Pollution status and ecological risk of heavy metals in the soils of five land-use types in a typical sewage irrigation area, eastern China



Chunfang Li • Jianfei Cao • Lei Yao • Quanyuan Wu • Jianshu Lv

Received: 21 January 2020 / Accepted: 24 May 2020 / Published online: 30 June 2020 © Springer Nature Switzerland AG 2020

Abstract Sewage irrigation has been widespread in the water shortage area of eastern China and inevitably tends to result in heavy metal accumulation in soils. A total of 148 surface soil samples from five land-use types were collected in Longkou, a typical sewage irrigation area of China, and As, Cd, Cu, Pb, and Zn concentrations were determined. The Nemerow index method and improved fuzzy comprehensive evaluation method were used to examine the pollution status of heavy metals. The potential ecological risk was evaluated by the Hakanson model by adjusting the assessment threshold, and its spatial distribution was interpolated using geostatistical techniques. As, Cd, Cu, Pb, and Zn accumulated in different amounts in the five land-use types. Urban industrial land and mining land were moderately polluted, irrigated land was slightly polluted, orchards were minimally polluted, and bare land was at a safe level of pollution. Cd exhibited high percentages of strong and severe levels of potential ecological risks. For Cd, irrigated land, orchard, and bare land mainly presented moderate risks, whereas urban industrial land and mining land mainly presented high risks. The comprehensive ecological risk of the five

C. Li · J. Cao · L. Yao · Q. Wu (⊠) · J. Lv (⊠) College of Geography and Environment, Shandong Normal University, Jinan, Shandong, China e-mail: wqy6420582@163.com e-mail: 1vjianshu@126.com

C. Li

College of communications and civil engineering, Shandong Jiaotong University, Jinan, Shandong, China

heavy metals was at a severe level for all tested land-use classes except for bare land.

Keywords Land-use type \cdot Heavy metal \cdot Pollution status \cdot Ecological risk \cdot Soil

Introduction

Soil is the foundation of all terrestrial ecosystems (Boquete et al. 2014). If the soil environment is polluted, then the normal function of the associated ecosystem will be affected. Polluted soil will cause ecosystem structures and functions to change, and "soil \rightarrow body \rightarrow plants" and "soil \rightarrow water \rightarrow human body" cycles allow pollution to be indirectly absorbed by the human body (Mineev 1996; Mohammed et al. 2011; An et al. 2016), thus threatening human health. Attention must be paid to soil environmental quality and ecological environment safety to ensure that human health is not threatened by construction associated with national progress, and important human-oriented principles must be adhered to. In addition, soil heavy metal pollution is a type of soil environmental pollution.

The physical properties (texture, porosity, structure, thermal property, and ploughing) and chemical properties (absorbency, acid-base, buffering, and nutrient) of the same type of soil under different land uses are different (Oleszek et al. 2003; Qishlaqi et al. 2009). In recent years, many studies have focused on the physical and chemical properties of soil under different land uses. Liang et al. (2018) analysed the differences in soil physical properties

of five typical land uses in the lower reaches of Tibet's Nyang River Basin and found obvious differences in the soil mass, porosity, and water holding capacity of the different land uses. Amirian Chakan et al. (2017) found that the distribution of soil organic carbon (SOC) stocks varied laterally and vertically under different land-use types. Shougrakpam et al. (2010) studied the macroporosity of different land use and land cover soils in northeastern India. Digital image analysis shows that there are significant differences in infiltration patterns under different land use, land cover, and tillage modes. Tang and Han (2017) researched the characteristics of heavy metals in soils under different land uses in a typical karst area of southwestern China. Shang et al. (2015) studied the distribution and influencing factors of Se under different land-use types in the Yangtze River Basin. Zheng et al. (2005) analysed Beijing vegetable and paddy fields, orchards, green land, wheat, and natural soil and found that different land uses had significant effects on the accumulation of lead in the soil. The characteristics of heavy metals in soil are related to the physical and chemical properties of the soil. Differences in soil properties caused by different land-use patterns may also affect the content of heavy metals in soil. Many studies have focused on heavy metal pollution and the ecological risks to soil under single land-use types, and they have primarily concentrated on farmland (Lv et al. 2012; Zou et al. 2018), urban lands (Liang et al. 2017; Zhang et al. 2019), mining areas (Lu et al.2014; Chen et al. 2018, Wei et al. 2017), industrial parks and their surroundings (Luo et al. 2017), landfills (Zhao et al. 2015) and sewage treatment plants (Luo et al. 2016), and areas along rivers (Ghrefat et al. 2012). However, studies on soil heavy metal pollution and ecological risk associated with different land-use types in the same area are scarce. For the few studies that have explored the pollution and ecological risk caused by heavy metals in soils under different land-use types in the same area (Fernández et al. 2001, Zhang et al. 2017; Bartkowiak et al. 2017), the diversity of land-use types and the universality of the evaluation methods require additional investigation.

The Hakanson potential ecological risk index method evaluates the potential ecological risk of sediment in water bodies (Ma et al. 2011a, b). Many scholars have improved the method (Hou et al. 2011) and used it as a potential ecological risk assessment of soil heavy metals. However, in the previous studies, the evaluation criteria were not adjusted according to the type and toxicity of heavy metals. In recent years, various types of land uses have occurred in the northern plain of Longkou city due to the rapid development of industry and agriculture, and heavy metals have accumulated in the soil. We evaluated the ecological risks of Cu, Pb, Zn, Cd, and As in irrigated land, orchards, urban industrial land, mining land, and bare land to adequately understand the pollution status and ecological risk of soil heavy metals in this area. This study is intended to provide a reference for soil management and regulation.

Materials and methods

Study area

The study area is located in the northern plain area of Longkou city, Shandong Province (120°13'-120°44'E, 37°27'-37°47'N), with a total area of 409 km², of which the agricultural area with irrigation is approximately 280 km². The study area has a temperate monsoon climate, the annual average temperature is approximately 12 °C, and the average annual rainfall is approximately 600 mm. It is surrounded by the sea on three sides and has the same soil-forming conditions in all areas. The soil pH value ranges from 6.5 to 7.5. The types of soil forms include brown loam, cinnamon soil, and aeolian sand. The main type of soil is brown loam, and the largest river is the Huangshui River. The crops planted in this area are mainly wheat, maize, apple, and grape. The area is rich in mineral resources, such as coal, gold, lead, zinc, kaolin, fluorite, and placer, and many other industrial industries, such as mining, foundry, and paper-making, occur there. Longkou city was established in 1975, and with the implementation of the policy of reform and opening up, industrial and mining enterprises developed rapidly. By 1984, the area changed to using sewage irrigation. Sewage irrigation has occurred for 30 years. Untreated industrial and domestic drainage has irrigated farmland through the Huangshui River, Yongwen River, and its branches. After 2002, the sewage irrigation area was altered to a reclaimed water irrigation area. The reclaimed water discharged from the Longkou and Huangcheng sewage treatment plants also enters farmland through the Huangshui River, Yongwen River, and other branches.

Sample collection

The land-use map of Longkou city was used to determine the different land-use types and the proportion of the main pollution sources in the study area, with sampling points in the subsurface soil at depths of 0~20 cm. According to the proportion of land-use types, 148 sampling points were established, including 8 samples in bare land, 21 samples in urban industrial land, 24 samples in mining land, 44 samples in orchards, and 49 samples in irrigated agricultural land. The sample plots were 30 m \times 30 m, and the sampling process involved clearing the surface soil using a wooden spatula and collecting the soil at 5 points on the diagonal. Dirt and other debris in the soil were removed, and the soil was mixed. Then, 0.5 kg of soil from each quartile was placed in a clean cloth bag, and the sampling point location information was recorded (Fig. 1).

After air-drying at room temperature, the soil samples were screened through a 2-mm sieve, and then, an agate mortar was used to finely grind the soil so that it could pass through a 0.074-mm sieve to determine the total amount of heavy metals. In the laboratory, HSO4-HNO3-HF was used to digest the soil to analyse the As, Cd, Cu, Pb, and Zn contents. The Cd content was determined by graphite furnace atomic absorption spectrometry (GF-AAS, Thermo Scientific, USA). The contents of Cu, Pb, and Zn were determined by inductively coupled plasma atomic emission spectrometry (ICP-OES, Thermo Scientific, USA). The content of As was determined by hydride generation atomic fluorescence spectrometry (HG-AFS, Thermo Scientific, USA). The detection limits of As, Cd, Cu, Pb, and Zn were 0.1 0.01, 0.3, 0.3, and 0.5, respectively. All the reagents in the analysis were superior grade pure according to the national standard of soil samples (GBW7401), and the recovery rate was $100\% \pm 10\%$ (Lv et al. 2018). The accuracy and precision of the analysis method were tested(Ministry of ecology and environment of the People's Republic of China 2014), and the results were all in accordance with the requirements.(Ministry of environmental protection of the People's Republic of China 2014), Ministry of land and resourses of the People's Republic of China 2006, 2016)

Method of evaluating environmental pollution

Many methods of evaluating heavy metal pollution are available, and they can be divided into two categories based on study objectives: evaluation methods based on heavy metals, which include the single factor index, Nemerow index, enrichment factor, geoaccumulation index, and potential ecological hazard index, and evaluation methods based on sampling points, which include modelling methods, such as the fuzzy comprehensive evaluation, grey clustering model, and analytic hierarchy process (AHP). In addition, with the development of geographic information technology, evaluations based on GIS and geostatistics have also become popular. In this paper, the single factor and Nemerow index methods were used to evaluate each heavy metal, and the improved fuzzy comprehensive evaluation method was used to evaluate each sampling point. Finally, the results of the above two methods were analysed based on a GIS and geostatistics approach.

Single factor and Nemerow index methods

The single factor pollution index method is used to evaluate the degree of pollution of a single pollution factor, and it takes the background value of soil elements as the evaluation standard. This index is expressed as follows:

$$P_i = C_i / S_i \tag{1}$$

where P_i is the pollution index number of a pollution factor at sampling point, C_i is the actual measured value of the pollution factor, and S_i is the background standard value of the pollution factor. The background values of the soil environment obtained by Dai et al. in the eastern part of Shandong Province in 2011 were used in this study. A value of $P_i > 1.0$ indicates that the content of heavy metals in the soil has exceeded the standard, i.e. the soil has been polluted, and $P_i \leq 1.0$ indicates that the heavy metal content is in the background value range, i.e. the soil is not contaminated. Higher values of P_i correspond to an increasing severity of heavy metal pollution. Based on the single factor index, the Nemerow index method, which is a comprehensive evaluation method of heavy metal pollution, was used, and the calculation formula is as follows:

$$\mathbf{P} = \sqrt{\frac{Pi_{max}^2 + Pi_{ave}^2}{2}} \tag{2}$$

where *P* is the Nemerow comprehensive pollution index of the sampling point and Pi_{max} and Pi_{ave} are the maximum and average values of the i factor pollution index, respectively. The values of $P \le 0.7, 0.7 < P \le 1, 1 < P \le 2, 2 < P \le$

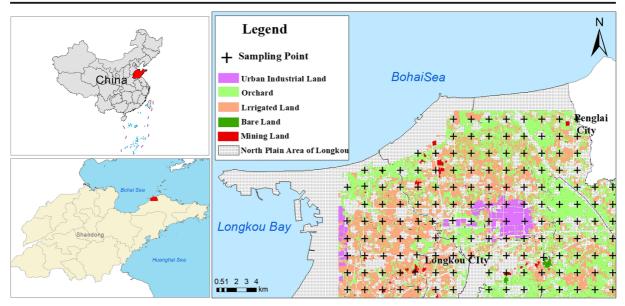


Fig. 1 Location of the study area with sampling sites

3 and P > 3 divide pollution into five categories from low to high: safe level of pollution, minimal pollution, light pollution, medium pollution, and heavy pollution, respectively.

Fuzzy comprehensive evaluation method

The fuzzy mathematical model and the evaluation factor weights were improved to assess the soil heavy metal pollution, and a fuzzy comprehensive evaluation method based on the double weight factor of pollutant concentration and toxicity was proposed.

$$\mathbf{B} = \mathbf{R} \cdot \mathbf{W} \tag{3}$$

where W represents the degree of membership of each evaluation factor to the evaluation grade; R represents the vector of the weight of each evaluation factor; and B represents the membership degree of the evaluation samples to evaluation grades (Dou et al. 2007).

(1) The portrayal of the membership degree was represented by the S shape function:

$$W(x, a, b) = \begin{cases} 1 & x \le S_{ij} \\ 2\left[\frac{x - S_{ij}}{x - S_{ij+1}}\right]^2 & S_{ij} < x \le \frac{S_{ij} + S_{ij+1}}{2} \\ 1 - 2\left[\frac{x - S_{ij}}{x - S_{ij+1}}\right]^2 & \frac{S_{ij} + S_{ij+1}}{2} < x \le S_{ij+1} & (4) \\ 0 & S_{ij+1} < x \end{cases}$$

where *x* is the measured concentration of the pollution factor; S_{ij} is the *j* index of the *i* pollution factor; and S_{ij+1} is the *j* + *1* index of the *i* pollution factor. The evaluation criteria of the background value and critical content of soil heavy metals were used in this evaluation (Table 1).

(2) The weight vector R of each evaluation factor was determined.

The excessive pollutant concentration method is commonly used to determine the weight of heavy metal pollution. Considering the different heavy metal pollutants and the toxicity of different individual pollutants, the concentration of pollutants that exceeds the standard weighting method must cover a proportion of low concentrations of toxicity. In addition, the toxicity of heavy metals at the level of the weight calculation can better reflect the concentration and toxicity of heavy metals. Lin used the heavy metal toxicity coefficient to determine the accuracy of toxicology evaluation data to provide a reference for research on heavy metal toxicity indexes, as shown in Table 2. Therefore, the concentration of pollutants and the grade of toxicity can be weighed and then normalized, and the calculation formula is as follows:

$$r_i = \frac{C_i/f_i}{\sum\limits_{i=1}^n C_i/f_i}$$
(5)

Table 1 Classification standard of soil heavy metal pollution (mg·kg⁻¹) (Dou et al. 2007)

Pollution factor	Safe I	Minimal II	Light III	Moderate IV	Heavy V	
Cu	28.37	40.63	120	280	400	
Pb	23.35	36.09	150	350	500	
Zn	83.68	116.75	240	560	800	
Cd	0.1204	0.2523	0.6	1.4	2	
As	6.71	8.92	13	30	45	

$$C_{i} = \frac{x_{ij} / \sum_{j=1}^{m} S_{ij}}{\sum_{i=1}^{n} \frac{x_{ij}}{\sum_{j=1}^{m} S_{ij}}}$$
(6)

where x_i is the measured concentration of pollution factor *i*; f_i is the toxicity grade index of pollution factor *i*; and r_i is the weight value of pollution factor *i*. In addition, $\sum_{i=1}^{n} C_i = 1$, $\sum_{i=1}^{n} r_i = 1$.

Potential ecological assessment methods

In this study, the potential ecological risk index (E) and the comprehensive potential ecological hazard index (RI) of each heavy metal were calculated using the Hakanson potential ecological risk assessment method. The calculation formula is as follows:

$$E_i^j = T_i \times P_i^j = T_i \times \frac{C_i}{S_i} \tag{7}$$

$$RI_j = \sum_{i=1}^n E_i^j \tag{8}$$

where E_i^j is the potential ecological risk index of the *j* pollution factor at the j sampling point. The toxic response coefficient of T_i to i heavy metals is the response to the toxicity of heavy metals and the sensitivity of biological pollution to the heavy metals; in this study, the values obtained from the study of Lin were selected (Table 2). In the study, P_i^j is the pollution index of the *i*

Table 2 Biological toxicity index of heavy metals

Pollution factor	Cu	Pb	Zn	Cd	As
Toxicity index	3	15	1	30	20

pollution factor at the *j* sampling point, C_i is the measured content of the pollution factor, and S_i is the background standard value of the pollution factor; the background value of the soil environment was from investigations by Dai et al. RI_j is the comprehensive ecological risk index at sampling point *j*.

Considering the accuracy and universality of Hakanson's potential ecological risk assessment results and the toxicity and species of heavy metals, the classification criteria of the ecological risk of soil heavy metals were adjusted. The specific methods are as follows.

(1) The grading value of the unit's toxicity coefficient was determined:

$$\mathrm{RI} = \frac{l_1}{TC} \tag{9}$$

(2) The threshold value of the first level was calculated:

$$L_1 = \sum_{i=1}^5 TC \times RI \tag{10}$$

where l_1 is the first level classification boundary value of Hakanson (150); *TC* is the total value of the toxicity coefficient of the 8 pollutants in the model (133); *RI* is the unit toxicity coefficient classification value (1.13); L_1 is an adjusted first level boundary value; and $\sum_{i=1}^{5} TC$ is the sum of the toxicity coefficients of 5 heavy metals. The most toxic heavy metal in this study was Cd (30),

and it was used as the first level threshold value for the

🖄 Springer

adjusted single factor ecological risk. The total toxic coefficient of the 5 heavy metals was 69, and after adjusting the comprehensive potential ecological risk of the first level, the limit value was 80, which was 2 times the upper limit value. Then, the classification standard of the potential ecological risk of heavy metals in soil was reclassified (Table 3).

Risk visualization and statistical methods

This study calculated the potential ecological risk of heavy metals using the ordinary kriging spatial interpolation method, which provides a visual expression of risk with the help of ArcGIS 10.2 (ESRI Inc.). Combined with the land-use status map of the study area, the proportion of potential ecological risk of heavy metals in each land-use type was calculated.

Results and analysis

Descriptive statistics of heavy metal contents

Descriptive statistics of the 5 heavy metals in the sampling sites in the study area were determined (Table 4). The mean value of each heavy metal was greater than its background value (Dai et al. 2011), indicating that these elements are enriched and that the environment and ecosystem in this area may be at risk. The 5 heavy metals exceeded the standards at a rate above 90% and in the following order Cu > Zn > Cd = As > Pb. Cu exceeded the standard at a rate of 100%, which was the highest among the metals. This result is consistent with the results of the previous research (Li et al.2017) in this area. Cu and Zn were significantly affected by human activities in farmland, and the sampling sites in irrigated land and orchards accounted for 65.52% of the area, which further verified the findings of the previous studies.

The soil heavy metal concentrations in the study region all showed certain variation characteristics, with variation coefficients ranging from 0.14 to 0.47 (Table 4). Cu had the highest variation coefficient, and Cd had the lowest. Dimensionalzing a variation coefficient can better reflect the fluctuations in heavy metal content. A larger coefficient of variation corresponds to a higher degree of variation in the heavy metal elements and a more uneven spatial distribution of the heavy metal content. Therefore, the spatial distribution of Cu

content was the most discrete, and the spatial distribution of Cd was relatively uniform.

Evaluation and analysis of heavy metal pollution

The degree of pollution of each heavy metal was evaluated by the single factor pollution index method and the Nemerow index method. The comprehensive pollution degree of each sample was evaluated by the improved fuzzy comprehensive evaluation method. The results of the index method are shown in Table 5. The pollution index of Cu was the largest, and the Nemerow index reached 7.059. Cu and Pb were classified as occurring at heavy pollution levels with the highest pollution degree among all metals, followed by Zn and As with a pollution index of 0.53~3.35 and an NMO index of 2~3, representing a moderate degree of pollution.

According to the fuzzy comprehensive evaluation method, the toxicity coefficients of the pollutants were weighted based on the calculated pollution weight. Although Cu and Pb presented the highest pollution degrees, the pollution degrees of each sampling point calculated by the fuzzy comprehensive evaluation method were not high because of their low toxicity coefficients (Table 5). Similarly, the pollution degree of Zn was not high, but the toxicity coefficient was large; the pollution degree of the sampling points based on the fuzzy comprehensive evaluation was not low. The results obtained by different methods were different and reasonable. As Table 6 shows, the pollution degree of the sampling points under the different land-use types was obtained by the fuzzy comprehensive evaluation method. Twenty-nine points of moderate pollution were distributed in urban industrial land and mining land, with 18 of the 24 points on mining land having moderate pollution. The urban industrial land and mining land sampling points were all slightly polluted; the pollution levels of the orchard and irrigated land were at the safe and minimal levels; and 8 points in bare land were at the safe level. Overall, there were no severe pollution levels in Longkou city.

Potential ecological risk assessment for single factors

In this study, the potential ecological risk assessment of the 5 heavy metals was evaluated according to the improved Hakanson potential ecological risk assessment criteria. To intuitively express the spatial

Ecological risk	1	2	3	4	5
Level	Slight	Moderate	Severe	Strong	Very strong
Ei	< 30	30-60	60–120	120-240	>=240
RI _i	< 80	80–160	160-320	>= 320	_

Table 3 Improved classification standard for the ecological risk assessment of heavy metals in soil

distribution of the ecological risk level, the evaluation results were interpolated by ordinary kriging. The potential ecological risk values of Cu, Pb, and Zn conformed to a normal distribution and were used directly for variogram fitting and spatial interpolation. As and Cd must be fitted and interpolated after a logarithmic transformation. Table 7 shows the variation function model and parameter statistics of the ecological risk of the 5 heavy metals. Cd conformed to the Gaussian model, Cu and Zn were best fit by the spherical model, and As and Pb were best fit by the exponential model. The effective ranges of the five elements ranged from 4887 to 7593 m; the residual sum of squares were all close to 0; and the determination coefficients were above 0.613. These fitting results satisfied the requirements of model selection.

In terms of ecological risk level, 4 levels of ecological risk were observed in the study area (Table 8, Fig. 2). As, Cu, Pb, and Zn were at the moderate risk and slight risk levels. Zn presented a slight risk, which increased to moderate in the northern part of the study area. Cu also presented a moderate risk level in the northern part of the area and a slight risk in the other areas. Pb presented a moderate risk level over a relatively scattered area but mainly in the eastern and western regions, with a slight risk in the central and northern regions. In contrast, As presented a moderate risk level over a relatively wide area that included the entire eastern and northwestern regions, and only small areas in the northern and southwestern regions were at a minor

 Table 4
 Descriptive statistics of the heavy metals in soil

risk. Cd presented the largest potential ecological risk with severe and strong risk levels in the eastern and northwestern parts of the central region, forming a high-risk zone, and the other areas had a moderate ecological risk, with essentially no areas at a minor risk level.

To study the ecological risk of soil heavy metals in different land-use types, we used ArcGIS 10.2 to extract and analyse the interpolated risk of each heavy metal according to various land-use types. Figure 3 presents the results of the extraction of Cd with the highest potential ecological risk grade, and Table 8 shows the area statistics of each risk grade under each land-use type. Cu, Zn, Pb, and As had a potential ecological risk of grade 1. The land area proportion from large to small was as follows: irrigated land, Cu (98.62%) > Zn(97.55%) > Pb (96.45%) > As (34.62%); orchard, Zn (95.73%) > Cu (95.68%) > Pb (92.48%) > As(49.84%); urban industrial land, Cu (100%) = Zn(100%) > Pb (98.53%) > As (60.35%); mining land, Zn (100%) > Cu (84.09%) > Pb (72.44%) > As (59.19%); and bare land, Cu (99.79%) > Zn (98.07%) > Pb (89.89%) > As (34.88%). As, Cu, Pb, and Zn had only two potential ecological risk levels for all five land-use types. For Cd, which corresponded to more severe pollution, most of the irrigated land, orchard, and bare land presented two levels of moderate risk. Most of the urban industrial land and mining land were at level three or the severe risk grade, and 30.52% of the mining land was at level four or the strong risk grade; thus, these areas

Heavy metal	$\begin{array}{l} \text{Minimum} \\ (\text{mg} \cdot \text{kg}^{-1}) \end{array}$	$\begin{array}{l} Maximum \\ (mg \cdot kg^{-1}) \end{array}$	$\frac{\text{Mean}}{(\text{mg} \cdot \text{kg}^{-1})}$	Variation coefficient	Background $(mg \cdot kg^{-1})$	Exceeding standard rate (%)
Cu	20.34	50.86	35.30	0.47	19.6	100
Pb	15.75	63.92	35.08	0.38	25.4	93.2
Zn	53.99	104.32	77.89	0.18	56.1	95.9
Cd	0.049	0.42	0.20	0.14	0.11	94.7
As	3.63	10.26	7.96	0.19	6.3	94.7

Heavy metal	Single factor pollution index			Nemerow index	Degree of pollution	Background value (mg/kg)	
	Minimum Maximum		Mean				
Cu	0.74	9.87	1.52	7.06	Heavy	19.60	
Pb	0.45	7.70	1.42	5.54	Heavy	25.40	
Zn	0.53	3.35	1.12	2.49	Moderate	56.10	
Cd	1.26	8.06	3.05	6.10	Heavy	0.11	
As	0.72	3.25	1.28	2.47	Moderate	6.30	

 Table 5
 Assessment of soil heavy metal pollution

present a relatively higher potential ecological risk. Bare land, however, was mainly below a level three risk and presented a relatively low degree of ecological risk.

Comprehensive potential ecological risk assessment

The risk of pollution produced by the synergistic effect of the 5 heavy metals was expressed by the comprehensive potential ecological risk index, which reflected the overall ecological risk level of the study area. As shown in Fig. 4, the overall ecological risk level in the study area was essentially above the moderate risk grade; the serious risk grade was mainly distributed in the middle and the southeastern and northwestern parts diagonally; and the low risk areas were scattered in the north, northeast, and southwest of the study area.

According to the type of land use (Table 9, Fig. 5), the area percentage of each class at a minor risk was below 10%, especially for urban industrial land, for which the slight risk levels only accounted for 0.01%. The risk level of irrigated land and bare land did not reach level 1 (slight). The potential comprehensive ecological risk of heavy metals in irrigated land, orchards, mining land, and urban industrial land was mostly at the severe level. The order according to the area occupation ratio was irrigated land (80.53%) > orchard (70.89%) > mining land (61.34%) > urban industrial land (54.17%). However, the comprehensive ecological risk of heavy metals in bare land was mostly at level 2 (moderate). Although there was a strong risk grade for the orchard, the proportion of the area was very small at only 6.97% of that area. The total potential ecological risk of heavy metals at level 4 (strong) for the urban industrial land and mining land was 38.85% and 34.52%, respectively, indicating that these two land-use types presented relatively serious pollution.

Discussion

Soil pollution assessments and ecological risk assessments are two effective methods of exploring the environmental problems caused by soil heavy metals. In this paper, the environmental pollution and ecological risk of five heavy metals in soils with different patterns were evaluated and analysed using these two methods. The following results were observed based on the evaluation and analysis.

(1) Because pollution assessments vary, the evaluation results obtained by the Nemerow index method and improved fuzzy comprehensive evaluation method were also different. The Nemerow index method was used to evaluate the pollution level of each heavy metal in Longkou. Thus, 5 elements were evaluated in this study, with 2 elements

Table 6	Number of sampling	g points with	different pollution	levels for the 5	and-use types
---------	--------------------	---------------	---------------------	------------------	---------------

Degree of pollution	Urban industrial land	Mining land	Orchard	Irrigated land	Bare land	Total
Moderate	11	18	0	0	0	29
Light	10	6	22	31	0	69
Minimal	0	0	24	18	0	42
Safe	0	0	0	0	8	8

 Table 7
 Variation function model of the ecological risk of heavy metals

Heavy metal	Model	Effective range/m	Residual sum of squares	Determination coefficients (R ²)
Cu	Spherical model	7593	0.012	0.912
Pb	Exponential model	6832	0.036	0.613
Zn	Spherical model	6160	0.003	0.675
Cd	Gaussian Model	7494	0.006	0.779
As	Exponential model	4887	0.015	0.738

presenting moderate pollution and 3 elements presenting serious pollution, indicating that the soil is seriously polluted by heavy metals. The fuzzy comprehensive evaluation is a method of evaluating the pollution status of sampling points. According to the results, the highest pollution level was moderate pollution. The results indicate that 29 of 148 points presented moderate pollution, 69 sampling points presented light pollution, and the remaining points were at the minimal and safe levels. Therefore, soil heavy metal pollution throughout the whole area is not serious. As a result, the degree of pollution varies based on the subject of analysis. Therefore, when evaluating soil environmental pollution, a variety of methods should be used to

Table 8 Single factor potential ecological risk grade area ratios of soil heavy metals for the 5 land-use types

Heavy metal	Land type	Level 1 (slight)		Level 2 (moderate)		Level 3 (severe)		Level 4 (strong)	
		Area	Percentage	Area	Percentage	Area	Percentage	Area	Percentage
Cu	Irrigated land	113.75	98.62%	1.59	1.38%	_	_	_	_
cu	Orchard	117.60	95.68%	5.30	4.32%	-	-	_	-
	Urban industrial land	14.77		_	-	_	-	-	-
	Mining land	1.94	84.09%	0.37	15.91%	_	-	-	-
	Bare land	0.92	99.79%	0.002	0.21%	_	-	-	-
Pb	Irrigated land	111.27	96.45%	4.08	3.55%	_	-	-	-
	Orchard	113.66	92.48%	9.24	7.52%	_	-	-	-
	Urban industrial land	14.55	98.53%	0.22	1.47%	-	-	-	-
	Mining land	1.67	72.44%	0.63	27.56%	_	-	-	-
	Bare land	0.83	89.89%	0.09	10.10%	_	-	-	-
Zn	Irrigated land	112.53	97.55%	2.82	2.45%	_	-	-	-
	Orchard	117.66	95.73%	5.24	4.27%	_	-	-	-
	Urban industrial land	14.77	100%	-	-	-	_	-	-
	Mining land	2.31	100%	_	-	_	-	-	-
	Bare land	0.91	98.07%	0.02	1.93%	_	-	-	-
Cd	Irrigated land	16.56	14.35%	84.81	84.81%				
	Orchard	25.49	20.72%	81.62	66.35%				
	Urban industrial land	0.37	37.15%	4.09	27.25%	13.72	11.89	0.26	0.23%
	Mining land	0.06	2.49%	0.15	6.42%	15.36	12.49%	0.53	0.43%
	Bare land	0.14	14.67%	0.78	85.32%	7.31	49.51%	2.98	20.20%
As	Irrigated land	39.94	34.62%	75.41	65.38%	1.40	60.56%	0.70	30.52%
	Orchard	61.26	49.84%	61.64	50.16%	4.72E-05	0.51%	_	_
	Urban industrial land	8.91	60.35%	5.85	39.65%	-	-	_	-
	Mining land	1.36	59.19%	0.94	40.81%	-	-	_	-
	Bare land	0.32	34.88%	0.60	65.11%	_	_	-	_



Fig. 2 Evaluation of the potential ecological risk classes for the 5 heavy metals

ensure that a comprehensive evaluation from multiple perspectives is performed.

(2) The ecological risk of single factors was small, and the comprehensive ecological risk was high. Although the ecological risk of a single heavy metal to the soil environment was low, the comprehensive potential ecological risk represented the sum of multiple elements and single factor ecological risk. As the number of heavy metal species increased, the potential ecological risk to the soil in the study area increased. The four heavy metals, Cu, Zn, Pb, and As, were all below the moderate risk level, and their spatial distribution area was less than 20%. After considering the comprehensive risk of these metals, the total ecological risk was primarily higher than

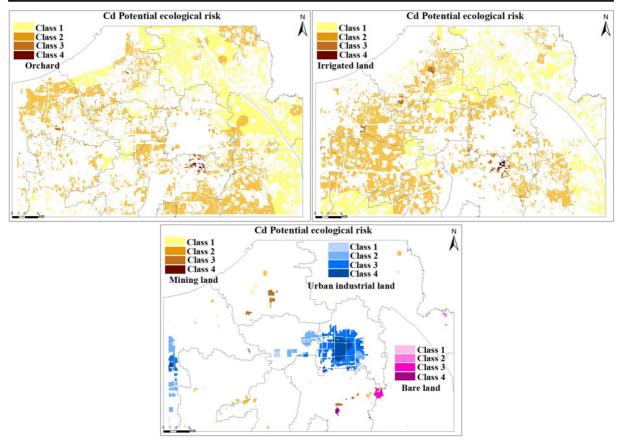


Fig. 3 Potential ecological risk levels of Cd for various land-use types

severe. In this paper, only five heavy metals were evaluated by the Hakanson potential ecological risk index. The comprehensive potential ecological risk index is not a single factor ecological index of all heavy metals and can only partly reflect the ecological risk of the study area; thus, further studies are warranted.

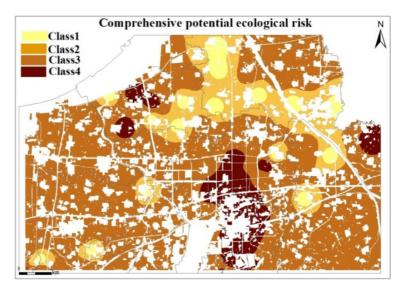


Fig. 4 Comprehensive potential ecological risk assessment

Heavy metal	Level 1 (slight)		Level 2 (moderate)		Level 3 (severe)		Level 4 (strong)	
	Area	Percentage	Area	Percentage	Area	Percentage	Area	Percentage
Irrigated land	1.16	1.00%	21.30	18.47%	92.89	80.53%	_	_
Orchard	1.94	1.57%	25.27	20.56%	87.13	70.89%	8.57	6.97%
Urban Industrial land	0.0028	0.01%	1.03	6.95%	8.00	54.17%	5.74	38.85%
Mining land	0.056	2.41%	0.040	1.72%	1.42	61.34%	0.80	34.52%
Bare land	0.05	5.53%	0.59	64.61%	0.28	29.85%	—	-

Table 9 Comprehensive potential ecological risk level area ratios of heavy metals for the 5 land-use types

(3) Although a heavy metal may present a serious pollution risk, the single factor ecological risks were not high. In the study, the Nemerow index indicated that Cu had a heavy pollution level, whereas the ecological risk caused by Cu was moderate in a small part of the northern part of the study area and was slight in other areas. Thus, although, individually, the heavy metals caused a certain degree of pollution in the soil environment of the study area, they have not yet caused changes to the soil ecology.

(4) For the 5 land-use types, the single factor and the comprehensive ecological risk were also high. The pollution level of urban industrial land and mining land was above moderate, and the spatial distribution of the ecological risk above severe accounted for more than 90% of the area. The comparison of heavy metal pollution and ecological risk

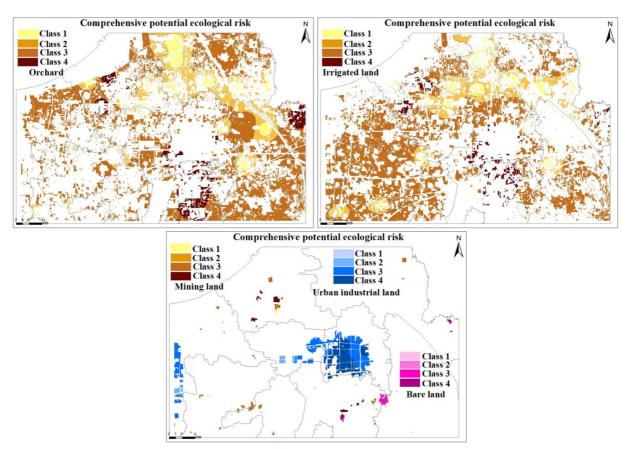


Fig. 5 Comprehensive potential ecological risk class for each land-use type

assessment for different land-use types can be used as the basis for soil pollution source analysis to better understand heavy metal enrichment and influencing factors.

Conclusions

The contents of Cu, Pb, Zn, Cd, and As in Longkou all exceeded the background values of the area, and the proportion that the standard was exceeded was over 90%, with Cu exceeding the standard at a rate of 100%. Based on the index method, each element had a certain degree of pollution, and the pollution index of Cu was the largest, followed by that of Pb and Cd. Thus, Cu, Pb, and Cd all had severe pollution levels, while As and Zn were classified at moderate pollution levels. The results of the fuzzy comprehensive evaluation showed that the degree of pollution at the sampling sites corresponding to different land uses was different. Urban industrial land and mining land were mainly characterized by moderate pollution, and irrigated land was mainly characterized by light pollution; thus, the pollution levels for these land-use types were above the minimal level. Orchard had relatively less pollution, with most of the land at the minimal level, whereas bare soil was almost free of heavy metal pollution.

Cd had the most serious impact on the soil environment, and the ecological risk to urban industrial land and mining land was especially high and categorized at strong and severe levels. The other land-use types were at a moderate level of risk. The potential ecological risks of Pb, Zn, Cd, and As were below the moderate level. The comprehensive potential ecological risk of heavy metals indicates that the study area mainly corresponded to a severe pollution level. Irrigated land, orchard, mining land, and urban industrial land accounted for more than 50% of the total potential ecological risk and severe pollution areas. Urban industrial land and mining land presented significant potential ecological risks from heavy metals in the soil. Thus, these areas should receive additional attention.

Funding information This study was supported by the National Natural Science Foundation of China (No. 41371395 and 41601549) and the Key Technology Research and Development Program of Shandong (2019GSF109034).

This work was also financed by the National Natural Science Foundation of China (41701206), the Natural Science Foundation of Shandong Province (ZR2017BD011), and the China Postdoctoral Science Foundation (2017 M622256).

References

- Amirian Chakan, A., Taghizadeh-Mehrjardi, R., Kerry, R., & Kumar, S. (2017). Spatial 3D distribution of soil organic carbon under different land use types. *Environmental Monitoring and Assessment, 189*, 131.
- An, J., Gong, X., Chen, H., & Wei, S. (2016). Temporal and spatial characteristics and health risk assessments of heavy metal pollution in soils of Shenfu irrigation area. *Journal of Agro Environment Science*, 35(1), 37–44.
- Bartkowiak, A., Lemanowicz, J., & Hulisz, P. (2017). Ecological risk assessment of heavy metals in salt-affected soils in the Natura 2000 area (Ciechocinek, north-central Poland). *Environmental Science and Pollution Research International*, 24(35), 27175–27187.
- Boquete, M. T., Aboal, J. R., Carballeira, A., & Fernández, J. A. (2014). Effect of age on the heavy metal concentration in segments of Pseudoscleropodium purum and the biomonitoring of atmospheric deposition of metals. *Atmospheric Environment*, 86, 28–34.
- Chen, Y., Jiang, X., Yong, W., & Zhuang, D. (2018). Assessment of ecological environment and human health of heavy metals in mining area based on GIS. *Acta Scientiae Circumstantiae*, 04, 1642–1652.
- Dai J, Pang X, Yu C, Wang C, Wang Z, Hu 2011. Geochemical baselines and background values and element enrichment characteristics in soils in eastern Shandong Province. Geochimica, 40(6):577–587.
- Dou, L., Zhou, Y., & Wang, X. (2007). Improvement and application of a fuzzy mathematical model for assessment of heavy metal pollution in soil. *Chinese Journal of Soil Science*, 38(1), 101–105.
- Fernández, J., & Carballeira, A. (2001). Evaluation of contamination, by different elements, in terrestrial mosses. Archives of Environmental Contamination and Toxicology, 40(4), 461– 468.
- Ghrefat, H. A., Yusuf, N., Jamarh, A., & Nazzal, J. (2012). Fractionation and risk assessment of heavy metals in soil samples collected along Zerqa River, Jordan. *Environmental Earth Sciences*, 66(1), 199–208.
- Hou, Q., Ma, J., Wang, X., & Duan, H. (2011). Bioavailability and potential ecological risk of soil heavy metals in kindergartens, Kaifeng City. *Environmental Sciences*, 32(6), 1764– 1771.
- Li, C., Wang, F., Cao, W., et al. (2017). Source analysis, spatial distribution and pollution assessment of heavy metals in sewage irrigation area farmland soils of Longkou City. *Environmental Sciences*, *38*(3), 1018–1026.
- Liang, L., Yu, S., Zhang, C., et al. (2017). Spatial distribution and ecological risk assessment of heavy metals in Yongkang City. *Journal of Zhejiang A & F University*, 34(6), 972–982.
- Liang, B., Nie, X., Yang, D., et al. (2018). Differences in soil physical properties of 5 typical land use types in downstream of the Niyang River in Tibet. *Journal of Northwest A&F University (Nat. Sci. Ed)*, 01, 1–9.

- Lu, S., Wang, Y., & He, L. (2014). Heavy metal pollution and ecological risk assessment of the paddy soils around a Pb-Zn mine in Huize country. *Ecology and Environmental Sciences*, 23(11), 1832–1838.
- Luo, Y., Bian, Q., Li, Y., et al. (2016). Evaluation of heavy metal contamination and its potential ecological risk to the soil in a river sewage district of Mianyang. *Chemical research and application*, 28(6), 783–788.
- Luo, C., Bi, J., Xiao, G., & Zhang, F. (2017). Pollution characteristics and assessment of heavy metals in soil of different industry zones of Ningdong Base in Ningxia, China. *Ecology and Environmental Sciences*, 26(7), 1221–1227.
- Lv, J., Bi, C., Chen, Z., et al. (2012). Distribution and ecological risk assessment of polycyclic aromatic hydrocarbons in agricultural soil of the Chongming Island in Shanghai. *Environmental Sciences*, 33(12), 4270–4275.
- Lv, J., Xia, Q., Yan, T., Zhang, M., et al. (2018). Identifying the sources, spatial distributions, and pollution status of heavy metals in soils from the southern coast of Laizhou bay, eastern China. *Human and Ecological Risk Assessment: An International Journal*, 1–15.
- Ma, J., Wang, X., Hou, Q., & Duan, H. (2011a). Pollution and potential ecological risk of heavy metals in surface dust on urban kindergartens. *Geographical Research*, 30(3), 486– 495.
- Ma, T., Zhao, D., Zeng, J., Yan, W., Jiang, C., & Ding, W. (2011b). Potential ecological risk assessment of heavy metal pollutants in surface sediments of the lakes in Nanjing. *Journal of Ecology and Rural Environment*, 27(6), 37–42.
- Mineev VG 1996. Accumulation and transformation of heavy metals HM within the 'soil-plants' system inprolonged agrochemical trials. Fertilizers and environment: Proceedings of the international symposium 'fertilizers and environment', held in Salamanca, Spain, 26–29, September, 66 :539–540.
- Ministry of ecology and environment of the People's Republic of China. (2014). Technical guidelines for site soil remediation. *HJ*, *25*, 4.
- Ministry of environmental protection of the People's Republic of China. (2014). Technical guidelines for risk assessment of contaminated sites. *HJ*, 25, 3.
- Ministry of land and resources of the people's republic of China (2006). The specification of testing quality management for geological laboratories, DZ0130.
- Ministry of land and resources of the people's republic of China (2016). Geochemistry evaluation standard for land quality, DZ/T 0295.
- Mohammed, A. S., Kapri, A., & Goel, R. (2011). Heavy metal pollution: Source, impact, and remedies. *Biomanagement of metal-contaminated soils*, 20, 1–28.

- Oleszek, W., Terelak, H., Maliszewska-Kordybach, B., et al. (2003). Soil, food and agroproduct contamination monitoring in Poland. *Polish Journal of Environmental Studies*, *12*(3), 261–268.
- Qishlaqi, A., Moore, F., & Forghani, G. (2009). Characterization of metal pollution in soils under two land use patterns in the Angouran region, NW Iran; a study based on multivariate data analysis. *Hazardous Materials*, *172*(1), 374–384.
- Shang, J., Luo, W., Wu, G., et al. (2015). Spatial distribution of se in soils from different land use types and its influencing factors within the Yanghe watershed, China. *Environmental Sciences*, 36(1), 301–308.
- Shougrakpam, S., Sarkar, R., & Dutta, S. (2010). An experimental investigation to characterise soil macroporosity under different land use and land covers of northeast India. *Journal of Earth System Science*, 119(5), 655–674.
- Tang, Y., & Han, G. (2017). Characteristics of heavy metals in soils under different land use in a typical karst area, Southwest China. Acta Geochim, 36(3), 515–518.
- Wei, Y., Zhou, C., Wang, J., Fan, Z., & Liu, G. (2017). Distribution and ecological risk assessment of 6 typical trace elements in mining. *Journal of university of science and technology of China*, 47(5), 413–420.
- Zhang, Y., Wu, F., Zhang, X., & Cao, N. (2017). Pollution characteristics and ecological risk assessment of heavy metals in three land use types on the southern loess plateau, China. *Environmental Monitoring and Assessment*, 189(9), 470.
- Zhang, Z., Anwar, M., & Zibibula, S. (2019). Pollution assessment and health risks evaluation of (metalloid) heavy metals in urban street dust of 58 cities in China. *Environmental Science* and Pollution Research, 26(1), 126–140.
- Zhao, X., Huang, Y., Li, J., et al. (2015). Environmental levels, spatial distribution, sources and potential ecological risk of heavy metals in soils surrounding a large solid waste incinerator. *Ecology and Environmental Sciences*, 24(6), 1013– 1021.
- Zheng, Y., Chen, T., Chen, H., et al. (2005). Lead accumulation in soils under different land use types in Beijing City. Acta Geograhica Sinica, 60(5), 791–797.
- Zou, J., Liu, X., Dai, W., & Luan, Y. (2018). Pollution assessment of heavy metal accumulation in the farmland soils of Beijing's suburbs. *Environmental Science and Pollution Research*, 1–10.

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.