Journal of Hazardous Materials 423 (2022) 127116

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Determination of priority control factors for the management of soil trace metal(loid)s based on source-oriented health risk assessment

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ARTICLE INFO

Editor: Dr. S Nan

Keywords:
Source apportionment
Monte Carlo simulation
Positive matrix factorization
Urban soil

ABSTRACT

Trace metal(loid)s (TMs) in soils can seriously threaten the ecological environment and human health. With the limitation of resources and costs, determining priority control factor is critical for managing soil TM pollution. To explore the pollution characteristics, source apportionment, and human health risk of TMs, a total of 209 surface soil samples were collected from Anqing City, China. Results showed that all the average values of TM concentration, except for Cr, were higher than their corresponding background value. Using a Positive matrix factorization model coupled with Correlation analysis, four sources (including agricultural sources, atmospheric deposition sources, industrial sources, and natural sources) were identified as the determinants for the accumulation of soil TMs, with the contribution rates of 12.4%, 8.1%, 64.1%, and 15.4%, respectively. The assessment of probabilistic health risks revealed that Non- carcinogenic risks of all populations were acceptable ($HI < 1$), while Carcinogenic risks were all at a high level ($TCR > 10E\text{-}04$). Agricultural pollution and As were identified as priority control factors, according to the analysis results of the relationship among TMs, pollution sources and health risks. Our findings provide scientific support for decision-makers to formulate target control policies and reduce management costs of soil pollution.

1. Introduction

With the rapid development of urban industrialization, many trace metal(loid)s (TMs) have been discharged into the urban environmental media, such as the atmosphere, water, and soil. Due to their cumulative, persistent, and toxic characteristics (Ali et al., 2019), TMs can pose a serious threat to the ecological environment and human health (Han et al., 2021; Kamani et al., 2018; Yuan et al., 2020; Zhang et al., 2020), and thus have attracted widespread concern.

Human activities (such as industrial production, traffic emissions, pesticide and fertilizer application) can accumulate TMs in soils (Nagajyoti et al., 2010). These TMs can enter human body through three exposure ways (including oral ingestion, oral and nose inhalation, and dermal contact), and eventually cause harm to human health (Huang et al.,2018b). Therefore, identifying the sources of TMs and analyzing the potential health risks are critical for managing soil TM pollution (Han et al., 2021).

Previously, many studies focused on the assessment of the concentration-oriented health risks (Singh and Kumar, 2017), but this cannot effectively distinguish the impact of natural sources and anthropogenic sources on health risks. Considering the uncontrollability of the natural sources, the key to soil TM pollution control is to restrict the discharge of anthropogenic pollution. Therefore, the source-oriented risk assessment is essential for decision-makers to develop mitigation strategies for human risks. At present, many researchers have realized its significance and gradually use the source-oriented method to assess health risks (Liu et al., 2018a, 2018b). The use of receptor model for health risk assessment is the mainstream of current research (Huang et al., 2021a, 2021b, 2018a, 2018b). However, some receptor models (such as Principal component analysis, Factor analysis, and Cluster analysis) cannot obtain the non-negative results and handle the process data below the detection level (Li et al., 2018; Paatero and Tapper, 1994;

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https://doi.org/10.1016/j.jhazmat.2021.127116
Received 23 May 2021; Received in revised form 20 August 2021; Accepted 31 August 2021
Available online 2 September 2021

Huang et al., 2021a), which are not conducive to source-oriented assessment. Many studies have shown that the Positive matrix factorization (PMF) model recommended by U.S. Environmental Protection Agency (US EPA) is an effective measure to make up for this deficiency (Fei et al., 2020; Guan et al., 2018; Paatero, 1997). Traditional health risk assessment (HRA) models (Khan et al., 2016) mainly rely on specific fixed exposure parameters and pollutant concentrations (Liu et al., 2021). However, this assessment assuming the same parameters for all populations may lead to inaccurate assessment results. Fortunately, some uncertainty analysis models (such as Monte Carlo simulation, Bayesian analysis, and Meta-analysis) have shown great potential in exposure assessment and have begun to be applied for health risk assessment (Armstrong et al., 2004; Islam et al., 2019; Yang et al., 2010). Compared with other uncertainty models, Monte Carlo simulation is an effective probabilistic risk analysis method, which requires fewer data samples and can evaluate the possibility of exceeding the guide threshold (Karami et al., 2019; Tong et al., 2018). However, there are few studies on the application of Monte Carlo simulation in risk allocation so far. Hence, based on the combination of Monte Carlo simulation, PMF model, and HRA model, we attempt to develop a Probabilistic source-oriented risk (PSOR) model to (1) identify and quantify the critical pollution sources and (2) assess the health risks in this study.

Previous studies have shown that pollution sources can cause different health risks due to different TM concentrations and toxic reaction factors (Fu et al., 2013; Lin et al., 2018; Liu et al., 2018a; Huang et al., 2018b; Islam et al., 2015; Wei et al., 2015; Wu et al., 2020; Zhang et al., 2019). Controlling and managing these pollution sources and elements usually requires considerable resources and costs (Pu et al., 2019; Wang, 2018). However, because of the limited availability of resources and costs, not all TMs and pollution sources can be effectively and simultaneously controlled (Men et al., 2020). Therefore, determining priority control factors (including TMs and pollution sources) is a highly critical step for preventing and controlling soil pollution (Chao, 2019). The key to determining priority control factor is to sort out the relationship among metals, pollution sources, and health risks. However, as far as we know, surprisingly little attention has been devoted to exploring this relationship systematically. Hence, we aim to reveal the impact of TMs and pollution sources on health risks and determine the priority control factor that contribute the most to health risks in this study.

Anqing City is a typical industrialized city in the Yangtze River economic belt of China. Due to the highly developed industrialization in the past few decades, most urban soils has been polluted by TMs. However, little attention has been paid to preventing and controlling health risks caused by soil TMs in such rapidly developing and densely populated cities. Therefore, taking Anqing City as a case, this study aimed (1) to explore pollution characteristics of TMs in soils, (2) to quantitatively analyze pollution sources of TMs by using Pearson correlation analysis and PMF model, (3) to develop a PSOR model and assess health risks caused by TMs, and (4) to determine the priority control factor by investigating the relationship among TMs, pollution sources, and health risks.

2. Materials and methods

2.1. Study area and soil sampling

Anqing City (30°29′N-30°41′N, 116°57′E-117°14′E) is located at the junction of the middle and lower reaches of the Yangtze River, with an area of 13,589 km^2 and a population of 5.28 million. It is a highly industrialized city with petrochemical plants, electroplating plants, steel plants, and many other chemical factories. The study area is rich in natural resources (such as minerals, land, and organisms), and mineral smelting is the primary industry. A total of 209 surface soil (0–20 cm) samples were collected from the urban areas, and all geographical locations of the sampling sites were recorded using handheld GPS (Fig. 1). According to the five-point sampling in the specification of GB/T 36197-2018 (Huang et al., 2021b; Jia et al., 2019; MARA, 2018), each representative sample was a mixture of five equal-weight sub-samples collected from five locations. After been removing the larger stone, grassroots and other impurities, the original mass of each sample was not less than 1 kg. All collected samples were kept in polyethylene bags and sent to the laboratory for further analysis.

2.2. Measurement method of TM concentration

The collected soil samples were air-dried to constant weight at room temperature and then sieved using a 2 mm nylon mesh. About 50 g soil sample was grounded in a mortar to pass a 120-mesh nylon mesh. The soil sample (about 0.5 g) was first digested in an acid mixture of HNO_3 (60%) and $HClO_4$ (60%) (2:1), and then digested with microwave (Sardans and Peñuelas, 2005). The Cd, Pb, Cr, Ni, As, Cu and Zn concentrations were measured by Inductively coupled plasma mass spectrometry (ICP-MS), with the detection limits of 0.01 mg/kg, 0.1 mg/kg, 0.1 mg/kg, 0.1 mg/kg, 0.01 mg/kg, 0.1 mg/kg, and 0.5 mg/kg, respectively. The Hg concentration was monitored by a cold atomic absorption mercury meter (JKG-205). To avoid the error of impurities, the blank solution was prepared by deionized water without soil samples, following the same steps and conditions as the sample digestion. Blank measurements were performed in every 20 soil samples and repeated three times for each blank sample (Alsbou and Al-Khashman, 2018).

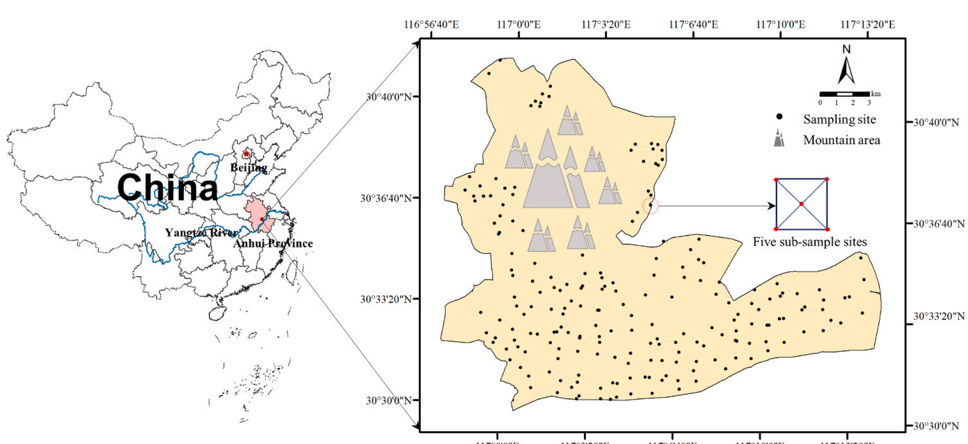

Fig. 1. Location and sampling sites of the study area.

2.3. Positive matrix factorization model

As a multivariate factor analysis tool, PMF model (Paatero, 1997) has been gradually used in the source analysis of TMs in soils (Lv, 2019). The sampling data matrix (X) is divided into a factor distribution matrix (G) and factor contribution matrix (F) to identify the source types that contribute to the sample (Paatero, 1997), which can be expressed as:

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

where X_{ij} is the concentration of the ith element measured in the jth sample, G_{ik} is the source profile for ith element for kth source factor, F_{kj} is the contribution matrix of kth source factor for jth samples, and E_{ij} is the residual error matrix. Meanwhile, the PMF model can obtain the factor contribution through the minimum objective function Q:

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\frac{E_{ij}^2}{U_{ij}^2} \tag{2}$$

where U_{ij} denotes the ith element uncertainty of the jth sample. The uncertainty is calculated based on the sample concentration and the method detection limit (MDL).

When the sample concentration is less than or equal to the MDL, the sample concentration is replaced by 1/2 MDL and the uncertainty (u) can be calculated by Eq. (3) (Paatero, 1997):

$$u = \frac{5}{6} \times \text{MDL} \tag{3}$$

When the sample concentration is higher than the MDL, the uncertainty (u) follows Eq. (4):

$$u = \sqrt{(\sigma \times \text{concentration})^2 + (\text{MDL})^2} \tag{4}$$

where σ is the relative standard deviation which is calculated by the ratio of standard deviation to average.

2.4. Human health risk assessment model

The human health risks, including Carcinogenic risk (CR) and Non-carcinogenic risk (NCR), are generally calculated by the HRA model (US EPA, 1996). CR is an assessment probability of an individual suffering from cancer due to long-term exposure to a particular pollutant or mixtures of pollutants (Kamarehie et al., 2019), while NCR is more associated with chronic exposure include genetic and teratogenic effects (Huang et al., 2021b). To assess the health risk caused by soil TMs, the susceptive population was divided into three groups: adult females, adult males, and children. In general, unlike atmospheric particles, TMs in soils can hardly reach the human body through inhalation (Zhao and Duan, 2014). Therefore, the average daily exposure doses (ADDs) only consider the two main exposure pathways (including direct oral ingestion and dermal contact), and it is calculated as follows:

$$\text{ADD}_{\text{dermal}} = \frac{C_{\text{soil}} \times \text{SA} \times \text{AF} \times \text{ABF} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \tag{5}$$

$$\text{ADD}_{\text{dermal}} = \frac{C_{\text{soil}} \times \text{SA} \times \text{AF} \times \text{ABF} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \tag{6}$$

where C_{soil} is the concentration (mg/kg) of TMs. The exposure parameters of BW, ED, SA, AF, IngR, ABF, AT, and EF were shown in Table S1.

The Carcinogenic risk is assessed by the total Carcinogenic risk (TCR) of TMs, which is calculated by Eq. (7) (Wu et al., 2020):

$$\text{TCR} = \sum CR_i = \sum (\text{ADD}_i \times SF_i) \tag{7}$$

where CR_i is the Carcinogenic risk (CR) of each TM, and SF_i is the slope factors value of each TM (Table S3). If TCR > 10^{-4}, it indicates that the risk is unacceptable; and if TCR < 10^{-6}, it indicates the opposite (Wu et al., 2015).

The Non-carcinogenic risk is assessed by the total Hazard indices (HI) of TMs, which is calculated by Eq. (8) (Wu et al., 2020):

$$\text{HI} = \sum HQ_i = \sum \frac{\text{ADD}_i}{RfD_i} \tag{8}$$

where HQ_i is the Hazard quotient (HQ) of each TM and RfD_i is the corresponding reference does of each TM (Table S3). If HI > 1, it indicates possible adverse health effects, and HI < 1 means no apparent risk to human body (Wu et al., 2015).

2.5. Hybrid model combing PMF and HRA

Health risks from different sources are quantitatively assessed by a hybrid model that combines the PMF and HRA models. The hybrid model is performed in the following four steps:

1) Analyze the potential source contribution of each TM based on the PMF model;
2) Estimate the concentration of the ith element in the jth sample from the kth source by Eq. (9):

$$C_{ij}^k = F_{ij}^k \times X_{ij} \tag{9}$$

where C_{ij}^k is the concentration (mg/kg) of the ith element from the kth source in the jth sample, and F_{ij}^k is the estimated contribution rate of the ith element from the kth source in the jth sample, and X_{ij} is the measured concentration (mg/kg) of the ith element in the jth sample.

3) Fit the probability density curve of the ith element from the kth source in all samples by Monte Carlo simulator. Probability density functions (PDFs) of TMs concentration from each source with Monte Carlo simulator were shown in Table S2.
4) Assess the human health risk quantitatively from different sources with the Monte Carlo simulator. ADDs of the ith element from the kth source in the jth sample are estimated by Eqs. (10–11):

$$\text{ADD}_{\text{ij,dermal}}^k = \frac{C_{ij}^k \times \text{SA} \times \text{AF} \times \text{ABF} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \tag{10}$$

$$\text{ADD}_{\text{ij,ingestion}}^k = \frac{C_{ij}^k \times \text{IngR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \tag{11}$$

The exposure parameters of BW, ED, SA, AF, IngR, ABF, AT, and EF were shown in Table S1. The Carcinogenic risks for different sources are determined as Eq. (12):

$$\text{CR}_{\text{ij,n}}^k = \left(\text{ADD}_{\text{ij,n}}^k \times \text{SF}_i\right) \tag{12}$$

where $CR_{ij,n}^k$ is the Carcinogenic risk on the nth exposure pathway from the kth source of the ith metal in the jth sample, and SF_i is the slope factor of each TM (Table S3). The Non-carcinogenic risks for different sources determined as Eq. (13):

$$\text{HI} = \text{HQ}_{\text{ij,n}}^k = \frac{\text{ADD}_{\text{ij,n}}^k}{\text{RfD}_i} \tag{13}$$

where $HQ_{ij,n}^k$ is the Hazard quotient on the nth exposure pathway from the kth source of the ith metal in the jth sample, and RfD_i is the corresponding reference does of each TM (Table S3).

2.6. Statistical analysis

The statistical analysis was conducted by using IBM SPSS v17.0 (IBM, USA). The bitmap of the sampling site was constructed with ArcGIS v10.7 (Liu et al., 2018a). Pearson correlation analysis and descriptive statistics were performed with R software environment. Pollution source analysis was conducted by following the PMF 5.0 (US EPA, 2016). The probabilistic health risks of TMs and pollution source were analyzed by Monte Carlo simulation. Oracle Crystal Ball software was used for simulation, and the number of iterations was set to 10,000 (Karami et al., 2019).

3. Results

3.1. Descriptive statistics of soil trace metal(loid)s

As shown in Table 1, the mean concentrations of seven TMs were 0.34 mg/kg (Cd), 46.01 mg/kg (Pb), 12.91 mg/kg (As), 25.73 mg/kg (Cu), 90.02 mg/kg (Zn), 59.51 mg/kg (Cr), and 0.32 mg/kg (Hg), respectively. The ranking of the mean concentration for all TMs was as follows: Zn > Cr > Pb > Cu > As > Cd > Hg. According to the Anhui Province soil environmental quality standard, all the average concentrations of TMs except for Cr were higher than the background values (Fig. S1). The over-standard rate of each TM was as follows: Hg > Cd > Pb > Zn > As > Cu > Cr. It is worth noting that the average concentration of Hg (700%) had a high over-standard rate. Among all TMs, the average and maximum concentrations of Cd, Pb, Cu, and Hg did not exceed their corresponding guide values. Besides, although the average concentration of As did not exceed the guide value, the maximum concentration was 2.53 times the guide value (accounting for 14.4% of all samples).

3.2. Source analysis of trace metal(loid)s in soils

Pearson correlation analysis was used to identify the correlation between different TMs, and PMF model was then used to identify and quantify the potential sources of TMs. Four factors finally determined by using PMF model coupled with Correlation analysis (Fig. 2). Correlation analysis results showed that there were significant positive correlations between Zn and Cd, and between Zn and Cu. This suggested that these elements might come from the same source. Hg and Pb also presented a positive correlation, while Hg and Zn, Hg and Cu, Hg and Cr presented a negative correlation, respectively. This showed that Hg and the other TMs might not come from the same source.

The PMF model was run 20 times, and ultimately four factors were determined (Fig. S2). Factor I had the strongest relationship with As (89.2%). The main elements of Factor II were Pb (62.0%) and Hg (100%), suggesting that these two elements converged in a common source. Factor III was mainly related to Cd (96.1%), Cu (83%), and Zn (80.3%), which indicated that these TMs were closely related and may have similar sources. Factor IV showed the highest scores for Cr (90.7%).

3.3. Health risk assessment based on Monte Carlo simulation

3.3.1. Concentration-oriented health risk assessment

By using Monte Carlo simulation (Section S1), Fig. 3 showed the probability distribution of Carcinogenic risk and Non-carcinogenic risk in different populations (children, adult females, adult males). Compared with adult females and adult males, children suffered more serious Carcinogenic risk, and the order of mean TCR value for three populations was as follows: children (8.30E-05) > adult females (4.02E-05) > adult males (1.81E-05). Additionally, nearly 98.8% (children), 97.5% (adult females), and 95.5% (adult males) of TCR values exceeded 1.0E-06. The high over-standard rate indicated the Carcinogenic risk was non-negligible in this study area. As for the Non-carcinogenic risk, the average HI value for children, adult females, and adult males were 1.93E-01, 5.03E-02, and 2.72E-02, respectively. According to the probability distribution, merely 0.2% of HI values for children surpassed the threshold of 1, and adults' HI values were within a safe range. It indicated that these TMs hardly posed a significant Non-carcinogenic risk to the health of children and adults. Overall, children suffered from higher Carcinogenic risk and Non-carcinogenic risk than adults. Hence, the health risk assessment in this study area should pay more attention to children.

3.3.2. Source-oriented carcinogenic risk assessment

The results of source-oriented risk assessment showed that Factor I was the primary anthropogenic source of Carcinogenic risk (Fig. 4a). The average risk values of Factor I were 1.17E-05, which was 11.7 times higher than the acceptable threshold (1E-06). For specific TMs, the contribution rates of As in Factor I and Cd in Factor III to CR are 96.8% and 95.9% (Fig. 4b and Figs. S3a-S3b). However, the Carcinogenic risk caused by As in Factor I cannot be ignored because the probability of exceeding the risk value reach 73.5%. In addition, the CR value caused by Pb in all sources did not exceed 1E-06 (Fig. S3), but Pb in Factor II contributed most to CR (62.0%).

3.3.3. Source-oriented non-carcinogenic risk assessment

The probability distributions of the Non-carcinogenic risk of different sources were shown in Fig. 5. Obviously, the Non-carcinogenic risks of different sources were different, and the risk values (the 95th percentile) of all sources were below the acceptable threshold (HI = 1), indicating no potential Non-carcinogenic risk (Fig. 5a). For specific sources, Factor I had a more significant contribution to HI value than other sources. The mean contribution rate for different sources decreased in the following order: Factor I > Factor IV > Factor II > Factor III. Although Cd, Cu, Zn, and Pb in Factor III mainly contributed HQ values, they contributed the least to the HI value of children. The risk values (the 95th percentile) of TMs from all sources were lower than the acceptable threshold (HQ = 1), indicating that all TMs posed negligible Non-carcinogenic risk to children.

4. Discussion

4.1. The pollution characteristic of trace metal(loid)s in soils

The concentrations of all TMs (except for Cr) exceeded that of the background value, indicating that TMs significantly polluted the soil in the study area. Previous studies showed that the anthropogenic activities (such as agricultural activities, traffic emission, and industrial production) might cause high over-standard values for Hg and Cd (Huang et al., 2015; Wang et al., 2019b). Besides, TMs with the

Table 1
Statistical summary of trace metal(loid) concentration (mg/kg) in soils of the study area.

Statistics	Cd	Pb	As	Cu	Zn	Cr	Hg
Mean	0.34	46.01	12.91	25.73	90.02	59.51	0.32
Median	0.20	34.85	11.25	22.29	69.30	56,96	0.11
Max	3.39	292.00	50.55	669.00	569.00	239.35	2.38
Min	0.01	8.24	0.33	0.71	4.02	0.10	0.002
SD	0.44	34.63	9.39	46.34	84.96	32.91	0.45
ABV	0.097	26.6	9.0	20.4	62.0	66.5	0.04
GV	20.0	400.0	20.0	2000.0	n/a	n/a	8.0

Abbreviations: SD, standard deviation; ABV, the background value of TMs in Anhui Province of China; GV, guide values of Soil environmental quality (GB36600-2018); n/a, not available.

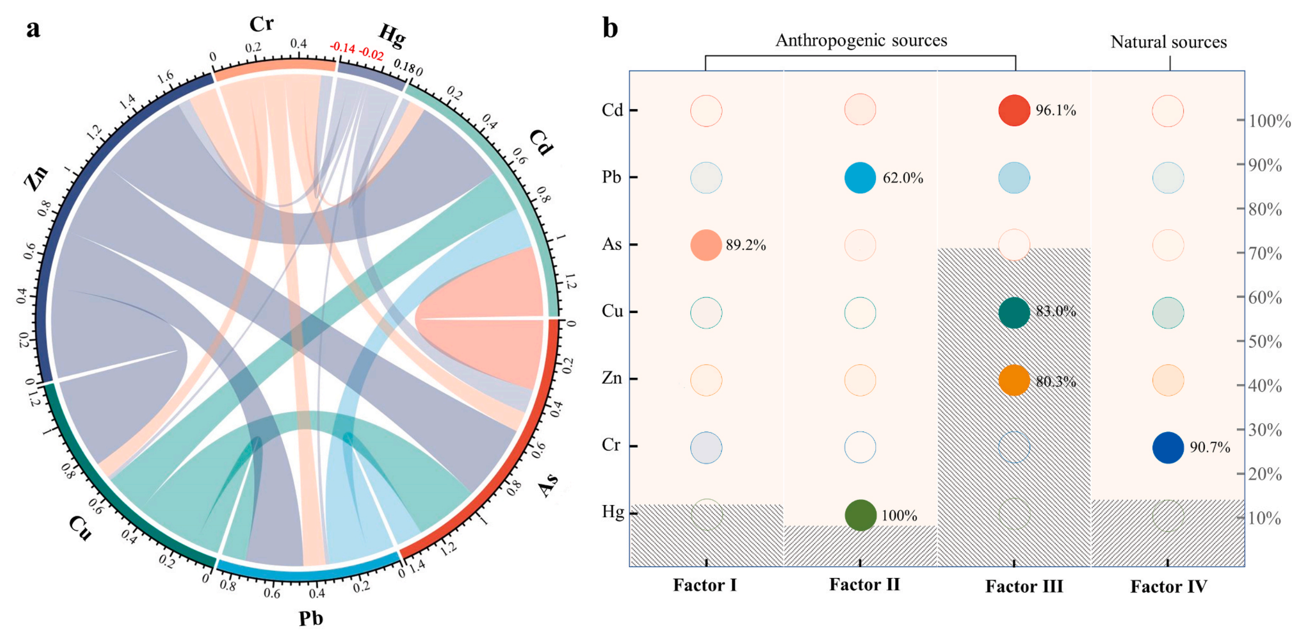

Fig. 2. Source analysis of trace metal(loid)s (TMs) in soils of the study area by combing a) Pearson correlation analysis and b) Positive definite matrix factorization (PMF). The width of each TM was shown to denote the correlation coefficient. The red scale is used to indicate a negative correlation between two TMs, while the black scale is for a positive correlation. The histogram is used to represent the percentage of each factor. Different color gradients are used to indicate the proportions of each TM for different factors. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

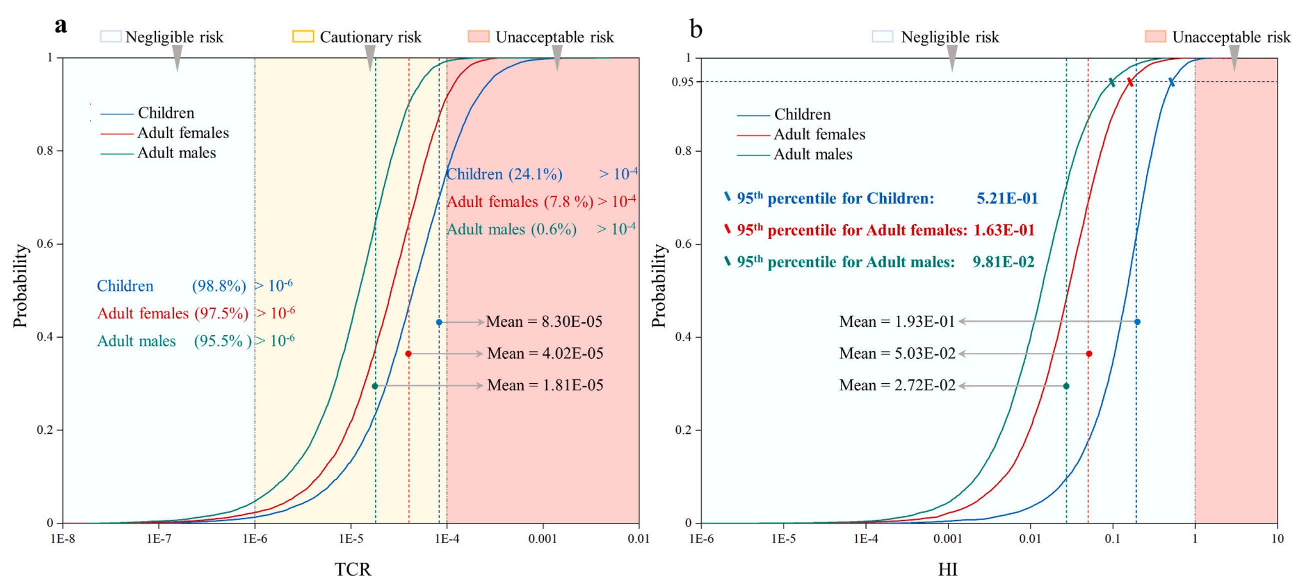

Fig. 3. The probability distribution for Carcinogenic risk and Non-carcinogenic risk: a) Total carcinogenic risk (TCR); b) Hazard index (HI). The red, blue, and green curves represented the probability distribution of adult females, children, and adult males, respectively. The black dotted line represented the acceptable threshold (TCR = 1E-06) and cautionary threshold (TCR = 1E-04) in Carcinogenic risk, and acceptable threshold (HI = 1) in Non-carcinogenic risk, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

concentration exceeding the screening value (namely, the guide value for soil pollution) may cause risks to human health (MEE, 2018). Therefore, whether the TM concentration exceed the corresponding guide values provides the essential basis for assessing the harm to human health (Guan et al., 2018). The concentrations of Cd, Cu, Pb, and Hg at all samples were below their corresponding guide values, indicating that these TMs were less likely to pose risks to human health. However, the As concentration in 14.4% of the samples exceeded the guide values, showing that As may pose a risk to human health. Therefore, local pollutant management and control should pay more attention to As pollution.

4.2. Source interpretation of trace metal(loid)s by using correlation analysis and PMF model

To obtain a reasonable and quantifiable explanation for the sources of TMs, the Correlation analysis and PMF model were conducted. In the PMF processing, the main indicators for PMF error evaluation are as follows: (1) more than 80% of the factors are mapped to the base factor in bootstrap (BS), (2) no factor swap was occurred for $Q_{max} = 4$ in displacement of factor elements (DISP), and (3) the objective function Q was minimum and stable bootstrap enhanced by displacement (BS-DISP) (Heidari et al., 2021; Norris et al., 2014). Furthermore, scaled residual

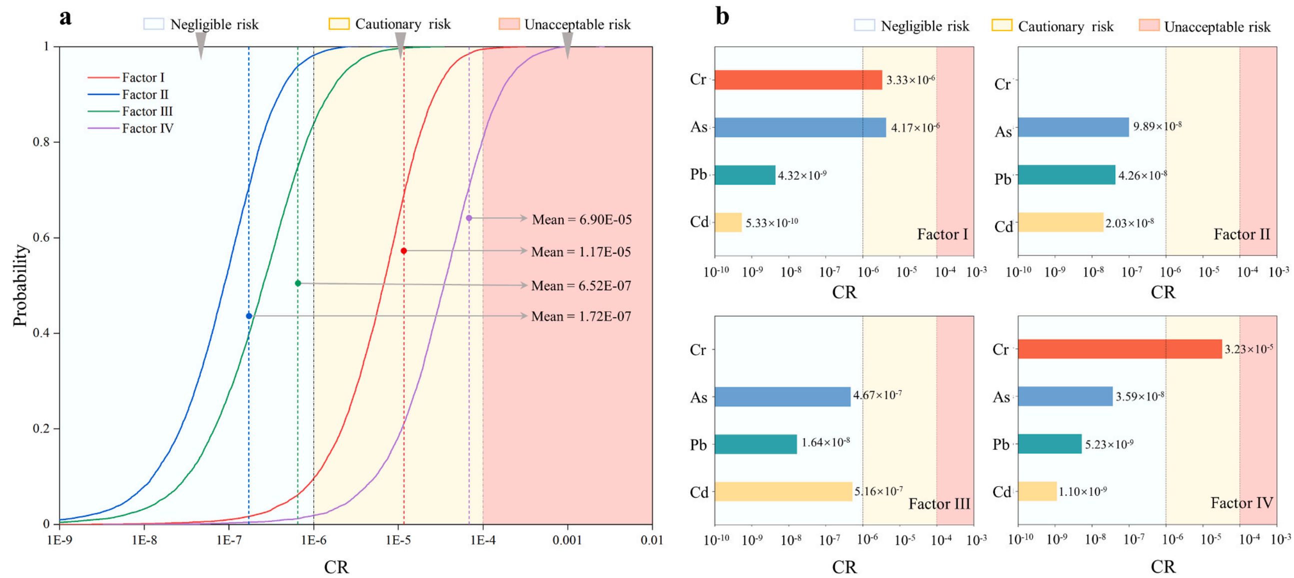

Fig. 4. (a) The probability distribution of Total carcinogenic risk (TCR) and the Carcinogenic risk (CR) of each TM based on different sources for children. a) Probability distribution of Total carcinogenic risk (TCR) based on probabilistic source-oriented risk (PSOR) model; b) the Carcinogenic risk (CR) of each TM based on different sources. The red, blue, green, or purple curves represented the probability distribution of Factor I, Factor II, Factor III, and Factor IV, respectively. The 1E-06 and 1E-04 of the black line represented the acceptable threshold and cautionary threshold in Carcinogenic risk. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

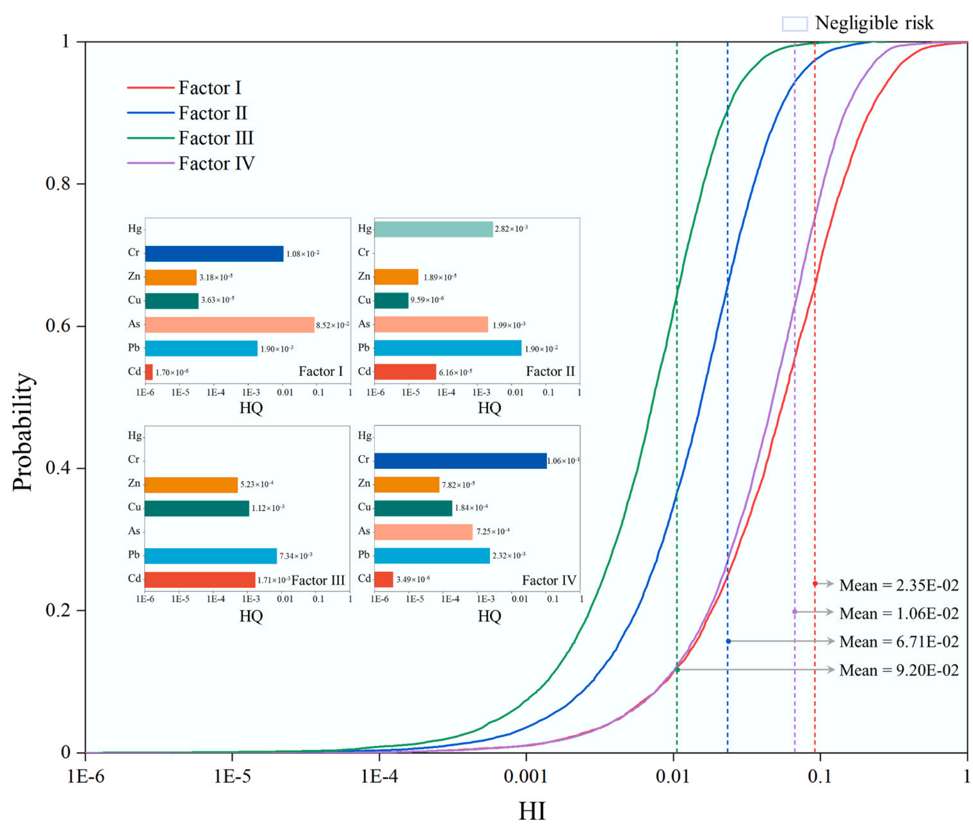

Fig. 5. The probability distribution of Hazard risk (HI) based on probabilistic source-oriented risk (PSOR) model, and the Hazard quotient (HQ) based on different sources for children. The red, blue, green and purple curves represented the probability distribution of Factor I, Factor II, Factor III, and Factor IV, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

values after standardization were within ±3, and the signal-to-noise (S/N) ratio of all TMs exceeded 2 (Guan et al., 2018). Therefore, the concentration data in the present study can be used for the PMF model, and the processing result was reasonable and reliable. According to the PMF processing result, four factors (named Factor I, Factor II, Factor III, and Factor IV) were finally determined, with the contribution rate of 12.4%, 8.1%, 64.1%, and 13.4%, respectively.

Factor I was mainly characterized by As (89.2%). The mean concentration of As exceeded the background value, showing the anthropogenic sources might contribute to the accumulation of As. Some previous studies have reported that As may be allocated to agricultural production (Fei et al., 2020; Zhang et al., 2018). For example, inorganic arsenic (calcium arsenate, sodium arsenate, lead arsenate) was proved to be widely used in fertilizers to increase crop production (Lu et al., 2012). Liang et al. (2015) also found that As was present in pesticides. Previous studies have shown that long-term agricultural activities and greening management (such as the over-use of fertilizers and pesticides) might lead to the accumulation of As in soils (Huang et al., 2021b; Jiang et al., 2017; Liu et al., 2017). There are many agricultural lands around the study area. Meanwhile, the green coverage in the study area is about 4169 ha, accounting for 40.1% of the whole study area. It was a common phenomenon to use fertilizers and pesticides in local farming and daily greening management. According to the 2019 local statistics yearbook (AQBS, 2019), 194,892 tons of fertilizers and 8001 tons of pesticides were used in 2018. Excessive use of fertilizers and pesticides in agricultural and greening practices could accumulate TMs in soils (Luo et al., 2009). Therefore, it could be inferred that Factor I was agricultural sources.

Factor II was dominated by Pb (62.0%) and Hg (100%). Automobile fuel containing Pb has been confirmed, so Pb is often regarded as the mark of traffic emissions and atmospheric deposition (Cai et al., 2015; Guan et al., 2018; Wang et al., 2019b). Previous studies have reported that automobile exhaust accounted for approximately two-thirds of the global lead emission over the past few decades (Fei et al., 2020). In addition, coal mining waste gas produced by combustion and fuel was another important source of Pb (Cui et al., 2018). Pb could diffuse into the atmosphere by burning fuel and then entered the soils through atmospheric deposition (Duan et al., 2020). According to the Anqing Traffic Statistics Yearbook (AQBS, 2019), the local road density and the number of vehicles were 172.7 (km/100 km^2) and 491,539 in 2018. The burning of a large number of fossil fuels and the increase in the number of vehicles would lead to increased Pb pollution. On the other hand, anthropogenic activities might cause the high over-standard rate of Hg (about 700% in this study). The correlation between Hg and other TMs (except for Pb) was weak or negative, indicating that Hg had different sources from them. Previous studies have supposed that Hg accumulation was mainly associated with coal combustion and traffic emission (Lv, 2019; Rachwał et al., 2015). Hg was easily volatilized into the atmosphere through fuel combustion because of its high volatility (Vejahati et al., 2010; Wang et al., 2012), which was different from other TMs. Hg in the atmosphere could enter the soil through dry and wet atmospheric deposition (Lv et al., 2013). With the increase in the number of vehicles and the development of industry in the study area, more and more industrial waste gas and automobile exhaust enter the atmosphere, causing the Hg and Pb concentrations to gradually increase. Consequently, Factor II could be explained as atmospheric deposition sources caused by fuel combustion and traffic emissions.

Factor III had high factor loading values for Cd (96.1%), Cu (83.0%), and Zn (80.3%). Pearson correlation analysis also proved that there were significant positive correlations between Pb and Cd, and between Zn and Cu. This indicated that these groups of TMs may come from the same pollution source. Many previous studies have described that Cd, Cu, and Zn were associated with industrial production. Firstly, Khan et al. (2016) confirmed that fossil fuel consumption, metal smelting, and waste incineration industry can cause the accumulation of Cd in environment media. The industry in the study area was dominated by coal mining and metal smelting. The exhaust gas was mainly produced in the combustion process and coal mining, and the industrial wastewater came from the metal smelting process. The statistics data (AQBS, 2019) showed that the industrial waste gas emission and wastewater in 2018 were 111.54 billion cubic meters and 28.56 million tons. Secondly, Ha et al. (2014) have shown that the Cu accumulation was related to industrial activities, such as coal-fired power and smelting plants. Previous studies have also found that Cu was mainly released in industries such as copper-zinc mining and smelting (Zhang et al., 2010), and copper compounds were also produced in electroplating industry (Yi et al., 2011). These processes could emit massive Cu-containing sewage and dust, which would cause soil pollution. Thirdly, (Li et al., 2020b) have showed that Zn was widely used in the galvanizing industry and zinc mining because of the corrosion resistance and excellent mechanical properties. Meanwhile, Zn was closely related to industrial discharge, such as mining and coal power plants (Liu et al., 2018a). In addition, Zn has also been reported to be used as a battery in the automotive and machinery manufacturing industry (Chen et al., 2008; Liu et al., 2017; Wang et al., 2020). Similar studies have also verified that Cu, Zn, and Cd were related to industrial activities (Dong et al., 2019; Huang et al., 2021b; Li et al., 2021, 2020b; Wang et al., 2019b). Therefore, Factor III can be interpreted as industrial sources.

Factor IV was exclusively dominated by Cr and explained 90.7% of the mean concentration in the PMF model. Cr showed a lower mean concentration than the background value, indicating that there is less contribution from external sources. Previously, Cr has been confirmed that it was generally widely present in soil parent materials and pedogenic processes (Huang et al., 2021b; Jin et al., 2019; Lu et al., 2012; Zhang et al., 2018). Hence, Factor IV can be interpreted as natural sources.

In recent years, many studies on TM pollution have been gradually focused on the quantitative source analysis based on receptor models (Huang et al., 2021b; Liu et al., 2018a), which combined mathematical-statistical analysis models with the information of pollutants in the environment to analyze the sources of pollutants. Compared with other receptor models, the PMF model had a non-negative constraint on a load of each factor in the solution process (Lv, 2019; Zhang et al., 2018), so the source composition spectrum of each TM was interpretable and the source of pollutants could be identified quantitatively. In particular, it can handle the missing data and the erroneous data. Therefore, more and more studies have tried to apply the PMF model to the source apportionment of soil TM pollution (Guan et al., 2018; Liu et al., 2018a). In general, the result of PMF model must follow the principle of "results can be explained" in source interpretation, which required researchers to have rich data analysis experience and comprehensive understanding of pollution characteristics in the study area. However, in the practical applications of PMF model, due to the complex emission of pollution sources, the phenomenon of superimposition transmission is common. It is difficult for PMF model to effectively separate the pollution sources with strong collinearity, and then the identified factors are often a mixture of several types of emission sources to varying degrees. Therefore, in addition to the non-negative elements of all the matrices, we will consider adding more constraints to the model calculations in subsequent studies, such as limiting the factor contribution matrix (F) based on the known information of the emission source spectrum to obtain more reliable analysis results.

4.3. Relationship among trace metal(loid)s, pollution sources, and health risks

By using Monte Carlo simulation, the health risks of all local populations (including children, adult females, and adult males) were assessed. The results showed that HI value of children had only a 0.2% probability of exceeding the USEPA`s guide values (HI = 1), while both adult males and adult females were in a safe state. Previous studies had reported that when the HQ value in the 95th percentile is lower than the acceptable risk threshold (HQ = 1) for Non-carcinogenic risk, it could be considered that the health risk caused by the target TMs is lower than the acceptable level (Guo et al., 2021). Therefore, it can be inferred that Non-carcinogenic risks for all populations in the study area are negligible. For another, nearly 98.75% (children), 97.5% (adult females), and 95.5% (adult males) of TCR values exceed the acceptable threshold of 1E-06 (Fig. 3b). It is worth noting that children, adult females, and adult males have 24.1%, 7.8%, and 0.6% probability of being at an "unacceptable risk" level (TCR > 10E-04) (Fig. 3a), showing that the Carcinogenic risk in this area was non-negligible. A high probability of Carcinogenic risk may lead to some diseases such as lung cancer and liver cancer (Kamarehie et al., 2019).

In addition, children suffered from higher Carcinogenic and Non-carcinogenic risks than adults (Fig. 3), which might be related to their lower body weight and higher feeding rates. Children have a higher feeding rate outdoors and more accessible contact with the soils, so it is necessary to keep their hands and mouth clean and avoid "hand eating" behavior (Wang et al., 2019a). Consequently, the risk assessment of soil TMs should pay more attention to children.

The health risk assessment could only help people understand the risk level but cannot effectively help decision-makers control TM pollution sources (Men et al., 2020). The source-oriented risk assessment results (Figs. 4–6, S5–S6) indicated that the health risks caused by four pollution sources had the following characteristics: (1) The health risk for all three groups (adult males, adult females, and children) showed the same change trend; (2) The Carcinogenic risk for all populations presented a high-risk level, while the Non-carcinogenic risk was negligible. Considering the uncontrollability of the natural sources, the health risks caused by natural sources of TMs are not deeply explored in this study. Therefore, this study took children as an example, focusing on the relationship between the anthropogenic sources (industrial sources, agricultural sources, and atmospheric deposition sources) and Carcinogenic risk.

As shown in Fig. 6 and Table. S4, the contribution rate of three anthropogenic sources to Carcinogenic risk was ranked as follows: agricultural sources (86.2%) > industrial sources (12.7%) > atmospheric deposition sources (1.1%). Previous studies have shown that the health risks are closely related to the toxic coefficients of target TMs, in addition to the TM content of pollution sources (Huang et al., 2018b). Industrial sources which accounted for 64.1% of all pollution sources were mainly loaded on Cu (83.0%), Zn (80.3%), and Cd (96.1%), but contributed only 12.7% to the Carcinogenic risk. It was mainly due to the lower carcinogenic toxicity of Cu and Zn. Although industrial sources had a low probability of posing a Carcinogenic risk to children, more policies are still needed to prevent the risk caused by TMs, particularly the potentially carcinogenic element Cd. However, agricultural sources accounted for 12.4% of all pollution sources and became the largest risk source with a carcinogenic contribution rate of 86.2%, far exceeding industrial and atmospheric deposition sources. It was mainly due to the high loading (89.2%) and the high toxicity of As in agricultural sources. Existing studies have shown that As could cause damage to lipids and proteins by destroying free radicals (Valko et al., 2006), which would result in many diseases such as cancer of the lungs, kidneys, bladder and skin, and nervous system (Järup, 2003; Jomova et al., 2011). Hence, agricultural sources and As were determined as the priority pollution source and the priority pollution element in the study area.

As discussed above, agricultural sources in this study area are mainly related to various agricultural activities and greening management. It is counted that the local fertilizer consumption reached 194,892 tons in 2018, increasing 5513 tons over 2010. Excessive use of fertilizers and pesticides in agriculture and urban greening could lead to As accumulation (Men et al., 2021). Other studies also proved that using organic fertilizers instead of chemical fertilizers could effectively reduce the accumulation of TMs in soils (Beesley et al., 2010; Luo et al., 2009). In general, it is believed that reducing the using pesticides and chemical fertilizers could effectively alleviate the health risks of residents. Given sustainable development, we advocate the use of low-toxic organic fertilizers instead of chemical fertilizers. It is worth noting that the use of chemical fertilizers reached a peak in 2015 (223,594 tons) and has shown a downward trend in recent years, indicating that the residents have consciously reduce the use of chemical fertilizers in agricultural production and greening management.

In general, our research emphasized that the largest source of pollution should not be confused with the largest source of health risks in health risk assessment. For the first time, the concept of priority control factor was put forward in this study, which can help determine the priority control level of soil pollution sources and target TMs and help decision-makers formulate strategies to mitigate pollution risks and reduce management costs. Future research should link priority control factor with more specific anthropogenic activities (such as pH, traffic volume, gross domestic product, population density, industrial distribution, and land use) to better understand the source of TMs and help local governments make targeted decisions.

5. Conclusion

In conclusion, taken Anqing City as a case, we have carried out the research to assess the source-oriented human health risks of TMs in soils. The main findings were as follows: (1) By combing PMF model and Correlation analysis, the agricultural sources, atmospheric deposition

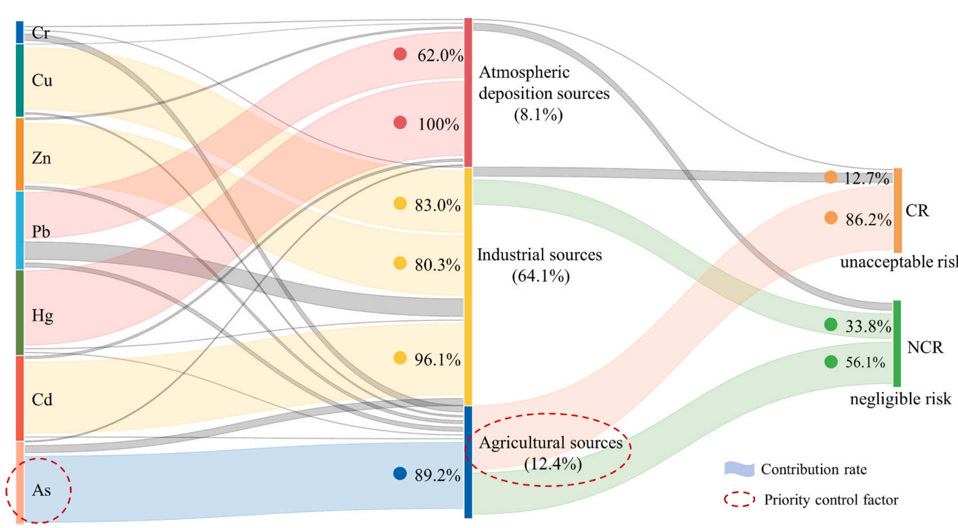

Fig. 6. Relationship among trace metal(loid)s, pollution sources, and health risks. The width of the curve represents the contribution rate. The curves of characteristic elements and characteristic pollution sources are marked in color. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

sources, and industrial sources were identified as the main anthropogenic sources; (2) Results showed that the Non-carcinogenic risks of all populations were acceptable, while the Carcinogenic risks were all at a high level; (3) Based on the developed PSOR model, we have quantitatively analyzed the relationship among TMs, pollution sources, and health risks. Then, agricultural sources and As were determined as the priority control factors in the study area.

In this study, for the first time, we put forward the concept of priority control factor for the management of soil TMs, which can help decision-makers to formulate target control policies and reduce management costs of soil pollution. We acknowledge the limitations of the present study because the health risks caused by natural sources of TMs are not deeply explored, while the higher background concentrations of TMs can also pose a threat to human health. Therefore, it is necessary to conduct research of TM pollution in the areas with high background concentration in the future. In addition, the models used in this study have certain limitations, and we will consider adding more constraints to the model calculations and linking priority control factor with more specific anthropogenic activities in future studies.

CRediT authorship contribution statement

Jiaxun Sun: Conceptualization, Experimental methodology, Formal analysis, Writing – original draft, Visualization, Validation and Software. **Menglu Zhao:** Visualization and Experimental methodology. **Jingling Huang:** Supervision. **Yafeng Liu:** Investigation. **Yuying Wu:** Resources. **Boya Cai:** Software. **Zhiwei Han:** Validation. **Honghui Huang:** Validation. **Zhengqiu Fan:** Funding acquisition and Writing-Editing.

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.

Acknowledgements

This work was supported by National Key Research and Development Program of China (No. 2018YFC1407501) and the Guangdong Provincial Key Laboratory of Fishery Ecology and Environment (No. FEEL-2017).

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2021.127116.

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