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Quantitative assessment and source apportionment of metal pollution in soil along Chao River

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ABSTRACT

Heavy metal pollution in surface soil is a serious threat to the drinking water source area, which might be washed into the reservoir by stormwater runoff. In this study, concentrations of 26 metals were measured for 15 surface soil samples taken along Chao River, which is the major inflow river for Miyun reservoir. The characteristics of metal concentrations were demonstrated and the pollution sources were analyzed by principal component analysis/multiple linear regression method and positive matrix factorization method. Results show that the mean concentrations of nickel (Ni), chromium (Cr), copper (Cu), and mercury (Hg) are larger than their standard values. Moderate risk of Hg pollution and lightly risk of lead (Pb) and Ni pollution are found in the study area. Anthropogenic behaviors of mining, vehicular emission, and coal combustion are the dominant metal pollution sources, which contribute to 33–35, 17–27, and 11–26% of the total metal pollution, respectively. Most of Hg, Pb, Cu, Cr, and manganese come from these sources. Natural source contributes to 22–29% of the total metal pollution, which is the main source of zinc and cadmium.

Keywords: Heavy metal pollution; Miyun reservoir; Principle component analysis; Source apportionment; Surface soil

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1. Introduction

Heavy metal pollution at drinking water source area has become a serious threat to the safety of urban water supply and human health in many places in China [1]. Various metal elements from many sources accumulate in the surface soil and can be flushed into the reservoirs and lakes nearby, which will either reach the drinking water supply plant or settle down at the bottom of the reservoir [2]. This will increase the risk of heavy metal pollution for urban water supply. For example, mercury (Hg) concentration has kept rising in recent years in Lake Songhua, which is

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the drinking water source for JinLin and ChangChun city [3]. A high risk of heavy metal contamination, especially cadmium (Cd), was found near the Guanting reservoir [4].

Therefore, it is necessary to characterize the metal contamination status in surface soil and discerning the sources of metals. This will be helpful to control different pollution sources for the heavy metal pollution in soil.

Generally, metal pollution sources can be classified into two groups. One is the natural material group and the other is anthropogenic source group. Nowadays, the main method to indentify different sources is to investigate trace elements in samples. For example, a coexistence of high concentrations of iron (Fe) and manganese (Mn) usually relates to the existence of soil parent material [5,6]. Likewise, a close association between magnesium (Mg) and Mn or an association among chromium (Cr), nickel (Ni), aluminum (Al) and Fe also indicates that these elements may originate from the parental materials [7]. Similarly, a strong relationship between zinc (Zn) and Cd also implies the existence of nature sources [8].

On the other hand, there are many anthropogenic sources for metal elements in soil such as vehicle emission, coal combustion, sewage for irrigation, fertilizer, pesticide, and mining [9]. Usually, both Hg and arsenic (As) are tracer elements in coal combustion [10]. The existence of Cr, lead (Pb), and Ni is usually associated with vehicular emission [11]. In lots of fertilizers, Hg and Cd are common elements which can be used as tracer elements for fertilizers [12]. In Tariq's study [13], strong correlation among Zn, Cr, and calcium (Ca) was considered to originate from tannery effluents.

Currently, many approaches including principal component analysis/multiple linear regression (PCA/MLR), positive matrix factorization (PMF), chemical mass balance, unmix method, etc. have been applied to identify different pollution sources. Different approaches need different kinds of data and use different basic theories. Therefore, the results of different methods might not be the same. As a result, the application of multi-methods in source apportionment study is necessary to scientifically evaluate the source identification results [14–18].

The objective of this study is to characterize the current pollution situation of metal elements in soil along the Chao River, which is the main upper stream of Miyun Reservoir. Since Miyun Reservoir is the main surface water supply source for Beijing, the results of this study will be useful to the pollution source management for Miyun reservoir.

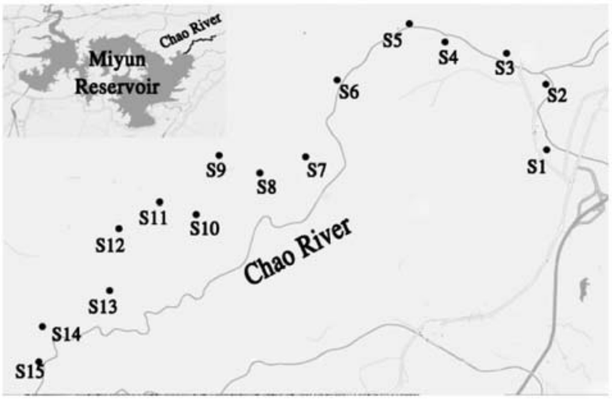

Fig. 1. Sampling site locations.

2. Materials and methods

2.1. Study area and sampling method

As shown in Fig. 1, Chao River is the main inflow reach of Miyun Reservoir. In this study, the surrounding land along the lower reach of Chao River is selected as study area, which contributes a large proportion of nonpoint source pollution to Chao River. Agriculture is the major land use in this area and the anthropogenic activities greatly influence the water quality of Miyun Reservoir.

A total of 15 sampling sites were selected, which were about evenly distributed along the Chao River as shown in Fig. 1. The detailed information of sampling sites is shown in Table 1. There are three kinds of land uses including grassland, forestland, and arable land. The grasslands of S2, S14, and S15 are usually drowned in the rainy season due to their short distance to the river. S3, S5, and S12 are forestlands. Among these sites, S3 and S5 are protective forest near the river to reduce the nonpoint source pollution to the river and S12 is a natural forest near the villages. Other sites are arable lands which include corn field (S1 S6, S8, S10, S11, and S13) and orchard field (S4, S7, and S9). In addition, topographical differences among sampling sites also exist. These sites have three kinds of topography including plain, sloping land, and terrace land.

Samples were taken in October 2011. At each sampling site, the top 15 cm surface soil was taken from three evenly distributed points within 1 m^2 area, then these three samples were mixed together to be the only sample for each sampling site. Samples were put into plastic bags and taken back to the laboratory for chemical analysis. All samples were air dried for 3 weeks before analysis.

Table 1
Sampling site description

Site No	Land use	Topography	Coordinate
S1	Arable land (corn field)	Plain	40°34´0.24´´N, 117°8´34.29´´E
S2	Grassland	Plain	40°34´53.73´´N, 117°8´27.48´´E
S3	Forestland	Plain	40°35´14.18´´N, 117°7´45.53´´E
S4	Arable land (orchard)	Terrace land	40°35´34.86´´N, 117°7´0.88´´E
S5	Forestland	Plain	40°35´35.09´´N, 117°6´7.94´´E
S6	Arable land (corn field)	Plain	40°35´32.57´´N, 117°5´30.60´´E
S7	Arable land (orchard)	Terrace land	40°34´11.87´´N, 117°4´15.98´´E
S8	Arable land (corn field)	Sloping land	40°34´12.66´´N, 117°3´10.29´´E
S9	Arable land (orchard)	Sloping land	40°34´22.83´´N, 117°2´26.81´´E
S10	Arable land (corn field)	Plain	40°33´39.18´´N, 117°2´18.65´´E
S11	Arable land (corn field)	Plain	40°33´49.39´´N, 117°1´42.78´´E
S12	Forestland	Sloping land	40°33´21.69´´N, 117°1´22.73´´E
S13	Arable land (corn field)	Plain	40°32´53.98´´N, 117°1´18.00´´E
S14	Grassland	Plain	40°32´23.59´´N, 117°1´3.16´´E
S15	Grassland	Plain	40°31´50.22´´N, 117°1´1.41´´E

2.2. Chemical analysis

Before chemical analysis, large stones and plant parts were removed by a sieve with an opening size of 2 mm. Then the sieved samples were grounded. The concentrations of 26 metal elements (As, barium (Ba), Cd, cerium (Ce), cobalt (Co), Cr, copper (Cu), Hg, lanthanum (La), Mn, molybdenum (Mo), niobium (Nb), Ni, Pb, antimony (Sb), scandium (Sc), titanium (Ti), thallium (Tl), yttrium (Y), Zn, aluminum oxide (Al_2O_3), ferric oxide (Fe_2O_3), magnesium oxide (MgO), calcium oxide (CaO), sodium monoxide (Na_2O), and potassium oxide (K_2O)) including heavy metals and geochemical parameters were measured on the grounded samples. Cd, Ce, Co, Cu, La, Mo, Nb, Ni, Pb, Sc, Tl, Y, and Zn were analyzed by inductively coupled plasma-mass spectrometry. Ba, Cr, Mn, Ti, Al_2O_3, Fe_2O_3, MgO, CaO, Na_2O, and K_2O were analyzed by inductively coupled plasma optical emission spectrometer. As, Hg, and Sb were measured by atomic fluorescence spectrometry method. Duplicate samples (one duplicate per 7 samples) were measured and geochemical reference materials (GSS17, GSS25, GSS26, GSS27) were used in the experiment for quality control. The average deviation from the reference materials is less than 5%.

2.3. Source apportionment analysis

2.3.1. PCA/MLR method

In this study, the PCA/MLR method was used with SPSS 18.0. Influential factors from different pollution sources were determined with PCA analysis [19,20]. After PCA analysis, the contribution rate of each factor was calculated by MLR method.

2.3.2. PMF method

To better understand the contribution of various pollution sources, PMF method was applied for source apportionment. Firstly, the uncertainty of the experimental data was evaluated by the fractional error and minimum detection limits. Secondly, the data were diagnosed by the signal-to-noise ratio and determined to be good, bad, or weak. Then the number of factors was obtained. Finally, the preliminary result will be regulated until the best result is obtained [21]. The EPA PMF 3.0 was used in this research.

3. Results and discussion

3.1. Heavy metal concentrations

Statistical summary of concentrations of the 26 metal elements is shown in Table 2. Ba, Mn, and Ti are quite enriched in the samples with mean concentrations higher than 500 mg/g, which are 620.63, 782.77, and 4668.27 mg/g, respectively. The concentrations of other metals are all less than 500 mg/g. The variation of the metal concentrations caused by site difference is represented by coefficient of variation (CV) values. Results show that the concentrations of most metal elements do not vary very much at different sites with CV values between 12 and 50% except Hg, which has a CV value of 86.95%. This indicates that Hg might come from different pollution sources than other

Table 2
Statistical summary of concentrations of 26 metal elements

	Min (μg/g)	Max (μg/g)	Mean (μg/g)	CV (%)	S.E. mean (±μg/g)
As	3.27	15.00	7.94	44.95	0.92
Ba	300.50	850.30	620.63	23.67	37.88
Cd	0.08	0.31	0.16	36.93	0.02
Ce	42.88	89.68	63.32	16.90	2.76
Co	9.47	32.09	18.60	28.17	1.35
Cr	51.38	283.30	127.71	49.08	16.18
Cu	17.05	68.52	40.42	39.55	4.13
Hg	0.06	0.76	0.22	86.95	0.49
La	20.38	44.75	31.09	18.44	1.48
Mn	548.60	1,326	782.77	24.32	49.14
Mo	0.32	1.00	0.69	27.60	0.05
Nb	8.41	15.03	11.44	17.34	0.51
Ni	16.14	95.10	45.97	42.08	5.00
Pb	9.87	38.17	21.98	29.28	1.66
Sb	0.31	1.02	0.66	33.81	0.06
Sc	9.22	23.10	13.08	23.58	0.80
Ti	3,438	6,390	4668.27	16.34	196.98
Tl	0.22	0.92	0.61	27.81	0.04
Y	18.19	41.57	26	22.10	1.49
Zn	54.71	115.90	81.06	21.36	4.44
Al_2O_3*	11.14	18.48	13.38	12.60	0.44
Fe_2O_3*	4.53	9.80	6.73	21.62	0.38
MgO*	1.41	3.72	2.17	31.83	0.18
CaO*	1.15	5.54	2.48	49.35	0.32
Na_2O*	1.29	3.60	2.48	22.51	0.14
K_2O*	1.10	2.85	2.33	22.29	0.13

*The unit for the min, max, mean, and S.E. mean of these metal oxides is %.

elements. The standard error of mean concentration of each element is represented by S.E. Mean values. The S.E. Mean values for most of the metals are much smaller than their mean values except Hg, which shows the representativeness of the mean values for most metal element. It is noticeable that the S.E. Mean value of Hg is more than two times of its mean value. This demonstrates the less representativeness of its mean. More attention should be paid on the statistical inference of Hg concentration.

The mean concentrations of eight heavy metals in this study are compared with the soil background concentrations in Beijing, soil quality standard, and the concentrations of soil in Shanghai as shown in Table 3. The sampling sites in Shanghai's study [23] are located in the drinking water conservation area of Shanghai, which is near Songjiang industrial area. Table 3 shows that most mean concentrations in this study are higher than the background values except Pb. The mean concentrations of Ni, Cr, Cu, and Hg

Table 3
Comparison of the mean concentrations of this study with soil background values, standard values, and other study

	As	Ni	Pb	Cd	Zn	Cr	Cu	Hg
Mean of this study (μg/g)	7.94	45.97	21.98	0.16	81.06	127.7	38.45	0.22
Background value (μg/g) [22]	7.09	26.8	24.6	0.119	57.5	29.8	18.7	0.08
Standard (GB15618-1995) (μg/g)	15	40	35	0.2	100	90	35	0.15
Over standard rate (%)	0	53.3	6.7	13.3	13.3	73.3	46.7	53.3
ShangHai [23]	6.31	24.04	30.14	0.16	103.79	65.75	30.66	0.33

are higher than the soil standard values. For example, the concentration of Cr is higher than the background value at all sites with the maximum value of 283.3 mg/g, which is about 20 times of the background value. The concentration of Cr exceeds the standard value at 11 sites in this study. Likewise, concentrations of Hg, Cu, and Ni are also higher than the background and standard values. These results indicate that there is a potential risk of heavy metal pollution in the soil of the study area, especially Cr and Hg. Compared with the study in Shanghai, the concentrations of Ni and Cr in this study are about two times of their concentrations in Shanghai. Concentrations of As and Cu are slightly higher. Meanwhile, concentrations of Pb, Zn, and Hg are slightly lower than those of Shanghai. These might be caused by more traffic pollution in Shanghai.

Metal concentration distribution at various sites is different for different metals. As, Hg, and Pb have a similar distribution with the maximum concentration at S15. The distributions of Ni and Cr are similar with high values at S4 and S6. These indicate that certain metal elements might come from similar sources and some of them might not have the same sources. It is noticeable that the S.E. Mean value of Hg is very large therefore more attention should be paid on this metal element.

Heavy metal concentrations demonstrate obvious difference for different land uses. The mean concentrations of As, Ni, Pb, Cd, Cr, Zn, Cu, and Hg at different land uses are presented in Table 4. Arable land use contains three terrains of plain, terrace, and sloping. Forestland use includes natural and protection forests. Heavy metal concentrations of As, Cd, Cr, Pb, and Zn of forestland are generally less than those of other land uses. This might be caused by the intense anthropogenic input at arable land and grassland. Therefore, enlarging forestland use is an effective method for heavy metal control at drinking water resource area. On the other hand, the concentrations of Hg, Pb, and As are highest at grassland especially at S14 and S15. Meanwhile, the concentrations of Cu and Cr are relatively low at these sites. The heavy metal concentrations at arable land are moderate compared with other land uses. The difference among different topographies at arable land is not obvious.

To evaluate the potential risk of heavy metals in the study area, Nemerow index [24] was calculated for each of the eight heavy metals by the following equations:

$$P_{ij} = C_{ij}/S_i \tag{1}$$

$$P_i = \sqrt{\frac{(\overline{P_{ij}})^2 + (\max(P_{ij}))^2}{2}} \tag{2}$$

where P_{ij} is the Nemerow index of pollutant i and observation j. C_{ij} is the concentration of pollutant i and observation j. S_i is the standard value of pollutant i. Max(P_{ij}) is the maximum value of P_{ij} for all observations and $\overline{P_{ij}}$ is the average value of P_{ij}. Based on the value of P_i, metal element i is classified to be unpolluted ($P_i < 1$), lightly polluted ($1 < P_i < 2$), moderate polluted $2 < P_i < 3$), and strongly polluted ($P_i > 3$). In this research, the Nemerow indexes of As, Ni, Pb, Cd, Zn, Cr, Cu, and Hg in the study area are calculated and presented in Fig. 2.

As Fig. 2 presents, the Nemerow index values of As, Pb, Cd, Zn, and Cu are less than 1, which suggests little impact on the environment from these elements. The Nemerow index values of Hg, Ni, Cr are larger than one with Hg's value even larger than two. This indicates that there is a lightly risk of Ni and Cr pollution, and a moderate risk of Hg pollution in the study area.

Table 4
Mean concentrations of heavy metals from different land uses

	Arable land			Grassland	Forestland	
	Plain	Terrace	Sloping		Natural forest	Protection forest
As	7.46	5.46	10.79	11.43	5.54	3.86
Cd	0.15	0.20	0.15	0.17	0.16	0.08
Cr	157.92	157.12	122.10	98.26	82.64	107.60
Cu	40.04	51.26	36.75	38.48	23.06	68.52
Hg	0.13	0.15	0.26	0.43	0.17	0.15
Ni	52.73	56.37	43.41	41.86	27.42	45.91
Pb	21.56	17.41	23.87	29.42	20.61	9.87
Zn	78.30	93.29	76.55	90.68	67.09	78.51

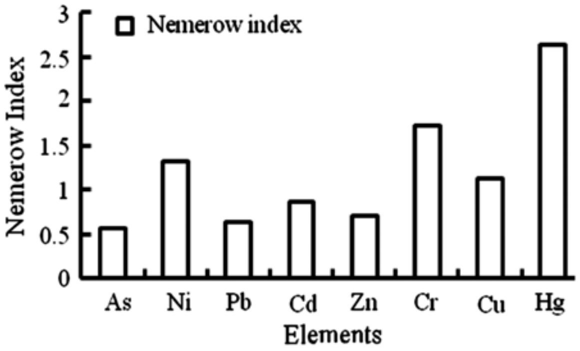

Fig. 2. Nemerow index values of eight heavy metals.

3.2. PCA/MLR analysis

To investigate the relationships among metals in the soil, Pearson correlation analysis was conducted. Results show that Co, Cr, Cu, Mn, Ni, Sc, and Zn have very significant correlations with Fe_2O_3 and MgO with the correlation coefficient of r larger than 0.7. Besides, Cd, Mn, and Zn have strong correlation with each other. Hg does not show any strong correlation with other elements.

Then PCA analysis using Varimax rotation was employed to find different sources of metals. The Eigen values were used to determine the number of components, which was curtained when the cumulative percentage variance was more than 80%. Trace elements of each component were obtained by the factor loadings to identify specific source. Using the data from all the 15 sites, four principal components (PC) were identified, accounting for 87% of the total variance in the dataset as shown in Table 5.

The first principle component (PC1) explains 31.2% of the total variance. It is strongly correlated with Pb, As, and Hg. The close association between Hg and As indicates a possible source of coal combustion and mining. There are some out-of-date mining areas in the upstream of Chao River, which is one of the main reasons for heavy metal pollution in the soil of Miyun basin, especially Pb and Hg pollution. These elements are flushed into Chao River from the mining area by runoff process, reaching Miyun reservoir and accumulate in the sediment. The concentrations of Pb and Hg are highest at grasslands, which are often drowned in the rainy season. Suspended solids with high Pb and Hg concentrations settle down at the drowned area. In addition, Pb and Ti usually have a strong correlation in natural environment, which is not found in this study area. All these results indicate that PC1 represents an anthropogenic factor especially mining behavior.

Table 5
Results of Varimax-rotated principal component analysis of all samples

	PC1	PC2	PC3	PC4
As	0.95	−0.15	0.05	−0.18
Ba	0.19	−0.31	0.87	0.05
Cd	0.22	0.88	0.12	0.17
Ce	0.83	0.37	−0.30	−0.04
Co	−0.21	0.44	0.59	0.62
Cr	−0.31	0.17	0.02	0.87
Cu	−0.26	0.26	0.53	0.71
Hg	0.74	0.02	0.27	−0.01
La	0.83	0.25	−0.37	−0.08
Mn	0.13	0.70	0.55	0.38
Mo	0.57	0.52	−0.29	0.14
Nb	0.90	−0.03	−0.09	−0.30
Ni	−0.21	0.24	0.13	0.90
Pb	0.89	0.12	−0.33	−0.03
Sb	0.91	−0.08	0.00	−0.29
Sc	−0.05	0.70	0.49	0.44
Ti	−0.17	0.20	0.85	0.25
Tl	0.84	−0.03	−0.47	−0.22
Y	0.31	0.80	0.44	0.06
Zn	0.29	0.73	0.26	0.44
Al_2O_3	−0.10	−0.06	0.66	0.32
Fe_2O_3	−0.13	0.34	0.50	0.69
MgO	−0.22	0.58	0.17	0.75
CaO	−0.23	0.79	−0.06	0.25
Na_2O	−0.07	−0.17	0.23	0.25
K_2O	0.60	−0.19	0.75	−0.15
Explained variance %	31.2	19.5	19.3	17.9
Cumulative %	31.2	50.7	69.9	87.8

PC2 is strongly correlated with Zn, Cd, MgO, and CaO, and accounts for 19.5% of the total variance. Mg and Ca are common mineral elements, such as calcite and dolomite. Besides, in nature, Zn and Cd are always strongly correlated with each other, which is true in this study as shown in Table 3. These indicate that PC2 represents natural sources.

About 19.3% of the total variance is explained by PC3, in which Ba, Cu, Mn, Ti, K_2O, and Co have a high loading. Al_2O_3 and Fe_2O_3 have a moderate loading. Previous studies showed that Cu and Mn were found in vehicular emissions and Co was a necessary material for auto-exhaust catalyst. As a result, PC3 might represent vehicular emission.

PC4 explains about 18% of total variance. Co, Cr, Cu, Ni, Fe_2O_3, and MgO are dominant metals associated with PC4. Based on related studies, Cr and Ni are commonly found in coal combustion from industry [25–27]. Therefore, PC4 represents the source of coal combustion.

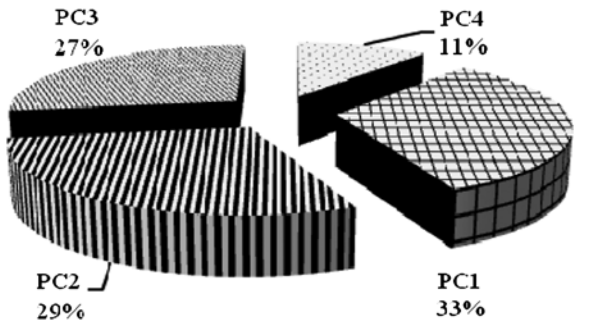

Fig. 3. Contribution proportion of each component calculated by PCA/MLR method.

MLR was employed to calculate the specific contribution rate of different sources after PCA. Factor scores were regressed on the metal concentration for each sample. Stepwise method was used in MLR. The contribution of each factor was calculated with regression coefficient. The regression result is shown as the following equation:

$$Z = 7.1 \times \text{PC1} + 6.4 \times \text{PC2} + 6 \times \text{PC3} + 2.3 \times \text{PC4} \quad (R^2 = 0.991) \tag{3}$$

where Z is the standardized normal deviates of metals.

The contribution proportion of each component is presented in Fig. 3. The contribution proportions of PC1, PC2, PC3, and PC4 are about 33, 29, 27, and 11%, respectively.

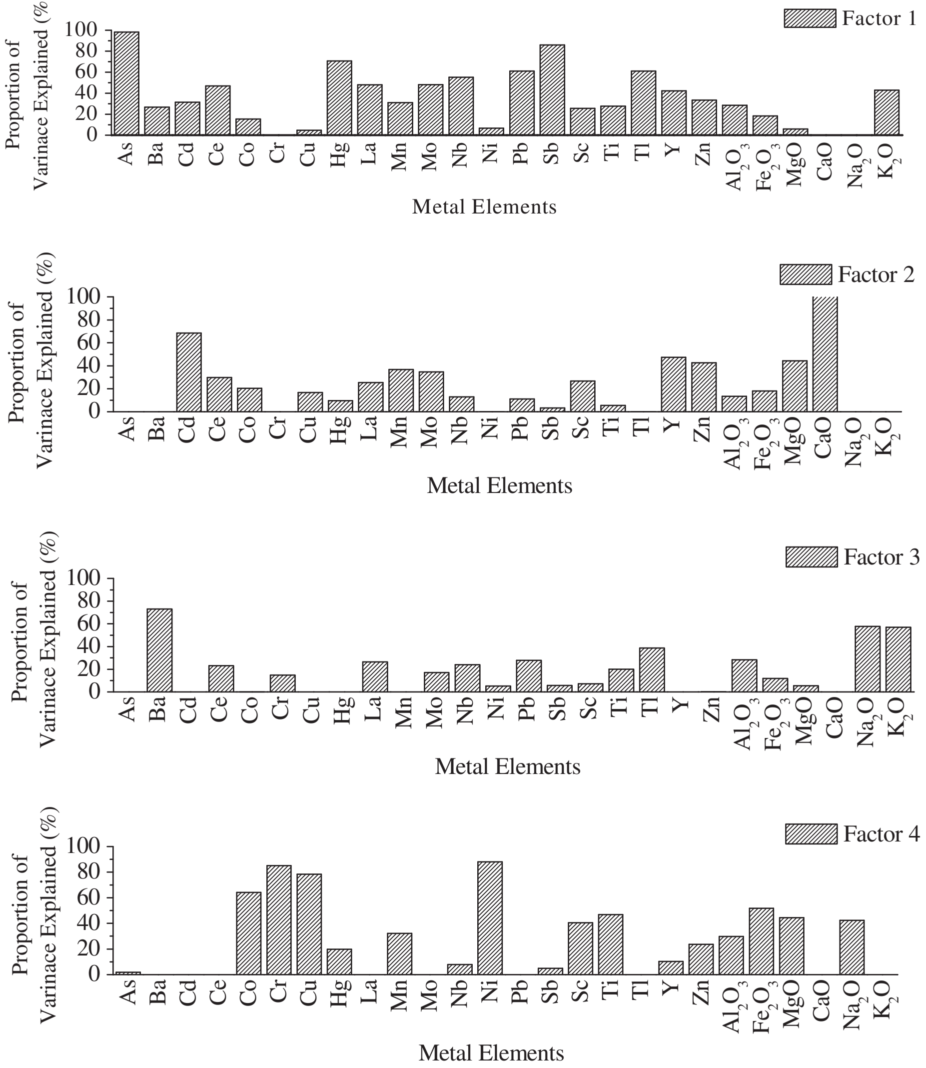

Fig. 4. The proportion of variance explained by each factor for 26 metal elements using PMF analysis.

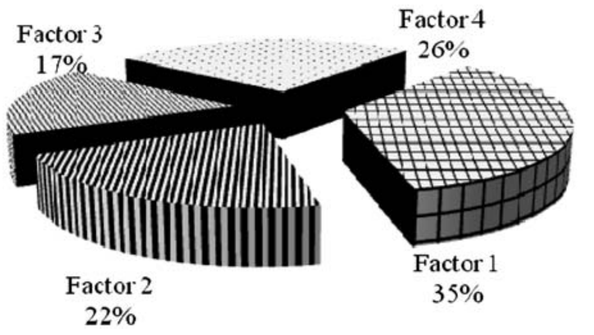

Fig. 5. The contribution proportion of each factor calculated by PMF analysis.

3.3. PMF analysis

To further investigate the contribution of different metal pollution sources, PMF analysis was conducted. The best fit was obtained with four factors. The qualitative identification of sources was carried out by the explained variation profile, which shows the proportion of variance explained by each factor for different metal elements (Fig. 4).

As shown in Fig. 4, factor 1 explains about 98% of As, 86% of Sb, and 71% of Hg. This result is similar to the PC1 determined by PCA. Therefore, factor 1 relates to the anthropogenic behavior especially mining. Likewise, about 69% of Cd, 44% of Zn, and almost 100% of CaO are explained by factor 2, which is similar to PC2. Factor 3 explains about 57% of Na_2O and K_2O. Factor 4 explains about 52% of Fe_2O_3 and 44% of MgO. The results of factors 3 and 4 are similar to those of PC3 and PC4, respectively.

The contribution proportion of each factor was calculated by the source contribution matrix obtained from PMF analysis. The result is shown in Fig. 5. The contribution proportions from factor 1, 2, 3, and 4 are 35, 22, 17, and 26% respectively, which are similar to those of PC1, PC2, PC3, and PC4 obtained from PCA/MLR analysis.

4. Conclusions

The mean concentrations of most metal elements in soil along the Chao River are higher than their background values. The mean concentrations of Hg, Cr, and Ni are even higher than their soil standard values. Based on the Nemerow index results, there is a moderate potential pollution risk of Hg, and a lightly potential pollution risk of Ni and Cr in the study area. PCA/MLR and PMF methods were used to study pollution sources. Four kinds of pollution sources are identified, which are mining source, natural source, vehicular emission, and coal combustion. The contribution proportion of each source is PC1 (factor 1) 33–35%, PC2 (factor2) 22–29%, PC3 (factor3) 17–27%, and PC4 (factor 4) 11–26%, respectively. These results suggest complex anthropogenic pollution condition in Miyun area. More attention should be paid on the management of mining, transportation, and coal combustion.

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