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Apportionment and risk assessment of heavy metals in sediments of the Chaiwen River flowing through the Xinwen mining area

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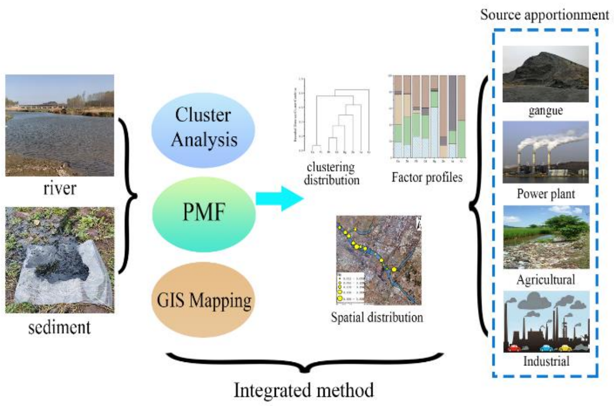
Graphical abstract



Abstract

The environmental quality of rivers is a comprehensive effect of the superposition of multiple sources, and distinguishing the contribution of coal mining areas to river is an important and interesting work. In order to explore the impact of heavy metal in the sediment and to distinguish the contribution patterns of various pollution sources in the Xinwen section of the Chaiwen River in Shandong Province of China, a cluster analysis and positive definite matrix factor analysis combined method were proposed and used to analyze the heavy metal content of Cu, Pb, Zn, Cd, Cr, Ni, Hg, and As in the sediment of the main channel of the Chaiwen River, and the ecological risk analysis was subsequently conducted. The results showed that the content of 8 heavy metals in the sediment of Chaiwen River exceeded the soil background value, with Cd and Hg exceeding the standard more severely. The spatial distribution of heavy metals was closely related to the distribution of pollution sources around the Chaiwen River.The sources of heavy metals were coal mining, agricultural pollution, industrial pollution, power plant pollution, and natural sources, with their respective contribution rates of 27.40%, 22.51%, 19.09%, 7.19%, and 23.81%, respectively. According to ecological risk assessment, Cd and Hg pollution in sediments was relatively severe. The results in the case study can provide a reference for the ecological environment management of coal mining areas in the lower reaches of the Yellow River.

Keywords: Coal mining area; River sediment; Heavy metals; Source Apportionment; Ecological risk assessment

1. Introduction

Due to rapid urbanisation and industrialisation, heavy metal pollution in river sediments has attracted great attention worldwide (Men *et al.* 2020; Xiang *et al.* 2022). Among a large number of pollutants in river sediments, heavy metals have become an important pollution factor affecting the environment of water bodies due to their easy accumulation, difficult degradation and strong toxicity (Li *et al.* 2022). At the same time, due to the poor hydrodynamic conditions and weak hydrodynamic exchange capacity of the estuarine waters, most of the heavy metals in the water body are finally enriched in the sediments through adsorption, desorption and deposition of suspended substances, which makes the river sediments become an important sink for heavy metals (Miranda *et al.* 2021). When the depositional environmental conditions change, heavy metals will be re-released into the river water environment causing "secondary pollution", which directly endangers the ecological environment (Zhang *et al.* 2019). Therefore, the analysis of heavy metal sources and ecological risks in river sediments has important research value for exploring the impact of human activities on the ecological environment and characterising the regional environmental quality and development trend. For example, if the concentration of iron in the soil is too high, it will affect the growth of rice (Hasan *et al.* 2022). In addition, high levels of heavy metals in soil pose a serious threat to human and animal health because heavy metal ions can readily enter the bodies of humans and animals through inhalation, dermal absorption or ingestion (Liu *et al.* 2022).

At present, the methods for tracing heavy metals can be mainly divided into two categories of methods: source identification and source analysis. Among them, source identification can make qualitative judgement on pollution sources, but cannot determine the contribution pattern of different pollution sources (Wang *et al.* 2023). Source identification methods mainly include principal component analysis, factor analysis and so on. Source analysis methods can calculate the contribution of different pollution sources to the environment of the study area (Shi *et al.* 2024), and these methods mainly include Positive Definite Factor Matrix (PMF) model (Wu *et al.* 2020), UNMIX model (Zang *et al.* 2022), isotope ratio method (Sun *et al.* 2011) and so on. In recent years, many researchers have analysed the contribution of different sources of pollution with the help of Positive Definite Factor Matrix Model (Li *et al.* 2018). The number of factors in this method is determined by human, and the number of factors can be optimised with the help of cluster analysis (Huang *et al.* 2015) and Principal component analysis (Liu *et al.* 2024).

Dawen River is the largest tributary of the lower reaches of the Yellow River, originating from the northern foot of the spinning mesa, converging with the waters of the Taishan Mountain Range and the Mengshan Branch, passing through the counties and cities of Laiwu and Xintai from the east to the west, and then injecting into the Dongping Lake, and then entering into the Yellow River after exiting from the mouth of the Chenshan Mountain (Wang *et al.* 2015). The main river channel is 239 kilometres long, with a watershed area of 9098 square kilometres. The Chaiwen River is an important tributary of the DaWen River. There are a variety of mineral resources in the river basin, among which coal reserves are relatively rich, and mining activities in some of the mines have a hundred years of history (Hua *et al.* 2018). However, long-term mining activities have eroded the regional ecological environment and caused drastic disturbance to the surrounding environment. Chaiwen River is one of the three major tributaries of the upper source of the Daven River, a tributary of the Yellow River revealing the impact pattern of mining activities on the environment of its upper watershed will provide important support in terms of investigating the environmental conditions of the mining areas in the Yellow River Basin, governance, and ecological environment restoration.

2. Study area

Chaiwen River is a major tributary of the upper reaches of the Da Wen River, most of the basin in the territory of Xintai City, with a total length of 116 km. Xintai City has a unique geomorphology, with three mountain ranges stretching almost parallel to each other from the north, centre and south of the city, from northwest to southeast, and intersecting in the eastern part of the city, with the overall shape of an E. The hills to the south of the central range are the plains to the north. The central mountain range is hilly in the south and plain in the north. The study area (117°40′9″～117°44′46″E, 35°53′46″～35°49′52″N) has a temperate continental monsoon climate, with an average annual temperature of 13.9°C and an average annual precipitation of 730.2 mm. The Chaiwen River basin is distributed with a number of coal mines, and there are a large number of residential areas, industrial zones, and agricultural land, which have a complicated impact on the river (Hua *et al.* 2018).

3. Sampling, experiments and methods

3.1. Sample collections and concentration determination

A total of 18 surficial river sediments samples were sampled in April 2023 within consecutive days along the Chaiwen River. GPS was used to locate the location of the sampling sites, and the specific sampling locations are shown in **Figure 1**. The samples collected in this study were near-bank sediments of the river, and the surface sediment samples were collected with stainless steel shovels and sealed with polyethylene bags. The samples were dried and crushed, and the sediment samples were digested using method of HNO_3-HF-$HClO_4$. The inductively coupled plasma mass spectrometer (ICP-MS) was used to analyze the concentration of Cu、Pb、Zn、Cd、Cr and Ni. And Hg, As concentration was determined by atomic fluorescence spectrometry (AFS).

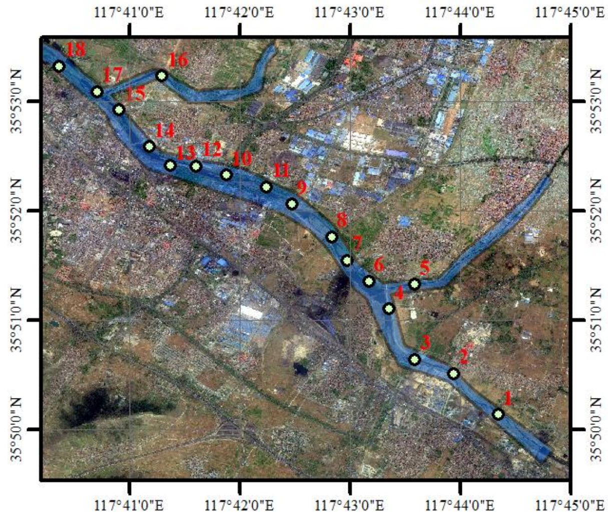


Figure 1. Location of study area and distribution of sampling sites

3.2. Source apportionment

3.2.1. Cluster analysis

Cluster analysis is a multivariate statistical method used to classify multiple research indicators and to distinguish similarities between different data sources (Bonetto *et al.* 2022; Frades and Matthiesen 2010). In the analysis of soil heavy metal pollution sources, the distance in the cluster analysis result graph indicates the correlation between heavy metal elements. The closer the distance, the higher the similarity and the stronger the correlation (Luo *et al.* 2021; Shokr *et al.* 2022). With the help of the results of cluster analysis, the number of pollution factors in the study area can be judged to provide a basis for predicting the number of PMF model factors.

3.2.2. PMF model

Positive definite matrix factorization (PMF) is a factor analysis method based on least squares, which is based on the least squares method, the decomposition matrix is not negatively constrained, and the standard deviation of the data can be used for optimization (Ambade *et al.* 2023). The basic principle of the PMF 2D model is to split the pollutant content matrix into a source component matrix and a source contribution matrix. The difference between the pollutant content matrix (actual data) and the split source component matrix and source contribution matrix (parsed data) forms the residual matrix (Zerizghi *et al.* 2022). In this research, PMF 5.0 was adopted to source apportionment of heavy metals in soils (EPA 2014).

$$X_{ij} = \sum_{k=1}^{p} G_{ik} F_{kj} + E_{ij}$$

where X_{ij} is the concentration of the j_{th} element in the i_{th} sample; F_{kj} is the concentration of the j_{th} element in the k_{th} source, G_{ik} is the contribution to i_{th} sample, and E_{ij} is the k_{th} source is the residual matrix.

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\left(\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}}\right) = \sum_{i=1}^{n}\sum_{j=1}^{m}\left(\frac{e_{ij}}{u_{ij}}\right)^2$$

$$u_{ij} = \begin{cases} \frac{5}{6} \times MDL, (c \le MDL) \\ \sqrt{(\sigma \times c)^2 + (0.5 \times MDL)^2}, (c > MDL) \end{cases}$$

3.2.3. *Ecological risk index evaluation*

The potential ecological risk index proposed by the Swedish scientist Hakanson (1980) is still widely used today as a diagnostic method for water pollution control (Hua *et al.* 2018; Min *et al.* 2013; Qian *et al.* 2017) Heavy metal background, load, and bioavailability are taken into account in the latent ecological index. The formula is as follows

Single heavy metal pollution index

$$C_f^i = C^i / C_n^i$$

Among them, C_f^i is the single heavy metal pollution index, C^i is the measured concentration of a heavy metal in coal gangue, C_n^i is the background value of heavy metal (BGV), and superscript i is the specific pollutant.

Potential ecological risk index

The potential ecological risk index combines environmental ecological effects with toxicological content, which can be used to evaluate potential environmental risks.

$$E_r^i = T_r^i \times C_f^i$$

where E_r^i is the potential ecological risk factor for each heavy metal, T_r^i is the toxicity factor of i_{th} heavy metal, Cu = Ni = Pb = 5, Cr = 2, Cd = 30, Zn = 1.

Comprehensive ecological risk index

$$RI = \sum E_r^i$$

Among them, RI is the comprehensive ecological risk index (**Table 1**).

Table 1. Potential ecological risk index level

E_r^i	RI	Potential ecological risk level
<40	<150	Low risk
40 ~ 80	150 ~ 300	Medium risk
80 ~ 160	300 ~ 600	Higher risk
160 ~ 320	>600	High risk
>320	—	Extremely high risk

4. Results and discussion

4.1. *Descriptive statistical analysis of heavy metals in sediments in the Chaiwen River Basin*

The heavy metal contents of the sediments in the study area are shown in **Table 2**. It can be seen from the table that the median values of Cu, Pb, Zn, Cd, Cr, Ni, Hg, and As were 36.2, 31, 168, 1.235, 101, 32, 0.184, and 7.715 mg/kg, respectively, and the median values of all the heavy metals were higher than the background values of the soils in Shandong Province. This suggests that mining activities around the Chaiwen River can lead to heavy metal pollution in the surrounding water system. The coefficients of variation (CoVs) (Li *et al.* 2020) for Cu, Cd, Hg and As were 63.09%, 63.85%, 53.08% and 265.2%, respectively, which are high, indicating that these heavy metals are highly influenced by human activities.

Table 2. Descriptive statistics of heavy metals in sediments (mg/kg)

variable	Cu	Pb	Zn	Cd	Cr	Ni	Hg	As
Median	36.2	31	168	1.24	101	32	0.184	7.7
Mean	40.2	32	178	1.40	105	33	0.194	20.8
Maximum	122.0	52	319	4.41	173	85	0.385	241.0
Minimum	16.5	15	64	0.54	32	14	0.051	3.2
Coefficient of variation (%)	63.09	29.94	40.48	63.85	35.85	45.52	53.08	265.2
Soil background value in Shandong Province	22.9	23	64	0.13	59	27	0.030	7.4

4.2. *Spatial distribution characteristics of heavy metals in sediments*

Using ArcGIS 10.8, the heavy metal content was graded and displayed to generate the spatial distribution map of heavy metals (**Figure 2**). Analysing **Figure. 2**, Cd and Hg showed similarity in spatial distribution, and the high value points were distributed in D12~D13, which were located near the gangue mountain, and might be related to gangue wastewater pollution. The point with the highest As content was D10, which was located near the power plant, indicating that the As pollution was related to the power plant. Cu, Pb, and Ni were similar in spatial distribution, and there were high value areas in D8~D9 and D12~D14, indicating that these three elements might have two sources of pollution. Cr was in the upper, middle, and middle levels, and there were two sources of heavy metals. Cu, Pb and Ni have similar spatial distribution, with high value areas in D8~D9 and D12~D14, suggesting that there may be two sources of pollution for these three elements.

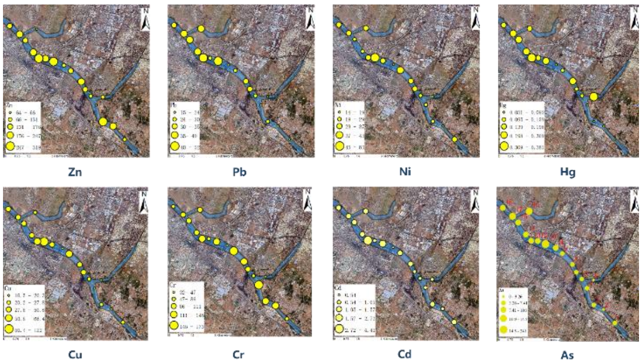


Figure 2. Spatial distribution of heavy metals in sediments

4.3. Analysis of the source of heavy metals in sediments

4.3.1. Cluster analysis

In recent years, many researchers have analyzed heavy metal pollution in the environment with the help of cluster analysis to obtain the links between different heavy metal elements (Jiang *et al.* 2021; Panda *et al.* 2020). At this stage, the research of cluster analysis in heavy metal pollution is mainly used for principal component analysis (Liu *et al.* 2020; Saravanan and Ramesh 2024). However, it can also determine the number of main pollution factors when extracting principal components, which can provide support for the determination of the number of factors in PMF analysis and optimize PMF analysis. In order to explore the correlation between various heavy metal elements in the sediments of the study area, the heavy metal data were clustered and analyzed to explore the correlation between different heavy metal elements (Tumuklu *et al.* 2023) (**Figure 3**). The correlation between Cd, Hg and As is very strong, and the correlation between Cd and Hg is particularly significant, which indicates that Cd-Hg and As are originated from two pollution sources, and the three elements may have the same pollution source, and the correlation between Cu, Pb and Ni is very significant, which indicates that they are strongly correlated with one source. The correlation between Cu, Pb and Ni is very significant, indicating that the three elements are strongly correlated with one source of pollution. In addition, the correlation between Zn and Cr is weak, indicating that they belong to two different sources of pollution.

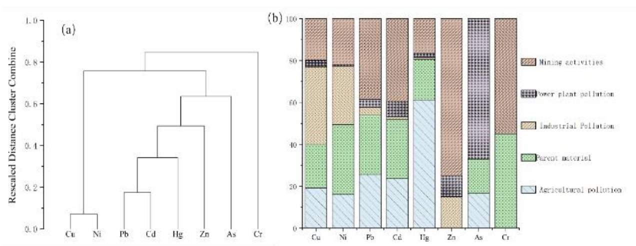


Figure 3. Sediment clustering distribution map(a) and factor profiles from the PMF model(b)

4.3.2. PMF model

Most PMF analyses use heavy metal content as a reference in determining the number of factors, but supporting evidence is lacking (Kim *et al.* 2023). In order to improve the scientific validity of the number of factors at the time of calculation and to provide effective support for PMF, we used cluster analysis to determine the number of major pollution sources in the study area. Based on the results of cluster analysis, in order to further analyze the contribution of each heavy metal source in the study area, the heavy metals in the study area were quantified with the help of PMF model. The heavy metal data and uncertainty data were imported into the EPA PMF 5.0 software, and the signal-to-noise ratios of the eight heavy metal elements were all much greater than 1, and they were classified as "Strong" (Wu *et al.* 2020). According to the results of cluster analysis, it can be assumed that the factor number range is 3~5, and 20 iterations were carried out in Robust mode, when the factor number is 5, A is less than 1.5, and most of the residual values are concentrated in -3~3. Finally, 5 factors were analyzed, and the results are shown in **Figure 4**

Factor 1 explained 22.51% of the heavy metal sources, with mercury contributing more at 61.21%. The mean ground level accumulation index of mercury indicated that the level of heavy metal pollution in the Chaiwen River Basin was moderate to severe and the correlation of mercury with lead and cadmium was significant at $P<0.01$. It was found that mercury was a significant indicator of industrial emissions. Atmospheric deposition is usually labelled as a significant source of lead accumulation in soil. Industrial activities can cause significant lead and mercury pollution (Chen *et al.* 2022; Lv 2019). Based on the distribution of heavy metal concentrations, it was found that the areas of high concentrations of lead and mercury were mostly concentrated near industrial parks and in the areas of main urban roads. Therefore, it is assumed that factor 1 may be related to industrial activities and atmospheric deposition.

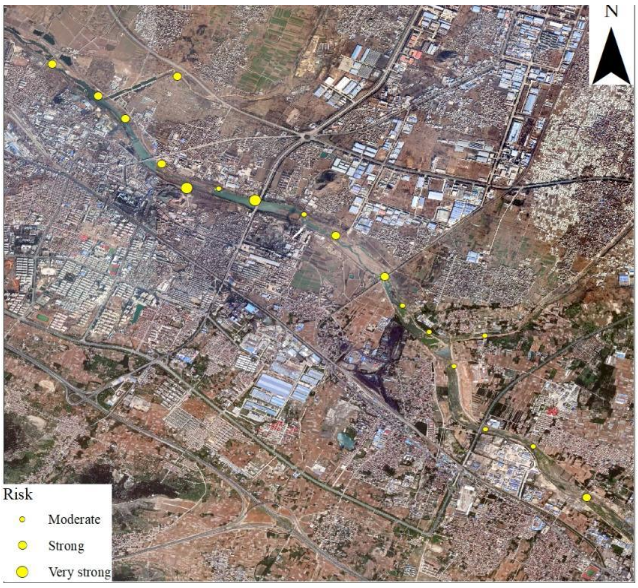


Figure 4 Spatial distribution of ecological risk from heavy metals

Factor 2 explains 23.81 per cent of the heavy metal sources, with a higher weighting for nickel. Previous studies have found that nickel is associated with the soil matrices that produce rocks and is widely present in the soil formation process. The low degree of variability of nickel indicates that it is less affected by human activities (Zang *et al.* 2022). In this study, the degree of Ni variability in the study area is at a low level, and in the evaluation method of the ground accumulation index, Ni belongs to the non-polluted level, and its average value is similar to the background value of soils in Shandong Province, which can be considered to come from the natural geological background according to previous studies. According to **Figure 4**, it can be seen that Factor 2 is distributed in each metal, so Factor 2 is defined as the parent material source.

Factor 3 explained 19.09% of the heavy metal sources. The highest contribution was made by Cu with 36.84 per cent. The results of correlation analysis showed that Cu and Zn were significantly correlated. Agricultural production is often highlighted as the main source of Zn, Cu and Cd accumulation in Chinese soils, with phosphate fertilisers, pesticides and organic fertilisers often containing considerable amounts of Zn, Cu and Cd. In the study area, crop production is often accompanied by high levels of fertiliser and pesticide use (Wang *et al.* 2022). Based on field surveys and spatial distribution of heavy metals, most of the areas with high levels of Cu and Zn pollution are located near agricultural land. Therefore, factor 3 may be related to agricultural surface sources.

Factor 4 explains 7.19% of the heavy metal sources, with As contributing 67.01% of the total. It was found that coal combustion leads to As pollution in different degrees, and also according to the spatial distribution of heavy metals, it was found that As has a significant enrichment around the power plant (Guo *et al.* 2021). The geoaccumulation index method shows that As is strongly to very strongly polluted near the power plant, while it is not polluted at other locations. Therefore, factor 4 was defined as a source of coal combustion and power plant pollution .

Table 3. Ecological risk classification

RI	**Risk level**
<150	Slight
150 ~ 300	Moderate
300 ~ 600	Strong
>600	Very strong

Table 4. Integrated ecological risk levels and sources of risk

	RI	**Risk level**	**Main sources**	**Secondary sources**
D1	310.22	Strong	Agricultural pollution	
D2	220.09	Moderate	Mining activities	
D3	298.74	Moderate	Mining activities	
D4	235.97	Moderate	Parent materials	
D5	225.83	Moderate	Agricultural pollution	
D6	219.16	Moderate	Parent materials	
D7	292.16	Moderate	Industrial Pollution	Parent material
D8	424.62	Strong	Agricultural Pollution	Mining activities
D9	390.38	Strong	Industrial Pollution	Parent materials
D10	1016.37	Very strong	Power plant pollution	
D11	155.55	Moderate	Mining activities	Industrial Pollution
D12	289.13	Moderate	Industrial Pollution	
D13	1103.88	Very strong	Mining activities	Agricultural Pollution
D14	400.29	Strong	Agricultural pollution	Mining activities
D15	398.24	Strong	Agricultural pollution	
D16	356.24	Strong	Agricultural pollution	Parent materials
D17	405.4	Strong	Agricultural Pollution	
D18	385.25	Strong	Industrial Pollution	Agricultural Pollution

Factor 5 explained 27.40% of the heavy metal sources, with Cr contributing more with 54.93%. The results of correlation analysis showed that Cr and none of the other heavy metals showed significant correlation, indicating that the source of Cr pollution was not similar to the other seven metals (Kim *et al.* 2023). According to the spatial distribution of heavy metals, Cr has obvious enrichment near the gangue mountain, and the ground accumulation index method shows that the pollution degree of Cr near the gangue mountain is from strong pollution to very strong pollution. The study shows that in the process of coal mining, coal mine dust, slag and groundwater outflow can bring out heavy metals such as chromium, causing Cr heavy metal pollution to the surrounding environment (Hua *et al.* 2018). Therefore, factor 5 is defined as the pollution caused by tailings and gangue (**Table 3**).

4.3.3. Ecological risk assessment

A comprehensive ecological risk assessment was carried out for 18 sites in the Chaiwen River Basin, and the comprehensive ecological risk level of each site was calculated (Liu *et al.* 2022; Song *et al.* 2023), and the main sources of heavy metals at different sites were analysed by combining the results of heavy metal source analysis and site investigation, and the results are shown in **Table 4**.

Comparing the calculated potential ecological risk factors with the potential ecological risk scale, it can be found that the potential ecological risk of Cd in the Chaiwen River Basin is high, and it can reach very high risk at individual points; comparing the comprehensive ecological risk index with the scale, it can be found that the Chaiwen River Basin as a whole presents a medium risk in the upstream, and a high risk in the downstream.

5. Conclusions

In this work, cluster analysis was combined with a positive definite factor matrix model to identify the sources of contaminants in river sediments and applied to an ecological risk assessment to identify the most significant sources of contamination in the sediments of the Chaiwen River. The concentration of heavy metals were all higher than their background values, especially, Cd and Hg were the most significant. The most important factor contributing to the pollution of the Chaiwen River was the historical impacts of the coal mining activities (27.40%), followed by industrial (22.51%) and agricultural (19.09%) activities which were also important sources of the contamination. In this study, the positive definite factor matrix analysis was optimized with the help of cluster analysis, which provided scientific support for determining the number of factors in PMF analysis. The method was also combined with ecological risk assessment to propose a method for identifying the main sources of pollution in polluted areas, which provides a method for subsequent targeted environmental management.

Author contributions

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Yuanhao Wang, Qiuhui Yao, Guangzhu Zhou. The first draft of the manuscript was written by Yuanhao Wang and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials

The authors confirm that the data supporting the findings of this study are available within the article.

Ethics approval and consent to participate

Not applicable

Consent for publication

Not applicable.

Competing interests

The authors have no relevant financial or non-financial interests to disclose.

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