Implementation of an Integrated Health Risk Assessment Coupled with Spatial Interpolation and Source Apportionment: A Case Study of Soil Heavy Metals, China

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Research Article

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Implementation of an integrated health risk assessment coupled with spatial interpolation and source apportionment: A case study of soil heavy metals, China

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Abstract

Soil heavy metal contaminated sites with multiple sources of pollution have caused worldwide public concern. However, the lack of correlation of risk assessment between source identification of heavy metal led to unclear direction of source governance. A methodology was established by combining source apportionment of human health risks with ecological enrichment to characterize source-identified risks of heavy metals based on Ordinary kriging interplotation. Principal component analysis (PCA) and positive matrix factorization (PMF) model were used to identify and classify potential sources of heavy metals synthetically. The integrated results were incorporated into the health risk model to evaluate potential non-carcinogenic and carcinogenic risk of soil heavy metals. A case study was conducted in Suzhou city of China. The results showed that concentrations of Cd and Hg were highly above the background values, accounting for percentages of 239.6% and 415.9% above background values, respectively. The source contributed human health risk index of As contributed 76.9% to non-carcinogenic risk by pollutant sources of

agriculture activities. The Non-cancer health risk index for children and adults was 1.08 and 1.00 respectively. The cancer health risk was 3.67E-03 for children and 3.97E-04 for adults. Cr originated from indutriy activities, accounting for 29.5% of total heavy metals, and constituted the largest carcinogenic impact on the population. This study provided a new insight for the treatment of mutil-sources of soil heavy metal pollution and also some reference value for the improvement of the risk assessment system.

Keywords:

Soil heavy metals; Spatial interpolation; Global data optimization; Quantitative source apportionment; Source-identified health risk

1. Introduction

Soil was the most important natural resource of a country and the material basis of the environment where human beings live on (Liang et al., 2017; Järup, 2003). However, with the increasing agricultural production, industrial activities and human activities, great pressure was put on the soil quality continually, resulting in direct soil pollution, especially soil heavy metal pollution (Ihedioha et al., 2017; Salmani-Ghabeshi et al., 2016; Luo et al., 2011). The heavy metal pollutants in soil mainly include Hg, Cd, Pb, Cr, Cu, Ni, Co, Sn and the metal-like As. Distinc from other pollution, heavy metal pollution was not easy to be leached with water and decomposed by microorganism in soil (Xie et al., 2016; Papa et al., 2010). There were four main ways in which heavy metals entered the soil: atmospheric deposition (natural and man-made), fertilizers containing metal impurities, industrial emission, and irrigation of contaminated water (Luo et al., 2015; Liu et al., 2017). Afterwards heavy metals entered the food chain through accumulation in plants and have obvious biological enrichment, posing risks to the ecosystem and human health (Han et al., 2006; Kurt-Karakus, 2012). In addition, heavy metal was often tightly bound to soil solids, and even small increases in the current concentration of heavy metals can have potentially adverse effects on soil quality (Islam et al., 2017; Noll, 2002). Due to the increasingly serious soil pollution, it was important to predict the accumulation trend of heavy metals in soil and understand the risk level of heavy metal pollution (Teng et al., 2010). Therefore, it

was of far-reaching significance to effectively assess the pollution situation, including potential local ecological risks, identify potential pollution sources, so as to provide information for soil pollution control (CSC, 2016). So far, many research groups had worked on the investigation of soil pollution (Couto et al., 2018; Mendoza et al., 2017). However, most of these studies focus on agricultural pollution or water pollution, and there were comparatively few studies on the soil pollution of abandoned dye factory (Gholizadeh et al., 2019; Xiao et al., 2019; Mohammadi et al., 2020;). The soil environment of many factories in the whole country of China was not optimistic and was serious in some areas (Chen et al., 2015). To strengthen environmental management, a series of policies were implemented to focus on seriously polluted enterprises and force the closure of the small and heavily polluted factories. The pollution of some heavy metals in the soil left by such closed enterprises deserved our attention. The site of the original production enterprises in the process of development, especially dyeing factories, may be polluted by heavy metals (Mao et al., 2019). Pollutants often existed in waste water and waste residue which may leak through the staging point, migration of elution infiltration into the soil and rivers (Wcislo et al., 2016). Therefore, the original location and the surroundings may be polluted, leading great harm to the ecological environment and human health. So it was necessary to scrap the dye factory for soil pollution characteristics, ecology and human health risk assessment. Scholars at home and abroad had carried out extensive studies on soil pollution characteristics and human health risk assessment in the surrounding areas such as coal production, transportation and storage, as well as coal-fired power plants, but there were few reports on the investigation and assessment of soil pollution in abandoned dyestuff factories (Jia et al., 2015). Therefore, it was necessary to further study the soil pollution in the abandoned dye factory area in order to make better use of the site and provide reference for the site remediation in the next step.

To reduce the hazard of soil heavy metals, source apportionment can be an effective tool to quantify the pollution sources of heavy metals in polluted sites (Jiang et al., 2019; Guan et al., 2018). Even if there were some studies on the classification of pollution sources of soil heavy metals. Studies organically combined the contribution rate of source analysis with human health risk had also been reported, but few about systematic relevance between them

(Xiao et al., 2019). In order to better understand the pollution level of heavy metals in soil, spatial interpolation method was often used to obtain the concentration distribution of heavy metals in soil. In addition, enrichment factor (EF) was also calculated to obtain the ecological enrichment information of soil pollution (Tian et al., 2017; Nazzal et al., 2016). It was essential to find out further information about the characteristic distribution, pollution sources and environmental risks of heavy metals in polluted sites (Jiang et al., 2017; Niu et al., 2020). Identifying potential pollution sources of heavy metals was the basic premise for designing targeted pollution control measures. Although source apportionment and environmental risk assessment of heavy metal pollution had been carried out in many studies and the study of source-specific health risk and potential ecological risk based on the contribution rate of source analysis was also been reported (Peng et al., 2016). However, they mainly focused on the heavy metals from atmospheric and urban river-lake system (Tapia-Gatica et al.,2020; Li et al., 2020). Metal pollution from abandoned dye factories had been scarely reported, let alone the overall impact of health risks on the study area. Positive matrix factorization (PMF) and Principal component analysis (PCA) was widely used for pollution source analysis in various media such as the atmosphere, hydrosphere and soil sphere (Liang et al., 2017a; Lang et al., 2015). It can be applied to apportion the potential sources of heavy metals (Norris et al., 2014). Compared with traditional multivariate statistical analysis methods, PMF was able to deal with the measurement uncertainty inherent in environmental data (Paatero and Tapper, 1994; Lv and Liu, 2019). Quantitative source analysis and its contribution research were helpful to identify the main pollution sources and quantified their contribution rate (Yang et al., 2019; Kumar et al., 2019).In addition, the city we studied, Suzhou, as a developed industrial area, the closure of polluting factories will inevitably leaded to the intensification of other human activities driven by the economy (Zhang et al., 2018). Therefore, there was the need to conduct the first detailed evaluation of the total effects of health risks on soil heavy metals associated with the areas related to a dye factory in the southeast of Suzhou by using multivariate statistical and spatial analysis methods.

This paper mainly dealt with: (i) the optimized spatial interpolation of heavy metal concentration to obtain global

data in the sampling sites with reference to the Soil Guide of China (National Environmental Protection Agency, 1995);(ii) Multivariate statistics and spatial distribution of ecological enrichment for analyzing the ecological pollution levels; (iii) identification of potential sources of soil heavy metals pollution and quantification of the proportions of various sources through combination of PCA and PMF models; (iv) Calculation of source-contributed human health risk index; (v) total effects of health risks on soil heavy metals. This study can provide reference for soil remediation and protection design in the study area and also reference for policy formulation and protection research on soil heavy metal pollution in other areas.

2. Materials and methods

2.1. Case study

2.1.1. Study area and sampling

Suzhou was located in the middle of the Yangtze river delta, the southeast of Jiangsu province, located at 119°55'E to 121°20'E, 30°47"N to 32°02'N, east of Shanghai, south of zhejiang, west of taihu lake, north of the Yangtze river, the total area of 8657.32 square kilometers. The city was low-lying and flat, with many rivers and lakes. Most of the water surface of taihu lake were in suzhou. Rivers, lakes and beaches accounted for 36.6% of the city's land area. Suzhou was a subtropical monsoon maritime climate, with an average temperature of 17.8℃ and precipitation of 1369.2mm in 2018. The prevailing wind direction was southeast wind all year along, with four distinct seasons, mild climate, abundant rainfall, fertile land, rich natural conditions and so on. In the shallow layer, the clay soil with slight deformation and high strength was mainly grey, with compact texture.

2.1.2. Sample collection and chemical analysis

According to the land use pattern, topographic features and perennial wind direction in the study area, 30 composite topsoil samples in the 0-7cm layer were randomly collected for measurement in Suzhou in October 2019 and the exact location of each point was recorded in **Fig .1**. The soil samples were taken back to the laboratory and naturally dried and ground crushing, first through a 20 mesh sieve for pH analysis, then used for the determination of physical

and chemical properties (Pal et al., 2019). The processed soil samples were dissolved in the mixture acid solution (HNO_3-HF-$HClO_4$) at a high temperature of 210℃ for 4h (Bryanin et al., 2019). Soil pH was determined with a ratio of 2:5 (w/v) soil/water mixture using a pH meter (Cheng et al. 2018). The soil organic matter (SOM) content was determined by the chromic acid titration method. The content of heavy metals As, Cd, Cr, Zn and Pb in soil samples were determined by by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS, PerkinElmer ELAN 9000). Atomic fluorescence photometer (AFS-230E) was used to analyze the content of Hg. Each batch of samples was evaluated by reagent blanks to reduce errors for quality assurance (Jiang et al., 2019).

To ensure the quality of analysis, several quality assurance and control methods were conducted as the following standard operating procedures. Blank reagents, duplicate samples and soil standard reference samples (GBW07401-07408, Beijing; National Center for Standard Materials of China) were operated in the Analytical and Testing Center of Suzhou research academy of North China Electric Power University. Chemical reagents are guaranteed reagents. The logarithmic deviation of the system was within the range of ±0.045 (Zhang et al., 2017), and the relative percentage difference between duplicate samples was within the range of ±7.50% (Han et al., 2006). Therefore, the precision and bias of the analysis were generally below 5%.

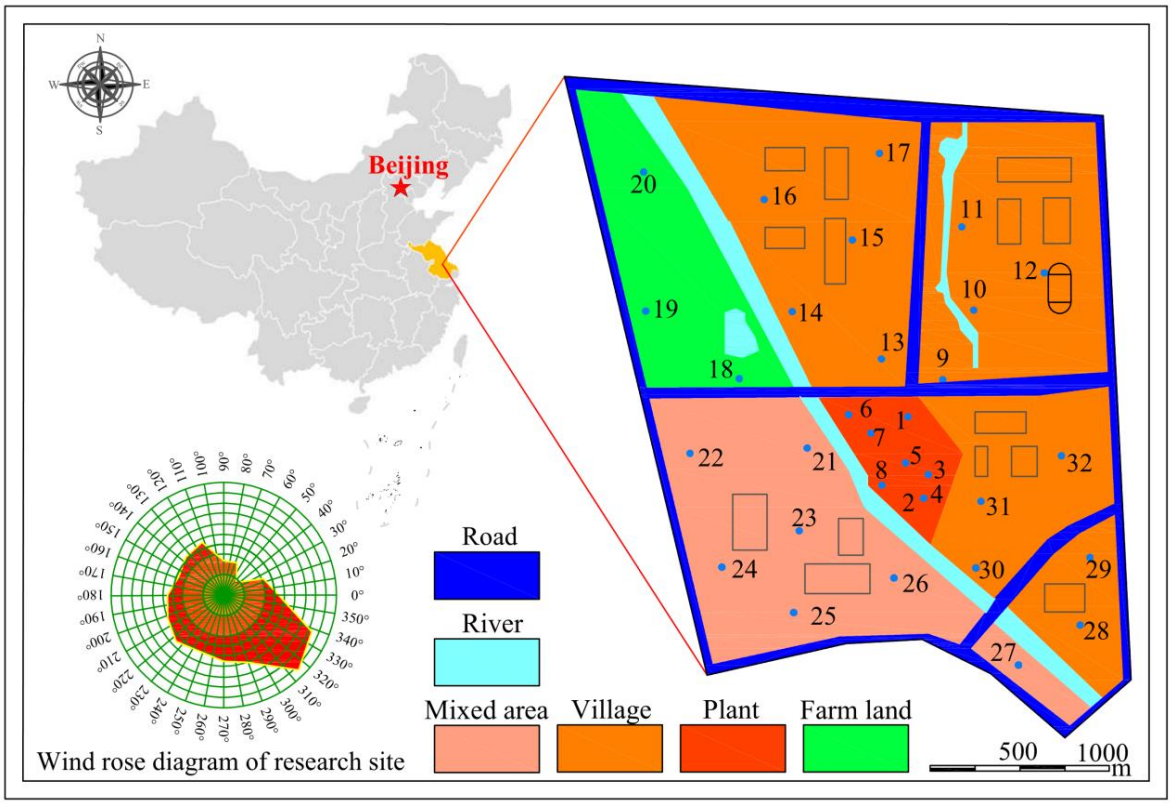


Fig. 1. Sampling sites of soil heavy metals in the study area.

2.1.3. Statistical analysis

Microsoft Excel (version 2010, USA) was used for calculations. The spatial distribution of soil heavy metals was mapped by the Surfer v.12.0 (Golden Software Inc., CO, USA) and ArcGIS 10.7 software. SPSS Statistics 25 (IBM Inc., CO,USA) was used to conduct the analyses and to obtain the relevant parameters. The data underwent by EPA-PMF 5.0 to assess the sources of soil heavy metals with a 95% confidence interval (significance $p < 0.05$). Crystal Ball Software (Version 2000, Decisioning, Denver, CO, USA) was employed for uncertainty analysis.

2.2. Overview of the framework

The city of Suzhou studied in this paper has a thriving ecotourism industry and there are abandoned industrial polluted sites that have not been treated. Therefore, it is necessary to pay attention to the environmental health risks in this region. The objective of this study is to conduct a combination of source analysis and health risk assessment methodology to characterize the soil heavy metal pollution levels of the sources and source-specific human health risk. The integrated framework of the method combined source apportionment and source-specific human health risk

assessment was shown in **Fig. 2**.

Firstly, the pollution sources is determined qualitatively by PCA analysis, and then the PMF source apportionment is carried out according to the obtained results to accurately define the types of sources and the distribution rate of pollution sources, which complement each other. Secondly, the spatial distribution of heavy metals in contaminated sites were studied by spatial interpolation based on the comparison of Ordinary kriging and inverse distance weighted. Besides, the contribution rate of pollution sources was used for risk sharing and the assessment results were incorporated into the health risk assessment model to identify heavy metal from identified sources. Thirdly, toxic metal with a large contribution from pollution sources and high risk level was screened out and the correlation and total effects of health risks have also been analyzed. Finally, we identify which sources of contamination need attention and which toxic metals need to be prioritized for removal or reduction of toxic emissions. Based on the above information, targeted risk reduction strategies can be developed. It provided a theoretical basis for the government to control heavy metal pollution scientifically (Huang et al., 2006; Luo et al., 2011).

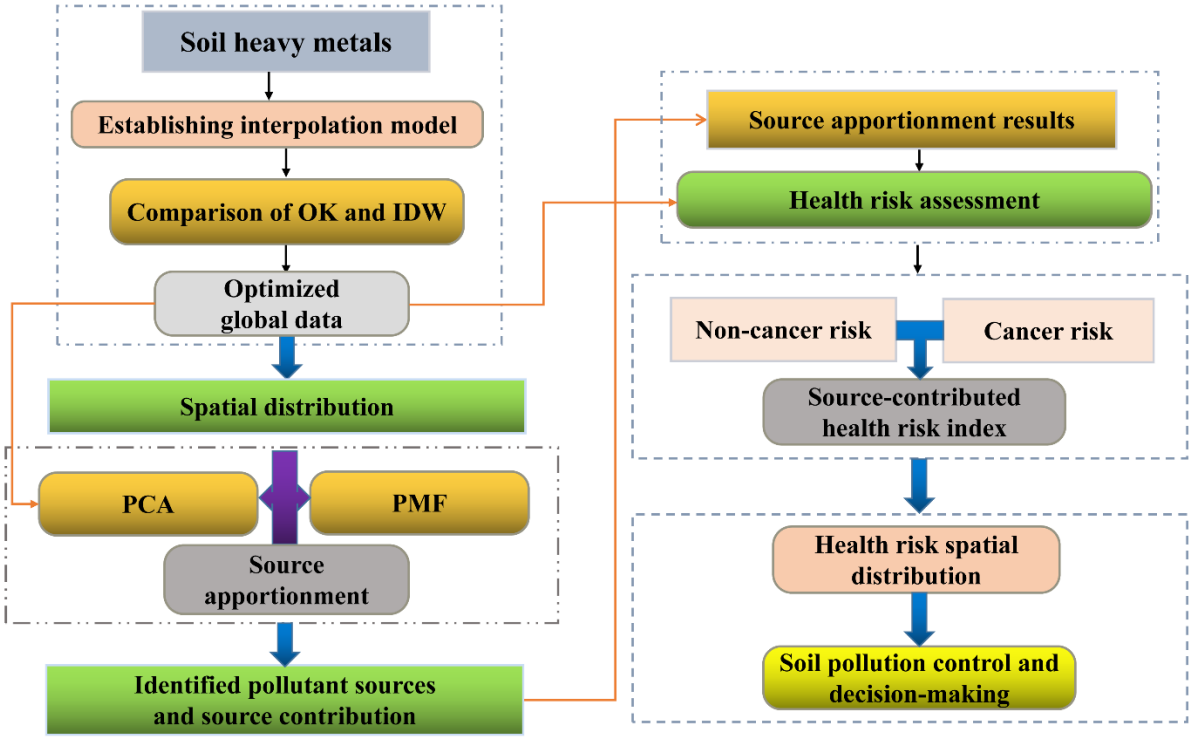


Fig. 2. Frameworks of the methodology for soil heavy metals.

2.3. Optimization of spatial interpolation for global data of heavy metals

Step 1: Ordinary kriging (OK)

Ordinary kriging was a type of kriging interpolation, which assumed that the property values were spatially stationary. OK interpolation routinely applied the weighted values of the attributes of known points as a predictor of the attributes of unknown points, which was mainly used to interpolate the parameters of un-sampled locations. The concentrations of soil heavy metals of the unknown point (x) was defined in **Eq. (1).**

$$C^t_{(x)} = \sum_{i=1}^{n} \lambda_i C^t_{(x_i)} \quad (1)$$

where, the $C^t_{(x)}$ was predicted concentration of soil heavy metals ***t*** (*t* = (As, Cd, Cr, Hg, Pb and Zn) at unknown site of ***x***; $\boldsymbol{C^t_{(x_i)}}$ was the content of metal *t* at point x_i (*i=1, 2...,* i,…j,... *n*);was the concentration at the sampling point $\boldsymbol{x_i}$; $\boldsymbol{\lambda_i}$ was the weight coefficient, which would be calculated by **Eq. (2).**

$$\begin{cases} \sum_{i=1}^{n}\sum_{j=1}^{n} \lambda_j \gamma(\boldsymbol{x_i}, \boldsymbol{x_j}) + \boldsymbol{\mu} = \gamma(\boldsymbol{x_i}, \boldsymbol{x}) \\ \sum_{i=1}^{n} \lambda_i = 1 \end{cases} \quad (2)$$

where, the $\boldsymbol{x_j}$ was another known point; $\boldsymbol{\mu}$ was the lagrangian constant; γ（$\boldsymbol{x_i}$,$\boldsymbol{x_j}$）was the semi-variogram function and could be obtained by **Eq. (3)**.

$$\gamma(h) = 1/2\, E\left[C^k_{(x)} - C^k_{(x+h)}\right]^2 \quad (3)$$

Where, *E* was the number of sample pairs at a separation vector *h*.

Step 2: Inverse distance weighted (IDW)

The IDW method has a wide range of applications in the field of geology, which can be simplified as the estimation process of calculating the spatial variables of the concentration of heavy metals at unmeasured locations by observing a large number of measured samples in the space close to the target location.

Compared with alternative interpolation methods, IDW will comprehensively analyze the acute values (outliers) within the data set and predict the values of unknown areas (Goix et al., 2013). In general, the more similar the soil characteristics between the adjacent and unknown locations, the better the prediction results (Zhou and Sha, 2013). The concentration of soil heavy metal $C^k_{(x)}$at the unknown point could be calculated by **Eq. (4)**.

$$C_x^k = \sum_{i=1}^{n} \frac{C_i^k}{D_i} \Big/ \sum_{i=1}^{n} \frac{1}{D_i} \quad (4)$$

Where, D_i ($i=1, 2..., n$) was the horizontal distance between interpolation point and reference point.

Step 3: Verification method

The accuracy of spatial interpolation methodology was verified by cross-validation model. The accuracy of both Ok and IDW interpolation methods was evaluated by examination of the errors of observed value (*OV*) and the predicted value (*PV*). The slope and determination coefficient (R^2) of fitting line between the *PV* and the *OV* approaching to 1 indicated that the method would be more accurate.

2.4. Mathematical models for source apportionment

The PCA model was utilised to identify the sources of soil heavy metal pollution qualitatively first, and then the PMF model was adopted for quantitative analysis to characterise the contribution of each source to soil heavy metal pollution. A combined analysis was eventually undertaken based on the types of pollution sources obtained from the two to accurately obtain the pollution sources and source contribution rates. The PCA/PMF model in combinatorial form can effectively validate the two with each other to obtain a thorough and comprehensive assessment.

2.4.1. Principal component analysis

Principal Component Analysis (PCA) was a statistical method that simplified the data structure primarily by dimensionality reduction, converting multiple variables into a small number of variables. PCA model could remodel the initial data into a collection of linearly freelance representations of every dimension through linear transformation, which can be used to extract the main feature components of data and was usually used for spatiality reduction of high-dimensional data. The application of PCA in environmental science has been relatively mature, which can effectively analyze the potential pollution sources and spatial distribution of heavy metals in soil (Xiao-Bo et al., 2009). PCA was often used to identificate the natural and anthropogenic sources of heavy metals by means of using chemical element principal factor loading (Cai et al., 2018; Hani and Pazira, 2011; Li et al., 2015).

2.4.2. Positive matrix factorization

The positive matrix factorization (PMF) method was one of the source analytical methods recommended by the U.S. Environmental Protection Agency (Paatero et al., 2010). PMF was a factor analysis method based on the least square method, which decomposed the matrix without negative constraints and can be optimized by means of standard deviation of data (Chen et al., 2011). It did not rely on the chemical composition spectrum analysis of pollution sources, but took the data sets of multiple soil samples and heavy metal elements as a matrix, and then decomposed the matrix into the contribution rate matrix and the source composition spectrum matrix (Dong et al., 2018; Franco et al., 2009). Through non negative constraint factor analysis and iterative calculation with the least square method, the objective function was minimized to solve chemical mass balance between the measured heavy metal concentration and pollution source (Guan et al., 2018). Compared with the traditional factor analysis method (such as FA-MLR), the PMF method could avoid the negative value in the result of matrix decomposition, so that the obtained source component spectrum and source contribution rate can be explained and had clear physical significance (Lv et al., 2019). In addition, the PMF used error estimates for each individual data point in order to cope with missing and inaccurate data more reasonably (Mamut et al., 2017). The data entered into the program include concentrations and equation-based uncertainties (Jing et al., 2014).

The PMF had a unique advantage because it could be used without source components as input. It was also applied to environmental data processing because it contained variable uncertainties associated with environmental sample measurements and forced all values to be non-negative (Xue et al., 2014). Compared with the traditional source apportionment methods, PMF was used to analyze pollution sources of heavy metals by weighing all data to analyze the contribution rate of target variables (Tian et al., 2018). The method based on matrix equation was as follows:

$$\boldsymbol{C_{ik}}=\sum_{1}^{p}\boldsymbol{g_{ip}f_{pk}}+\boldsymbol{e_{ik}} \quad \textbf{(5)}$$

$$\boldsymbol{Q}=\sum_{1}^{i}\sum_{1}^{k}(\boldsymbol{e_{ik}}/\boldsymbol{u_{ik}})^{2} \quad \textbf{(6)}$$

Where, the C_{ik} was the concentration of the k_{th} for soil heavy metals at the i_{th} sample; g_{ip} was the contribution of the p_{th} source to the i_{th} sample; f_{pk} was the concentration of the k_{th} for soil heavy metals in the p_{th} source; e_{ik} was the

deviation of the k_{th} for soil heavy metals at the i_{th} sample; Q was the objective function; u_{ik} was the uncertainty of the k_{th} for soil heavy metals at the i_{th} sample.

The contribution rate of source ***m*** was obtained by **Eq. (7)**.

$$g\% = \left(A_m / \sum_{m=1}^{p} A_m\right) \times 100\% \quad (7)$$

Where, the A_m referred to the regression coefficient for p_{th} source (Liu et al.,2018; Larsen and Baker, 2003).

2.5. Health risk assessment based on source-oriented model

Exposure to heavy metals may cause potential adverse effects on human health due to heavy metal toxicity. Therefore, certain risk assessment model was developed by the USPEA and used frequently for the risk assessment. There were three exposure routes contributed to the average daily dose (***ADD***) via ingestion, inhalation and dermal contact. Non-carcinogenic hazards for heavy metals and the three exposure routes were calculated by the hazard quotient (***HQ***) for two groups mainly including children and adults.

$$ADD_{ing} = C \times IngR \times EF \times ED / (BW \times AT) \times 10^{-6} \quad (9)$$

$$ADD_{inh} = C \times InhR \times EF \times ED / (PEF \times BW \times AT) \quad (10)$$

$$ADD_{dermal} = C \times SL \times SA \times ABS \times EF \times ED / (BW \times AT) \times 10^{-6} \quad (11)$$

$$HI = \sum_{i=1}^{3} HQ = \sum_{i=1}^{3} ADD_i / RfD_i \quad (12)$$

$$THI = \sum HI \quad (13)$$

$$CR_{sc}^{ti} = ADD_i \times SF_i \quad (14)$$

i represented each different exposure pathways of heavy metal entering the human body. ***RfD*** was the reference dose (mg/kg·d) and parameters of detailed description of ***RfD*** were shown in **Table S1**; The hazard index (***HI***) is the sum of ***HQs*** (**Eq. (12)**). The total hazard index (***THI***) is the sum of ***HI*** (**Eq. (13)**). CR_{sc}^{ti} represented the carcinogenic risk index of evaluating carcinogenic hazards of heavy metals (Zhang et al., 2018b). The other parameters were provided by supplementary material in **Table S2**.

To better clarify the impact of pollution sources of heavy metals on human health, quantitative source

apportionment was combined with health risk assessment. Quantitative evaluation of ***HI*** and ***THI*** contributed from each source were caculated by **Eq. (15) to (17)**.

$$HI^{k}=HQ^{k}_{ing}+HQ^{k}_{inh}+HQ^{k}_{der} \quad (15)$$

$$HI^{k,p}_{sc}=HI^{k}\times g^{k}_{p} \quad (16)$$

$$THI^{k,p}_{sc}=\sum_{1}^{k} HI^{k,p}_{sc} \quad (17)$$

Where $HI^{k,p}_{sc}$ was the source contributed hazard index of the k_{th} for soil heavy metals in the p_{th} source; $THI^{k,p}_{sc}$ was the total source contributed hazard index of soil heavy metals in the p_{th} source.

Carcinogenic risk (***CR***) for adults and children was evaluated to assess carcinogenic health effects on humans. When ***CR*** and ***TCR*** greater than 1×10^{-4} were considered as unacceptable, while ***CR*** and ***TCR*** lower than 1×10^{-6} was considered as no significant carcinogenic effects on humans.

$$TCR^{k}=CR^{k}_{ing}+CR^{k}_{inh}+CR^{k}_{der} \quad (18)$$

$$CR^{k,p}_{sc}=CR^{k}\times g^{k}_{p} \quad (19)$$

$$TCR^{k,p}_{sc}=\sum_{1}^{k} CR^{k,p}_{sc} \quad (20)$$

Where $CR^{k,p}_{sc}$ was the source contributed carcinogenic risk of the k_{th} for soil heavy metals in the p_{th} source; $TCR^{k,p}_{sc}$ was the total source contributed carcinogenic risk of all soil heavy metals in the p_{th} source.

Combined with quantitative source apportionment and risk assessment, the source-contributed health risk was calculated. Human health risks of the pollution sources corresponding to soil heavy metals would be clearly displayed, which was mainly to assess the hazards to human health of each specific source of pollution. The source-contributed health risk was calculated by **Eq. (21) to (22)**.

$$SCHI = HI^{k,p}_{sc} g\% \quad (21)$$

$$SCCR = CR^{k,p}_{sc} g\% \quad (22)$$

Where, the ***SCHI*** and ***SCCR*** represented the source-contributed non-carcinogenic and carcinogenic risk index of each sources, respectively.

3. Results and discussion

3.1. Description and optimized spatial interpolation of soil heavy metals

The pH value of the soil detected from the site studied was ranged from 7.03 to 7.57, the average value of which was 7.24. The summary statistics of heavy metals in soil from the site were presented in **Table 1**. The mean concentrations of different heavy metals decreased following the order: Cr > Zn > Pb > As > Cd > Hg. Compared with the background value, the mean concentrations of Cd, Hg and As were relatively higher than the background values (***BV***), accounting for percentages of 415.9%, 239.6% and 77.05% above ***BV***, respectively. Additionally, the highest coefficient of variation (***CV***) of heavy metal turned out to be Hg followed by As and Zn, which indicated that the extensive variation may be affected by multiple factors, especially the anthropogenic activities (Fu et al., 2014). Furthermore, the concentrations of heavy metals (Cr, Pb and Zn) were close to the background values and the ***CV*** of them were lower than 50%, indicating moderate variability in the study area.

Table 1

Summary statistics for heavy metal concentrations (mg kg^{-1}) in soils.

Element	Pb	Hg	Cr	Cd	As	Zn
Min	5.46	0.02	7.10	0.81	2.08	6.95
Max	81.57	1.51	80.59	28.95	56.88	84.32
Mean	28.42	0.18	71.03	0.65	17.75	69.45
Median	24.71	0.13	71.12	0.56	14.40	70.43
Variance	145.04	0.033	0.073	16.51	124.33	78.97
SD	12.04	0.18	0.27	4.06	11.15	65.63
CV	0.32	1	0.26	0.39	0.63	0.49
BV[a]	26.20	0.053	77.80	0.126	10.0	62.60
AV[b]	27.0	0.071	61.0	0.097	11.0	74.00
Chinese soil criteria[c]	250	2.5	150	0.3	30	200
Percent	8.50%	239.6%	0	415.9%	77.05%	10.94%

Note: GM- geometric mean; SD- standard deviation; CV- coefficient of variation; Percent- percentage above BV;

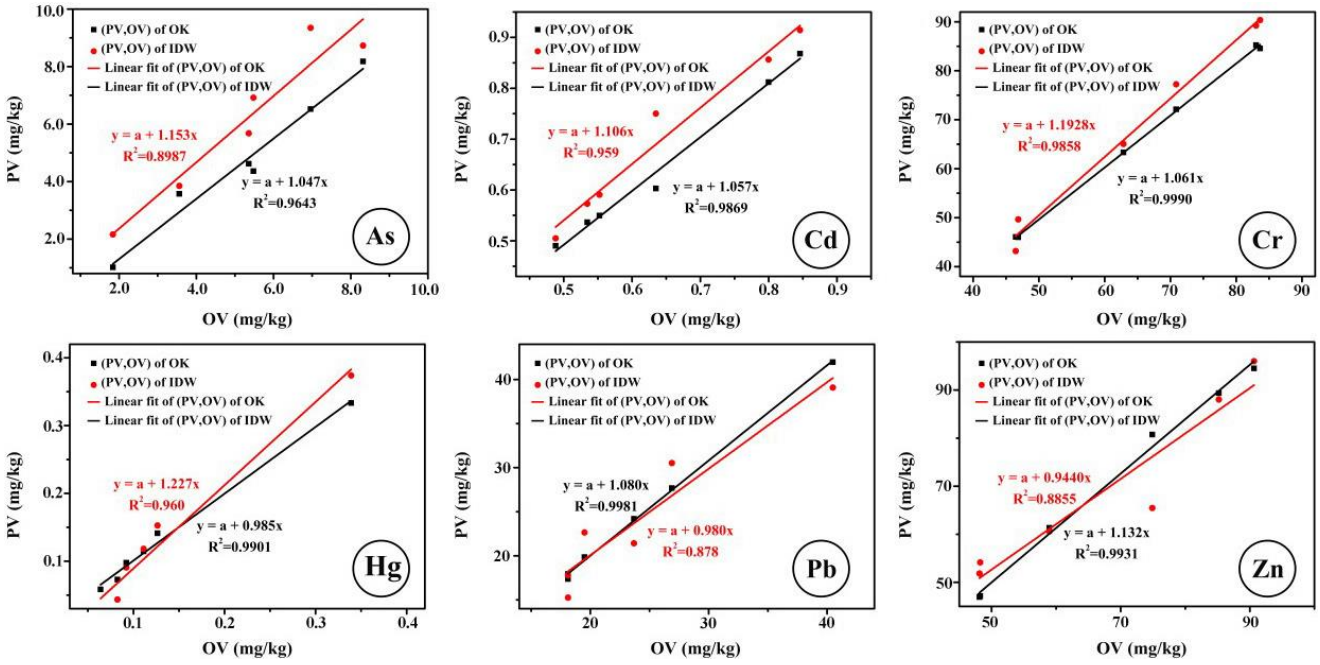
[a] BV-Background value of Jiangsu (Ma et al., 2015);

[b] AV-Average value of China (Wang et al. 2019a);

[c] Chinese soil criteria (CNEPA (1995)).

Trend analysis was performed according to the correlation coefficient between observed values (***OV***) and predicted value (***PV***) (***R***: square root of R^2) to test the accuracy of the prediction method on the spatial distribution. Scatter trend analysis of soil heavy metals for ***OV*** and ***PV*** could convey a linear relationship with high R^2 values ($0.85<R^2<1$).

272 R^2 can fit the correlation between the simulated concentration and the observed concentration of soil heavy metals in
273 the spatial interpolation models (Islam et al., 2018). The results shown in **Fig. 3** indicated that it was more reliable to
274 estimate the spatial distribution for the geological data colledted. The Results demonstrated that the ***PV*** and ***OV*** values
275 were more close in the linear fit of heavy metals extracted by the ***OK*** method. Therefore, ***OK*** can provide a more
276 accurate prediction of the spatial distribution characteristics of heavy metals than ***IDW***. ***OK*** allowed for the spatial
277 correlation of regional variables and the introduced variance function expressions to obtain globally optimal
278 interpolation data (Shahbeik et al., 2014). The introduced variance function can reduce the error in the spatial
279 interpolation results of heavy metals. Therefore, ***OK*** was more appropriate for the spatial distribution of the heavy
280 metals characterised in this study.



281

282 **Fig. 3.** Liner fitting for the predicted and observed values of soil heavy metals by *OK* and *IDW*.

283 Hence, the spatial distribution of the concentrations of heavy metals in the study area was depicted by means of
284 ***OK*** interpolation technique. As it can be seen from **Fig. 4**, the highly concentrated areas of spatial distribution of Cd
285 were mainly in the north and south-west of the study area. The concentrations of Pb and Zn had a similar tendency
286 for higher enrichment of the elements, mainly in the northwestern and southeastern part of the study area, which were

concentrated near the traffic roads. Meanwhile, the average concentrations of Zn and Pb were slightly higher than the background values for Jiangsu Province, mainly due to the proximity of this area to the northern part of the traffic roads in the study area, which could lead to higher levels of Zn and Pb when traffic flows increased. Areas of higher concentrations of Hg were present to the west and center of the dyeing plant. High concentrations of Cr were concentrated near the dye factory area and were close to background values. In addition, high concentrations of As were present around the north-western part of the study area, where agricultural land was located, particularly the overuse of fertilisers may lead to the accumulation of As.

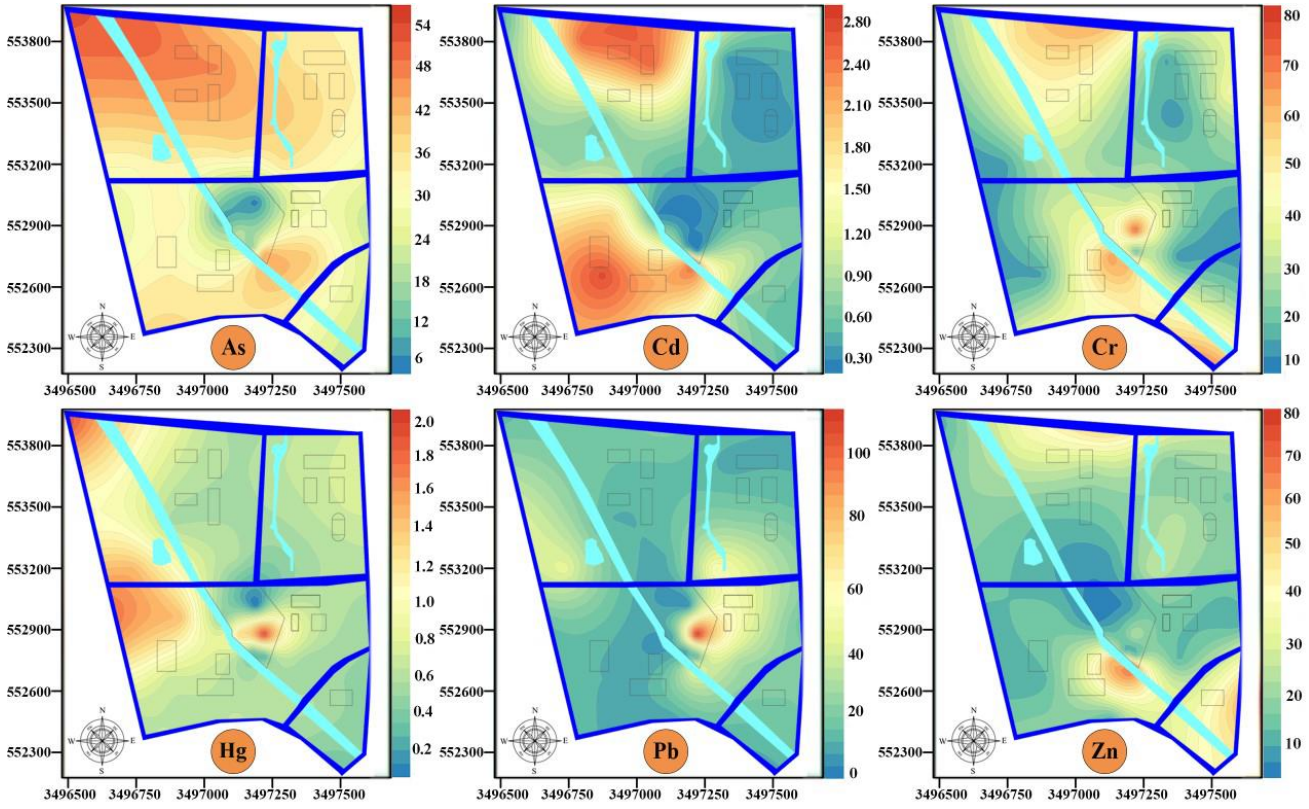


Fig. 4. Spatial distributions of soil heavy metals in the study area using the interpolation model of ***OK***.

3.2. Quantitative source apportionment in the study area

3.2.1. Pollution sources identification

The factor loading results by PCA for concentrations of soil heavy metals were showed in **Fig. 5a**. As shown in the firgure, four principal components were extracted to reflect the relationship of heavy metals in soil. The loading factors of heavy metals elements to the total variance was the main factor in the determination of major pollutants.

The first PC (PC1) in component matrix demonstates that the contaminationof Cr originated in the same sources perhaps, such as waste-water, industrial contamination, lithogenic processes and solid waste. The second PC (PC2) which consists of Zn and Pb can be defined as anthropogenic components, such as vehicle, industrial fumes which was a common source of lead contaminationin soil (Facchinelli et al., 2001). The high Cd level in the third PC (PC3) may be related to traffic activities and diesel exhaust. The high scores of PC4 mainly including As and Hg maybe related to the combination of industrial and agricultural activities (Luo et al., 2017). The rotation ofthe matrix displayed the same relationship between these heavy metals, which is useful to explain ambiguities in the component matrix.

3.2.2. pollution sources contribution

PMF model was implemented to obtain relevant data to better identify the sources and contributions of heavy metals in soil (Wang et al., 2019). Five factors were loaded and the percentage of the total number of species was presented in **Fig. 5b**. **Factor 1** accounted for a high percentage of Hg (56.6%) and Cr (32.7%), the high ***CV*** of which reflecting that they may be related to anthropogenic sources (Guan et al., 2018). Such as Hg emissions brought about by human activities and the emissions caused by waste treatment. On the other hand, Cr can be attributed to emissions of vehicle activities from the traffic roads (Parra etal., 2014). **Factor 2** was dominated by Pb and Zn, with a percentage of 44.8% and 55.6%, respectively. It was noted that the concentrations of Pb and Zn were close to the background values. High concentration distributions of Pb and Zn were mainly concentrated nearby the traffic road, which can be inferred that the contaminants were from traffic activities or fossil fuel. **Factor 3** explained 40.97% of the total species to As, which may be caused by utilization of agrochemicals (Xiao et al., 2019). Thus because As was the main element in the use of pesticides to farmland (Cai etal., 2015). The sharp concentration area of As was around the sampling site S18 nearby the farmland on the map (Salim et al., 2019). Previous studies had also confirmed that the enrichment of arsenic was due to the use of pesticides (Liang et al., 2017b). **Factor 4** was defined mainly by Cr accounting for 45.7% and the accumulation of Cr was mainly associated fine particles from the road area, including

atmospheric deposition, coal burning, printing and dyeing industry and metal processing (Wu et al., 2015). Cd accounted for a large proportion in **factor 5** with the percentage of 44.0%, the main sources of Cd pollution were lead-zinc mines, smelting of non-ferrous metals, electroplating and factories that use cadmium compounds as raw materials or catalysts, the same polluted sources as the factor 4 (Wang et al., 2019). The high ***CV*** value and higher mean concentration compared with the background value, showing the influence of artificial factor (Gao et al., 2018; Peng et al., 2017). According to the foregoing investigation, the textile and dyeing industries attributed high content of Cd from the emission of the sewage of dyeing activities (Wang et al., 2019; Luo et al., 2011; Sun et al., 2019). The high concentration area and high risk area of metal Cd were all around the dye factory as investigated. Therefore it was concluded that there were four sources of pollution dominating the contaminated site. Comparatively speaking, industrial activities occupied the largest contribution to soil heavy metals in the study area; followed by the human activities such as waste emission. The third largest contribution was made by agricultural activities, which were related to the farmland in the study area. Owing to the frequent utilization of vehicle and fuel near the roads, traffic activities accounted for the fourth largest contribution and the followed one was atmospheric deposition. The results were consistent with the current situation of the whole study area, with abandoned factories, developed transportation around and frequent agricultural activities, indicating the close relationship between the dye factory and its surrounding soil.

Therefore, the number of factors ought to be set to four within the PMF run by the results of PCA, at that purpose the simplest analysis results were achieved with a run variety of fifty. Additionally, the coefficients of the six heavy metal components shown in **Table S3** varied between 0.35 ~ 0.99, indicating that the overall fit of the PMF was sensible.

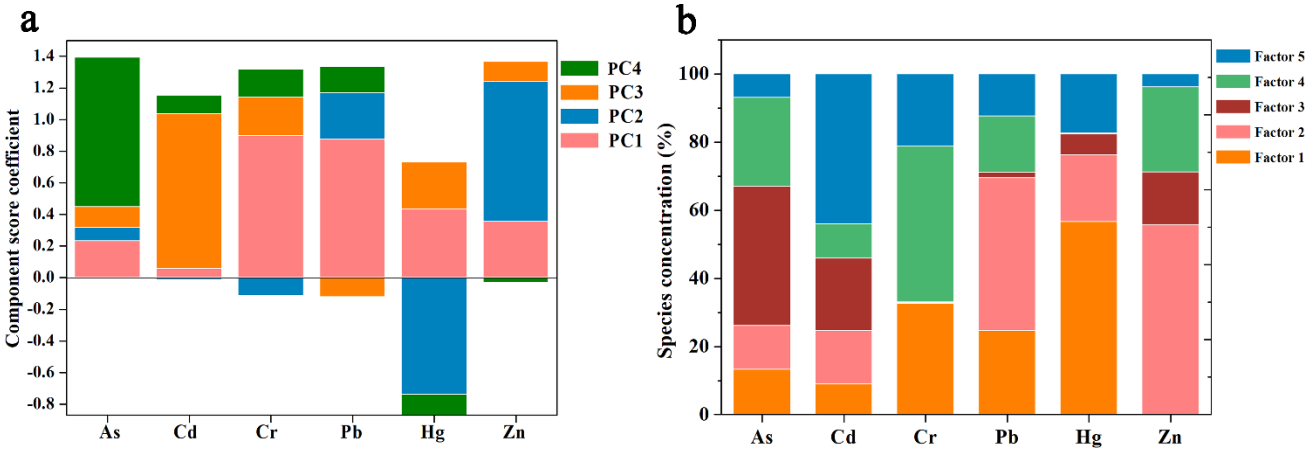


Fig. 5. (a) Component source coefficient by PCA analysis and **(b)** source contributions for each heavy metal estimated by PMF model.

3.3. Human risk assessment

3.3.1. Human risk assessment of heavy metals

The upper limit of 95% confidence intervals of regional survey data ($C_{95\%}$ UCL in mg/kg) was considered to evaluate of the maximum intake daily dose for human health risk assessment (USEPA, 1989). The original data of ***HQ***, ***HI*** and ***THI*** (adults and children) by means of classical health risk assessment were presented in **Table 2**. The highest average intake turned out to be the ingestion pathway and the order followed were: dermal contact > inhalation. In the ingestion pathway, average ***HQ*** ranged as: As > Cd > Pb > Hg > Cr > Zn. ***HQs*** for six heavy metals were less than 1 both for adults and children, indicating that there were no significant health risks. However, the ***THI*** value of children through ingestion pathway was 1.00, posing potential health threat to the children. The contribution of As to non-carcinogenic risk was 76.9%, which should be paid attention to in this region. The ***CR*** values of As, Cd, Cr, Hg, Zn and Pb were evaluated by health risk assessment and the results showed that ***CR*** index of the whole region for the six heavy metals were all less than 1.0E-06 through ingestion of exposure, indicating that there was no carcinogenic risk in this region. Therefore, the source-contributed assessment of health risks mainly focused on the evaluation of non-carcinogenic health risk. Then the modified assessment of human health risk for five heavy metals was performed based on source contribution (Li etal., 2020).

Table 2

Estimations of human health risk of soil heavy metals.

	Element	Pb	Hg	Cr	Cd	As	Zn	HI	THI
	C-95%	28.75	0.19	71.02	0.76	18.05	68.50		
	HQ_{ing}	1.13E-02	8.68E-04	4.70E-04	1.20E-02	8.24E-02	3.13E-04	1.07E-01	
Adults	HQ_{der}	4.46E-05	3.44E-06	1.97E-04	4.79E-05	3.28E-04	1.25E-05	6.33E-04	1.08E-01
	HQ_{inh}	1.10E-05	6.38E-07	3.46E-06	1.76E-04	2.96E-05	2.30E-07	2.21E-04	
	HQ_{ing}	1.05E-01	8.10E-03	4.39E-03	1.12E-01	7.69E-01	2.92E-03	1.00E+00	
Children	HQ_{der}	2.92E-05	2.25E-06	1.29E-04	3.14E-05	2.15E-04	8.16E-06	4.15E-04	1.00E+00
	HQ_{inh}	1.96E-05	1.14E-06	6.16E-06	3.14E-04	5.26E-05	4.10E-07	3.94E-04	

3.3.2. Source-contributed health risk assessment

The source contribution of ***SCCR*** and ***SCHI*** was calculated to evaluate the risk level of the source. The results of source analysis were incorporated into the health risk model to assess the potential hazards to human health by soil heavy metals. The calculation of source-contributed health risk assessment could demonstrate the relationship between pollutants sources and health risk, as shown in **table. 3**. The contributions of the carcinogenic risk assessment for the heavy metals were demonstrated in **Fig. 6a**. Cr originated from anthropogenic sources, of which Cr accounted for 5.79E-04. For As, with a high ***SCCR*** index of 4.50E-04, accounted for the major contributor in the agricultural activities. For Cr, industrial sources was the main source of pollution in the high ***SCCR*** area with a ***SCCR*** index of 1.09E-03. The carcinogenic risk allocation was consistent for adults and children, but the risk values of adults were lower than that for children. The ***TCRs*** of the sources were in the order of industry (6.42E-04), agriculture (4.59E-04), transportation (4.11E-04), and atmosphere (3.23E-04). The industrial sources had the highest ***TCR***, indicating harmful effects from the activities of the dye plant. There was a potential carcinogenic hazard in the study area, for which reasonable programmes should be developed to reduce or isolate the input or transport of industrial and agricultural sources. In addition, some activities, particularly industrial and agricultural activities, should be controlled to reduce population health and environmental risks.

The contributions of the non-carcinogenic risk assessment for the heavy metals were demonstrated in **Fig. 6b**. For Hg and Cr, industrial sources were the main contributors to ***SCHI***, accounting for 3.36E-01 and 3.97E-01,

respectively. For Pb and Zn, traffic activities was the largest contributor to ***SCHI***, accounting for 7.94E-02 and 2.45E-02, respectively. For As, the higher ***SCHI*** came from agricultural activities, with the risk values of 7.96E-02. For Cd, ***SCHI*** was higher from industrial sources at 6.80E-02. The largest contributor to ***THI*** was industry (6.85E-01), followed by traffic sources (4.89E-01), agriculture (4.38E-01), natural (3.44E-01). Industry sources had the highest ***THI*** and were mainly associated with emissions caused by waste treatment of the dye factory, which may contribute significant amounts of heavy metals and posed a threat to the local residents.

Table 3

Estimation of non-cancer (hazard index) and cancer risk (total cancer risk) of heavy metals from four sources.

	Children（aged 1-17）					**Adults（aged 18+）**				
	Factor 1	**Factor 2**	**Factor 3**	**Factor 4**	**Total factors**	**Factor 1**	**Factor 2**	**Factor 3**	**Factor 4**	**Total factors**
	hazard index and total hazard index					***hazard index and total hazard index***				
As	1.40E-01	1.00E-02	7.96E-02	5.04E-02	4.00E-01	1.51E-03	1.08E-03	9.64E-03	7.-7E-03	4.30E-02
Cd	1.35E-03	9.65E-04	8.65E-04	6.80E-02	3.86E-03	1.47E-04	1.05E-04	9.41E-05	4.40E-02	4.20E-04
Cr	3.97E-01	2.83E-03	2.54E-03	2.00E-02	1.13E+00	5.88E-02	1.20E-04	3.76E-03	2.96E-03	1.68E-01
Hg	3.36E-01	1.40E-02	2.15E-02	1.69E-02	9.59E-02	3.63E-02	2.59E-03	2.32E-03	1.82E-03	1.04E-02
Pb	1.11E-02	7.94E-02	4.11E-02	5.59E-04	3.17E-01	1.20E-02	8.54E-02	7.65E-03	6.01E-03	3.42E-02
Zn	2.03E-03	2.45E-02	1.30E-03	1.02E-03	5.81E-03	2.19E-04	1.56E-02	1.40E-04	1.10E-04	6.25E-04
Total hazard index	6.85E-01	4.89E-01	4.38E-01	3.44E-01	1.96E+00	8.98E-02	6.42E-02	5.75E-02	4.52E-02	2.57E-01
	Cancer risk of each heavy metals and total cancer risk					***Cancer risk of each heavy metal and total cancer risk***				
As	6.30E-05	4.50E-04	4.03E-06	3.17E-05	3.60E-04	6.82E-06	4.87E-06	4.37E-06	3.43E-06	3.90E-05
Cd	2.38E-10	1.70E-10	1.52E-10	3.20E-04	1.36E-09	1.34E-10	9.55E-11	8.56E-11	6.73E-11	7.64E-10
Cr	1.09E-03	4.13E-05	3.70E-05	2.91E-04	3.31E-03	6.26E-04	4.47E-05	4.01E-05	3.15E-05	3.58E-04
Hg	1.58E-08	1.13E-08	1.01E-08	7.96E-09	9.04E-08	8.89E-09	6.35E-09	5.69E-09	4.47E-09	5.08E-08
Total cancer risks	6.42E-04	4.59E-04	4.11E-04	3.23E-04	3.67E-03	6.95E-05	4.96E-05	4.45E-05	3.49E-05	3.97E-04

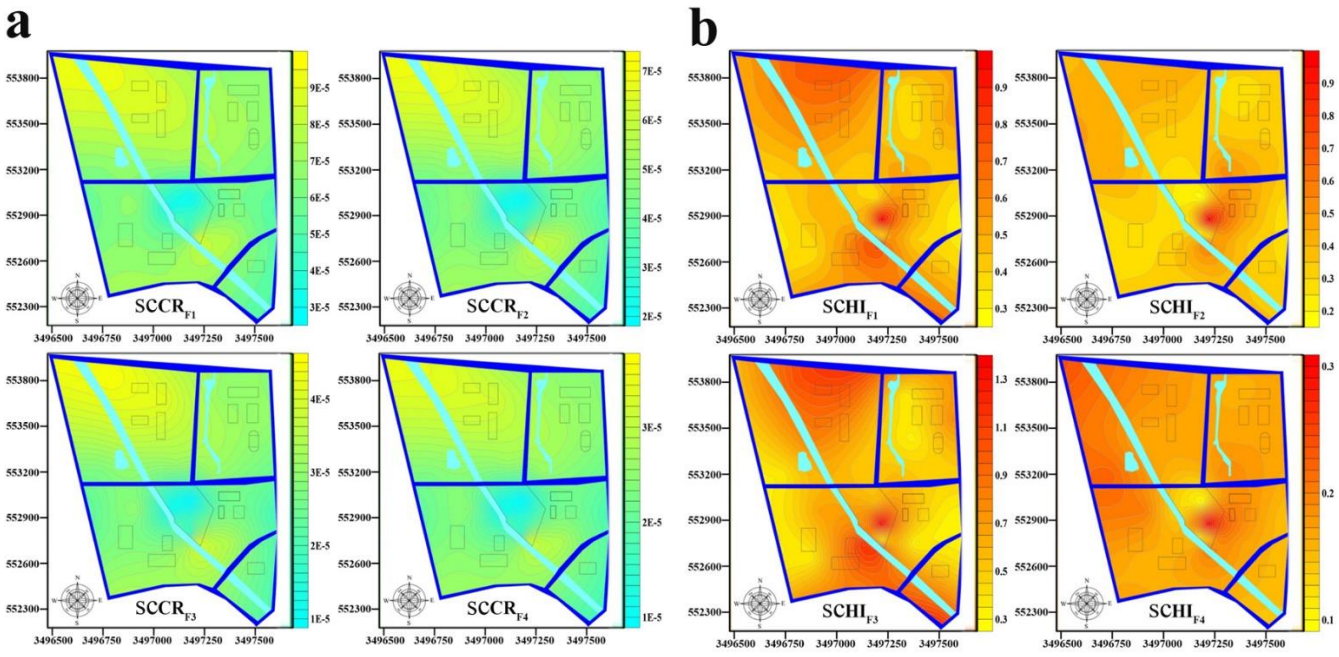


Fig. 6. Spatial distribution of source-contributed carcinogenic and non-carcinogenic risk assessment.

3.4. Disscussion of the methodology

To the traditional research, risks assessment under traceability of pollution sources was incorporated into the paper. The evaluation model of the risk allocation of pollution sources was constructed, which was constructive for the treatment of heavy metal soil pollution. It had a certain reference value for the supplement and improvement of the health risk assessment system. The health risk assessment based on source apportionment mainly included optimized spatial interpolation and human health risk. The pollution sources were quantified for health risk assessment, which was optimized compared with conventional risk model. It turned out that the traceability results of the two source-oriented risk assessment model were different as well as the risk contribution order of pollution sources. Source-oriented health risk analysis was at a low risk level, which was dominated by industrial activities. Therefore, Cr should be regarded as the target metal, the emission of which should be converted to hypotoxic elements. It can effectively prevent pollution and realize regional environmental management. Source-oriented human health risk of As was dominated by agricultural activities. So metal arsenic should be treated as a priority target metal for soil pollution. Effective control of metal emission or lowering the concentrations of toxic element can reduce the level of health risk in this region. It was concluded that traditional health risk assessment method may overlook the source

contributions of pollution, The traceability and contribution of certain pollution sources combined with health risk assessment could make up for these deficiencies. In the comparative study of PCA and PMF tracing, it was more reasonable to take a comprehensive source apportionment for consideration to set definite numbers for pollution sources. This study provided new insights for the treatment of complex sources and hazards of soil heavy metals.

4. Conclusions

It was of great significance to study the distribution of heavy metal content in abandoned factories and carry out the comprehensive health risk assessment of heavy metal, so as to provide the basis for the prevention and control of soil heavy metal pollution. The method was developed by integrating health risk assessment and source contribution based on source apportionment to study pollutant sources and risk levels. This method optimized spatial interpolation based on the global content data output so as to quantitatively analyze pollution level of the area intuitively. PCA and PMF model were arried out by auxiliary quantitative analysis on the determination of pollution sources accurately. A case study was conducted in an abandoned dye factory in Suzhou City, China, so as to implement the methodology. This study assessed spatial distribution characteristic and source-identified health risk of six heavy metals (Pb, Hg, Zn, Cr, Cd, As) in the study area. PMF model was utilized to apportion the sources of risk, combining a receptor model and a risk model. The source-specific human health risk of heavy metals for each source category was also discussed, in which some significant results were found. However, owing to anthropogenic influence, the area was contaminated by these heavy metals studied in some degree. Particularly, industrial activities were identified as the largest contributor by Cr, mainly associating with the discharge and disposal of pollutants from the dye factory in the area. The results of source-contributed health risk suggested that there was potential health threat to the children. As as agriculture activities contributed most to the risk level and would lead to higher risks in the study area. After comprehensive comparison, it was concluded that in the comparative study after tracing health risks, it was more reasonable to use comprehensive site risk assessment to treat regional pollution sources in this region. This study provided a new insight for the treatment of multi-sources of heavy metal and it was advised to reduce the total

emission in the process of industry and agriculture by regional structural reconstruction.

Credit authorship contribution statement

Fangfang miao: Data curation, Writing - original draft, Writing –review & editing. Yimei Zhang and Yu Li, performed the guiding of this paper with support from Yinzhuang Zhou. Qinglu Fang is mainly responsible for sorting out data and assisting in drawing. We confirm that this manuscript has not been published elsewhere and is not under consideration by another journal. We affirm that all authors have approved the submission and the study does not involve human and animal subjects.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The datasets generated and analyzed during the current study are not publicly available due to it is confidentiality but are available from the corresponding author on reasonable request.

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