



# The spatial distribution and source apportionment of heavy metals in soil of Shizuishan, China

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Received: 8 September 2022 / Accepted: 12 September 2023 / Published online: 26 September 2023  
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## Abstract

Environmental pollution of heavy metals in the typical coal industrial city should be paid more attentions nowadays. The spatial distribution and source apportionment of 8 heavy metals (i.e., Cd, Cr, Co, Zn, Ni, Cu, Pb and Mn) from topsoil samples (158) of Shizuishan city in Ningxia Hui Autonomous Region of China were investigated using principal component analysis/absolute principal component scores (PCA/APCS) receptor model and geographic information system (GIS). These results showed that the mean concentrations of Cd, Cr, Co, Zn, Ni, Cu and Pb were higher than their soil background values in Ningxia. 99.36% of soil samples were heavily polluted according to analysis of integrated Nemerow pollution index ( $P_N$ ), whereas 81.65% of soil samples exhibited the highly strong potential ecological risk by  $E_{RI}$  (the comprehensive of potential ecological risk index) values. The source apportionment results showed that eight heavy metals in soil were mainly from natural (32.39%), industrial (26.56%), traffic emission/coal consumption (20.18%) and atmospheric deposition source (12.73%). Typically, Zn, Mn and Ni were derived from natural source, whereas Cr and Co were mainly derived from industrial sources. Cu was from the multiple sources, whereas Pb and Cd were weighted primarily from traffic emission/coal consumption source and atmospheric deposition source, respectively. These findings were crucial for the prevention and control of heavy metals pollution in Shizuishan city.

**Keywords** Heavy metals · Spatial distribution · Source apportionment · Soil pollution

## Introduction

With the rapid development of industrialization and urbanization, more various heavy metals in the industrial cities were discharged into soil (Liu et al. 2013, 2023; Sun et al. 2019). Heavy metal in soils is an outstanding pollutant, which is from atmospheric deposition, livestock manures, fertilizers, sewage sludge and irrigation (Pan et al. 2016; Liang et al. 2017; Ran et al. 2021). The contamination of heavy metals in soil has attracted great environmental concern for their toxicity, persistence and bioaccumulation (Chen et al. 2015; Fang et al. 2015; Zhang et al. 2020). The excess heavy metals in soil could reduce crop productivity (Baek et al. 2012), due to the decrease of microbial activity

(Burges et al. 2015), the hindrance of nutrient uptake and accumulation (Hussain 2006) and the inhibition of crop root growth. More importantly, heavy metals have caused potential risk to human health through inhalation, ingestion and dermal absorption (Zhang et al. 2018; Hu et al. 2020). Cadmium (Cd) is one of the most toxic heavy metals to humans, which has harmful effects to structure and physiological and biochemical functions (Sarkar et al. 2013). Lead (Pb) can disrupt the neurological, skeletal, reproductive, hematopoietic, renal, and cardiovascular systems (Collin et al. 2022). Cobalt (Co) is considered a human carcinogen with the lung being a primary target (Smith et al. 2014). Copper (Cu), nickel (Ni), zinc (Zn), Manganese (Mn) and chromium (Cr) also have adverse effects on the human body when exposed to environments exceeding tolerated doses for a long time (Nriagu 2011; Mitra et al. 2022; Dey et al. 2023). Therefore, it is important and urgent to reveal the contaminations, ecological risks and sources identification of heavy metals in soils at the regional scales (Wang et al. 2019; Zhao et al. 2020; Zhang et al. 2023).

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The quantitative identification of potential sources was important for preventing and controlling the pollution of heavy metals in soil (Wang et al. 2015; Zhang et al. 2023). The multiple sources of heavy metals in soil included natural (e.g., pedogenic processes) and anthropogenic sources (e.g., mining, metallurgy, fossil fuel combustion, traffic, municipal solid waste) (Luo et al. 2015; Liu et al. 2018a). These multiple sources and various environmental factors greatly affected the spatial variability of soil heavy metals (Peng et al. 2019; Liu et al. 2021). Multivariate statistical approach and GIS method were frequently used to determine the spatial variability and possible sources of heavy metals in soils (Shan et al. 2013; Zhou et al. 2016; Liu et al. 2023), such as Chemical Mass Balance (CMB) model (Lan and Jiang 2013), Absolute Principal Component Scores (APCS) (Luo et al. 2015), UNMIX model (Lang et al. 2015), and Positive Matrix Factorization (PMF) (Liang et al. 2017). Among them, APCS as an effective tool has been successfully applied to identify independent sources and quantify the contributions of each identified source in numerous sources (Jin et al. 2019; Zhang et al. 2021).

Shizuishan is an important industrial city in the North-western of China, with generous industrial enterprises such as mining, chemical industry and machinery manufacturing (Fan et al. 2006; Xia et al. 2020). With the development of the Belt and Road Initiative, Shizuishan city strived to build an inland open economic experimental zone, but also introduced a large number of highly polluting enterprises (Liu et al. 2015). The discharge of waste (e.g., gas and solid waste) was being increasing continually from various industries such as coal, power, metallurgy, machinery manufacture, chemical industry, construction material (Wang et al.

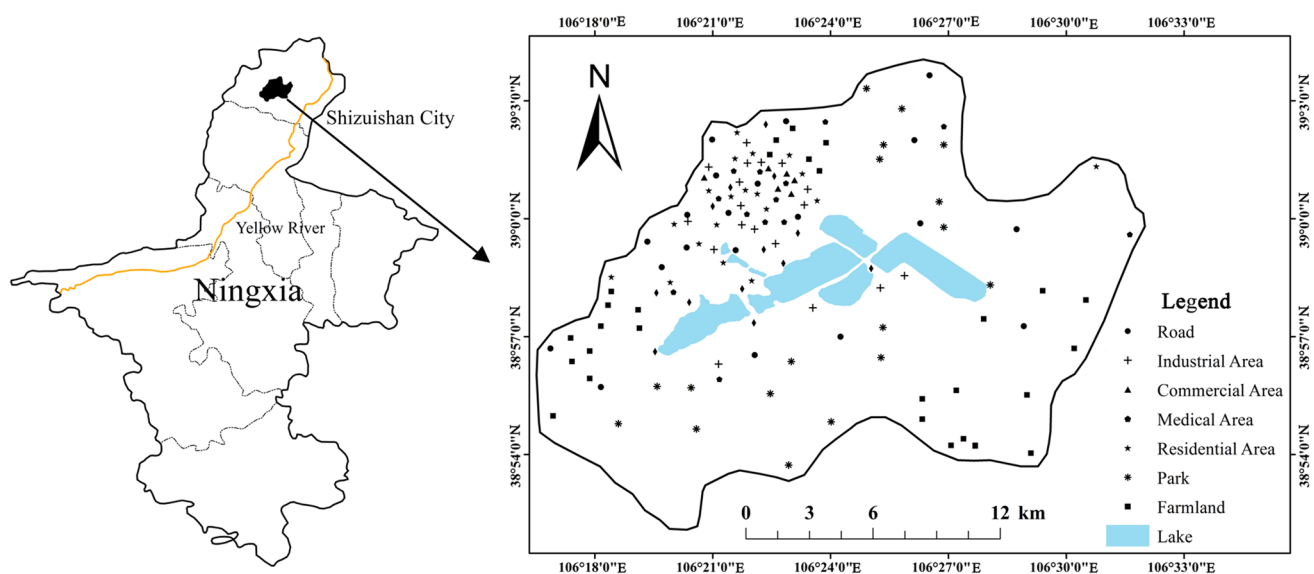
2021). Therefore, Shizuishan city was highly exposed to various heavy metal sources (Fan et al. 2013). The average concentrations of Cd, Cr, Zn, Cu and Pb in Shizuishan were higher than the local soil background values (Fan et al. 2013), particularly Cd as the most polluted element in these heavy metals (Wang et al. 2014, 2016). Most of the previous studies in China concentrated on the heavy metals in eastern or southern industrial areas (Liang et al. 2017; Sun et al. 2019), urban and agricultural regions (Zhu et al. 2017; Liu et al. 2021; Zhang et al. 2021). However, the contamination, ecological risks, spatial distribution and source identification of heavy metals in soil of Shizuishan city were not available.

The main objectives of the study were to (1) evaluate the contamination and ecological risks of eight heavy metals in soil from Shizuishan city, (2) identify sources and quantify the contributions of each identified source of heavy metals by APCS, and (3) investigate the spatial distribution of concentrations, contamination indices and potential sources of eight heavy metals. These results were useful for the prevention and reduction of heavy metals in soil of Shizuishan city.

## Materials and methods

### Geologic setting

Shizuishan city (105°58'–106°39' E, 38°21'–39°25' N, elevation of 1090–3475.9 m, respectively) is located in the upper and middle reaches of the Yellow River, the north of Ningxia Hui Autonomous Region, China (Fig. 1). The urban area of Shizuishan (~5310 km<sup>2</sup>) belongs to the typical temperate continental climate with annual average temperature



**Fig. 1** The location of the study area and distribution of the sampling sites

of 8.4–9.9 °C, average annual rainfall of 167.5–188.8 mm, and annual evaporation of 1708.7–2512.6 mm. The western areas of Shizuishan city are the Helan Mountain with steep hillsides, and the eastern areas are vast plain (e.g., piedmont pluvial plain, alluvial–proluvial plain and river–lake alluvial plain). The particle sizes of soil predominantly include sandy gravel, fine sand, silt and silt sand. The main types of soil are new alluvial, light sierozem, aeolian sandy and alluvial soil. The rich mineral resources include coal (e.g., 47 coal mines, proved reserves of  $24.3 \times 10^8$  t, total area of 2200 km<sup>2</sup>), silica, clay, aluminum and gold (Xia et al. 2020).

### Sampling and chemical analysis

A total 158 samples (0–20 cm) were collected from topsoil and then measured the concentrations of 8 heavy metals (i.e., Cd, Cr, Co, Zn, Ni, Cu, Pb, and Mn). The geographic coordinates of sampling sites were positioned by GPS (Fig. 1). Each sample was composed of a mixture of five subsamples collected from five spots of an area (100 m<sup>2</sup>) using wooden shovel. Each soil sample (~ 1 kg) was retained by quartering method and then stored in clean polyethylene bags and returned to the laboratory immediately. All soil samples were air-dried, removed debris, sieved through 2 mm polyethylene mesh, and then stored in glass bottles for content determination.

The soil samples were primarily digested with HNO<sub>3</sub>–HCl–HF–HClO<sub>4</sub>, and then the concentrations of heavy metals were determined by inductively coupled plasma atomic emission spectrometer (ICP-AES, HK-8100, Beijing Huake ETS Analysis Instrument Co Ltd, China). The accuracy of determinations was verified using the National Standard Reference Material of China (GSS-8). The recovery rates of Cd, Cr, Co, Zn, Ni, Cu, Pb and Mn were 101.6, 100.5, 102.1, 100.1, 99.8, 105.2, 96.9 and 102.3%, respectively. Detection limits of eight heavy metals were 0.003–0.005 mg/kg.

### Contamination assessment

#### Geoaccumulation index

The geoaccumulation index ( $I_{geo}$ ) as a geochemical criterion was used to evaluate contamination level of heavy metals in soils by comparing the concentrations of heavy metals to the background values (Liu et al. 2018a, b; Yang et al. 2018), which can be calculated by Eq. (1):

$$I_{geo} = \log_2 \left[ \frac{C_i}{1.5B_i} \right] \tag{1}$$

where  $C_i$  and  $B_i$  represent the measured concentration and background value of the metal  $i$  in soil, respectively, and the

coefficient of 1.5 is attributed to the small anthropogenic influences (Ochiagha et al. 2020).  $I_{geo}$  can be classified 7 types as follows: class 1 ( $I_{geo} < 0$ , practical no pollution), class 2 ( $I_{geo} = 0-1$ , no pollution to moderate pollution), class 3 ( $I_{geo} = 1-2$ , moderate pollution), class 4 ( $I_{geo} = 2-3$ , moderate to strong pollution), class 5 ( $I_{geo} = 3-4$ , strong pollution), class 6 ( $I_{geo} = 4-5$ , strong to extreme pollution) and class 7 ( $I_{geo} > 5$ , extreme pollution).

#### Contamination index

The single pollution index ( $P_i$ ) and the integrated Nemerow pollution index ( $P_N$ ) are applied to quantify the heavy metal contamination.  $P_i$  and  $P_N$  can be defined as Eqs. (2) and (3), respectively (Ma et al. 2018):

$$P_i = C_i/S_i \tag{2}$$

$$P_N = \sqrt{\frac{(\text{ave}P_i)^2 + (\text{max}P_i)^2}{2}} \tag{3}$$

where  $C_i$  and  $S_i$  are the measured concentration and reference values (i.e., average background values of soils) of heavy metal  $i$ , respectively. The  $\text{max}P_i$  and  $\text{ave}P_i$  are the maximum and average value of  $P_i$ , respectively.  $P_i$  can be classified as 3 types: low ( $P_i \leq 1$ ), moderate ( $1 < P_i \leq 3$ ) and high contamination ( $P_i > 3$ ).  $P_N$  is classified as 5 types: safety ( $P_N \leq 0.7$ ), warning ( $0.7 < P_N \leq 1$ ), light pollution ( $1 < P_N \leq 2$ ), moderate pollution ( $2 < P_N \leq 3$ ), and heavy pollution ( $P_N > 3$ ) (Zhu et al. 2017).

#### Potential ecological risk index

Potential ecological risk index (PERI) method can not only evaluate the ecological risk of single heavy metal, but also comprehensively understand the potential ecological risk of multiple heavy metals to soil (Hakanson 1980), which can be defined as follows:

$$C_i = C_r^i/C_b^i \tag{4}$$

$$E_i = T_i \times C_i \tag{5}$$

$$E_{RI} = \sum_{i=1}^n E_i \tag{6}$$

where  $C_i$  is the contamination coefficient of heavy metal  $i$ ,  $C_r^i$  and  $C_b^i$  are the measured and background concentrations of heavy metal  $i$ , respectively.  $E_i$  and  $T_i$  refer to the PERI and toxicity response coefficient for a single heavy metal  $i$ , respectively. The values of  $T_i$  were calculated as 30 (Cd), 2 (Cr), 5 (Co), 1 (Zn), 5 (Ni), 5 (Cu), 5 (Pb) and 1 (Mn)

(Hakanson 1980; Alahabadi and Malvandi 2018).  $E_{RI}$  is the comprehensive PERI of the heavy metals, whose classification standard is adjusted with the toxicity and quantity of elements.

According to Fernández and Carballeira (2001), the individual PERI of a single heavy metal can be divided as: the lowest ( $E_i = 1$ ), low ( $E_i < 30$ ), moderate ( $E_i = 30-60$ ), strong ( $E_i = 60-120$ ), very strong ( $E_i = 120-240$ ), highly strong ( $E_i > 240$ ). The comprehensive PERI of all heavy metals can be divided as: low ( $E_{RI} < 60$ ), moderate ( $E_{RI} = 60-120$ ), strong ( $E_{RI} = 120-240$ ), very strong ( $E_{RI} = 240-480$ ), highly strong ( $E_{RI} > 480$ ) (Fernández and Carballeira 2001; Chen et al. 2018).

## Source apportionment

### Multivariate statistical analysis

The correlation coefficients were used to measure the intensity of inter-relationship between heavy metals. The principal component analysis (PCA) was a mathematical process to identify the sources of heavy metals in soil.

### APCS-MLR (the absolute principal component scores and multivariate linear regression) receptor model

APCS-MLR receptor model was used to calculate the contributions of various sources for each heavy metal after determining the number and characteristics of possible sources (Singh et al. 2008). The calculation steps are as follows (Guo et al. 2004):

- ① Standardized all heavy metals:

$$Z_{ij} = \frac{C_{ij} - \bar{C}_i}{\sigma_i} \quad (7)$$

where  $Z_{ij}$  is the normalization of all heavy metals' concentrations,  $C_{ij}$  is the concentration of variable  $i$  in sample  $j$ ,  $\bar{C}_i$  and  $\sigma_i$  are the arithmetic mean concentration and standard deviation of variable  $i$ , respectively.

- ② Introduced an artificial sample (i.e.,  $C_{ij} = 0$ ) for all variables:

$$Z_{0i} = \frac{0 - \bar{C}_i}{\sigma_i} = -\frac{C_i}{\sigma_i} \quad (8)$$

- ③ The APCS for each component are estimated by subtracting the factor scores of this artificial sample from the factor scores variables, which are obtained from PCA by analysis of normalized heavy metals concentrations.

- ④ The multiple linear regression is carried out between each heavy metal concentration as dependent variable and APCS as independent variable. The resultant regression coefficients convert APCS into the contribution of each source to each sample:

$$C_i = b_{0i} + \sum_{p=1}^n (APCS_p \cdot b_{pi}) \quad (9)$$

where  $C_i$  is the concentration of heavy metal,  $b_{0i}$  is the constant of multiple regression for pollutant  $i$ ,  $b_{pi}$  is the coefficient of multiple regression of the source  $p$  for pollutant  $i$ , and  $APCS_p$  is the scaled value of the rotated factor  $p$  for the considered sample, which represents the contribution of source  $p$  to  $C_i$ .

### Methods of data analysis

Statistical analysis, PCA and multiple linear regression of heavy metal content in soil were carried out by SPSS18.0 and Excel 2007. The spatial distribution maps were completed by ArcGIS 10.0.

## Results and discussion

### Concentrations of heavy metals in soil

The descriptive statistical characteristics of eight heavy metals in topsoil were shown in Table 1. Variation coefficient of eight heavy metals ranged from 0.24 to 0.58, showing moderate degree of variability. According to KS (Kolmogorov–Smirnov) test of normality, the concentrations of Cd, Cr, Co and Zn were normally distributed ( $P > 0.05$ ), whereas the concentrations of Ni, Cu, Pb and Mn were lognormally distributed. The mean concentrations of Cd (4.15 mg/kg), Cr (203.18 mg/kg), Co (97.85 mg/kg), Zn (70.27 mg/kg), Ni (37.09 mg/kg), Cu (50.08 mg/kg), Pb (67.83 mg/kg) and Mn (457.97 mg/kg) were higher than their soil background values of Ningxia (CNEMC 1990), which was 37.70, 3.35, 8.51, 1.20, 1.01, 2.27, 3.29 and 0.87 times, and the standard ratios exceeded 97.47%, 98.73%, 99.37%, 71.52%, 41.14%, 96.84%, 96.20% and 22.78%, respectively. Compared with the soil environmental quality-risk control standard for soil contamination of agricultural land in China (MEEC 2018), only the concentration of Cd was higher than the corresponding risk intervention value (to guarantee and protect agricultural and human health). The heavy metal may cause harm to human health when the concentrations exceeded risk intervention values (Ma et al. 2020). Thus, Cd was the most harmful heavy metal in study area.

**Table 1** Descriptive statistical characteristic for eight heavy metals in soil in the study area

Heavy metals	Minimum (mg/kg)	Maximum (mg/kg)	Mean (mg/kg)	Standard deviation	Coefficient of variation	KS test	Background of Ningxia <sup>a</sup> (mg/kg)	Chinese soil criteria GB15618-2018 <sup>b</sup> (mg/kg) pH > 7.5
Cd	0.08	9.61	4.15	2.39	0.58	N	0.11	0.60
Cr	17.03	367.48	203.10	49.14	0.24	N	60.60	250
Co	8.15	167.99	97.85	25.34	0.26	N	11.50	–
Zn	3.57	155.80	70.27	28.01	0.40	N	58.80	300
Ni	6.81	138.82	37.09	18.07	0.49	LN	36.60	190
Cu	5.86	119.42	50.08	20.25	0.40	LN	22.10	100
Pb	6.04	193.54	67.83	38.31	0.56	LN	20.60	170
Mn	76.99	1012.66	457.97	146.58	0.32	LN	524	–

N normally distribution; LN lognormally distribution

<sup>a</sup>Data came from CNEMC (1990)

<sup>b</sup>Data came from MEEC (2018)

The pollution of Cd, Cr, Co, Ni, Cu and Pb in Shizuishan city was much higher than the other regions in China such as Guangrao County of northern Shandong Province (Lv 2019), Xiamen City (Luo et al. 2015), Puning City of south-east Guangdong Province (Wang et al. 2019), Tangshan City of Hebei Province (Sun et al. 2019) and Lin’an City of Zhejiang Province (Zhao et al. 2020) (Table 2). These results indicated that the soil was contaminated seriously by heavy metals, especially Cd in Shizuishan, which was at least eleven times higher than other regions in China.

### Contamination assessment

#### Geoaccumulation index

Figure 2a shows the class distribution of geoaccumulation index ( $I_{geo}$ ) of eight heavy metals. According to calculation of  $I_{geo}$ , Zn, Mn and Ni within 80% of sample sites were classified as class 1, indicating that most of soil were practically unpolluted by Zn, Mn and Ni. The  $I_{geo}$  values of Pb were dominated as class 2(38.61%) and 3(37.34%). The class of Cr (70.25% of class 3) and Cu (68.99% of class 2) was slightly lower than Co (87.34% of class 4), indicating moderate Cr/Cu pollution and strong Co pollution in the most of soil. However, the  $I_{geo}$  values of Cd ranged from

class 1 to 7 and more than 57% of samples were above class 6, indicating that soil was strongly or extremely polluted by Cd.

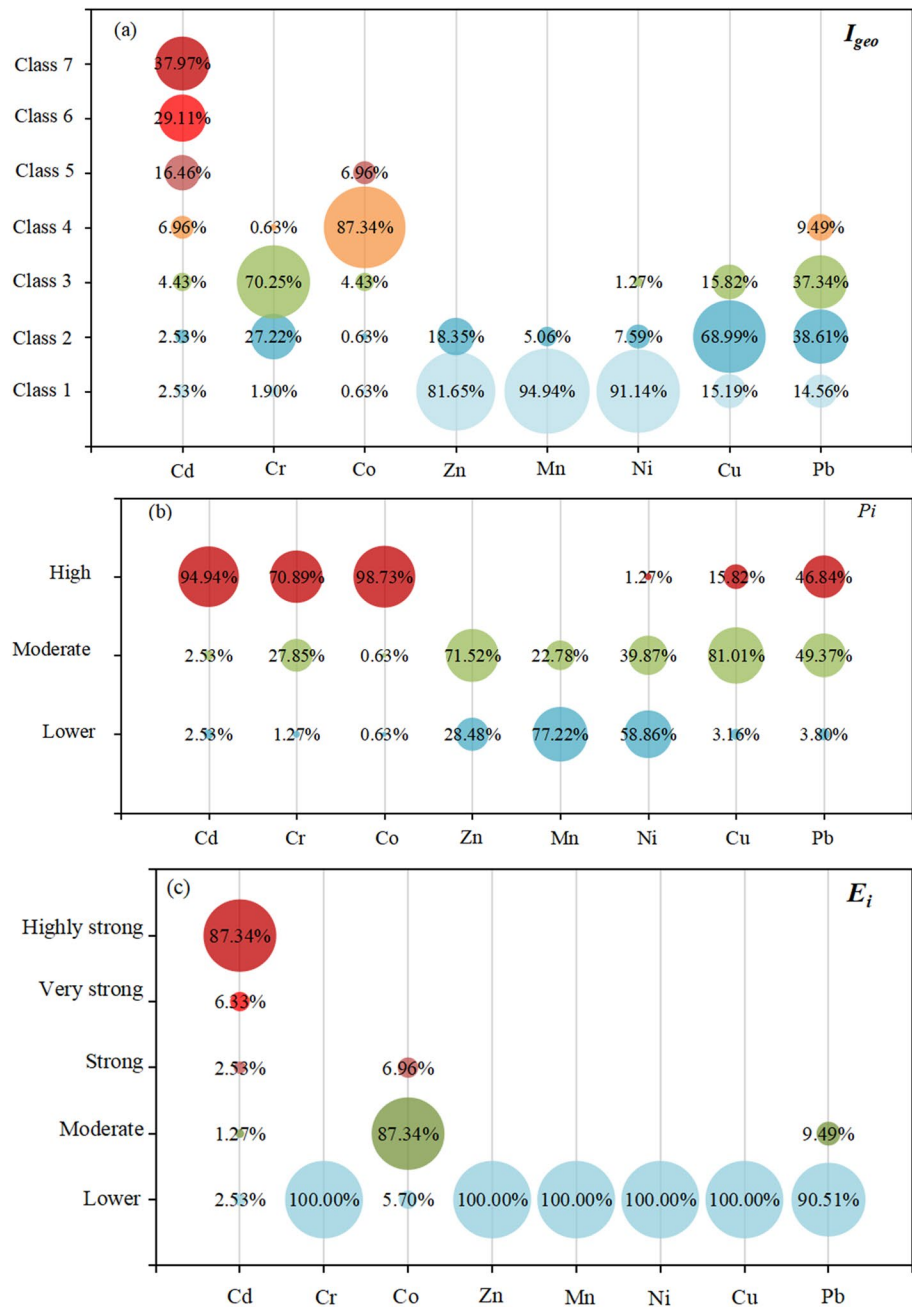
#### Contamination index

Figure 2b shows the contamination index ( $P_i$ ) of eight heavy metals in soil. There were some differences and similarities between the glass distribution of  $P_i$  values and  $I_{geo}$  values both corresponding to the background concentration of Ningxia. The mean of  $P_i$  values and  $I_{geo}$  values both showed the contamination levels of these heavy metals generally decreased in the order of: Cd > Co > Cr > Pb > Cu > Zn > Ni > Mn. The  $P_i$  values showed no high contamination for Zn, Mn and nearly all Ni in soil samples. The concentration of Cu showed moderate level at 81.01% sampling site, and high and moderate contamination for Pb presented in almost 50% sampling site, respectively. The study strongly suffered from the high contamination from Cd, Cr and Co in 94.94%, 70.89% and 98.73% of all sampling sites, a much higher percentage compared with  $I_{geo}$  values. In addition,  $P_N$  as a multi-element method was also used to evaluate overall soil quality according to Eq. 3. The high  $P_N$  values (from 1.28 to 62.45) indicated that 99.36% of soil was heavily polluted.

**Table 2** The mean values of soil heavy metals (mg/kg) in other regions of China

Regions	Cd	Cr	Co	Zn	Ni	Cu	Pb	Mn	References
Shizuishan city	4.15	203.10	97.85	70.27	37.09	50.08	67.83	457.97	This study
Guangrao County	0.13	68.10	11.90	67.20	28.80	24.00	22.60	586.00	Lv (2019)
Xiamen City	0.14	12.40	4.50	73.00	6.62	25.10	30.40	349.00	Luo et al. (2015)
Puning City	0.06	22.50	–	56.60	12.00	11.10	42.40	–	Wang et al. (2019)
Tangshan City	0.15	36.98	–	70.31	16.81	22.42	22.93	–	Sun et al. (2019)
Lin’an City	0.37	56.57	–	87.61	28.33	40.76	20.10	–	Zhao et al. (2020)

**Fig. 2** Class distribution of geoaccumulation ( $I_{geo}$ , **a**), contamination indexes ( $P_i$ , **b**) and single index of the ecological risk factor ( $E_i$ , **c**) of eight heavy metals in soil



**Potential ecological risk index**

The individual potential ecological risk index ( $E_i$ ) was used to assess the potential impact of single heavy metal to the ecosystem of Shizuishan city soils (Fig. 2c). Cr, Zn, Mn, Ni and Cu posed a low ecological risk in Shizuishan city, whereas Pb posed moderate and low ecological risks to 9.49% and 90.51% sampling sites, respectively. However, Co posed the strong and moderate ecological risks to 6.96% and 87.34% sampling sites, respectively. It was noted that Cd posed very strong (6.33% sampling

sites) and highly strong ecological risks (87.34% sampling sites). Thus, the low (1.90%), moderate (1.90%), strong (5.70%), very strong (8.86%) and highly strong (81.65% of sampling sites) potential ecological risks were obtained according to calculation of  $E_{RI}$ . Combined with the  $P_i$ ,  $I_{geo}$  and  $E_i$ , Shizuishan city existed the serious Cd pollution and extreme strong threat to the ecosystem. Liu et al. (2021) in southern Shandong Peninsula and Jin et al. (2019) in Shaoxing City also found that Cd was high potential ecological risk category.

## Spatial distribution of heavy metals and contamination index

The spatial distributions of eight heavy metals in topsoil were obtained by GIS method (Fig. 3). The similar varied tendencies of concentrations of four heavy metals (i.e., Mn, Ni, Zn and Cu) in soil were observed. The concentrations of four heavy metals in the west and north regions were significantly lower than those in the southeast regions, which was relative to accumulation of alluvium and residues. Besides, the accumulation of four heavy metals was also influenced by industrial activities from industrial development zone in the southeast of this area. The high Pb concentrations were observed in the south of Xinghai Lake, where a large coal chemical enterprise was located. Thus, the serious Pb pollution was influenced by coal chemical industrial activities such as combustion of coal, smelting of non-ferrous metal, production of chemical products and building materials. The highest Cd concentrations mainly appeared in northwest (i.e., Dawukou district of Shizuishan city), where administrative area of municipal government, the highly dense population and some industrial enterprises were present (Xia et al. 2020). Previous researchers found that the heavy metals often focused on these areas with frequent human activities and more waste emissions (Wang et al. 2019; Chai et al. 2021). Thus, the high distribution of Cd in the area was attributed to human activities. The low Co and Cr concentrations were mainly found in the northwest of the area, whereas the high values were irregularly distributed in the east, west and middle part of this area, where there were industrial areas. Thus, the high distribution of Co and Cr concentrations could be controlled by industrial activities.

The spatial distribution of  $P_N$  and  $E_{RI}$  values of eight heavy metals are shown in Fig. 4. The high values of  $P_N$  and  $E_{RI}$  in the northwest of the study area and a small region of southeast were observed, indicating serious environmental pollution and highly strong potential ecological risk in these regions. For Cd, the similar spatial distribution of  $P_N$  and  $E_{RI}$  suggested serious Cd pollution and significant influence on the spatial distribution of heavy metals.

## Source apportionment of heavy metals

### Correlation analysis

Correlation analysis is an effective method to identify the relationship between different heavy metals, and the high correlation coefficients may indicate the similar sources between different heavy metals (Micó et al. 2006). Pearson correlation coefficients of eight heavy metals in soils are listed in Table 3. The concentration of Cd was not significantly correlated with any other heavy metals, indicating Cd and other metals were not controlled by a single

factor. However, Cr, Co, Zn, Ni, Cu, and Mn were found significantly positively correlated with each other, indicating possible same sources for these heavy metals. Under these positively correlation, high correlations between Cr and Co (0.459), Zn and Mn (0.463), Mn and Ni (0.531) were observed. The only positive correlation of Pb with Cu ( $P < 0.01$ ) and Mn ( $P < 0.05$ ) indicated possible same origins. According to correlation analysis, eight heavy metals were divided into three groups: (i) Cr, Co, Zn, Ni, Cu and Mn; (ii) Cd and (iii) Pb. The complicated relationships of these heavy metals were influenced by various factors, and the comprehensive conclusion of correlation coefficients was not obtained. The areas with compound soil pollution were suitable for further traceability study using principal component analysis (Liu et al. 2023).

### Principal component analysis

To further examine the relationships between eight heavy metals and their sources, PCA and APCS were used to identify the sources of heavy metal in soil and to calculate the contributions of various sources. According to KMO and Bartlett sphericity test (KMO = 0.747,  $P < 0.05$ ), PCA was suitable to identify the sources of eight heavy metals due to the strong correlations of variables (Table 3). The orthogonal rotation and maximum variance methods were used to extract and rotate the factor load matrix orthogonally, respectively. Then factor extraction with eigenvalues ( $> 1$ ) was conducted for source identification. Four factors were extracted by PCA method, explaining 72.6% of the cumulative variance contribution (Table 4). The range of communalities of variables (0.534–0.993) indicated that four factors were generally represented the information contained in the source data. To further identify possible sources of heavy metals and some pollution hotspots in study area, spatial distribution maps of four principal components (PC1, PC2, PC3 and PC4) were visualized using GIS method (Fig. 5).

According to analysis of PC1, 25.84% of the total variance was mainly loaded on Zn (0.730), Ni (0.744) and Mn (0.844), and moderately loaded on Cu. In study area, the concentrations of Zn, Ni and Mn in most samples were less than or near the background levels. Mn was considered generally as the most abundant element in the lithosphere and the major element in soil (Lee et al. 2006; Chen and Lu 2018). It was also reported that the main source of Ni was derived from parent material (Cai et al. 2019; Sun et al. 2019). The high PC1 in the southeast of this area was matched well to the distribution of alluvium and residues. The variability of the elements seemed to be controlled by parent rocks. Thus, PC1 can be considered to the factor of natural source.

Approximately, 20.45% of the total variance had predominant loading of Cr and Co and moderate loading of

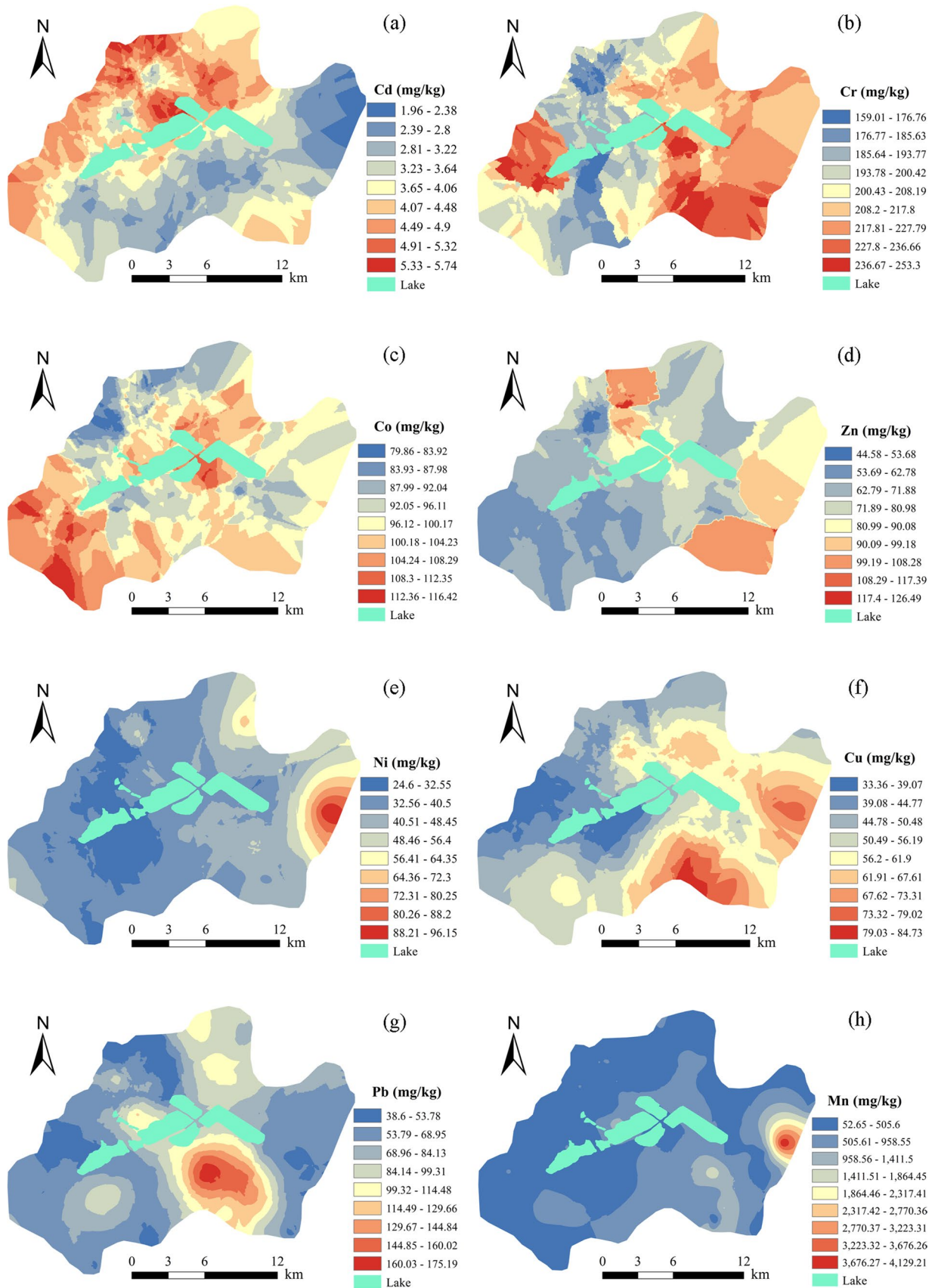
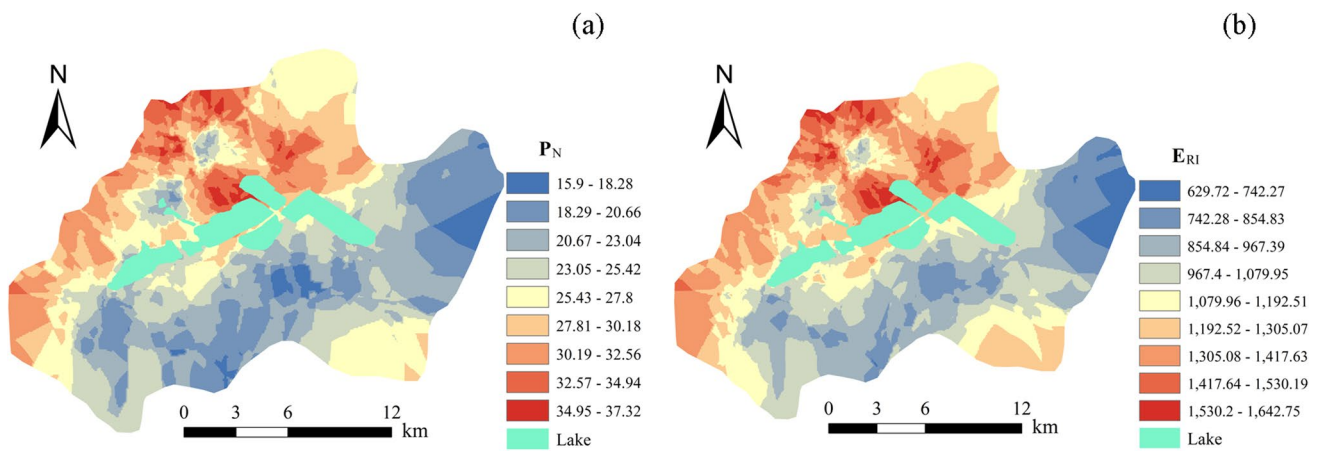


Fig. 3 Spatial distribution of eight heavy metals (a Cd; b Cr; c Co; d Zn; e Ni; f Cu; g Pb; h Mn) in soil





**Fig. 4** The spatial distribution of  $P_N$  (a) and  $E_{RI}$  (b) values of eight heavy metals in soils of Shizuishan city

**Table 3** Correlation coefficients between heavy metal elements in soils

	Cd	Cr	Co	Zn	Mn	Ni	Cu	Pb
Cd	1							
Cr	-0.042	1						
Co	-0.052	0.459**	1					
Zn	-0.018	0.289**	0.222**	1				
Mn	-0.096	0.208**	0.227**	0.463**	1			
Ni	-0.065	0.278**	0.251**	0.346**	0.536**	1		
Cu	-0.072	0.341**	0.349**	0.378**	0.431**	0.261**	1	
Pb	-0.095	0.046	0.143	0.035	0.184*	0.121	0.225**	1

\*, \*\* refer to correlation coefficients significant at  $P < 0.05$  and  $P < 0.01$  (two tailed), respectively

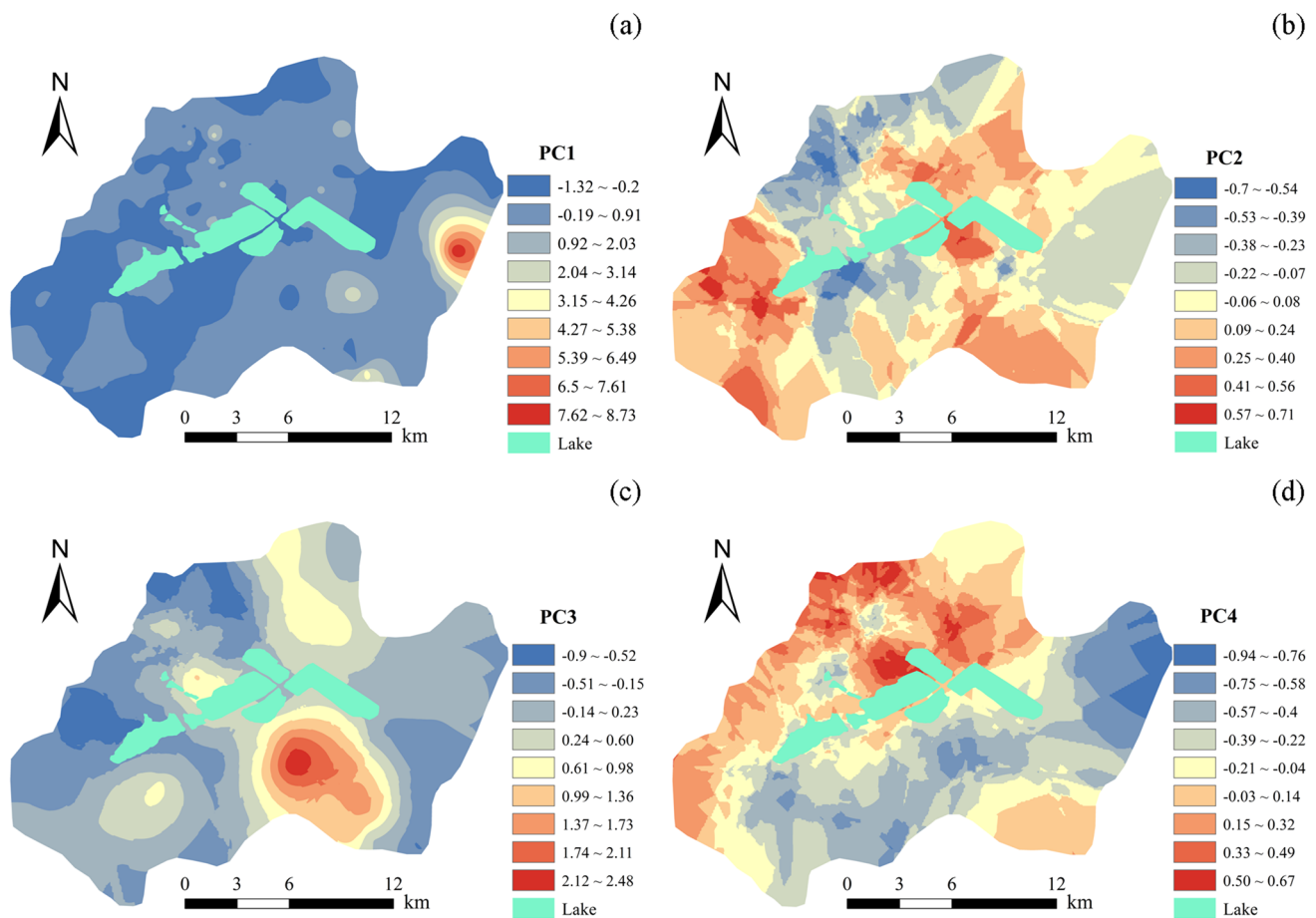
**Table 4** Rotated component matrix for heavy metals in soil

Elements	Component				Communalities
	PC1	PC2	PC3	PC4	
Cd	-0.043	-0.033	-0.045	<b>0.994</b>	0.993
Cr	0.185	<b>0.826</b>	-0.093	-0.022	0.726
Co	0.112	<b>0.821</b>	0.139	-0.022	0.707
Zn	<b>0.730</b>	0.215	-0.107	0.062	0.595
Mn	<b>0.844</b>	0.063	0.185	-0.067	0.755
Ni	<b>0.744</b>	0.132	0.039	-0.064	0.577
Cu	<b>0.464</b>	<b>0.456</b>	<b>0.333</b>	0.002	0.534
Pb	0.056	0.048	<b>0.955</b>	-0.047	0.920
Eigenvalues	2.068	1.636	1.100	1.004	
Percentage of variance/%	25.847	20.451	13.754	12.548	
Accumulative of variance/%	25.847	46.298	60.051	72.600	

The bold emphasized that this component was mainly loaded on these heavy metals

Cu according to PC2 analysis. It was noted that the mean concentrations of Cr and Co were much higher than their background values. Moreover, the concentrations of Co at 98.1% sampling sites and Cr at 15.33% sampling sites exceeded the risk intervention values. In previous studies, it was demonstrated that Cr inherited from parent materials

were lower than background values (Lv 2019; Sun et al. 2019). However, Zhang et al. (2016) based on 464 published papers reported that the mean concentrations of Cr in China (78.94 mg/kg) exceeded the background value (57.30 mg/kg) and the maximum was up to 3353.60 mg/kg, indicating introduction of Cr into soil by human activities. It was well



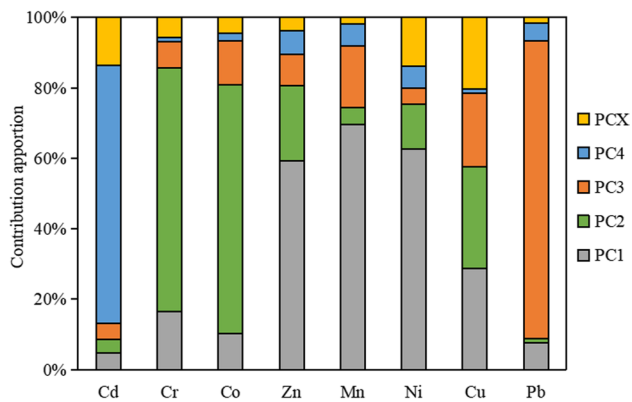
**Fig. 5** Spatial distributions of four principal components scores: **a** PC1, **b** PC2, **c** PC3 and **d** PC4

known that Shizuishan city was an important industrial city in northwest of China, featuring the frequent and complex industrial activities (e.g., coal mining and consumption, steel smelting, metal processing, thermal power and chemical industry) (Zhou and Li 2008). It was demonstrated that high Cr concentrations were likely associated with industrial activities such as Hexi Corridor (Guan et al. 2017), Jinchang mining city (Li et al. 2011), Shanxi Province (Pan et al. 2016), and Wuhan (Qu et al. 2013). Besides, the large source of Co was attributed to the contribution of steel production (Qu et al. 2013). According to the spatial distribution (Fig. 5), high PC2 in surrounding industrial areas was considered to the industrial sources.

Approximately, 13.75% of the total variance was predominantly loaded on Pb (0.955) according to analysis of PC3, and moderate load of Cu (0.333) was observed. Generally, Pb as the antiknock agent was added to gasoline to reduce the knocking tendency in vehicle engines (Chen and Lu 2018; Sun et al. 2019), thus vehicle exhaust was the most significant source of Pb in urban soil (Guo et al. 2004; Hu et al. 2018). Moreover, high PC3 was located in the south region of the area, where a large coal chemical enterprise

was located. The intensive activities of coal combustion were observed and plenty of cinder was dumped nearby area. Thus, the enrichment of Pb in soil was much associated with the coal consumption (Qu et al. 2013). In addition, Pb was extremely affected by anthropogenic sources such as industrial activities, traffic, mining, smelting and agricultural practice (Lv 2019). Cu was conventionally used as the vehicular braking system and automotive radiators because of the high corrosion resistance and thermal conductivity, so the high concentrations of Cu in soil were linked to traffic emissions (Chen and Lu 2018; Sun et al. 2019). Therefore, PC3 was considered to the traffic emission/coal consumption source.

Approximately, 12.54% of the variance was only predominantly loaded on Cd (0.994) according to analysis of PC4. Figure 5 shows the high and low PC4 in the northwest and southeast of the study area, respectively. The region of high PC4 was consistent with Dawukou District of Shizuishan city, where highly dense population and many industrial enterprises were present. Cd as an important addition was used in lubricating oil, wearing of tires and brakes (Duan and Tan 2013), so frequent industrial and traffic activities



**Fig. 6** Source contribution apportionment (natural source (PC1), industrial sources (PC2), traffic emission/coal consumption source (PC3), atmospheric deposition source (PC4) and unidentified sources (PCX)) of eight heavy metals by APCS-MLR model

could lead to the release of Cd into the environment, which was significant source of Cd (Liu et al. 2018b; Men et al. 2018). In addition, Cd accumulated nearby the iron and steel industries by atmospheric deposition and the adsorption of industrial dust particles (Bi et al. 2006). Therefore, PC4 was atmospheric deposition source.

Based on source identification, APCS-MLR model was applied to apportion the contributions of identified sources of heavy metal concentrations in soil (Fig. 6). The high  $R^2$  values of APCS-MLR model (e.g., Cd and Mn were 0.988 and 0.959, respectively) indicated the credible source apportionment results (Ma et al. 2018). The contributions of natural source (PC1, 32.39%), industrial sources (PC2, 26.56%), traffic emission/coal consumption source (PC3, 20.18%), atmospheric deposition source (PC4, 12.73%) and unidentified sources (PCX, 8.13%) were obtained by APCS-MLR model. PCX was mainly derived from wastewater, sewage and other potential pollution sources. It was observed that 67.61% of the heavy metals in soil were contributed to anthropogenic sources. PC1 predominantly contained Zn (59.34%), Mn (69.67%) and Ni (62.70%), whereas PC2 included Cr (69.10%) and Co (70.71%). PC3 and PC4 were primarily weighted on Pb (84.54%) and Cd (73.29%), respectively. It was noted that Cu exhibited the most complicated sources, including PC1 (28.65%), PC2 (28.87%), PC3 (20.90%), and PCX (20.28%). Thus, the serious pollution of heavy metals in Shizuishan city was attributed to the anthropogenic activities such as industrial sources and traffic emission/coal consumption source.

## Conclusions

In this study, the spatial distribution and source apportionment of heavy metals in soil of Shizuishan city were investigated by PCA/APCS and GIS method. The mean concentrations of Cd, Cr, Co, Zn, Ni, Cu and Pb were higher than their soil background values of Ningxia, and highest Cd was obtained. The spatial distribution showed that the most northwest and small region of southeast exhibits high  $P_N$  and  $E_{RI}$  values, indicating that 99.36% of the soil was heavily polluted by heavy metals, and 81.65% of soil had highly strong potential ecological risk through  $E_{RI}$  values. According to analysis of APCS-MLR and GIS method, Zn, Ni and Mn were predominantly derived from natural source, whereas Cr and Co were mainly derived from industrial source, Pb and Cu were from the traffic emission/coal consumption source and Cd was from atmospheric deposition source, which were mainly distributed in the southeast, industrial area, a large coal chemical enterprise and highly dense population/many industrial enterprises, respectively. These findings indicated that APCS-MLR approach and geostatistical method were an effective tool for source apportionment and identification of heavy metal pollution in soil, which had reference significance for the prevention and control of soil heavy metal pollution in some industrial cities.

**Acknowledgements** This work was supported by the National Natural Science Foundation of China (No. 41761049, 41867003), and National Key Research and Development Program of China (No. 2021BEG02011).

**Author contributions** YB and YW wrote the main manuscript text, and YZ and XL prepared the figures. All authors reviewed the manuscript.

**Funding** This research was funded by National Natural Science Foundation of China, Grant no [41761049, 41867003], Key Research and Development Program of Ningxia, Grant no [2021BEG02011].

**Data availability** All relevant data are within the paper.

## Declarations

**Conflict of interest** The authors declare no competing interests.

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