

Original Research Paper

Pollution Characteristics and Sources Analysis of Soil Heavy Metal in Taoyuan Coal Mine of Suzhou City

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Abstract: Soil is one of the most important resources and an important part of an ecosystem in the development of human survival. The purposes of this study are to investigate concentration characteristics and source analysis of heavy metals in the Taoyuan coal mine of Suzhou City. 30 soil samples were collected and characterized for metal concentration (Cd, Cr, Cu, Pb, Zn, and As), then the heavy metal pollution was evaluated by using the pollution load index and geoaccumulation index and the sources of heavy metals were analyzed. The results revealed that the mean concentrations, except Cr, were considerably higher than the reference values, especially Cd, Cu Pb, and Zn in study area soil, than in non-mining soil in Suzhou. The results of the geoaccumulation index (I_{geo}) showed the following sequence: Cd>Cu>Pb>Zn>As>Cr. At the same time, Cr and As of all samples were not contaminated, Zn and Pb were non-pollution to light pollution, and Cd and Cu were light pollutions to moderate pollution. The potential ecological risk (E_i) values of Cd had strong risk pollution and the other five metals belonged to a light ecological risk, the potential ecological risk (RI) mean implied moderate pollution for samples in the study area. The correlation analysis declared that the first principal component is dominated by Cd, Zn, and Pb and marked correlation indicates that they should be related to coal mining, smelting processing, weathering, leaching, and coal transportation. The second principal component was occupied by Cr, Cu, and As, the source of Cu may be affected by Zn and Pb, and Cr and As can be regarded as from the soil parent materials. It was believed that the heavy metals in the soil of the Taoyuan Coal Mine were polluted to a certain extent and the accumulation of Cd, Zn, and Pb were mainly affected by mining activities.

Keywords: Heavy Metal, Pollution Load Index, Geoaccumulation Index, Potential Ecological Risk, Suzhou City

Introduction

With the rapid advance of urbanization and industrialization, soil quality has become one of the core environmental issues. Soil is an important part of an ecosystem and the carrier of various pollutants, which contains specific environmental information for a good indicator of environmental quality (Jiang *et al.*, 2017; Hou *et al.*, 2019). As an important pollutant, heavy metals have attracted attention because of their characteristics of nondegradability, ability to easy enrichment, strong toxicity, and latency for long period in the soil environment (Chen *et al.*, 2018). Once entering the soil, they will migrate and transform with the changes of environmental media and then harm human health

through the food chain. Along with the development of the social economy and the gradual improvement of residents' living standards, people's demands on environmental quality, especially food safety, are also becoming higher and higher. So it is of great social value to understand the level of soil heavy metal content, pollution characteristics, and the risks to human health and the ecological environment caused by soil heavy metal (Xiong *et al.*, 2017). Therefore, soil heavy metal pollution is becoming one of the hot research issues. To date, various studies have demonstrated that the soil environment in different regions is contaminated to various degrees by heavy metals. The main contents of the

current studies involve the spatial distribution characteristics (An *et al.*, 2016), sources (Keshavarzi and Kumar, 2020), geochemical baseline (Cheng *et al.*, 2014; Li *et al.*, 2015), morphology and bioactivity (Zhao and Wang, 2020), transformation (Cong *et al.*, 2017) of heavy metals in soil. Over the past decades, coal mining played an important role in social-economic development and human activities. At the same time, coal resources have also brought heavy metal pollution in the process of promoting economic development because it makes minerals and wastes originally buried in the ground expose and cause heavy metal pollution in the soil due to weathering, transportation, leaching, and infiltration (Li *et al.*, 2010). For example, Li *et al.* (2010) found that heavy metal contents have obvious accumulation in gangue dump, industrial square, coal transfer station, transportation lines, and other surrounding farmland of coal mine area in southwest Shandong Province; showed that soil was seriously polluted by Hg and Cd in coal mine area of Guizhou Province; Cong *et al.* (2017) reported that the contents of heavy metal in soil decreased with the increase of distance from gangue dump in Haizhou coal mine. These studies provide theoretical and methodological support for the restoration of the regional ecological environment, but there are few studies on Suzhou City. Suzhou City is an important coal production base in Anhui Province even the China, the coal industry occupies an important position in the development of a national economy. In the process of accelerating the strategic rise of Northern Anhui Province, ecological environment problems brought by the coal industry are increasingly prominent, so the study of soil heavy metal pollution is of great significance for the sustainable development of the regional economy and the protection of the ecological environment in coal area of Suzhou City. Thus, soil samples were collected from the Taoyuan mine in Suzhou City to determine the concentration of heavy metals (Cd, Cr, Cu, Pb, Zn, and As) and assessed heavy metal pollution and enrichment using geoaccumulation index, pollution load index, and potential ecological risk method in this study and analyzed the sources of heavy metals.

Study Area

Suzhou City lies at a latitude of 33°18'~34°38'N and a longitude of 116°09'~118°10'E in the northeast of Anhui Province, China and covers an area of 9787 km² with a population of 65.656 billion people in 2018. The climate is a warm temperate sub-humid monsoon climate with a mean annual precipitation of about 850 mm and a mean annual temperature of about 14 ~ 14.5°C, cold winter and hot summer, and four distinct seasons. The plain area accounts for 90% of the total area, the north is dominated by yellow tide soil, while the south is mainly black mortar soil, the agricultural production is developed and it is a famous food production base and fruit production base. Suzhou coal mine, which is an important part of the Lianghui coal mine, lies at

the southern end of the Su-xu structure and belongs to the diassic coal measure strata, coal storage area of about 2000 km² and the predicted reserves of 60 × 10⁸t, is one of the 13 large coal bases planned by the state.

Materials and Methods

Sample Collection and Analysis

For the present investigation, a total of 30 topsoil samples (0-20 cm) were collected in November 2017 in Taoyuan coal mine of Suzhou around 100 ~ 200 m distance. About 1 kg of soil obtained by the four-part method at each sampling site was taken back to the laboratory. After soils were air-dried in the laboratory, removing stones and plant impurities, the samples were ground into powder and passed through a 2 and 0.149 mm nylon sieve for measuring.

For measurement of total metal concentrations, each soil sample weighed 0.5 g was placed digester and digested in a microwave accelerated reaction system (Bergh of MWS-3) at 220°C for 20 min with concentrated HNO₃, HCl, H₂SO₄, and HClO₄ until all solids are dissolved. After cooling, deionized water was added to bring the final volume to 50 mL. Cd and Pb were determined by atomic absorption spectrophotometer (TAS-990 FG), Zn, Cr, and Cu were determined by the flame method of atomic absorption spectrophotometer (TAS-990 FG), and As was determined by atomic fluorescence photometer (pf 6-3). To ensure the accuracy of the measurement results during these measurements, quality control shall be carried out according to the standard soil reference materials (GBW 07403 and GBW 07404, National Standard Detection Research Center, Beijing, China). The way to do that is two blank samples should be added for each batch of samples and recalibrated for every 10 samples.

Assessment Methods

Pollution Load Index (PLI) Method

The pollution load index method is a useful tool to assess the degree of enrichment of heavy metals, which contains a variety of heavy metal elements and each heavy metal is quantitatively evaluated to judge its contribution to environmental pollution (Pang *et al.*, 2014; Tomlinson *et al.*, 1980). Determination of *PLI* involves the calculation of the concentration factor obtained by dividing the measured concentration of an element by the background concentration of the same element in shale (Ali *et al.*, 2015). The calculation formula is as follows:

$$PI = C_n / B_n \quad (1)$$

$$PLI = \sqrt[n]{PI_1 \times PI_2 \times PI_3 \dots PI_n} \quad (2)$$

where C_n denotes the measured concentration of heavy metal in the sample, B_n denotes the environmental

geochemical background value of the corresponding heavy metal, soil background value of non-mining soil in Suzhou City was selected as the reference standard in this study. The *PI* denotes a single factor index, which is the basis of another environmental quality index, environmental quality classification, and comprehensive evaluation. The *PI* is classified as When $PI < 1$ light pollution; $1 \leq PI < 2$ moderate pollutions; $2 \leq PI < 5$ serious pollution; $PI \geq 5$ extreme pollutions. The *PLI* denotes the pollution load index, pollution will exist when the *PLI* is more than one. If $PLI < 1$, non-pollution; $1 \leq PLI < 2$, light pollution; $2 \leq PLI < 3$, moderate pollution; $PLI \geq 3$, high pollution.

Geoaccumulation Index (I_{geo}) Method

The geoaccumulation index I_{geo} a method that was originally proposed by Muller (1979) is to assess enrichment degree by heavy metal using Eq. 3 since 1969. Compared with other methods, this method takes into account not only the environmental geochemical background value and the impact of human activities on the environment but also the changes in the environmental background value that may be caused by natural geological processes (Zhang *et al.*, 2018).

$$I_{geo} = \log_2 \left[\left(\frac{C_n}{k B_n} \right) \right] \quad (3)$$

where I_{geo} is the geoaccumulation index, the pollution classification standard is listed in Table 1. C_n denotes the measured concentration of heavy metal in the sample, B_n denotes the environmental geochemical background value of the corresponding heavy metal, soil background value of non-mining soil in Suzhou City was selected as the reference standard in this study and 1.5 is the background matrix correction factor due to lithogenic effects.

Potential Ecological Risk Method

The potential ecological risk method was developed by Hakanson (1980) to assess heavy metal pollution in the sediments 1980, which benefits from the inclusion of a toxic response factor and relative enrichment factor for a given substance (Liu *et al.*, 2020). The potential ecological risk index can be calculated by the following formula:

$$RI = \sum_{i=1}^n E_r^i = \sum_{i=1}^n T_r^i \cdot PI = \sum_{i=1}^n T_r^i \cdot C_i / C_{i0} \quad (4)$$

where the *RI* is composite potential ecological risk index and E_r^i is single potential ecological risk index, according to Hakanson, the grading standards E_r and *RI* values are shown in Table 2; T_r^i denotes the toxic-response factor (i.e., Cd = 30, Pb = Cu = 5; Cr = 2; As = 10; and Zn = 1).

Statistical analysis of data was carried out in SPSS and all graphs were made in Sigma plot.

Results and Analysis

Concentrations of Heavy Metals Analysis

The descriptive statistics of the total Cd, Cr, Cu, Zn, Pb, and As concentrations in the Taoyuan mine, as well as background values in Suzhou non-mining soil, are presented in Table 3. The concentrations of Cd, Cr, Cu, Zn, Pb and As varied from 0.66~7.46, 70.32~239.67, 55.76~129.75, 23.47~54.20, 109.18~916.15, 13.28 ~ 19.76 mg/kg and the mean concentrations were 1.11, 123.12, 90.44, 28.95, 169.42 and 16.94 mg/kg, respectively. The mean concentration of Cr was lower than the reference value in Suzhou non-mining soil, whereas those of Cd, Cu, Zn, Pb, and As were considerably more than the reference values in Suzhou non-mining soil, especially Cd, Cu, Pb, and Zn, which were 5.82, 5.39, 3.14 and 2.75 times higher than those in Suzhou non-mining soil, respectively. Thus, the pollution of heavy metals of topsoil in the Taoyuan mine presented a significant enrichment of heavy metals by Cd, Cu Pb, and Zn. Variation Coefficient (CV) can reflect sample data differences in spatial distribution and the discrimination of heavy metal content by human activities (Zhang *et al.*, 2016). The variation coefficient of Cd (88.19%) and Cu (84.40%) in soils was relatively higher than the other heavy metals. This high variation coefficient illustrated wide variations of heavy metals, which suggested that they were more likely to be affected by anthropogenic activities in the study area.

Pollution Load Index of Heavy Metal

The results of *PI* and *PLI* were calculated and presented in Fig. 1 and 2. It can be seen from Fig. 1 and 2 that the *PI* values of Cd, Cr, Cu, Pb, Zn, and As calculated were observed in the range of 3.46 to 7.81, 0.42 to 1.44, 3.32 to 7.73, 2.55 to 5.89, 1.77 to 3.51 and 0.90 to 1.34 with the mean 4.76, 0.74, 5.39, 3.14, 2.37 and 1.15, respectively. The order of the single factor of heavy metals was Cu > Cd > Pb > Zn > As > Cr. Among them, the *PI* values of As were almost always less than 1, which illustrates soil for light pollution and the *PI* values of most Cr were slightly greater than 1, indicating moderate pollution. For Zn, 70% of soil sampling sites belonged to the serious study area. However, the pollution of Cu, Cd, and Pb is relatively serious, especially since all the indexes of Cu and Cd are greater than 3, which there were 26.67 and 63.33% of the samples to be extremely serious, respectively. The *PLI* values varied from 1.82 to 3.12, revealing that nearly all samples of the study area were light to moderate pollution by heavy metals. And the mean of *PLI* is 2.32 for moderate pollution.

Geoaccumulation Index of Heavy Metal

The I_{geo} results of heavy metals in the study area were presented in Fig. 3. The calculated values told us the

means of heavy metals were 1.63, -1.07, 1.81, 1.05, 0.64, and -0.39, respectively, indicating the following sequence: Cd>Cu>Pb>Zn>As>Cr. Fig. 3 is revealed that Cr and As elements of all soil samples in the study area were not contaminated with I_{geo} values less than zero. All the samples could be considered as non-pollution to light pollution for Zn with I_{geo} values varying from 0 to 1 and 10% Zn was light pollution. For Pb, there were about 50% of them non-pollution to light pollution and light pollution, respectively. The most serious polluted elements were Cd and Cu, all the soil samples of Cd and Cu were greater than lightly polluted, 13 Cd and 27% Cu were light pollutions to moderate pollution, respectively.

Potential Ecological Risk of Heavy Metal

Table 4 shows the potential ecological risk factors (E_i) of different heavy metals in the surface soils analyzed in this study. The results calculated using equation 4 decreased in an order of Cd > Cu > Pb > Zn > As > Cr. Cd with a mean of 142.91 had a strong risk of pollution and the other five metals means were all less than 40 had a light ecological risk, which declared that Cd with a high contribution rate in ecological risk is the main element of heavy metal pollution in Taoyuan mine soil. The values of RI were found to be high and varied between 168.42 and 322.85 (about 97% of samples in study area less than 300) with an average value of 210.40, indicating that the potential ecological Risk (RI) was in moderate pollution status. From the sampling data, the points with large RI values are mostly distributed in the areas close to the coal mining area, indicating mining, processing, and transportation of coal have a great influence on soil heavy metals in the Taoyuan coal mining area.

Source Analysis of Heavy Metals

The descriptive analysis of heavy metal contents shows that, except Cr, the concentrations of the other five heavy metals are higher than the reference values of non-mining soil and the high variation coefficient illustrated a wide variety of heavy metals for Cd (88.19%) and Cu (84.40%), which declared that soil in the study area was disturbed to varying degrees by human activity. To further analyze the source of soil heavy metals in Taoyuan coal mining, a statistical analysis of the sample data was conducted and calculated the Pearson correlation coefficient of elements using SPSS 21 statistical software. The Pearson correlation coefficient of heavy metals and p -value for statistical hypothesis testing are shown in Table 5. The principal component analysis is a powerful tool for driving mechanisms, source analysis, pattern recognition, and assessment of other data. This method can simplify the research problem by linear fitting a large amount of data interacting together and concentrating the information on several principal factors. Heavy metal data in this study were analyzed using principal component analysis. At the same time, a multi-factor analysis was performed and two principal component factors were extracted according to the principle that the characteristic root was greater than 1 (Fig. 4). As observed from Table 5 and Fig. 4, the contribution of the first and second principal component factors of the total variance are 50.34 and 19.20%, respectively and the accumulative variance contribution rate reached 69.54%. The two principal components occupy most of the data information, which indicates that the sample data in the study area can be analyzed by factor analysis.

Table 1: Pollution classification standard of geoaccumulation index

Ranks	Range	Pollution level
0	$I_{geo} \leq 0$	Non-pollution
1	$0 < I_{geo} \leq 1$	Non to light pollution
2	$1 < I_{geo} \leq 2$	Light pollution
3	$2 < I_{geo} \leq 3$	Light to moderate pollution
4	$3 < I_{geo} \leq 4$	Moderate pollution
5	$4 < I_{geo} \leq 5$	Moderate to strong pollution
6	$I_{geo} > 5$	Strong pollution

Table 2: Classification of potential ecological risk of soil heavy metal

	Light pollution	Moderate pollution	Strong pollution	Serious pollution	Extreme pollution
(E_i)	$E_i < 40$	$40 \leq E_i < 80$	$80 \leq E_i < 160$	$160 \leq E_i < 320$	$E_i \geq 320$
(RI)	$RI < 150$	$150 \leq RI < 300$	$300 \leq RI < 600$	$RI \geq 600$	

Table 3: Descriptive statistics of heavy metal concentrations in the study area soils unit: mg/kg

Element	Minimum	Maximum	Mean	SD	CV/%	Reference value
Cd	0.66	7.46	1.11	0.97	88.19	0.19
Cr	70.32	239.67	123.12	34.79	28.26	166.50
Cu	55.76	129.75	90.44	19.63	21.70	16.78
Pb	23.47	54.20	28.95	5.44	18.79	9.21
Zn	109.18	916.15	169.42	142.99	84.40	61.68
As	13.28	19.76	16.94	1.54	9.09	14.71

Table 4: Potential ecological risk indexes of heavy metals in topsoil sampling sites

Samples	E_{Cd}	E_{Cr}	E_{Cu}	E_{Pb}	E_{Zn}	E_{As}	RI	Samples	E_{Cd}	E_{Cr}	E_{Cu}	E_{Pb}	E_{Zn}	E_{As}	RI
1	125.37	1.35	18.85	14.60	8.85	12.65	181.67	16	138.95	1.16	27.17	15.68	10.43	12.79	206.18
2	111.47	1.49	21.78	16.05	9.74	12.14	172.67	17	168.63	1.62	28.74	17.06	12.89	12.21	241.15
3	123.79	1.06	17.68	13.29	10.79	10.94	177.56	18	129.79	1.44	25.92	15.04	11.02	11.27	194.49
4	130.42	1.10	16.62	14.36	8.97	10.80	182.26	19	129.16	1.66	25.72	16.40	12.79	10.85	196.58
5	129.32	0.84	18.15	13.64	9.28	10.75	181.98	20	158.53	1.46	29.57	17.83	12.24	10.65	230.28
6	103.89	1.62	27.50	12.74	9.66	13.00	168.42	21	116.53	2.24	28.74	16.80	11.62	10.50	186.43
7	133.89	1.02	21.79	14.77	9.77	12.68	193.92	22	134.53	1.46	29.98	15.93	12.84	9.03	203.76
8	145.42	0.90	23.41	14.63	11.64	12.15	208.16	23	126.63	1.81	29.22	14.39	11.90	11.11	195.07
9	143.21	1.09	35.97	16.07	11.21	11.09	218.65	24	141.16	1.67	29.14	16.85	12.13	9.74	210.69
10	234.32	1.12	24.20	14.67	10.23	10.59	295.12	25	107.53	1.77	29.83	13.80	12.43	11.75	177.10
11	150.47	1.01	22.86	15.83	12.57	13.43	216.17	26	203.21	1.67	32.75	15.83	13.50	12.30	279.27
12	147.00	2.88	22.11	13.06	9.80	12.37	207.22	27	230.68	1.61	32.45	29.43	17.53	11.16	322.85
13	107.68	1.87	34.30	13.93	16.23	12.13	186.15	28	152.68	1.69	34.12	16.07	14.84	11.81	231.21
14	120.63	1.57	23.01	14.69	10.90	11.24	182.04	29	197.37	1.44	35.67	18.73	14.79	10.36	278.36
15	122.21	1.52	22.55	13.82	9.73	10.86	180.69	30	122.68	1.22	38.66	15.50	14.95	13.00	206.03
Mean	142.91	1.48	26.95	15.72	11.84	11.51	210.40								

Table 5: Correlation of elements in Taoyuan coal mining of Suzhou City

Element	Cd	Cr	Cu	Pb	Zn	As
Cd	1					
Cr	0.04	1				
Cu	0.204	0.241	1			
Pb	0.897	0.039	0.387	1		
Zn	0.979	0.089	0.306	0.902	1	
As	-0.085	-0.045	-0.035	-0.195	-0.067	1

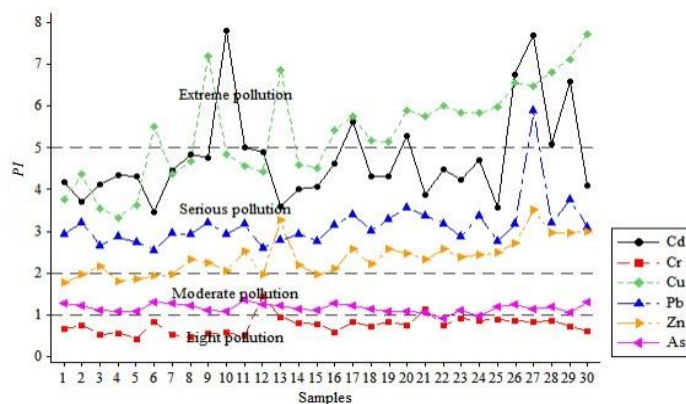


Fig. 1: PI index of heavy metals

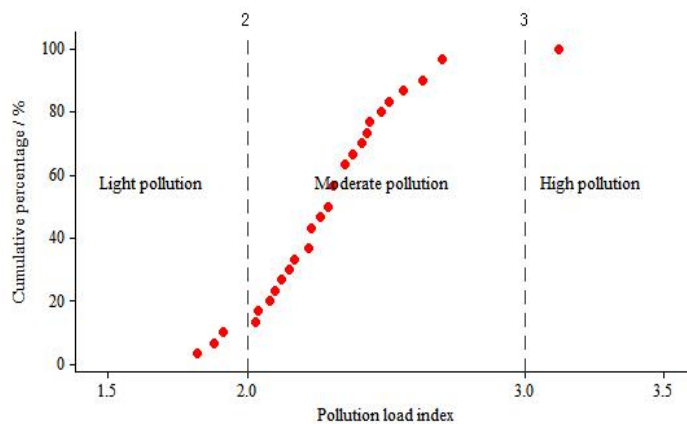


Fig. 2: Pollution load index of heavy metal

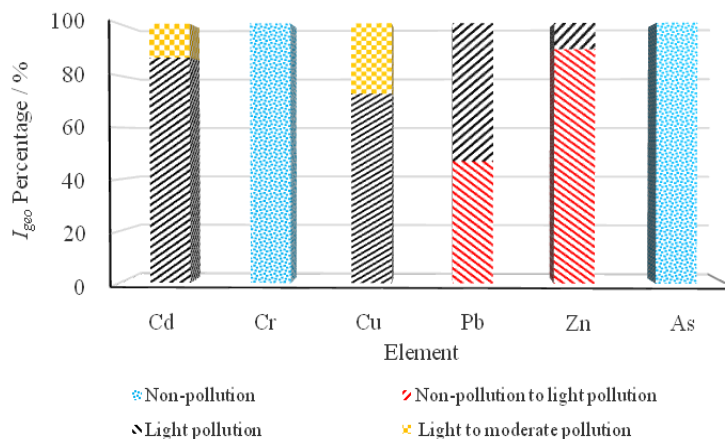


Fig. 3: Cumulative percentage of geoaccumulation index of heavy metal

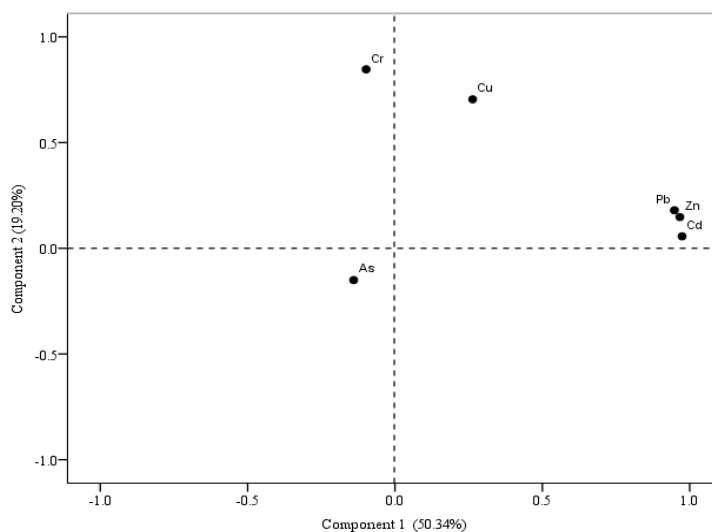


Fig. 4: Load plots of principal component factors in Taoyuan coal mining of Suzhou City

Cd (0.952), Zn (0.970), and Pb (0.961) have a large load in the first principal component and there is a strong positive correlation among the three elements, Cd shows a marked positive correlation with Zn (0.979), Pb (0.897) and Pb also shows marked positive correlation with Zn (0.902), which illustrate that the concentrations of heavy metals in Taoyuan coal mining soils probably originated from the same pollutant sources or had similar chemical properties. According to the previous descriptive data, the concentrations of these elements were greater than the regional soil reference values and the large coefficient of variation, indicating that their sources were mainly affected by human activities. Therefore, the elements Zn Pb and Cd should be related to coal mining, smelting processing, weathering, leaching, and coal transportation.

Cr (0.841) and Cu (0.605) have a large load in the second principal component, Cr and As have no

significant correlation with the first principal component factors, whereas Cu shows a better positive correlation with Zn (0.306), Pb (0.387), indicating the concentration of Cu is affected by these two elements. So the source of Cu may be released by emissions from coal transportation, wear of tires, or coal industry activities. The contents of Cr and As, which were low in the soil and the pollution evaluation of heavy metals were pollution-free or mild pollution, can be regarded As from the soil parent materials.

Conclusion

Except for Cr, concentrations of the other five elements in the study area soil were higher than the reference values in Suzhou non-mining soil, especially Cd, Cu Pb, and Zn with 5.82, 5.39, 3.14, and 2.75 times the reference values. The variation coefficient of Cd (88.19%) and Cu (84.40%) in soils was relatively high, illustrating more likely to be

affected by anthropogenic activities in the study area.

The PI values of As and Cr were slightly lightly polluted, however, the pollution of Cu, Cd, Zn, and Pb is relatively serious, especially for Cu and Cd. The PLI values varied from 1.82 to 3.12 revealing that nearly all of the study area was moderate to serious pollution by heavy metals. The results of the Igeo declared all soil samples could be considered as non-pollution for Cr and As, non-pollution to light pollution for Zn and Pb, while values of Cd and Cu illustrated light pollution to moderate pollution. The potential ecological risk (Ei) values of Cd with the mean of 142.91 had a strong risk of pollution and the other five metals belonged to a light ecological risk. All samples' RI values were higher than 150 and the average value was 210.40, indicating that the potential ecological risk (RI) was moderate pollution status considering the total of the studied metals. All the results reveal that heavy metal contamination has occurred in this region, especially Cd, Cu, and Pb should be paid attention to according to the enrichment factor or their potential ecological risk index.

The factor analysis revealed that two principal component factors were extracted with a variance contribution rate of 50.34 and 19.20% respectively. The first factor was dominated by Cd, Zn, and Pb and there is a strong positive correlation among the three elements, indicating that they should be related to coal mining, smelting processing, weathering, leaching, and coal transportation. The second principal component was occupied by Cr and Cu which the source of Cu may be affected by Zn and Pb and the contents of Cr and As can be regarded As from the soil parent materials.

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Ethics

This article is original and contains unpublished material. The corresponding author confirms that all of the other authors have read and approved the manuscript and that no ethical issues are involved.

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