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Original Research

Geochemical Baseline of Heavy Metals in Topsoil on Local Scale and Its Application

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Abstract

Bengbu is the central city of Northern Anhui Province in the Huai River Basin, China. No relevant research has yet discussed the geochemical baseline of heavy metals in topsoil of Bengbu. In this study, 139 topsoil samples were collected and analyzed for heavy metal content from different districts in Bengbu main urban area, and the methods of normalization and cumulative frequency distribution (CFD) were used to determine the geochemical baseline. The results showed that the mean baseline values of Cr, Cu, Ni, Pb, Zn, Mn, and As were 40.10, 30.77, 31.41, 30.30, 54.23, 422.92, and 1.21 mg/kg, respectively. Based on these values, baseline factor pollution index (BFPI) and geo-accumulation index (I_{geo}) were applied to assess the enrichment level of the heavy metals. The mean indices (I_{geo} and K_i) for Cr, Cu, Ni, Pb, Zn, Mn, and As were all $I_{geo} < 0$ and $K_i \leq 1.2$, indicating there was no heavy metal accumulation in study area as a whole. I_{geo} showed that 1.44% and 0.72% of the samples for Zn and Pb were moderately accumulated, and BFPI showed that the high accumulation rates of Zn, As, Pb and Cu in the samples accounted for 6.47%, 4.32%, 1.44% and 0.72%, respectively, the enrichment of heavy metals at individual sampling points needs attention.

Keywords: heavy metals, geochemical baseline, accumulation, Bengbu

Introduction

Soil heavy metal pollution will not only directly affect the structure and function of soil ecosystem, but also cause serious harm to human health through the food chain [1-3], so it is particularly important to investigate heavy metal contents in soils and then evaluate the pollution level. Traditional assessment generally uses geochemical background values as a reference. However, due to the wide range of human activities, the real background values are often difficult to obtain, moreover, the natural abundance of heavy metals in original soil varies in different regions, so it is not scientific to evaluate the accumulation degree of heavy metals in soil with the same standards [4].

The environmental geochemical baseline refers to the natural abundance of an element in soil when a local data set is used as a reference. It can not only reflect the content of elements in the regional environment, but also reflect the disturbance caused by natural or human factors [5-6]. It is defined as the upper limit of geochemical background or the lower limit of the impact of human activities [7-8]. It is an important reference for evaluating the enrichment of heavy metals in the earth's surface by human activities.

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A large number of studies have been carried out on the geochemical baseline of soil heavy metals at home and abroad. Wang J. et al. determined geochemical baseline values based on ralative cumulative frequency in Pacific sector of Arctic Ocean and Bering Strait surface sediments [9]. Nunes José Rato et al. studied the available levels of Cd, Cr, Cu, Ni, Pb, and Zn in surface horizons of agricultural soils in a typical European Mediterranean region, and established the geochemical baseline concentration model [10]. Insight from geochemical baselines and source apportionment, Tian Kang et al. reported environmental capacity of heavy metals in intensive agricultural soils from Jitai Town, South of Shouguang City, China [11]. Sun Houyun et al. assessed soil heavy metal pollution in the Luanhe River [12]. Tong Dekai et al. showed that Cd, As, Cu, Pb and Zn were the main pollution in soils of an onferrous metal mining area in Guangxi, China. As and Cu were related to ore mining; Cd, Pb and Zn were combined pollution; Hg, Cr and Ni were mainly derived from nature [13].

Bengbu is an important comprehensive industrial base in Anhui Province. It has a wide range of industries, however, due to the lack of research on geochemical baseline, previous pollution evaluation was all based on local background values, thus an objective standard of soil pollution assessment has not yet been formed in this area. Therefore, our study takes Bengbu main urban area for example, through the sampling and analysis of the topsoil, the geochemical baseline of heavy metals is determined by the methods of normalization and CFD curve. The results can be used as a standard for geoaccumulation assessment, and provide scientific reference for the soil environmental protection.

Material and Methods

Study Area

Bengbu is located in the Northern Anhui Province in the middle reaches of the Huai River Basin, with latitude of 32°43’ to 33°30’ N and longitude of 116°45’ to 118°04’ E. It includes 4 districts of the main urban area (Longzihu, Bengshan, Yuhui, and Huaishang) and 3 counties (Huaiyuan, Guzhen, and Wuhe), covering an area of 5951 square kilometers. Bengbu is a transitional zone between the humid monsoon climate in the north subtropical zone and the semi humid monsoon climate in the south temperate zone, which has the characteristics of both climate zones. The monsoon is significant, with four distinct seasons, mild climate and moderate rainfall. The territory is dominated by plains and scattered hills in the south.

Sample Collection and Analysis

A total of 139 sampling locations were selected in four districts of Bengbu main urban area (Fig. 1). At each sampling location, 5 topsoil (0-20 cm) subsamples were collected at random and merged to give a composite sample of roughly 1 kg. Meanwhile, the longitude and latitude of the sampling sites were recorded with

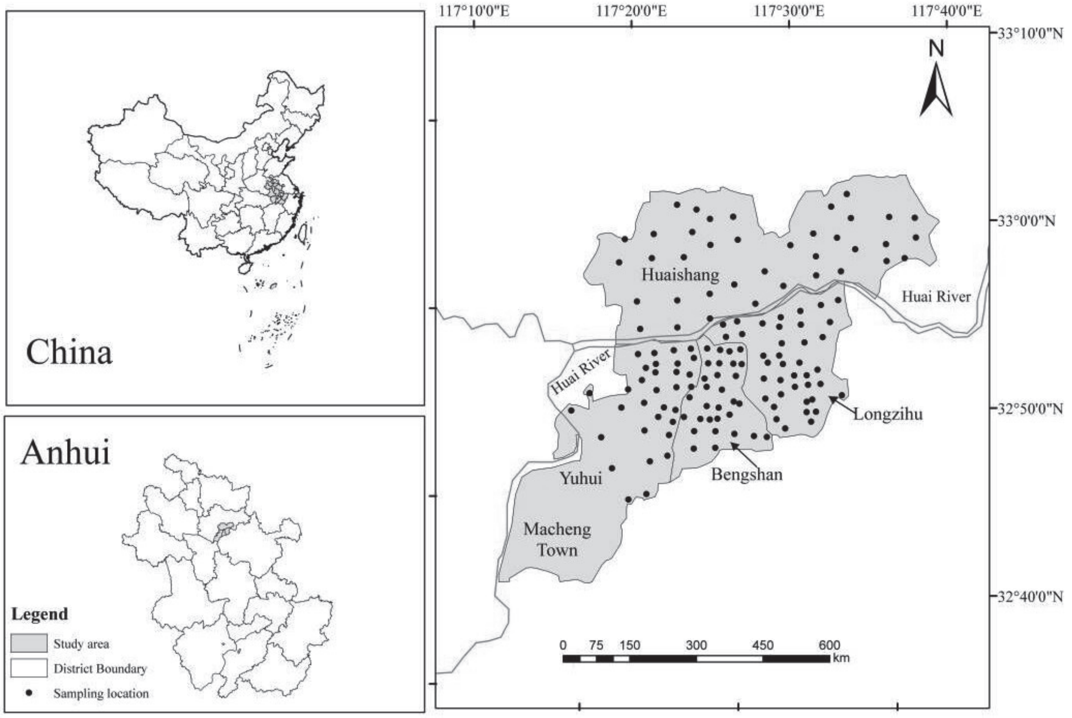

Fig.1. Sampling locations for study area.

handheld GPS. During the sampling process, the soil samples were placed in a clean polyethylene bags and labeled. After being brought back to the laboratory, the soil samples were air-dried at room temperature and sieved through 0.149 mm sieves for the next analysis.

The test elements included Cr, Cu, Ni, Pb, Zn, Mn, and As. The concentrations of As in soil samples were determined by atomic fluorescence spectrometer (Ruili, AF-610E, China), and the other elements were determined by atomic absorption spectrophotometer (TAS990, China). In order to ensure the accuracy, each batch of samples added two blank samples, and the national standard material was used for qualitative analysis.

Determination Method of Geochemical Baseline Value

Normalization

The basic idea of normalization is to take normalizing elements as the reference, and establish the equation to determine the geochemical baseline according to the linear correlation between them and each metal. It can compensate for grain-size and mineralogy effects on trace element concentrations. The baseline model can be expressed in the following form:

$$C_m = aC_n + b \tag{1}$$

Where C_m is the measured concentration of contaminated elements in the sample, C_n is the concentration of selected normalizing element, a and b are regression constants.

The 95% confidence interval was estimated by means of the SPSS Procedure, thus defining a range of data variability around each significant regression line. It is generally believed that the scatter points within the 95% confidence interval are not affected by man-made pollution and can represent the baseline range, so it is necessary to eliminate those points outside the 95% confidence interval to make the determination of geochemical baseline more reasonable. Using this method, Lu Xinzhe et al. obtained geochemical baselines of heavy metals (Cd, Cr, Cu, Hg, Ni, Pb and Zn), and evaluated the accumulation and potential ecological hazard risks of heavy metals in agricultural soil around a city in eastern Zhejiang [14]. For densely populated areas, Chen Song et al. established linear regression models between the heavy metal and Fe of soils, then determined the baseline values of Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Hg and Pb [15].

Cumulative Frequency Distribution (CFD)

Bauer et al. introduced the CFD curve into soil quality assessment to determine geochemical baseline value [16]. This method adopts normal decimal coordinates, and there may be two inflection points on this curve. The lower point represents the upper limit of the baseline value, and the average content of the element below this point (data within the 95% confidence interval) is determined as the baseline value, and the higher point represents the lower limit of anomaly, that is, the part affected by human activities. The part between the two inflection points is uncertain whether it is related to human activities. If the curve is approximately straight, the measured sample concentration itself may represent the baseline range. CFD is a very common method often applied by scholars. Niu Siping et al. determined the baseline values of Ni, Zn, Pb, and Cr using this method, then assessed the contamination levels in agricultural soils [17]. Wang Shuhang et al. calculated the baseline concentrations of metals (Cr, Ni, Cu, Zn, As, Cd, Hg, Pb) in surface sediments of Lihu Lake, based on these data, the enrichment degree and the pollution source of heavy metals in the medium were analyzed [18].

Baseline Factor Pollution Index (BFPI)

For assessing the contamination of heavy metals in soil, BFPI for selected heavy metals in each sample site was calculated, and is given in formula (2):

$$K_i = \frac{C_i}{B_i} \tag{2}$$

Where K_i is the accumulation index of heavy metal i, C_i and B_i are the investigated concentration and baseline value of heavy metal i in local soil, respectively. Pollution categories based on BFPI are divided into four categories [19]: no accumulation ($K_i \leq 1.2$); low accumulation ($1.2 < K_i \leq 1.5$); moderate accumulation ($1.5 < K_i \leq 2$); and high accumulation ($K_i > 2$). This method can make a specific evaluation of the pollution degree of each sample site.

Geoaccumulation Index (I_{geo})

I_{geo} was proposed by Muller in 1969 for quantitative evaluation of sediments [20]. This method is widely used to evaluate the accumulation degree of heavy metals in topsoil [21-25]. I_{geo} not only reflects the natural change of heavy metal distribution, but also can judge the impact of human activities on the environment. It is an important quantitative index with the expression as follows:

$$I_{geo} = \log_2\left(\frac{C_n}{1.5B_n}\right) \tag{3}$$

Where C_n is the measured content of heavy metal in the soil; B_n is geochemistry baseline value of a given metal; 1.5 is correction factor used to account for possible variations. The classification criteria of

Table 1. Classification criteria of geoaccumulation index.

The range of I_{geo}	Level	Pollution degree
$I_{geo}<0$	0	No polluted
$0\leq I_{geo}<1$	1	Unpolluted to moderately polluted
$1\leq I_{geo}<2$	2	Moderately polluted
$2\leq I_{geo}<3$	3	Moderately to heavily polluted
$3\leq I_{geo}<4$	4	Heavily polluted
$4\leq I_{geo}<5$	5	Heavily to extremely polluted
$I_{geo}\geq 5$	6	Extremely polluted

accumulation level can be classified into seven grades [26], as shown in Table 1.

Results and Discussion

Overview of Heavy Metal Concentrations

The descriptive statistics of heavy metal concentrations in Bengbu main urban area were shown in Table 2. The average concentrations of Cr, Cu, Ni, Pb, Zn, Mn, and As in topsoils were 40.94, 32.24, 32.28, 30.87, 60.70, 421.35, 1.25 mg/kg, respectively. Compared with the background value of Anhui Province, Cu, Ni, Pb, Zn, and Mn concentrations were higher than the background levels at some sampling sites, and the exceedance rates were 99.3%, 63.3%, 69.1%, 30.2% and 9.4%, respectively. As and Cr concentrations were lower than background values at all sampling sites. The coefficient of variation (CV) for heavy metals ranged from 18.5% to 45% for the topsoil samples, and the metals (Pb, Zn, and As) had comparatively high variations (CV>30%) with the following order: Zn>Pb>As, indicating human activities affected the distribution of some heavy metals. For some elements, such as As (high variation and low concentration) and Cr (low variation and concentration), due to regional differences, it is obviously not very scientific to adopt a unified background value, so it is particularly important to establish local baseline value.

The Determination of Geochemical Baseline Values

The central key factor of normalization is the selection of normalizing element. It must be an important constituent of one or more of the major trace metal carriers and reflect their granular variability in the sediments [27]. In general, normalizing element should follow the principles below: 1) it mainly comes from natural parent material, lacks obvious man-made source, and the mass fraction is sensitive to man-made input; 2) its concentration can be measured accurately and has obvious correlation with other elements'. Combined with previous studies, Al, Fe, Li, Rb, Cs, Ti, Mn and V were usually selected as normalizing element for analysis [28-30]. It is mainly considered that Fe has relatively stable properties (high abundance, low solubility, difficult leaching and migration, and low bioavailability). Through the basic geological survey and environmental survey in the study area, there is lower anthropogenic input of Fe in the topsoil of Bengbu main urban area. Therefore, Fe is selected as the normalizing element, the regression linear equation is established using the points within the 95% confidence interval to determine the baseline value.

The scatterplot matrix between heavy metals and Fe can be seen in Fig. 2, and the linear relationships are determined in the form of $C_m = aC_n + b$ (Table 3), where C_m is the predicted metal value and C_n is the Fe concentration in the sample. It can be seen from Fig. 2 that the concentration of heavy metals has a good linear correlation with Fe in topsoil of this area. Except for the metals (Pb and Zn), the other elements have relatively high correlation coefficient. Therefore, it is feasible to choose Fe as normalizing element. On the basis of above formula $C_m= aC_n+ b$, geochemical baseline value for each element was computed finally, the result was (in mg/kg): 40.93, 31.04, 32.32, 29.82, 54.21, 420.83, and 1.186 for Cr, Cu, Ni, Pb, Zn, Mn, and As, respectively.

Table 2. Summary statistics of heavy metal concentrations (n = 139).

Element	Cr	Cu	Ni	Pb	Zn	Mn	As
^{a}Min	14.65	19.17	14.91	3.67	29.20	109.70	0.045
^{a}Max	64.65	71.82	53.51	138	219.69	659.81	2.9145
^{a}Arithmetic mean	40.94	32.24	32.28	30.87	60.70	421.35	1.25
^{a}Geometric mean	40.17	31.47	31.54	28.42	56.70	411.86	1.14
^{a}Median	40.4	30.62	32.08	30.67	52.44	419.41	1.1742
CV %	18.5	24	21	44	45	19.8	38
Background value	66.5	20.4	29.8	26.6	62	530	9

^{a}The units of min, max, mean, geometric mean, and median: mg/kg

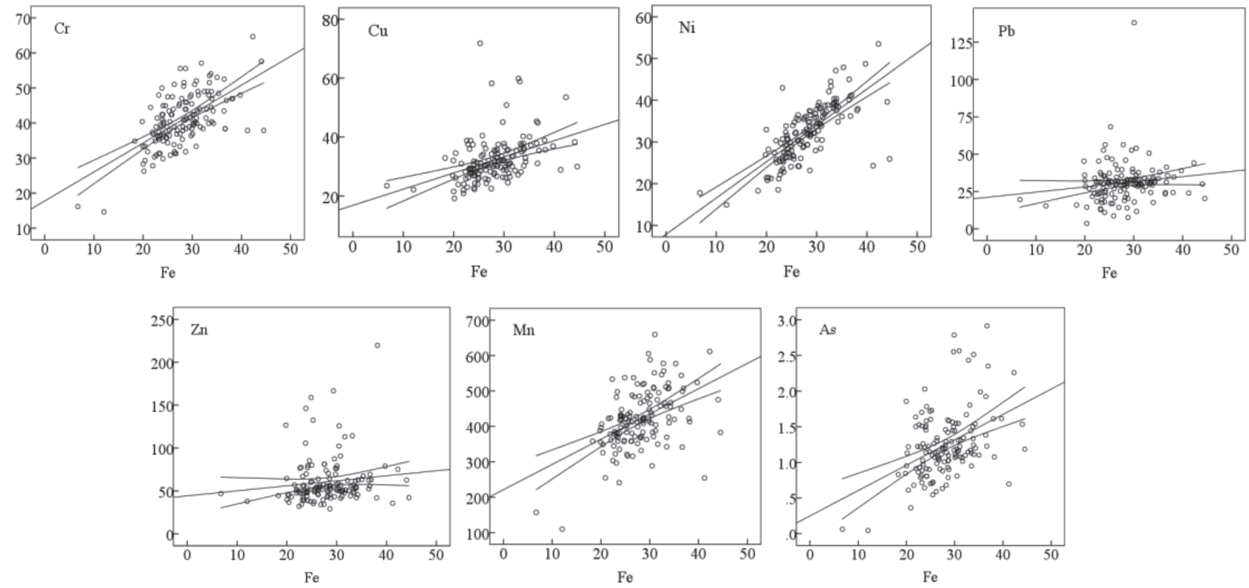

Fig. 2. Scatterplot matrix between metals (mg/kg) and Fe (g/kg).

The frequency count and CFD curve for heavy metals were shown in Fig. 3. The inflection point can be found, and the environmental geochemistry baseline for Cr, Cu, Ni, Pb, Zn, Mn, and As were 39.26, 30.49, 30.5, 30.77, 54.26, 425, and 1.225 mg/kg, respectively. The results were also listed in Table 3 for comparison. As can be seen from the table, the baseline values calculated by normalization and the CFD curve were very close to each other, and the mean values of the two methods were obtained as local geochemical baseline values.

The estimated baseline contents were compared with that of several places in China and abroad, it was found that there were certain regional differences (Table 4). The estimated baseline content of As was much lower than that of other localities listed in the table. Due to the lack of sufficient data, the baseline level of Mn was not so clear, but its value was significantly lower than that in Chengde, Hebei Province. Meanwhile, the determined baseline concentration of Cr was similar to that in Jiedong, lower than other cities in China, but higher than two regions abroad. As for the rest heavy metals (Cu, Ni, Pb, and Zn), their contents were basically at a medium level in contrast to those reported in other places. It can be observed that the baseline values of resource-oriented cities were relatively high, such as Fuxing and Tongling, and the main reason probably had more to do with the soil type and parent material.

Heavy Metal Pollution Assessment

Using the determined baselines, the BFPI was introduced to evaluate the accumulation level of heavy metals in topsoil. The K_i values for seven metals from the samples were shown in Table 5. According to the classification standard of K_i, an K_i value less than 1.2 means the enrichment of heavy metals is mainly

Table 3. Geochemical baseline values determined by different methods.

Element	Normalization				CFD	[a]Mean value of baseline
	Regression equation	R	P	[a]Baseline value	[a]Baseline value	
Cr	Cr = 0.826 Fe + 17.770	0.617	0.01	40.93	39.26	40.10
Cu	Cu = 0.460 Fe + 18.223	0.496	0.01	31.04	30.49	30.77
Ni	Ni = 1.116 Fe + 1.169	0.822	0.01	32.32	30.5	31.41
Pb	Pb = 0.323 Fe + 20.757	0.182	0.05	29.82	30.77	30.30
Zn	Zn = 0.416 Fe + 42.545	0.188	0.05	54.21	54.25	54.23
Mn	Mn = 7.795 Fe + 203.126	0.543	0.01	420.83	425	422.92
As	As = 0.028 Fe + 0.406	0.420	0.01	1.186	1.225	1.21

[a]The unit of baseline value: mg/kg.

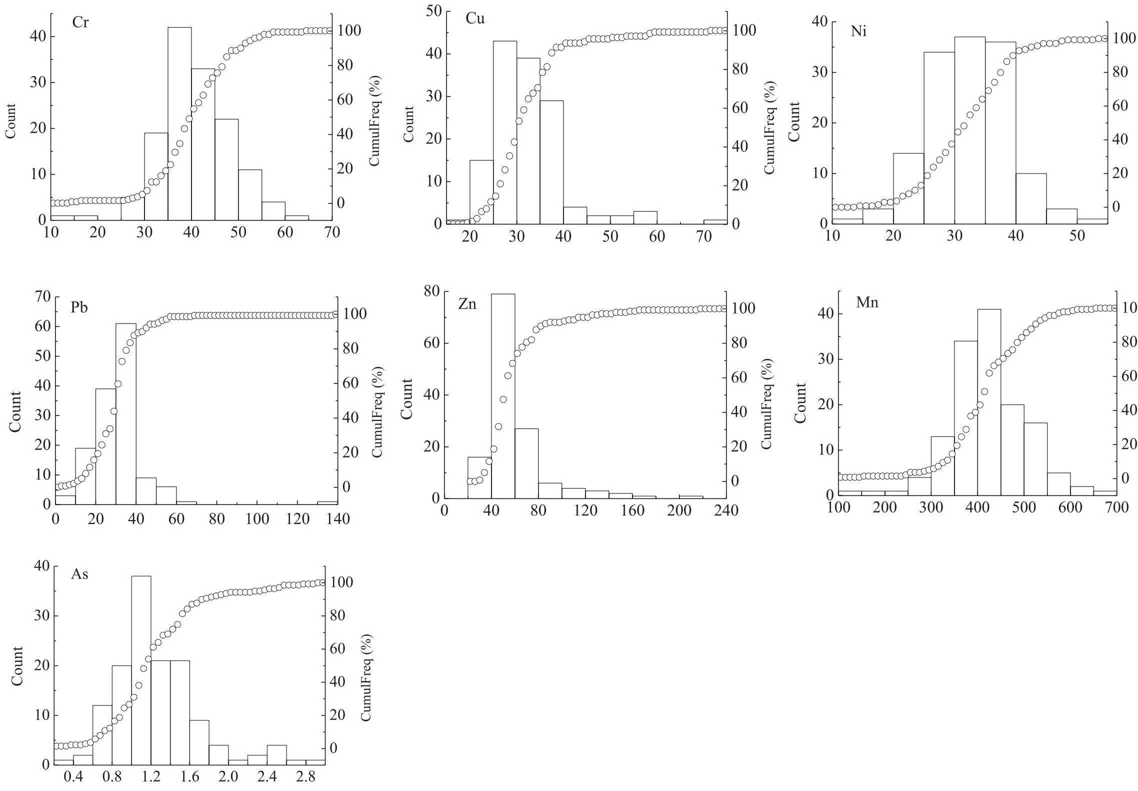

Fig. 3. Frequency count and the CFD curves for heavy metals (mg/kg).

from natural input, there is no pollution in soils; an K_i value greater than 1.2 means the enrichment of heavy metals is related to human activity. The higher the K_i value is, The greater the impact of human activities. As can be seen in Table 6, on the whole, the mean K_i values of Cr, Cu, Ni, Pb, Zn, Mn and As were all lower than 1.2, indicating that these elements were mainly from natural input, no pollution in study area. In detail, the proportion of samples with different pollution levels as shown in Fig. 4. The proportion of moderate accumulation of Cr, Cu, Ni, Pb, Zn, Mn and As were 0.72, 3.6, 2.88, 6.47, 3.6, 0.72 and 4.32%, respectively, and high accumulation of Cu, Pb, Zn, and As were 0.72, 1.44, 6.47, and 4.32%, respectively. The enrichment of Zn and As at individual sampling points needs attention.

The I_{geo} of the sample was calculated according to formula (2), the mean I_{geo} of Cr, Cu, Ni, Pb, Zn,

Table 4. Comparison of baseline values of heavy metals (mg/kg) reported in literature.

City/Country	Heavy metals						
	Cr	Cu	Ni	Pb	Zn	Mn	As
Fuxin, Liaoning, China[a]	145.21	42.86	89.34	60.55	92.23	-	4.17
Chengde, Hebei, China[b]	50.78	19.59	24.99	23.31	72.65	754.48	8.53
Shouguang, Shandong, China[c]	62.84	21.17	28.38	19.25	59.60	-	7.67
Tongling, Anhui, China[d]	73.50	47.70	25.20	39	113	-	24.30
Suzhou, Anhui, China[e]	65.22	16.97	-	19.43	48.66	-	9.07
Jiedong, Guangdong, China[f]	39.91	14.22	12.70	47.62	64.54	-	11.48
Chongqing, China[g]	66.78	25.45	29.90	26.18	78.44	-	5.83
Karachi, Pakistan[h]	12.9	36.31	-	56.23	123.03	-	-
Andes Mountain Range, Peru[i] (Cajamarca-Huancavelica, Peru)	8.26	22.20	56.97	44.87	47.42	-	27.50
This study	40.10	30.77	31.41	30.30	54.23	422.92	1.21

Note: The data sources of [a-i] are from literature [31], [12], [11], [32], [1], [33], [7], [34], and [35].

Table 5. The statistical result of K_i value of each element.

Element	Cr	Cu	Ni	Pb	Zn	Mn	As
Min	0.37	0.62	0.47	0.12	0.54	0.26	0.04
Max	1.61	2.33	1.70	4.55	4.05	1.56	2.41
Average	1.02	1.05	1.03	1.02	1.12	1.00	1.03

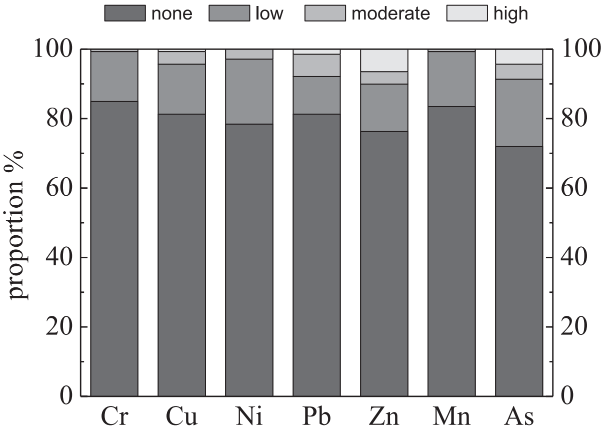

Fig. 4. Frequency histogram based on BFPI.

Mn, As were -0.58, -0.55, -0.58, -0.68, -0.52, -0.62, -0.67, respectively, and the frequency histogram of accumulation degree of each heavy metal are shown in Table 6 and Fig. 5. As can be seen from them, the heavy metals (Cr, Cu, Mn, Ni and As) showed no pollution to moderate pollution, with respective proportion of Cr (99.28%, 0.72%), Cu (95.68%, 4.32%), Mn (99.28%, 0.72%), Ni (97.12%, 2.88%) and As (91.37%, 8.63%). For Pb and Zn, the proportions were 92.09% and 89.93% for level 0 (unpolluted), 7.19% and 8.63% for level 1 (unpolluted to moderately polluted), 0.72% and 1.44% for level 2 (moderately polluted). On the whole, as to the heavy metals studied, I_{geo} assessment is basically consistent with the evaluation result of BFPI, the surface soil is relatively clean in study area.

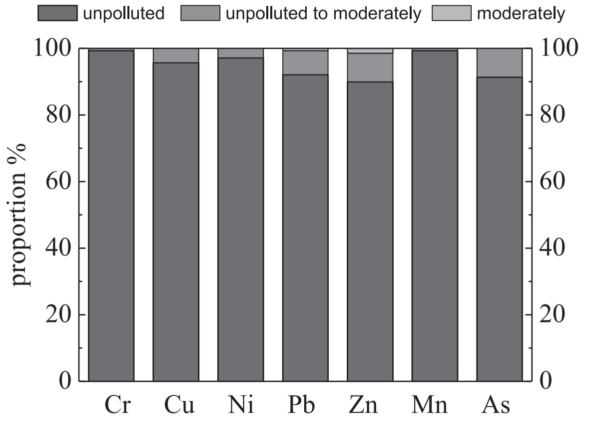

Fig. 5. Frequency histogram based on I_{geo}.

Conclusions

This study established local geochemical baseline model to assess the accumulation level of heavy metals. Normalization was used to determine the baseline values combining the method of CFD curve. The results showed that the geochemical baseline values of Cr, Cu, Ni, Pb, Zn, Mn, and As were 40.10, 30.77, 31.41, 30.30, 54.23, 422.92, and 1.21mg/kg, respectively, which objectively reflected the natural abundance of heavy metals in surface soil. Based on the determined geochemical baseline, BFPI and I_{geo} were applied to evaluate the accumulation level of heavy metals. The mean values of two indicators (I_{geo} and K_i) for all elements were lower than 0 and 1.2, respectively, indicating an uncontaminated level. But specifically, 1.44% and 0.72% of the Zn and Pb samples were moderately accumulated for I_{geo} assessment, and 6.47%, 4.32%, 1.44%, and 0.72% of Zn, As, Pb, and Cu samples were highly accumulated, Thus, the accumulation of heavy metals at individual sites needs attention in study area. By calculating BFPI and I_{geo} of heavy metals in topsoil of Bengbu main urban area, it can be seen that there is no obvious man-made pollution in this area. Bengbu is located in the middle and lower reaches of Huaihe River, and the soil type and soil forming parent material are basically the same as those in the surrounding areas. Therefore, the geochemical baseline of heavy metals in Bengbu main urban area can be used as an important reference for the middle and lower reaches of Huaihe River.

Table 6. The statistical result of I_{geo} value of each element.

Element	Cr	Cu	Ni	Pb	Zn	Mn	As
Min	-2.04	-1.27	-1.66	-3.63	-1.48	-2.53	-5.33
Max	0.10	0.64	0.18	1.60	1.43	0.06	0.68
Average	-0.58	-0.55	-0.58	-0.68	-0.52	-0.62	-0.67

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Conflict of Interest

The authors declare no conflict of interest.

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