchment area of 571 km². The elevation of the river bottom slopes from west to east, and the slope of the channel is 0.6‰ (Li ZH et al., 2010). There are ten tailings ponds, two copper mill plants, one mining factory, one smelting plant, and one fishpond along the river bank within the research area. The tailings pond is flat, mainly wet discharged, constructed wetlands and drainage ditches consisting of the reed arranged near the tailings pond. Wastewater is discharged into a drainage ditch, flows into Daye Lake via Daye Dagang, and finally flows into the Yangtze River. The farmland in the research area is relatively scattered and small, mainly dry land for planting vegetables.

3. Material and method

3.1. Collection and handling of samples

In April 2021, 25 sampling sites were arranged in the sediment and sampled in the research area (Fig. 1). About 1 kg overlying deposit (0–20 cm) was sampled using grab bucket dredges and then packed into PE sampling bags and marked. All samples were sent to the laboratory of the 1st Geological Team of the Hubei Geological Bureau for handling and testing.

After the samples had been sent to the laboratory, stones, plant residue, and other sundries were removed. Samples were dried naturally under room temperature and then ground and sieved to test the content of heavy metals. Cu, Zn, Cr, Ni, Cd, and Pb were tested by using an ICP mass spectrometer (JCYQ-MS-01), and the test method was specified in HJ 803-2016; as was tested by using an atomic fluorescence spectrophotometer (JCYQ-AFS-03) and the test method was specified in DZ/T 0279.13-2016. Hg was tested using an atomic fluorescence spectrophotometer (JCYQ-AFS-01), and the test method was specified in DZ/T 0279.17-2016. The method detection limits of each element, measurement error of standard substance, and relative tolerance of repeat analysis should all meet the requirements of DZ/T0130.3-2006, the specification of testing quality management for geological laboratories.

3.2. Evaluation method

3.2.1. Geo-accumulation index (I_{geo})

The geo-accumulation index demonstrates characteristics of change for the natural distribution of sediment and heavy metal in soil, which also highlights the historical accumulation of heavy metal contamination and reflects

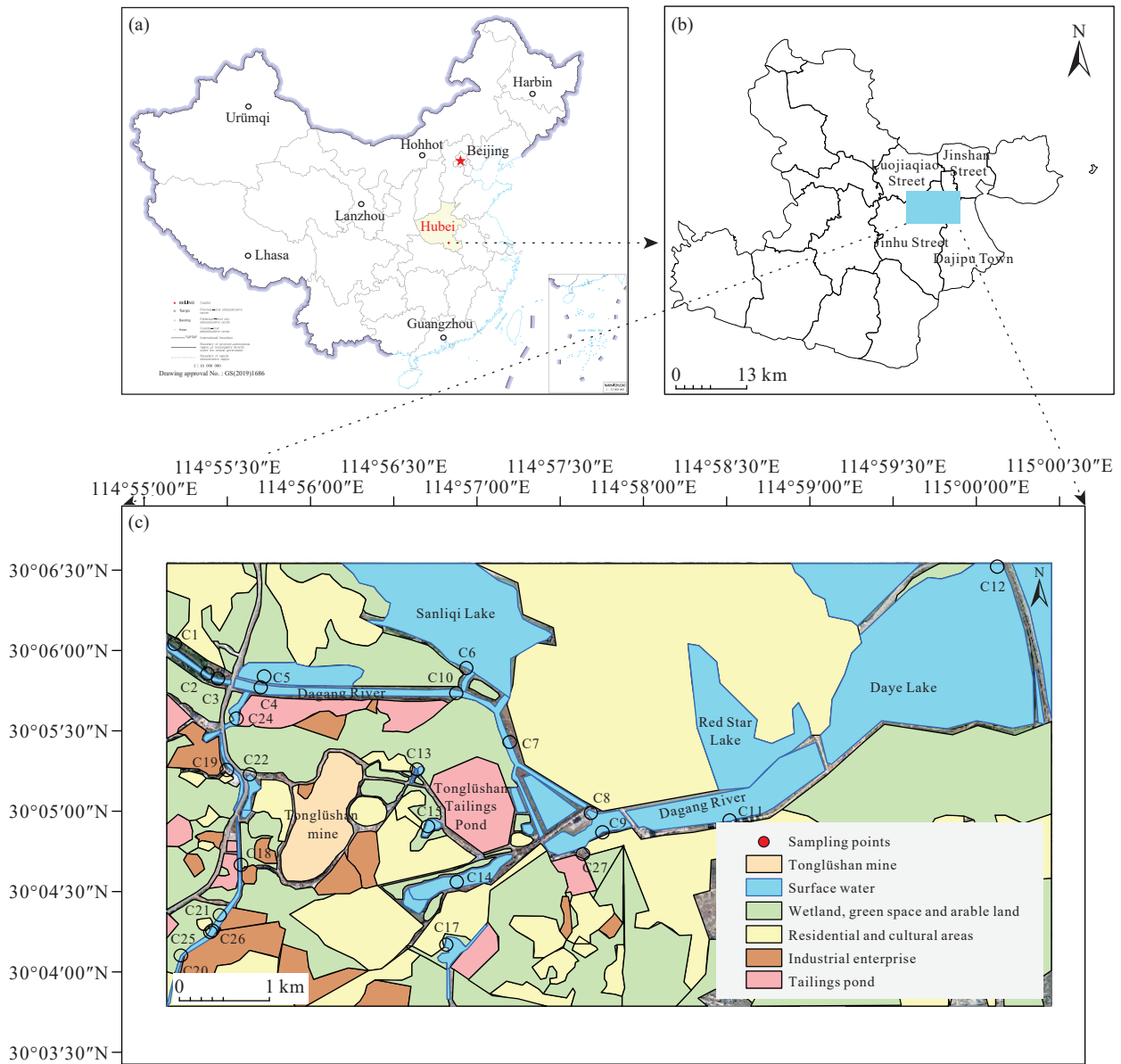


Fig. 1. Sketch map for distribution of sampling site. a–location of Daye City, China; b–location of Tonglùshan mine area in Daye City; c–location of sediment sampling sites at the Tonglùshan mine area.

influences of natural geology and anthropogenic activity on heavy metal contamination. It is widely used for evaluating the accumulated contamination status of heavy metals in sediment, soil, and other substances (Müller G, 1969; Cai YS et al., 2020; Yakovlev E et al., 2020). The expression of the method is shown below:

$$I_{geo} = \log_2(C_n / KB_n) \tag{1}$$

where: C_n is the measured concentration of heavy metal element n , mg/kg; B_n is the background value of the corresponding heavy metal element, mg/kg. The selection of background value is an essential factor, and the self-background value of the sediment of the area shall be used (Teng YG et al., 2002). Therefore, the topsoil background value of the Ezhou-Huangshi area is used in the research (Zhu LQ et al., 2020); constant K is a constant set for natural fluctuation of the content of heavy metal elements during the

diagenetic process; take K as 1.5 (Yang A et al., 2020). I_{geo} represents the contamination level of sediment (Table 1).

3.2.2. The mean sediment quality guideline-quotient (SQG-Q)

The mean sediment quality guideline-quotient combines the level of heavy metal and its influence on the bentonitic organism to comprehensively evaluate sediment’s ecological risk (O’Connor TP, 2004), which could reflect the ecological effect of the research area to some extent.

The expression of the method is shown below:

$$SQG - Q = \frac{\sum_{i=1}^n (PEL - Q)}{n} \tag{2}$$

$$(PEL - Q)_i = \frac{C_i}{PEL_i} \tag{3}$$

where: $(PEL - Q)_i$ is the probable effect level coefficient of

heavy metal i ; C_i is the measured level of heavy metal i ; PEL_i is the probable effect level of heavy metal i (Macdonald DD et al., 2000); n is the quantity of metal element. Using the calculated SQG-Q coefficient, the biological risk of heavy metal contamination in the research area could be evaluated (Li RZ et al., 2013). Generally, if $SQG-Q \leq 0.1$, the area is basically not polluted by heavy metals, with the lowest potential unfavourable biotoxicity effect; if $0.1 < SQG-Q < 1.0$, the area has a moderate potential unfavourable biotoxicity effect; if $SQG-Q \geq 1.0$, the area has the extremely high potential unfavourable biotoxicity effect (Meilina H et al., 2021).

3.3. Data processing method

Statistical analysis of data, correlation analysis of Pearson, clustering analysis, and PCA analysis are made using SPSS V26.0. Data processing and distribution map drawing of heavy metal contamination level is made using WPS and Origin 2021. Sampling sites, the concentration of heavy metal, ecological risk index, and other spatial distribution maps are drawn by using ArcGIS10.7 and CorelDRAW X4.

4. Result and analysis

4.1. Pollution characteristics of heavy metals in the sediment

4.1.1. Concentration characteristics of heavy metal in the sediment

It can be seen from Table 2 that in the sediment, Zn has the highest average heavy metal concentration and Hg is the lowest, and the variable coefficient of all elements is higher than 1, which is obviously influenced by anthropogenic pollution sources. Compared with the upper continental crust

(UCC) (Taylor SR and McLennan SM, 1995), the topsoil background value of the Ezhou-Huangshi area (Zhu LQ et al., 2020), average value of Southern China stream sediment (Cheng ZZ et al., 2011), background value of sediment of Yangtze River basin (Gao H et al., 2001), the average value of Zhujiang River sediment (Zhao G et al., 2017), the heavy metal concentration of sediment within the research area is higher. Compared with the average value of Xiang River sediment (Sheng WK et al., 2019), the heavy metal concentration (except for Cr) of sediment within the research area is higher.

Measuring the threshold of the mass basis of sediment SQGs (Macdonald DD et al., 2000) includes threshold effect level (TEL) and probable effect level (PEL) (Tang L et al., 2017). If the pollutant concentration is lower than the TEL value, there are few negative effects on the benthonic organism. If the pollutant concentration is higher than the PEL value, negative effects appear frequently. If the pollutant concentration is between the TEL value and the PEL value, negative effects probably appear (Yuan SF et al., 2020). It is known by comparing the average heavy metal concentration of the sediment with the TEL and PEL value that only when Hg concentration is higher than TEL and lower than PEL, the negative effects may appear; as the concentration of Cd, Cu, As, Zn, Pb, Ni, and Cr are 9.69–1.07 times than the PEL value, negative effects appear probably.

To sum up, heavy metal in the sediment of the research area is greatly influenced by humans, and there are different degrees of enrichment. The content of heavy metals is higher than the background value, and that of many typical southern rivers, and heavy metals in the sediment pose a threat to the aquatic system of the research area. Therefore, for heavy metals in the sediment of the research area, attention shall be

Table 1. Geo-accumulation index and contamination grading.

Item	Grading of contamination level						
	Uncontaminated	Uncontaminated to moderately contaminated	Moderately contaminated	Moderately to strongly contaminated	Strongly contaminated	Strongly to extremely contaminated	Extremely contaminated
I_{geo}	≤ 0	$>0, \leq 1$	$>1, \leq 2$	$>2, \leq 3$	$>3, \leq 4$	$>4, \leq 5$	>5
Level	0	1	2	3	4	5	6

Table 2. Concentration characteristics of heavy metal in the sediment of Tonglushan mining area.

Type	Element/(mg/kg)	Cr	Ni	Cu	Pb	Zn	Cd	As	Hg
The research area	Average value	118.46	52.53	873.86	271.86	1204.01	48.28	99.14	0.34
	Maximum	644.96	515.37	4950.14	3830.62	11638.9	747.26	782.51	2.02
	Minimum	38.14	13.10	93.30	34.58	107.65	0.56	19.48	0.06
	Standard deviation	131.74	95.04	1079.61	732.45	2389.19	152.72	160.92	0.39
	Variable coefficient	1.11	1.81	1.24	2.69	1.98	3.16	1.62	1.13
	Background value	77.83	30.06	29.69	30.55	19.72	0.195	9.9	0.071
	Sample size	25	25	25	25	25	25	25	25
UCC		35	20	25	20	71	0.098	1.5	–
The average value of the Southern China water system		0.23	29	25	32.3	81	0.23	13.1	0.075
Background value of sediment of Yangtze river basin		50	21.37	21.23	45.05	62.92	0.061	8.30	0.036
The average value of Xiang River sediment		133	32	36	98	187	7.5	52.2	0.24
The average value of Zhujiang River sediment		78.37	–	46.76	49.66	143.1	0.46	21.99	0.13
TEL		43.4	22.7	31.6	35.8	121.0	0.99	9.79	0.18
PEL		111.0	48.6	149.0	128.0	459.0	4.98	33.0	1.06

emphatically given to the content of Cd and Cu, and then As and Zn.

4.1.2. Distribution characteristics of heavy metals in the sediment

It can be seen from Fig. 2 that the maximum of Cr appears at sampling site C18, which is 8.29 times the background value. The utmost Zn, Pb, and Hg appear at sampling site C20, the concentration of which is respectively 590.21 times, 125.39 times, and 28.48 times than the background value. The maximum of Cd, Cu, As, and Ni all appear at sampling site C27, the concentration of which is respectively 3832.13 times,

166.73 times, 79.04 times, and 17.14 times than the background value. C18 and C20 are both located at the southwest branch (Xiaozhahe) of Daye Dagang, and C27 is located at the southeast branch (Zhaobao hu draining pump station) of Daye Dagang. All three sampling sites are located downstream of the drain outlet of a tailings pond, which is greatly influenced by discharged pollutants of the draining outlet.

The main channel of Daye Dagang flows through the north end of the research area from west to east, and the variation tendency of heavy metal concentration is generally stable. However, due to the influence of influent branches, the

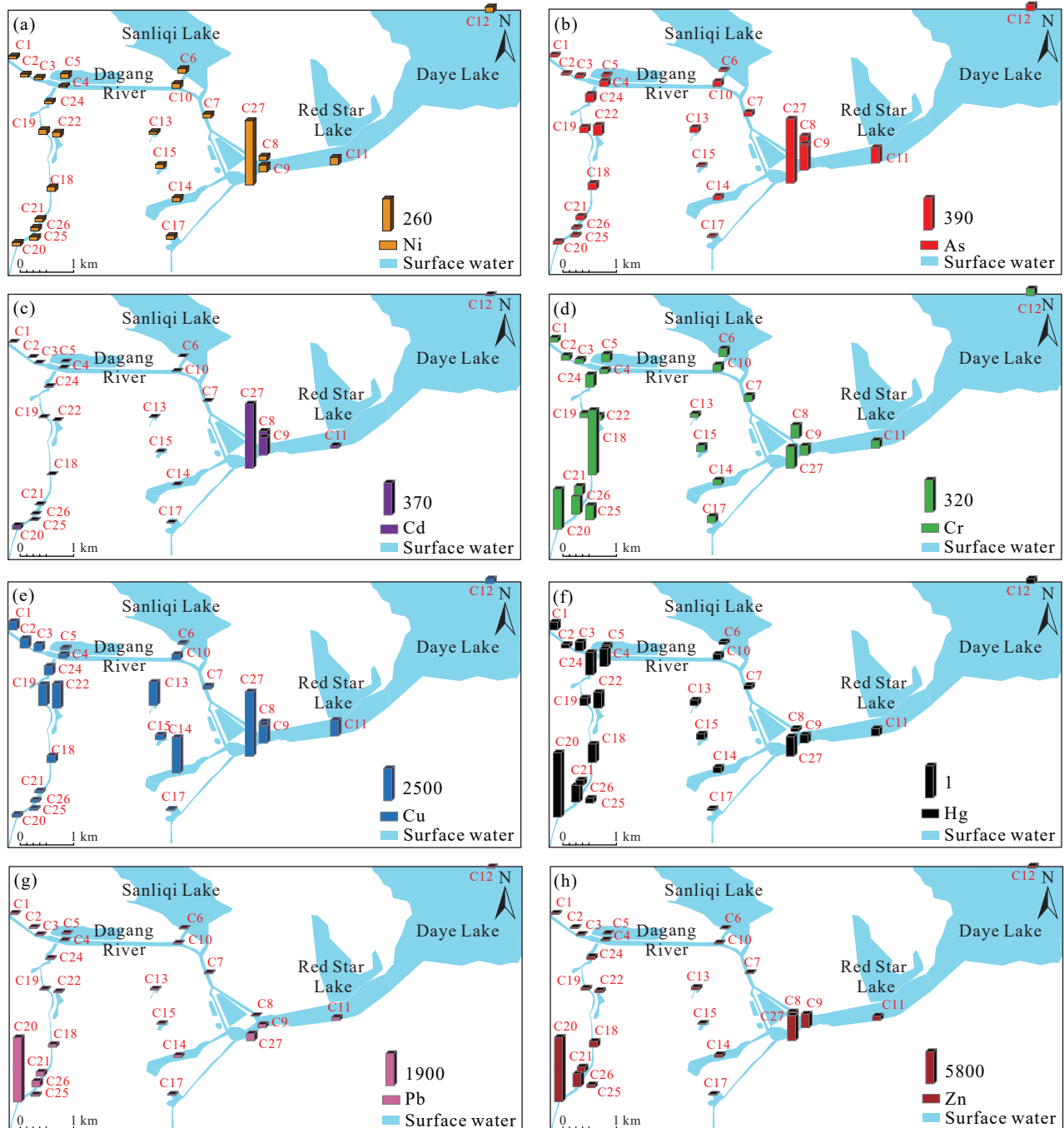


Fig. 2. Spatial distribution map of heavy metal concentration in the sediment. a–Ni concentration; b–As concentration; c–Cd concentration; d–Cr concentration; e–Cu concentration; f–Hg concentration; g–Pb concentration; h–Zn concentration.

heavy metal concentration of sampling sites downstream of the estuary fluctuates to some extent. C4 and C11 are sampling sites downstream estuaries of the southwest branch Xiaozhahe and southeast branch. The content of Hg and As at sampling site C4 increase more obviously than that of the upstream sampling site, and the content of Hg is obviously higher than that of other sampling sites of Daye Dagang. As the southeast branch of Daye Dagang influences sampling site C11, the content of Ni, As, Cd, Cu, Zn, and Pb is higher than that of other sampling sites of Daye Dagang.

The heavy metal concentration of the southeast branch of Daye Dagang takes on a variation tendency from low to high from south to north, and the maximum heavy metal concentration appears at sampling site C27.

Heavy metal distribution of each sampling site at the southwest branch Xiaozhahe of Daye Dagang differs obviously. The content of Cd, Hg, Pb, and Zn at C20, the content of Cr at C18, and the content of Ni, As, and Cu at C22 are obviously higher than that of other sampling sites at

Xiaozhahe.

The content of Cd, As, Zn, Pb, Ni, Cr, and Hg in the sediment of the west and southwest pond of Tonglūshan tailings pond is low, while the content of Cu is high.

Fig. 3 shows a contrastive analysis between the heavy metal concentration of each sampling site in the sediment and the TEL, PEL threshold value of the mass basis in the sediment. For sampling sites with a heavy metal concentration lower than TEL, heavy metal concentration from high to low is Hg (58%) > Ni (16%) > Cr (12%) > Pb (4%) = Zn (4%) = Cd (4%) > Cu (0%) > As (0%), which is of the lowest potential unfavourable biotoxicity effect, and negative effects are less likely to appear. For sampling sites with a heavy metal concentration higher than TEL and lower than PEL, heavy metal concentration from high to low is Ni (72%) > Pb (64%) > Cr (60%) > Zn (52%) > Cd (48%) > Hg (44%) > As (36%) > Cu (24%), and negative effects may appear. For sampling sites with a heavy metal concentration higher than PEL, heavy metal concentration from high to low is Cu (76%) >

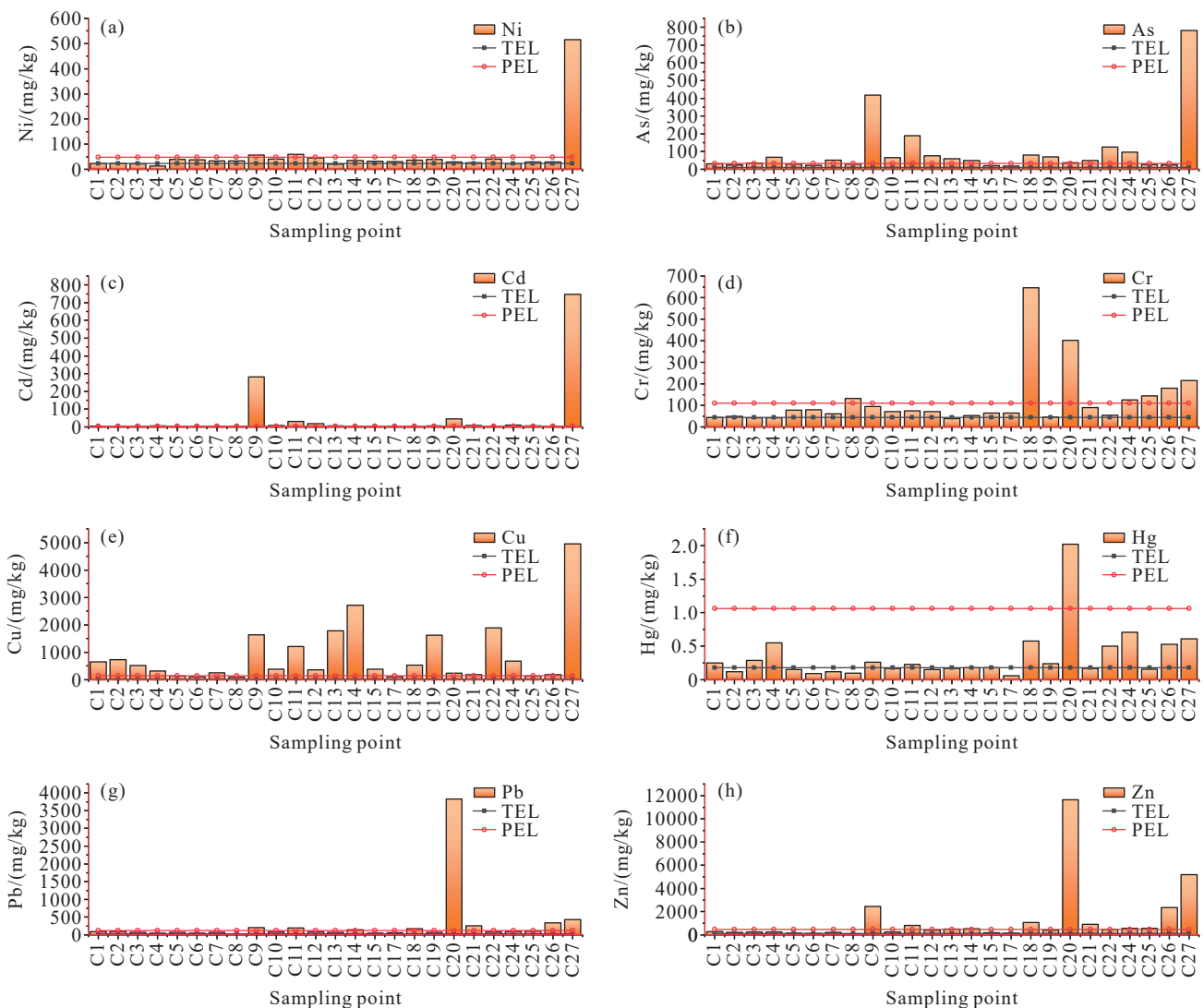


Fig. 3. Compares each sampling site's heavy metal sediment concentration and TEL, PEL. a–Ni vs. TEL and PEL; b–As vs. TEL and PEL; c–Cd vs. TEL and PEL; d–Cr vs. TEL and PEL; e–Cu vs. TEL and PEL; f–Hg vs. TEL and PEL; g–Pb vs. TEL and PEL; h–Zn vs. TEL and PEL.

As (64%) > Cd (48%) > Zn (44%) > Pb (32%) > Cr (28%) > Ni (12%) > Hg (4%), and negative effects are liable to appear.

4.2. Evaluation of heavy metal contamination and ecological risk of the sediment

4.2.1. Evaluation of heavy metal contamination of sediments

It can be seen from Fig. 4 and Fig. 5 that the range of I_{geo} value of Cr is -1.61 to 2.47 (the average value is -0.44), and the range of I_{geo} value of Ni is -1.78 to 3.51 (the average value is -0.35). The I_{geo} value of Cu range is 1.07 to 6.80 (the average value is 3.42), and the range of I_{geo} value of Pb is -0.41 to 6.39 (the average value is 1.33). The range of I_{geo} value of Zn is 1.86 to 8.62 (the average value is 4.07), and the range of I_{geo} value of Cd is 0.95 to 11.32 (the average value is 4.43). The range of I_{geo} value of As is 0.39 to 5.72 (the average value is 1.91), and the range of I_{geo} value of Hg is -0.88 to 4.25 (the average value is 1.18). Therefore, the average geo-accumulation index of heavy metal in the sediment from the largest to the smallest is: $I_{geo}(Cd) > I_{geo}(Zn) > I_{geo}(Cu) > I_{geo}(As) > I_{geo}(Pb) > I_{geo}(Hg) > I_{geo}(Ni) > I_{geo}(Cr)$, and mild outlier appears in Zn, As, and Cr, and extreme outlier appear in Cd, Pb, and Ni.

Geo-accumulation indexes of Cr, Ni, Pb, and Hg of all sampling sites are all above the clean level, 4% of the sampling sites (C20) have Pb contamination extremely, and none of the sampling sites has extremely Cr, Ni, and Hg

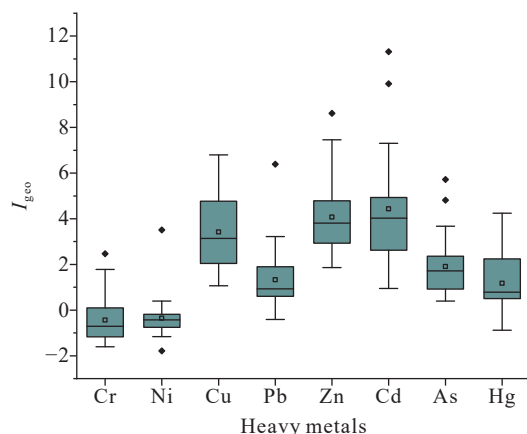


Fig. 4. Evaluation statistics of heavy metal contamination in the sediment of the Tonglūshan mining area, China.

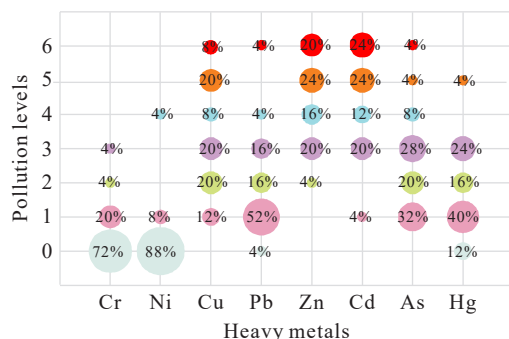


Fig. 5. Grade distribution of heavy metal contamination in the sediment of the Tonglūshan mining area, China.

contamination. Geo-accumulation indexes of Cd, Cu, and As of all sampling sites are all above the mild contamination level. 24% of the sampling sites (C9, C11, C12, C20, C24, and C27) have extremely Cd contamination. 8% of the sampling sites (C14 and C27) have extremely Cu contamination. And 4% of the sampling sites (C27) have extremely As contamination. Geo-accumulation indexes of Zn of all sampling sites are all above the level of partial moderate contamination, and 20% of the sampling sites (C18, C20, C26, and C27) have extremely contamination.

4.2.2. Ecological risk evaluation of heavy metal in the sediment

Table 3 shows the SQG-Q of heavy metal at each sampling site of the sediment, as well as the relative contribution. The range for ecological risk SQG-Q of heavy metals in the sediment at each sampling site is 0.46 – 29.41 . Among these nine sampling sites, C3–C8, C15, C17, and C25, have potential biotoxicity effects of moderate level. SQG-Q values of other sampling sites are all larger than 1.0, which have extremely high potential unfavourable biotoxicity effects. The maximum SQG-Q value appears at C27, followed by that of sampling sites C9 and C20 (Fig. 6). It is indicated that sediment of the research area has an extremely high biological risk, and 64% of the sampling sites take on extremely high potential biotoxicity effects, which is mainly located in Xiaozhahe, the southeast branch of Daye Dagang, as well as the afflux reach.

The unit contribution rate for the average toxicity of each heavy metal from large to low is $Cd > Cu > As > Zn > Pb > Cr > Ni > Hg$. As can be seen, the unit contribution rate for the average toxicity of Cd in the sediment is relatively high, which is 37.52%, and Cu is ranked second.

4.3. Analysis of the source of heavy metal in the sediment

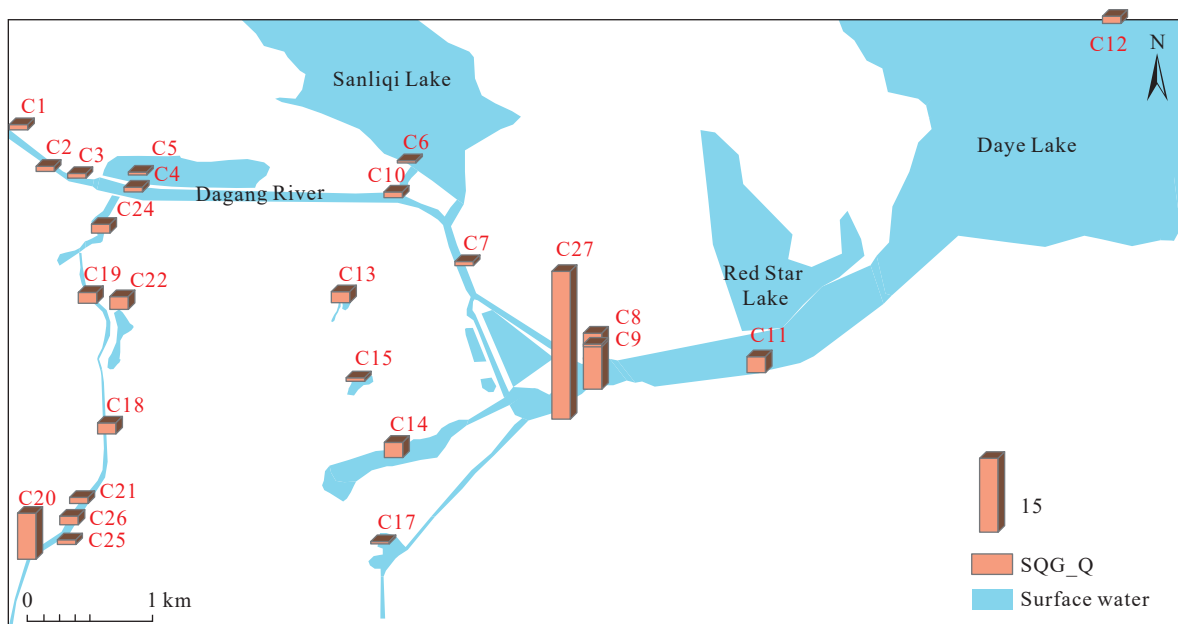
Fig. 7 is an R-type clustering analysis (inter-group connection, Pearson correlation) of heavy metals in the sediment. Heavy metals in the sediment could be generally divided into two types: The first type is Ni, Cd, As, and Cu, and the second type is Pb, Zn, Hg, and Cr.

Table 4 shows the principal component analysis of heavy metal in the sediment. Fig. 8 shows the loading diagram of the main components of heavy metal in the sediment. The Kaiser Meyer Olkin value is 0.659 (>0.5), Bartlett test P is 0.000 (<0.05), and the result of the principal component analysis is reliable (Field AP, 2009), which is suitable for source recognition. For the two principal components with an accumulative contribution rate reached 85.866% in the principal component analysis, the variance contribution rate of the first principal component is 50.642%, and Cd, As, Ni, and Cu are of large factor loading. It can be seen from the correlation coefficient matrix (Fig. 9) of each index that four heavy metals ($r > 0.781$, $p < 0.01$) are significantly positively correlated, and the content of which all exceed the background value of the Yangtze River basin, which is same to the first type of R-type clustering analysis.

The variance contribution rate of the second principal component is 35.224%, and Hg, Pb, Zn, and Cr are of high

Table 3. SQG-Q of heavy metal at sampling sites in the sediment and the relative contribution (%).

Sampling site	(PEL-Q) _{Cr}	(PEL-Q) _{Ni}	(PEL-Q) _{Cu}	(PEL-Q) _{Pb}	(PEL-Q) _{Zn}	(PEL-Q) _{Cd}	(PEL-Q) _{As}	(PEL-Q) _{Hg}	SQG-Q
C1	0.39	0.48	4.37	0.70	0.59	0.65	0.94	0.24	1.04
C2	0.44	0.47	4.92	0.69	0.49	0.31	0.73	0.11	1.02
C3	0.38	0.45	3.53	0.56	0.53	0.62	1.05	0.28	0.92
C4	0.38	0.27	2.14	0.39	0.58	1.21	2.08	0.52	0.95
C5	0.70	0.80	0.94	0.59	0.35	0.26	0.85	0.14	0.58
C6	0.72	0.78	0.74	0.43	0.28	0.11	0.69	0.08	0.48
C7	0.55	0.69	1.73	0.58	0.48	0.86	1.57	0.11	0.82
C8	1.19	0.70	0.63	0.27	0.23	0.31	0.85	0.10	0.53
C9	0.86	1.16	10.95	1.57	5.34	56.47	12.65	0.25	11.16
C10	0.64	0.83	2.63	0.63	0.54	1.79	2.01	0.16	1.16
C11	0.68	1.22	8.15	1.49	1.79	6.17	5.75	0.21	3.18
C12	0.64	0.91	2.45	0.68	0.88	3.58	2.31	0.14	1.45
C13	0.34	0.42	11.96	0.54	1.09	1.06	1.83	0.16	2.18
C14	0.47	0.72	18.27	1.11	1.18	0.96	1.47	0.17	3.04
C15	0.58	0.66	2.56	0.41	0.39	0.36	0.62	0.17	0.72
C17	0.58	0.63	0.69	0.47	0.34	0.34	0.59	0.05	0.46
C18	5.81	0.74	3.59	1.33	2.32	0.73	2.43	0.55	2.19
C19	0.42	0.81	10.89	0.54	0.90	1.26	2.13	0.23	2.15
C20	3.62	0.61	1.60	29.93	25.36	9.25	1.09	1.91	9.17
C21	0.81	0.55	1.22	1.97	2.02	1.61	1.48	0.16	1.23
C22	0.49	0.82	12.70	0.66	1.00	0.29	3.78	0.47	2.53
C24	1.39	0.46	4.58	0.72	1.23	2.06	2.94	0.67	1.75
C25	1.61	0.61	0.92	0.82	1.22	1.31	0.81	0.15	0.93
C26	2.00	0.60	1.23	2.68	5.11	0.76	0.73	0.50	1.70
C27	2.40	10.60	33.22	3.34	11.34	150.05	23.71	0.58	29.41
Relative contribution	4.35	4.18	22.70	8.22	10.15	37.52	11.63	1.26	

**Fig. 6.** Spatial distribution map of the biological risk index of the sediment.

factor loading. It can be seen from the correlation coefficient matrix (Fig. 9) of each index that three heavy metals, Pb, Zn, and Hg ($r > 0.894$, $p < 0.01$), are significantly positively correlated, and Cr, Zn, and Hg ($r > 0.524$, $p < 0.01$) are significantly positively correlated, and Cr and Pb ($r > 0.894$, $p < 0.05$) are significantly positively correlated. Their content exceeds the background value, the same as the first type of R-

type clustering analysis.

Q-type clustering analysis reflects spatial distribution characteristics of the content of heavy metal pollutants in the sediment, which could be used for evaluating the similarity of each heavy metal pollution of each sampling site. Fig. 10 shows a Q-type clustering analysis of the sampling site of the sediment. Sampling sites of the sediment could be divided

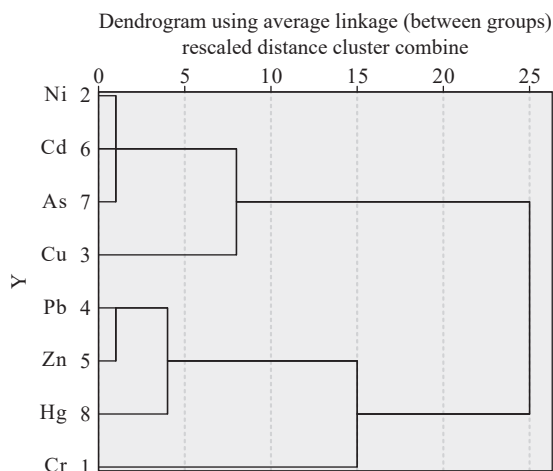


Fig. 7. R-type clustering analysis of heavy metal of the sediment.

Table 4. The main result of component analysis of heavy metals in the sediment of the Tonglūshan mining area, China.

Element	Component	
	1	2
Cd	0.969	0.147
As	0.967	0.082
Ni	0.953	0.102
Cu	0.89	-0.025
Hg	0.046	0.957
Pb	-0.056	0.956
Zn	0.269	0.936
Cr	0.054	0.687
Variance contribution rate/%	50.642	35.224
Accumulative contribution/%	50.642	85.866

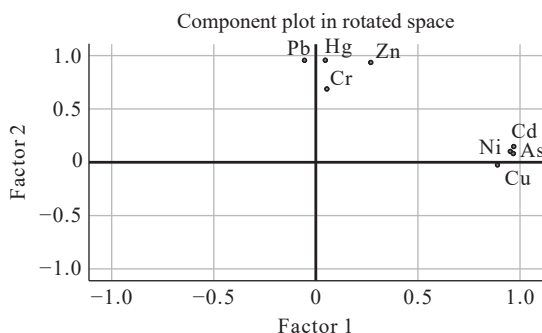


Fig. 8. Loading diagram of principal component of heavy metals in the sediment of the Tonglūshan mining area, China.

into four types. The first type includes C1 to C15, C17, C19, C21 to 22, and C24 to 26; the second type is C18; the third is C20; the fourth is C27.

5. Discussion and suggestion

5.1. Contamination characteristics of the sediment

The result of this research shows that the content of heavy metal elements Cr, Cu, Pb, Zn, Cd, As, Hg and Ni in the sediment of the research area are relatively high, which is higher than the background value and average content of heavy metal in the sediment of many typical southern rivers.

Some researches indicate that the content of heavy metal element Cr, Cu, Pb, Zn, Cd, As, and Hg in the sediment of Daye Lake basin is respectively 9.74–206.00 mg/kg, 5.48–1414.31 mg/kg, 31.25–596.88 mg/kg, 14.19–1320.20 mg/kg, 0–206.00 mg/kg, 6.15–1051.26 mg/kg and 0–2.92 mg/kg (Zhou GQ et al., 2016). It could be seen by comparison that the maximum concentration values of As and Hg in this research are lower than the above results, while concentration values of other heavy metal elements are higher than the above research result, indicating accumulation of Cr, Cu, Pb, Zn, Cd, and Ni is intensified. The results of the geo-accumulation index method and the mean sediment quality guideline-quotient show that heavy metals have extremely polluted sediment of the research area, and heavy metal Cd is the most extremely contaminated, with the strongest biotoxicity, which is consistent with other research findings (Zhou GQ et al., 2016; Fang YM et al., 2017).

From the distribution, there are obvious regional differences in the content of heavy metals in the sediment of the research area. The maximum value of each heavy metal is

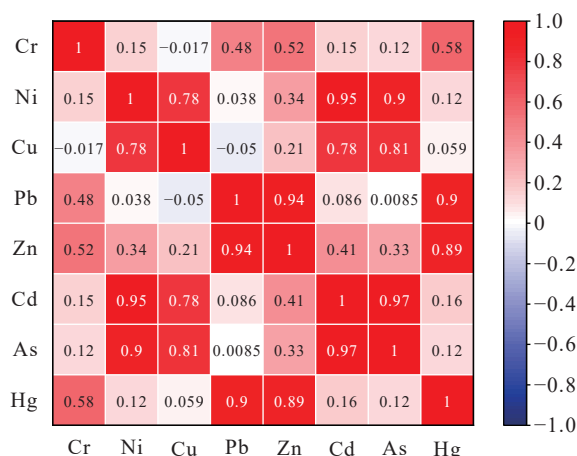


Fig. 9. Correlation analysis of heavy metals in the sediment of the Tonglūshan mining area, China.

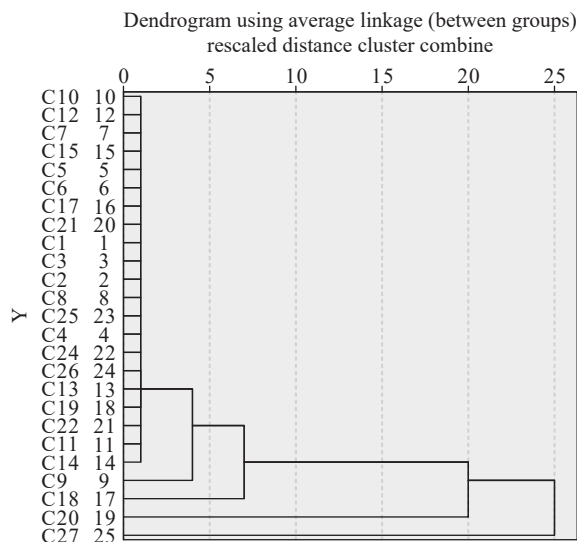


Fig. 10. Q-type clustering analysis of the sampling site in the sediment of the Tonglūshan mining area, China.

respective distributed at sampling sites C18 and C20 of the southwest branch (Xiaozhahe) of Daye Dagang and sampling site C27 of the southeast branch (Zhaobaohu draining pump station), and the content of As and Hg at sampling site C4 downstream estuary of the two branches is obviously higher than that of other sampling sites upstream main channel of Daye Dagang. The content of Hg is obviously higher than that of other sampling sites of the main channel of Daye Dagang. The content of Ni, As, Cd, Cu, Zn, and Pb at sampling site C11 is obviously higher than that of other sampling sites of the main channel of Daye Dagang. The mean sediment quality guideline quotient indicates that 64% of the sampling sites distributed at the southwest branch Xiaozhahe of Daye Dagang, the southeast branch, and the affluent reach are of extremely high biological risk. It shows that the accumulation of heavy metals in the sediment and the ecological risks of the two branches are both higher than that of the main channel of Daye Dagang, which obviously influences the accumulation of heavy metals in the sediment and the ecological risk level of the main channel of Daye Dagang. Some researches show that most fracture surface of the Daye Lake basin is slightly polluted by heavy metal. Still, some branches, such as Donggang and Sanliqihu, are extremely polluted (Zhou GQ et al., 2016). The waters and sediment of the Daye Lake basin have been polluted by heavy metal at different level, among which Sanliqihu and the east and west ports of Luoqiao is polluted mostly (Fang YM et al., 2017). It is basically consistent with the result of the research that branches are more seriously polluted than main channels, and it influences the contamination status of the main channel.

The C27 sampling site of the southeast branch (Zhaobaohu draining pump station) of Daye Dagang is located downstream scupper of a tailings pond. There are smelting plants, copper mine beneficiation companies, a tailings pond, and a fishpond along banks of the southwest branch Xiaozhahe, so the distribution of heavy metal at each sampling site differs greatly; sampling sites with the maximum value are C18, C20, and C22. C18 and C20 are both located downstream scupper of tailings ponds, and C22 is located at an enclosed fishpond on the east side of Xiaozhahe. Three sampling sites, C18, C20, and C27, at two branches, are greatly influenced by the drainage of tailings ponds and tailings washed down by rains. As can be seen, the content of heavy metals in the sediment is greatly influenced by the distance from the contamination source of mining activities. Sampling sites C27 and C20 at two branches have the highest potential biotoxicity effects of heavy metals.

To sum up, the contamination level and potential biotoxicity effects of heavy metals in the sediment of the research area are extremely high, and attention shall be particularly paid to the heavy metal element Cd. As for contamination distribution, contamination of the branched that receives each kind of industrial wastewater is getting worse and worse, and attention shall be paid to contamination discharging of tailings ponds upstream of C27 and C20.

5.2. Analysis of sediment source

In virtue of clustering analysis and principal component analysis, the first type of heavy metal and the first type of principal component in the sediment mainly include four metals Cd, As, Ni, and Cu. Correlation analysis shows these four heavy metals significantly correlate with each other, so they have a common source. Cd and Zn are significantly correlated, suggesting Cd is diverse; Cu is negatively correlated with Cr and Pb, suggesting Cu's source differs from that of Cr and Pb.

Studies show that Ni and Cu mainly exist as sulfide and sulfate minerals. Sulfide has low solubility, so the transport mode of which in waters of nature is mainly adsorbing and complexation (Li J et al., 2018). Due to sulfide in sediment, heavy metal Cd is liable to be immobilized in the sediment, thus causing enrichment of Cd. Furthermore, elementary substances Cu and As are rarely found in the natural environment, mainly existing in the form of minerals, which is also the composition of pesticides and chemical fertilizers (Xia B et al., 2014). As the research area is located in a compact district of Cu-Fe tailings pond and Cu-Au tailings pond of Tonglūshan mining area, Cd, As, and other elements usually symbioses with Cu. There is little agricultural acreage in the research area, and pesticide and chemical fertilizer is rarely used, so beneficiation wastewater and tailings pond are probably the main sources of these heavy metals.

Sampling site C22 is located on the east bank of the southwest branch Xiaozhahe of Daye Dagang, which is an enclosed fishpond. As heavy metals can be easily adsorbed on suspended particles in waters, and they can't be degraded, so heavy metals go into aquatic water in various ways, such as atmospheric bulk deposition, surface infiltration, farming activity, etc. finally, go into sediment through absorption, sedimentation, burying and other processes. The research shows that C22 has an extremely high potential for unfavourable biotoxicity effects. Heavy metal's maximum probable effect level coefficient is Cu, followed by As and Zn. Cu and Zn are essential elements for the survival and production of the farmed organism, which are widely added in fishery feed (Shi QX et al., 2015). In addition, the usage of copper sulfate and other fish medicine in cultivation will result in an increasing escalation of Cu in the cultivation environment (Liu M et al., 2019). To this end, C22 is greatly influenced by beneficiation, aquaculture feed, fish medicine, etc.

The second type of heavy metal and the second type of principal component in the sediment mainly include four metals, Hg, Pb, Zn, and Cr. Correlation analysis shows these four heavy metals significantly correlate with each other, so they have a common source.

The background value of Hg in nature is not so high, which is 0.03–0.07 mg/kg in cultivated soil and 0.030–0.034 mg/kg in clay, and the content in water is usually below the determination level (Liu RP et al., 2021). Studies have shown that Hg is mainly derived from coal burning and cement

manufacturing, and coal burning contributes 25% to Hg (Jiang X et al., 2019; Xu J et al., 2015). According to the investigation, element Hg generated during the production of smelting plants in the research area and cement companies 1 km away from the south of the research area could be transferred via atmospheric transmission under the prevailing southeast wind, causing heavy metal accumulation of downwind direction. Studies have shown that the content of Pb and Hg in the sediment of Ballinger Lake had increased by three orders of magnitudes during ten years of smelting of Tacoma copper smeltery, indicating main contamination source of Pb and Hg in the sediment of the lake is the discharged pollutant of smelting industry (Gray JE et al., 2013). Researches also show that Pb is usually regarded as a specific pollutant of transportation (Milenkovic B et al., 2015). An increase in vehicles, tire-wearing of cars, burning of engine lubricating oil, discharging of tail gas, etc., will all increase Pb and Cr in soil and sediment (Li L et al., 2012). Heavy vehicles will especially discharge a large amount of Pb when running (Jiang X et al., 2019). Research has shown that Cr is liable to be transferred and converted, reflecting the influence of the recent discharging of human beings. It mainly comes from metallurgy, electroplating, leather making, machinery manufacturing, and other industries (Davidson CM et al., 1994; Xu YD et al., 2015). Therefore, the second principal component mainly reflects the influence of smelting companies and vehicles in the research area on the distribution of heavy metals in the sediment.

In summary, heavy metals in the sediment of the research area are probably derived in two ways, Cd, As, Ni, and Cu are probably derived from beneficiation wastewater and discharged pollutants of the tailings pond of the research area, and Pb, Zn, Hg, and Cr are probably derived from discharged pollutant of smelting companies and vehicles of the research area. Besides, the heavy metal of sampling site C22 is perhaps influenced by aquaculture feed and fish medicine.

The Q-type clustering analysis of sampling sites of the sediment shows that sampling sites of the sediment could be divided into four types. And the contamination status of sampling sites of the same type is similar. The contamination intensity of different types are different: The greater difference between the two types, the greater difference in heavy metal contamination level. The second type, C18, and the third type, C20, are located at the southwest branch of Xiaozhahe of Daye Dagang, and the fourth type, C27, is located at the southeast branch of Daye Dagang. Thus, heavy metal contamination levels in the two branches are higher than that of the main channel of Daye Dagang. It is consistent with the distribution characteristics of the heavy metal distribution of this research.

Furthermore, sampling sites of the second, third, and fourth types are all located downstream drain outlets of tailings ponds. Upstream tailings ponds greatly influence heavy metal content in the sediment of such three sampling sites. Some researchers once proposed that deposition of Cd in mine wastewater was mainly controlled by hydrologic

conditions and distance away from the drain outlet (He Y et al., 2011). It can be seen that deposition of each kind of heavy metal in the sediment of the research area is mainly controlled by the distance away from the drain outlet of tailings ponds.

5.3. Suggestion

The following controlling measures are proposed to reduce heavy metal contamination levels in the sediment of the Tonglūshan mining area.

The first is to strengthen source control to meet discharging standards. Strengthen capacity building for online monitoring of drain outlets of companies and tailings ponds, strengthen supervision to completely eradicate illegal discharging, and reduce discharging of pollutants from the source.

The second is to strengthen the monitoring of heavy metal pollution in the sediment by using advanced techniques, proving the theoretical basis for further contamination abatement.

The third is to carry out river dredging work per the monitoring data and dispose of sediments safely to reduce secondary contamination caused by improper disposition of sediment.

The fourth is to strengthen the construction of sewage treatment works and make every effort to treat as many pollutants should be treated as possible.

6. Conclusion

Sediment in the research area has been extremely polluted by heavy metals, with extremely high biological risk, especially since the heavy metal pollution level in the sediment of the southwest branch (Xiaozhahe) and southeast branch of Daye Dagang is both obviously higher than that of the main channel of Daye Dagang. Moreover, heavy metal accumulation is obviously higher than the existing Daye Lake basin research results. The source of heavy metal in the sediment is closely related to discharging of smelting plants, copper beneficiation companies, tailings ponds, and vehicles within the research area. The research results figure out key contamination sources, zones, and elements for the governance of heavy metals in the sediment of the research area.

Presently, a systematic monitoring network is lacking in the survey of heavy metals in the sediment of the research area, posing a challenge to precious early warning and late forecast.

This research suggests local governments strengthen capacity construction on the management of contamination discharging when developing the economy, paying more attention to the influence of heavy metals on the aquatic ecological environment, and strengthening source control and public awareness.

CRedit authorship contribution statement

Jing Wang, Ai-Fang Chen, Bo Wang, Qi-Bin Zhao, and

Guan-Nan Liu conceived the presented idea. Xin-Xin Zhang, Xiao Xiao, and Jin-Nan Cao collected samples and analyzed the data. Jing Wang wrote the manuscript with input from all authors.

Declaration of competing interest

The authors declare no conflicts of interest.

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