Journal of Hydrosciences and Environment
Available online at http://jhe.usb.ac.ir/

ISSN: 2345-5608

Relationship Between Morphometric Stream Characteristics and Sediment Heavy Metal Pollution in a Mining-Influenced Watershed

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ARTICLE INFO

Article history:

Received: 12 Jan 2020
Accepted: 29 Mar 2020

Keywords:
Fluvial Sediment,
Stream Slope,
Stream Length,
Zn -Pb mine.

ABSTRACT

Streams located in watershed environments act as a principal component of transportation and reservoir of heavy metals evacuated by mining activities. This study evaluated the relationship between morphometric stream characteristics and variations in the rate of sediment heavy metal pollution in a mining-influenced watershed. The study area was a mining site located in the Anguran region, Zanjan province, Iran. Three rivers were selected that exhibited significant differences in terms of path length and slope. Approximately 1500 g sediment was collected from downstream positions of Kaka River (R1), Allah Lochay River (R2), and Shurab River (R3). Total zinc (Zn), nickel (Ni), lead (Pb), and cadmium (Cd) concentrations and particle size distribution were measured. The pollution load index (PLI) and geo-Accumulation Index (Igeo) were then calculated. Based on the results, the sediment particle size distribution significantly decreased in response to an increase in the river length and slope (R1>R2>R3). Stream path length and slope are factors determining the size of bed particles transported by a river. The streams with longer path lengths and steeper slopes could transport finer fractions toward depositional positions. Further, the pollution level of the Kaka River sediment was the highest compared with other rivers. The highest Igeo value for the measured heavy metals was also observed in the Kaka River sediment, followed by the sediments of Allah Luchay and Shurab rivers. The regression results illustrated that the heavy metal concentrations were significantly positively correlated with clay and silt contents. These findings unequivocally showed that watershed environments exhibit a high vulnerability to mining activities because the streams can facilitate the transportation of sediment polluted by heavy metals across the landscape. On the other hand, the streams with higher lengths and slopes surrounding mining sites increase the pollution transfer rate and cause a higher level of sediment heavy metal pollution.

1. Introduction

Heavy metals are natural components contained in the earth's crust (Sun et al., 2018). Some of these harmful elements are necessary for the normal metabolic activities of humans, animals, and plants. The development of urbanization, industrialization, and human activities (e.g., wastewater irrigation and fertilizers, industrial production, traffic, and municipal compost) considerably stimulated soil heavy metal pollution (Lin et al., 2018; He et al., 2017; Fei et al., 2017). Mining activities are one of the most common land uses that can be an important source of soil and sediment heavy metal pollution (Pu et al., 2019; Marrugo-Negrete et al., 2017). Mining operations threaten environment health through different pathways such as flux of dust contained in heavy metals into the atmosphere and production of a great amount of acidic drainage containing heavy metals (Sun et al., 2018). Mining is recognized as the most critical anthropogenic activity that significantly affects the quality of soil, sediment, and water resources. Although, mining activities cause the economic development of a region, the lack of environmental management can result in a negative effect of this land use on the quality of sediment, soil, and water. The drainage networks remain a principal reservoir of heavy metals evacuated by mining activities. Further, they are recognized as the second source of pollution, depending on the surrounding physicochemical conditions (Omwene et al., 2018; Wu et al., 2019). These elements are easily dissolved and transported by water and thereby entered within the ecosystem and food chains. Sediments are a preferential indicator for assessing the level of water pollution and environment health. Nowadays, stream sediment evaluation contains ecological and geochemical risks created by the distribution of harmful elements. Streams provide a condition for the transportation of sediments polluted by heavy metals excavated by mining activities toward the downslope areas. This condition can be

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DOR: 20.1001.1.23455608.2020.4.7.5.4

an important agent in the spatial distribution of these harmful elements. On the watershed scale, the downslope positions act as a reservoir for the accumulation of heavy metals. Although, mining activity occurs within a small spatial scale, the presence of drainage networks surrounding mining sites, especially the mines located within watersheds, can be a principal factor in the distributing of harmful elements across broader spatial scales. Topographic characteristics of drainage networks, such as their slope and length, significantly affect the pollution level of different stream parts. Stream path length and slope can be a factor determining the size distribution of sediment particles. Streams with longer path lengths transport finer fractions toward depositional positions (Pyrce and Ashmore, 2003). On the other hand, the accumulation of heavy metals has a significant relationship with particle size distribution. As a result, different slope positions that a river crosses and variations in the stream path length, can exhibit significant differences in terms of the sediment pollution levels by heavy metals. The assessment of stream sediment quality surrounding mining sites is an important step toward terrestrial and aquatic ecosystem management.

Over the past decade, many studies investigated the role of mining activity in soil and water pollution (Lu et al., 2019; Mao et al., 2019; Siddiqui and Pandey, 2019; Marrugo-Negrete et al., 2017; Duncan et al., 2018; Fei et al., 2017; Pu et al., 2019). These studies comprehensively evaluated changes in physicochemical properties of soil and water resources surrounding mining sites and their impacts on the environment. Gyamfi et al. (2019) studied the quality of water and soil in a typical artisanal mining community. They found that the stream had a high concentration of Fe and Mn above acceptable the World Health Organization, and therefore, the soil was significantly polluted by those metals. Lu et al. (2019) evaluated the soil and water pollution caused by mining activity around an abandoned sphalerite mine. They found that harmful elements in the mining area display apparent non-carcinogenic health risks to children while the risks to adults are not equally obvious. Mao et al. (2019) provide a comprehensive analysis of dissolved heavy metal pollution characteristics and factors controlling pollution levels during the three different water flow regimes of the Lhasa River basin, Tibet. Siddiqui and Pandey (2019) investigated eight heavy metals, including Cr, Cd, Cu, Ni, Pb, Zn, Mn, and Fe, in water and bed sediment of the Ganga River. Their findings provide a detailed watershed-scale database on heavy metal concentration in water and bed sediment, the magnitude of contamination, and ecological risks to aquatic organisms in the Ganga River, India. Pu et al. (2019) illustrated the effects of copper mining on heavy metal contamination in a rice agrosystem in the Xiaojiang River Basin, China. Duncan et al. (2018) exhibited the heavy metal pollution levels in the sediment belonging to the Pra Basin of Ghana from 27 sampling points during the dry and wet seasons using the geo-accumulation index, enrichment factor, and pollution load index. Sun et al. (2018) evaluated how pollution could decrease with increasing distance from the mining district. They found that heavy metal pollution of farmland soils displays a high potential ecological risk. Marrugo-Negrete et al. (2017) studied the concentration of heavy metals in 83 agricultural soils irrigated by the Sinú River, Colombia, influenced by mining areas upstream. Fei et al. (2017) investigated the temporal and spatial distribution of antimony and related heavy metal contaminants (lead, zinc, and arsenic) and the exposure risks for the population of the Yuxi river basin, China.

There are numerous metal mines in various parts of Iran. The Anguran Zn -Pb mine is one of the most important mining sites in Zanjan province. Despite extensive research on the pollution and ecological risk, and environmental problems generated by mining operations (as explained above), the impact of topographic stream characteristics on the level of stream sediment heavy metal pollution influenced by mining activities are poorly covered. Therefore, in this study, to better understand the effect of variation in morphometric stream characteristics on heavy metal pollution levels of sediments transported by rivers, we investigated changes in sediment pollution levels along streams with different morphometric characteristics, including slope and length. The overall objective of this research was to evaluate the relationship between topographic stream characteristics, including path length and slope, and the levels of zinc (Zn), nickel (Ni), lead (Pb), and cadmium (Cd) pollution of sediment transported by rivers.

2. Materials and methods

2.1. Study area and field Work

The studied region is located surrounding a mining site located in the Anguran, Mahneshan County, Zanjan Province, northwestern Iran (Figure 1). The region's climate is cold and semi-arid with a mean annual temperature between 9 and 17 °C and approximately 400 mm mean annual precipitation. The average elevation of the Anguran is approximately 3,000 m.a.s.l. According to the aim of the present study, we firstly selected three rivers surrounding the Anguran Zn -Pb mine, including Shurab, Kaka, and Allah Luchay rivers. These rivers exhibited significant differences in terms of path length and slope. The Shurab River is a seasonal river with a length of 18 km and a slope of 6.5%. The Kaka River is a perennial stream with a length of 30 km and a slope of 7.3%. The Allah Luchay River is a seasonal river with a length of 20 km and a slope of 5.5%.

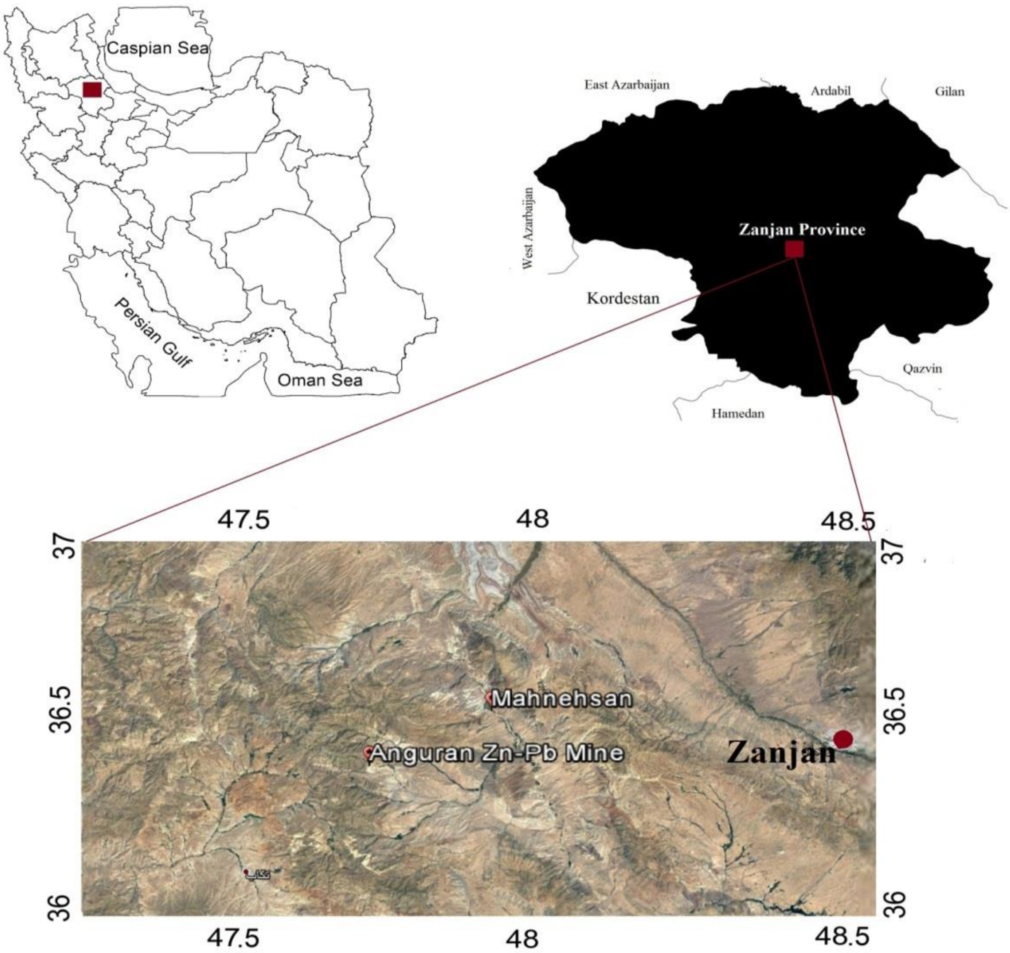

Fig. 1. Location of the mine in the Anguran-Zanjan

2.2. Data Collection and Statistical analysis

To evaluate the impacts of the topographic river characteristics on sediment heavy metal pollution, approximately 1500 g bed sediment was collected from downstream positions of Kaka River (R1), and Allah Luchay River (R2), and Shurab River (R3). The samples were air-dried at 25°C and sieved through a 2 mm steel sieve. After sieving, the sediments were equally divided into three portions. The weight of each portion was considered 500 ± 1 g, (three replicates per sediment). Afterward, the prepared samples were acid digested by 65% HNO3, and Total Zn, Ni, Pb, and Cd concentrations were then analyzed by atomic absorption spectrophotometer (Sposito et al., 1982). The particle size fractions of each sample were also determined by the hydrometer method (Gee and Bauder, 1986).

To assess the pollution status of sediment and the level of heavy metal contamination in the different sampling sites, we calculated the pollution load index (PLI, equation 1) (Soares et al. 1999) and the geo-accumulation index (Igeo, equation 2) (Muller, 1981), respectively.

$$\text{PLI}=\left(\frac{C_{n1}}{B_{n1}} \times \frac{C_{n2}}{B_{n2}} \times \ldots \frac{C_{n_i}}{B_{n_i}}\right)\frac{1}{1} \quad (1)$$

where $\frac{C_{n1}}{B_{n1}}$ is the ratio of measured metals to their background values and i is the number of metals. This index compares the heavy metal pollution status between the different sampling sites. PLI value = 0 shows perfection, PLI < 1 shows no pollution, and PLI > 1 indicates pollution (Sojka et al., 2019; Awosusi and Adisa, 2020).

$$\text{Igeo}= log_{2\,(\frac{Cn}{1.5B_n})} \quad (2)$$

where Cn is the measured heavy metal concentration in the sediment. B_n is the geochemical background concentration of the metal (n). 1.5 is the background matrix correction factor due to the lithogenic effects. This factor evaluates the level of heavy metal contamination in the sediment sample. Values of this index are classified into 7 classes (Muller, 1981), including < 0-uncontaminated, 0-1-uncontaminated to moderately contaminated, 1-2-moderately contaminated, 2-3-moderately to heavily contaminated, 3-4-strongly contaminated, 4-5-heavily to extremely contaminated, and > 5-extremely contaminated.

We first performed an analysis of variance and a Tukey *post hoc* test to compare the level of heavy metal pollution of sediments collected from downstream positions between the rivers. We examined whether the sediments belonging to the streams with different topographic characteristics (path length and slope) exhibit significant differences in the rate of heavy metal pollution. Therefore, a regression analysis was employed to examine the relationship between changes in the topographic stream characteristics, particle size distribution, and sediment heavy metal pollution rate.

3. Results

Analysis of variance (ANOVA) for the heavy metal concentrations and particle size distribution at the different river types is presented in Table 1. These results showed that the river types, including different slopes and lengths had a significant effect on the Zn, Ni, Pb, and Cd concentrations and distribution of clay, silt, and sand in the river sediments.

The statistical results (Tukey *post hoc* test) of the physical and chemical sediment properties illustrated the appearance of different rates ($P < 0.05$) of sediment heavy metal pollution and particle size distribution within downstream positions of three rivers (Table 2 & Figure 2). The highest heavy metal concentrations were seen in bed sediments belonging to the Kaka River (R1). The Allah Luchay (R2) and Shurab (R3) rivers illustrated the lowest heavy metals concentrations compared with the Kaka River.

As shown in Table 3, the sediments belonging to all the rivers showed heavy metal pollution and the pollution level of the Kaka River sediment (R1) was the highest (PLI > 1) compared with other rivers. According to the geo-accumulation index (Igeo), the sediments of all the rivers were strongly polluted with Pb and Cd, and moderately polluted with Zn. However, there was no Ni pollution in the sediments. Additionally, the highest Igeo value for the measured heavy metals was observed in the Kaka River sediment (R1), followed by the sediments of Allah Luchay (R2) and Shurab (R3) Rivers.

Table 1. Coefficient of variation and analysis of variance, showing the effect of the river types on the sediment heavy metal concentrations and particle size distribution

Source of variation	df	Mean square						
		Zn	Ni	Pb	Cd	Clay	Silt	Sand
River type	2	24166^{**}	220^{*}	6185^{**}	1.73^{**}	38.78^{**}	10.33^{*}	91.01^{*}
Coefficient of variation (CV)		2.73	11.67	6.63	7.51	8.74	18.43	13.25

Note: * and ** p-values were significant at 0.01 and 0.05 levels, respectively

Table 2. Comparison of heavy metals concentrations between the different streams

River sediment	Zn	Ni	Pb	Cd
R1	385.92 ± 12.45^{a}	24.5 ± 3.11^{a}	185.98 ± 9.54^{a}	2.78 ± 0.92^{a}
R2	235.92 ± 7.28^{b}	10.54 ± 2.46^{b}	107.31 ± 6.81^{b}	1.47 ± 0.71^{b}
R3	220.33 ± 9.31^{c}	9.31 ± 1.98^{b}	102.04 ± 7.55^{b}	1.39 ± 0.52^{b}

Note: All values are defined based on mean ± standard deviation. Different lowercase letters show statistically significant differences (*P* < 0.05) between different streams; R1, Kaka River; R2, Allah Luchay River; R3, Shurab River.

Table 3. Results of the pollution load index and geo-accumulation index

River sediment	Pollution load index (PLI)	Geo-accumulation index (I_{geo})			
		Zn	Ni	Pb	Cd
R1	6.94	1.87	-0.3	4.63	4.21
R2	3.69	1.16	-1.5	3.83	3.29
R3	3.43	1.06	-1.6	3.76	3.21

The particle size distribution, including fractions of clay, silt, and sand, exhibited a statistically significant difference ($P < 0.05$) between the sediments belonging to the different rivers (Figure 2). The greater value of fine fractions, including clay and silt, was observed in the downstream sediments of the Kaka River (R1) compared with other rivers. Whereas, the sediments belonging to the Allah Luchay (R2) and Shurab (R3) rivers showed the highest value of coarse fractions compared with the Kaka River (R1).

Further, the results of the regression analysis explain how variation in the particle size distribution of sediments transported by the streams with different path lengths and slopes could encourage significant differences in terms of the heavy metals concentrations excavated by mining activity (Figure 3). The results revealed a significant and positive correlation ($P < 0.05$) between the measured elements, including Zn (R^2=0.85; R^2=0.74), Ni (R^2=0.75; R^2=0.93), Pb (R^2=0.86; R^2=0.71), and Cd (R^2=0.82; R^2=0.78) and fine earth fraction of clay and silt in the studied stream sediments. In contrast, coarse fraction, including sand, gave a negative and non-significant correlation with the metals concentrations in bed sediments.

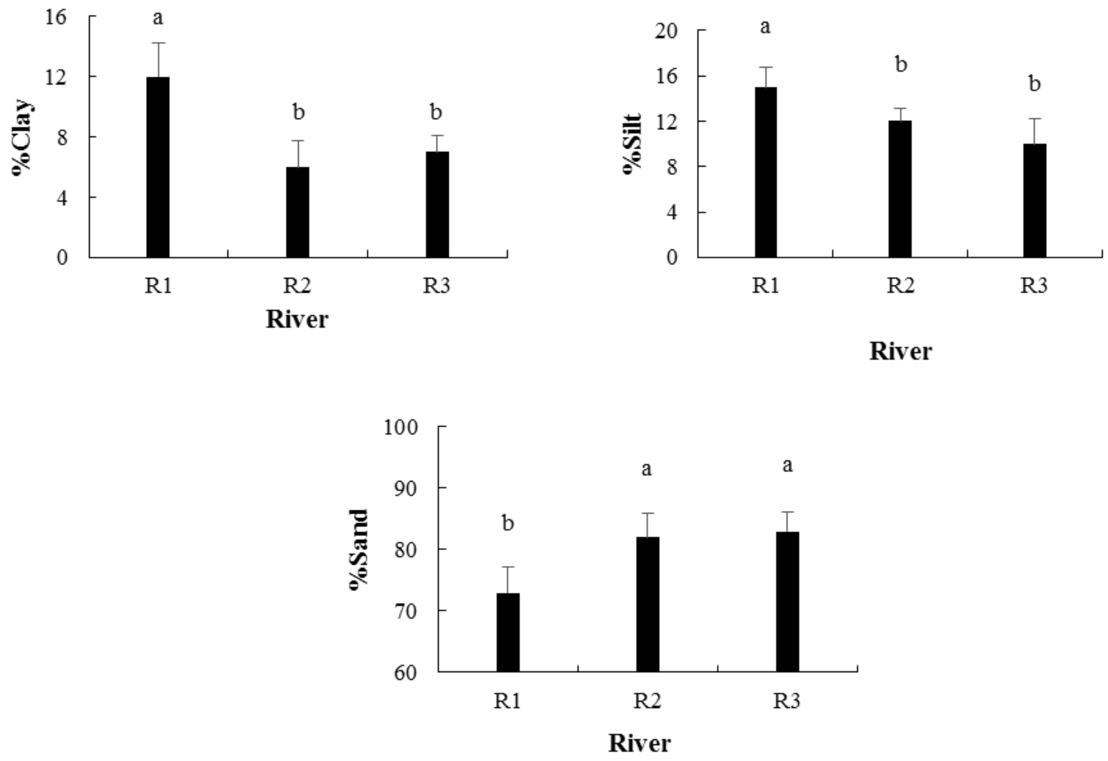

Fig. 2. Comparison of the particle size distribution between the different bed sediments. Lowercase letters show statistically significant differences ($P < 0.05$) between different streams; R1, Kaka River; R2, Allah Luchay River; R3, Shurab River

4. Discussion

The findings of this research showed that how variation in topographic stream characteristics such as stream path length and slope could significantly affect the pollution level of the sediments transported by the rivers. Stream path length can be a factor determining the particle size of sediment transported by a river. This characteristic defines as the total distance of the transportation path of a particle from initial entrainment to final sedimentation. Previous studies proved that larger bed particles mobilize shorter path lengths (Pyrce and Ashmore, 2003). This means that streams that have longer path length can transport finer fractions toward depositional positions. On the other hand, the accumulation of heavy metals has a significant relationship with the particle size distribution of sediment (He et al., 2016; Kalantzi et al., 2013). Our findings showed that variation in the length of a stream could cause significant differences in the rate of sediment pollution by the heavy metals. The higher value of the heavy metal concentrations and the pollution load index in the Kaka River was likely due to the longer length of this stream compared with other studied rivers. An increase in the stream path length and slope enhances the river's ability to transport finer fractions. The Allah Luchay and Shurab Rivers with a shorter path length exhibited a higher amount of coarse fractions compared with the Kaka River, while the Kaka River showed a higher accumulation of fine fractions.

Some studies proved that particle size distribution plays a crucial role in the control of heavy metal concentrations within the stream sediments (Roussiez et al., 2005; Zhang et al., 2002). Sediment constituents affect the behavior of the metals in the rivers. The metal concentrations within the streams are likely controlled by the amount of oxides and clays contained in the sediments. The presence of these constituents is known to form highly stable complexes with heavy metals (Zhao et al., 2014). The sediments with a higher amount of finer fractions, such as clay and silt, effectively absorb heavy metals because the pollutants can bond with fine fractions to contribute toward the formation of aggregation (Sarkar et al., 2004; Brooks and Mahnken, 2003). The findings of the regression analysis confirm that sediments with particle size lower than < 0.063 mm have a high potential of the heavy metals concentrations, as other

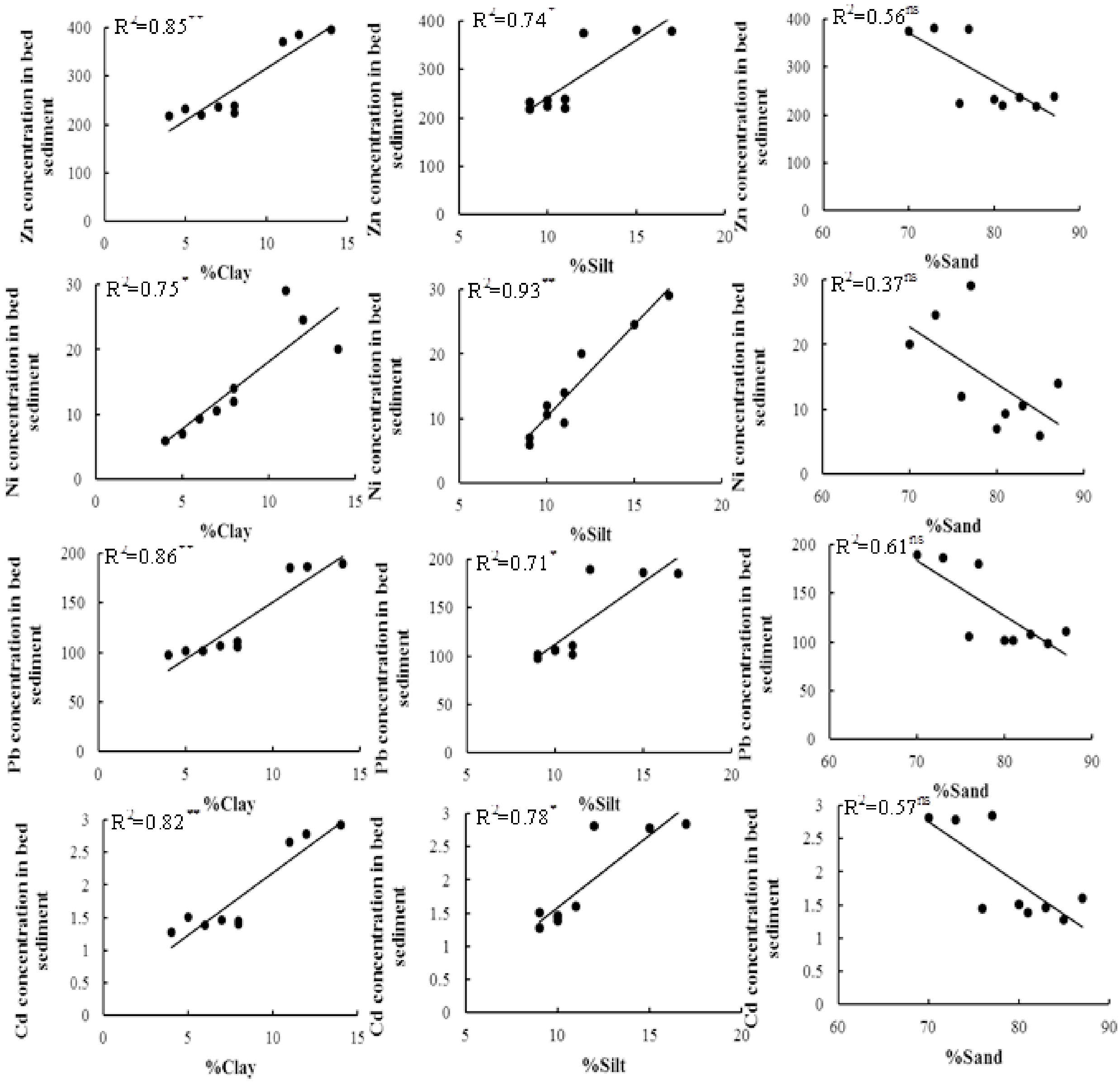

Fig. 3. Relationship between the particle size distribution (%Clay, %Silt, and %Sand) and sediment heavy metals concentrations. The stars show $P < 0.05$ (*) and $P < 0.01$ (**).

studies noted (He et al., 2016; Zhang et al., 2013). This positive relationship is likely because the fine fractions have a large specific surface area and high content of organic matter and clay fraction (Kalantzi et al., 2013).

Mao et al. (2019) provide a comprehensive analysis of dissolved heavy metal pollution characteristics and factors controlling pollution levels during the three different water flow regimes of the Lhasa River basin, Tibet. They found that the river water showed alkaline characteristics, which may affect heavy metal elements in a dissolved fraction. Also, their results illustrated that Mn, Cd, Cu, and Zn originated from the natural geological background, whereas Pb originated from mining drainage. Siddiqui and Pandey (2019) investigated eight heavy metals, including Cr, Cd, Cu, Ni, Pb, Zn, Mn, and Fe, in bed sediment within the Ganga River. Their findings provided a detailed watershed-scale database on heavy metal concentration in bed sediment, the magnitude of contamination, and ecological risks to aquatic organisms in the Ganga River, India. They proved that in the middle and lower reaches, Cr, Cd, Pb, Ni, Cu, and Fe in the water had exceeded the regulatory limit. Although headword river stretch seemed very less contaminated, the middle and lower reaches showed moderately to the heavily contaminated range. Marrugo-Negrete et al. (2017) studied the concentration of heavy metals in agricultural soils irrigated by the Sinú River, Colombia, influenced by mining areas upstream. Their findings suggested that soil contamination was mainly derived from agricultural practices, which was caused probably by atmospheric and river flow transport from upstream gold mining. Fei et al. (2017) investigated the temporal and spatial distribution of antimony and related heavy metal contaminants (lead, zinc, and arsenic), and the exposure risks for the population of the Yuxi river basin, China. The results of this investigation showed that the antimony concentrations in the river sediments in the midstream and downstream regions significantly exceeded the regulatory limit. Pu et al. (2019) pointed out the effects of copper mining on heavy metal contamination in a rice agrosystem in the Xiaojiang River Basin, China. Their findings showed that copper mining had a crucial effect on heavy metals in a rice agrosystem near the copper mining area. Long-term wastewater irrigation, especially the deposition of sediments, lowered the soil pH and induced the accumulation of heavy metals in the fluvial soils along the Xiaojiang River.

Despite extensive studies on the role of mining activities in the quality of soil and water resources, less research investigated the effect of variations in morphometric stream characteristics on heavy metal pollution levels of sediments transported by rivers surrounding mining sites. The drainage networks remain a principal reservoir of heavy metals evacuated by mining activities. Rivers are easily transported sediment polluted by heavy metals toward the downslope areas. Therefore, sediments are a critical indicator for assessing the level of water pollution and environment health. On a watershed scale, the positions of downslope act as a reservoir for the accumulation of heavy metals. Although, mining activity occurs on a small spatial scale, the presence of drainage networks surrounding mining sites can be a principal factor in the distributing of these harmful elements on broader spatial scales. As a result, mining activity in the watershed environments can be a serious threat to agriculture productions. Further, the streams with higher length and slope surrounding mining sites increase the pollution transfer rate and cause a higher level of sediment heavy metal pollution. Therefore, the assessment of the quality of river sediment surrounding mining sites is an important step toward terrestrial and aquatic ecosystem management.

5. Conclusion

In the present study, to better understand the effect of variation in morphometric stream characteristics on heavy metal pollution of sediments transported by rivers, we investigated changes in the levels of sediment pollution along streams with different morphometric characteristics, including different slopes and lengths. The results illustrated that the sediments belonging to all the rivers had heavy metal pollution induced by mining activity. However, the pollution level of the Kaka River sediment (with the greatest slope and length) was the highest compared with other rivers. Further, the geo-accumulation index illustrated that the sediments of all the rivers were strongly polluted with Pb and Cd, and moderately polluted with Zn. However, the highest Igeo value for the measured heavy metals was observed in the sediments belonging to rivers with the highest length and slope. Our findings unequivocally showed that watershed environments exhibit a high vulnerability to mining activities because the streams can facilitate the transportation of sediment polluted by heavy metals across the landscape. On the other hand, the streams with higher lengths and slopes surrounding mining sites increase the pollution transfer rate and cause a higher level of sediment heavy metal pollution.

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