



# Risk sources quantitative appointment of ecological environment and human health in farmland soils: a case study on Jiuyuan District in China

Huilan Zhang · Min Zhang · Yueting Wu · Juan Tang · Shiyu Cheng · Yilin Wei · Ying Liu

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**Abstract** Heavy metals (HMs) in farmland soils lead to adverse influences on ecosystem and human health. Despite that, data on quantitative risk from different sources are still scarce. In this study, 100 farmland soil samples in Jiuyuan District were collected and analyzed for selected HMs (As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V and Zn) content characteristics and pollution statuses. The positive matrix factorization (PMF) model combined with the Nemerow integrated risk index (NIRI) and human health risk assessment (HHRA) was used to quantitatively identify the primary risk sources. The results indicated that the mean contents or median values (mg/kg) of 10 HMs were all higher than the background values. The contamination factor (CF) and pollution load index (PLI) revealed that the soil was severely polluted. Based on PMF, the main source of HM pollution was anthropogenic activities,

accounting for 78.91%. Sewage irrigation represented the biggest input but was not associated with the highest risk. The results of PMF-based NIRI and PMF-based HHRA showed that the chemical fertilizers and pesticides were the largest and priority risk sources with contribution rates of 38.10% to ecological risk and 34.61 and 32.82% to non-carcinogenic and carcinogenic risk, respectively. In addition, non-carcinogenic risk of children was higher than that of adults, while the carcinogenic risk was the opposite. The integrated approaches were beneficial for priority risk quantification from different sources and can provide direct risk information and effective policy recommendations for management and control of key risk sources.

**Keywords** Risks quantification · Priority · Source apportionment · Heavy metals · Farmland soils

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H. Zhang · M. Zhang · Y. Wu · J. Tang · S. Cheng · Y. Wei · Y. Liu (✉)  
College of Life and Environmental Sciences, Minzu University of China, Beijing 100081, China  
e-mail: liuying4300@163.com

Y. Liu  
Beijing Engineering Research Center of Food Environment and Public Health, Minzu University of China, Beijing 100081, China

## Introduction

Heavy metals (HMs) are being increasingly studied as an active field of multidisciplinary environmental problems (Pourret & Bollinger, 2018; Wang et al., 2020). It has attracted extensive attention of the public all over the world from the vision of environment and human health (Tóth et al., 2016; Wang et al., 2020). It is well known that more and more HMs have entered the agricultural ecosystem environment due to

frequent human activities, such as industrial processes, waste treatment, the application of agrochemicals and wastewater irrigation (Luque-Espinar et al., 2018; Osman, 2014). Moreover, determining the pollution sources of HMs in farmland soils is the basis to protect soil quality and make sustainable management strategies (Lu et al., 2012), and further identifying the risk sources of HMs is the prerequisite for risk control of farmland soils. Therefore, it is very necessary to determine the potential pollution sources and quantitatively identify ecological and human health risk sources of HMs in farmland soils.

The literature indicated that HMs in soils produced serious interference on ecological function (Liu et al., 2018; Mahbub et al., 2017) and had multiple effects on the availability and functions of soil in the ecosystem (Nielsen, 1997). Society of Environmental Toxicology and Chemistry (SETAC, 2018) pointed out that human health risk assessment determined the nature and possibility of harmful effects occurring to organisms (such as humans, animals or plants). In recent years, the study of ecological and human health risks on HMs in soils mainly has focused on the contents level (Giri & Singh, 2017; Kolo, 2018). It cannot provide specific and priority control pollution sources, which may result in the failure to implement targeted pollution control measures. Above problem had been solved by using the methods of quantitative source appointment, among them, positive matrix factorization (PMF) was widely used to quantify the soil pollution sources (Gan et al., 2018; Guan et al., 2018; Zhang et al., 2018). However, HMs produced by different pollution sources had different toxicity and risk levels (Jiang et al., 2017). This indicated that the pollution source, which contributed the most to the HMs contents, did not necessarily pose the greatest risk (Liu et al., 2018). Thus, it is a key and necessary to obtain the priority risk source based on the contribution of each pollution source to the potential risk, rather than content levels of HMs (Jiang et al., 2020).

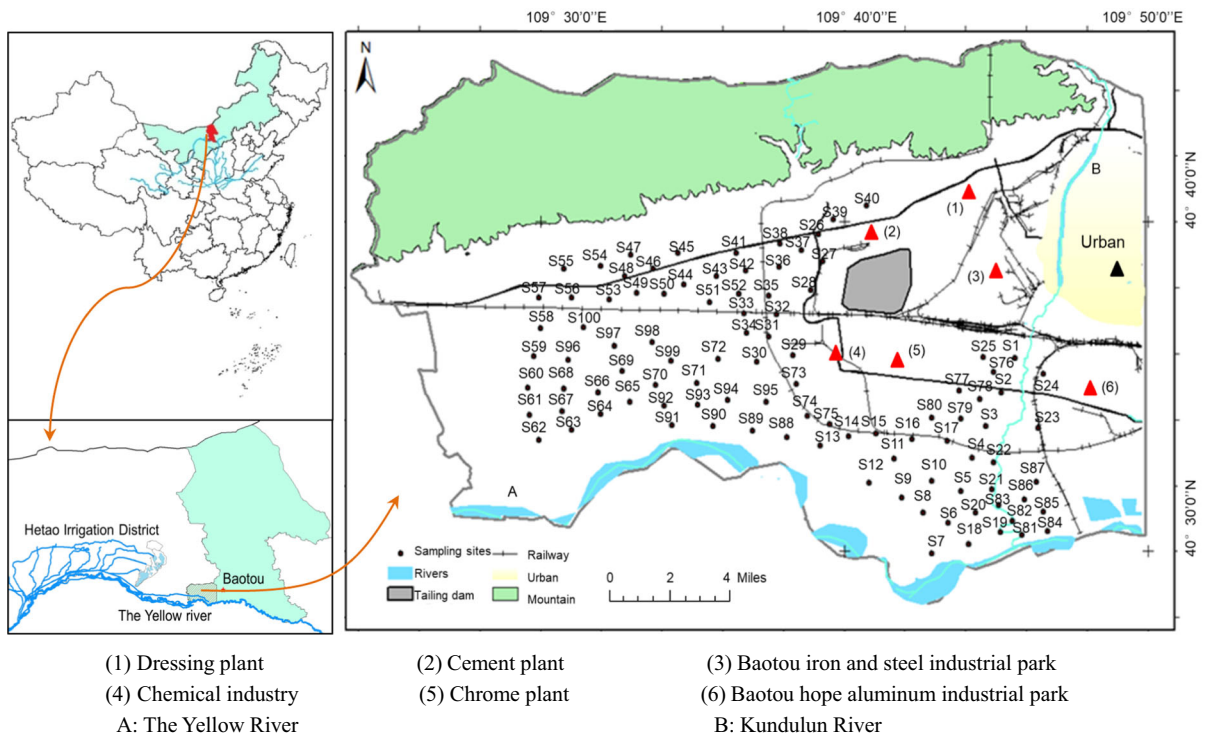
Jiuyuan District, located in the west of the Inner Mongolia Autonomous Region of China, and the north of the Yellow River, is the core area of the economic zone of “Hu Bao Yin Yu” and the urban agglomeration of “Hu Bao E Yu.” The current study on HMs in soils mainly focused on the distribution and source appointment in urban, industrial and park soils (Dai et al., 2017; Huang et al., 2017), and only minimal information was reported about PMF within a regional scale. But it was

studied little to quantitatively identify and apportion the ecological and human health risks of HMs in farmland soils. Therefore, based on the understanding of pollution sources, this study conducted quantitative analysis of risk sources. The main objectives were to (1) study the content characteristics and pollution statuses of HMs in farmland soils; (2) quantitatively identify and apportion possible sources of HMs by using PMF; (3) conduct the methods (PMF-based NIRI and PMF-based HHRA) to quantify the ecological risks and human health risks from different pollution sources and confirm the priority risk source. The results provided an effective quantitative way for risk appointment and can help to reduce the ecological and human health risks caused by HMs in farmland soils by managing and controlling the priority risk source.

## Materials and methods

### Study area

The study area ( $109^{\circ}23'35''$ – $109^{\circ}49'52''$ E,  $40^{\circ}46'29''$ – $40^{\circ}28'30''$ N), having a permanent population of 300,000 and an area of 360 km<sup>2</sup>, is located in Jiuyuan District, the southwest of Baotou, north of the Yellow River and south of the Daqingshan Mountain Range (Fig. 1). There is a continental semi-arid climate with an average annual temperature of 7.2 °C and total annual precipitation of about 421.8 mm. The dominant wind direction is northwest wind. Kastanozem is the main soil type, and farmland is a main type of land use. Because of the proximity to desert, there are frequent gale ( $N > 17$  m/s), floating dust and sandstorm in this area from March to May. According to the 2019 Statistical Yearbook of Baotou, the agriculture of Jiuyuan District is relatively developed and mainly produces food crops (corn and wheat) and cash crops (vegetables). This area is also an important production base of vegetables (cucumber, tomato and garland chrysanthemum) in Baotou, which not only undertakes the consumption of local people, but also sells to all parts of the country. In the surrounding environment of the sampling area (Fig. 1), there are two major industrial parks, namely Baotou iron and steel industrial park (BISIP) and Baotou hope aluminum industrial park (BHAIP). BISIP is located in the west of the Kundulun River, mainly processes coal, copper, nickel, iron, chromium, manganese and other resources; at the southwest direction of BISIP, there is a



**Fig. 1** Location of study area and distribution of sampling sites (The data set of Chinese Map is provided by Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences (<http://www.resdc.cn>))

tailings pond, which put into use in 1965. Now, there are about 1.35 tons of tailings and 15 million cubic meters of waste water (Han et al., 2018). BHAIP is located to the east of the Kundulun River and south of the city. It is mainly made of electrolytic aluminum, and the production process produces heavy metal pollution such as nickel, chromium and other HMs (Zhang et al., 2016). The Kundulun River is the main industrial sewage river, flowing from north to south through Jiuyuan District. Both it and the Yellow River which flows through Baotou are the main sources of agricultural irrigation water in study area (Han et al., 2018; Si et al., 2015). More importantly, the sampling area also includes an intricate railway lines mainly used to transport mineral resources and a 110 National Highway which traverses the sampling area.

**Sample collection and analysis**

In April 2019, 100 topsoil (0–20 cm) samples were taken from farmland in Jiuyuan District (Fig. 1) and collected by using the chessboard layout method with a

sample spacing of about 2 km, each sample was a mixture of five subsamples within about 100m<sup>2</sup>, and 1.0 kg fresh subsample was collected in polyethylene bags by stainless steel auger and brought back to the laboratory. All soil samples were air-dried, fully mixed and screened. The samples were digested with HNO<sub>3</sub>-HF-H<sub>2</sub>O<sub>2</sub> by microwave, and the contents of 10 HMs were determined by inductively coupled plasma mass spectrometry (Agilent 7700 ICP-MS). The soil pH was measured according to the agricultural industry standard (NY/T1377-2007), and the quality assurance and quality control (QA/QC) measures were ensured by analysis of duplicate samples, standard reference materials and method blanks. The duplicate samples were determined randomly for 20% of the collected samples, and the relative standard deviations (RSD) were not more than 5%. The recoveries for 10 HMs of the Chinese standardized reference material (GBW07410) ranged from 92.45 to 105.21%. And the method detection limits were 0.016, 0.01, 0.004, 0.011, 0.084, 0.039, 0.001, 0.171, 0.003 and 0.003 μg/L for As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V and Zn, respectively.

## Pollution assessment

The contamination factor (CF) and pollution load index (PLI) were used to analyze pollution levels (Mamut et al., 2017). CF is defined as the following equation:

$$CF = \frac{C_i}{C_n}$$

where  $C_i$  is the content of heavy metal element  $i$  (mg/kg);  $C_n$  represents the local background value of heavy metal element  $i$  (mg/kg).

The PLI can be calculated as the following equations:

$$PLI = \sqrt[n]{CF_1 + CF_2 + \dots + CF_n}$$

where CF is the contamination factor above;  $n$  indicates the total number of heavy metal elements.

Quantitative ecological risk (PMF-based NIRI) and quantitative human health risk (PMF-based HHRA)

### Positive matrix factorization (PMF)

Positive matrix factorization (PMF) was an effective multivariate factor analysis tool used to quantify the contribution of pollution sources to the sample according to the composition of the sources (Paatero & Tapper, 1993). In this study, the PMF model was run using the content data (10 HMs in 100 soil samples) and the uncertainty data file (including sampling and analysis errors). It can be expressed as:

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

where  $X_{ij}$  is the sample content matrix;  $g_{ik}$  refers to the contribution of each factor to any given sample;  $f_{kj}$  represents chemical composition matrix of  $p$  source;  $e_{ij}$  is the residual matrix for each sample;  $Q$  is the sum of squares of the differences.

The uncertainty of the concentration was calculated using the following equation:

$$\begin{aligned} \text{Unc} &= \frac{5}{6} \text{MDLUnc} \\ &= \sqrt{(\text{ErrorFraction} \times C_i)^2 + (0.5 \times \text{MDL})^2} \end{aligned}$$

where  $C_i$  is the concentration of the heavy metal  $i$ , MDL is the species-specific method detection limit, and error fraction is a percentage of the measurement uncertainty.

### Quantitative ecological risk (PMF-based NIRI)

In order to conduct quantitative potential ecological risk assessment of HMs in farmland soils, the specific steps of the PMF-based NIRI were as follows:

Step 1: PMF model was used to estimate the contribution contents of each pollution source to HMs in each sample, and the calculation is as follows:

$$C_{jn}^l = C_{jn}^{l*} \times C_j$$

where  $C_{jn}^l$  is the mass contribution (mg/kg) of HM  $n$  from source  $l$  in the sample  $j$ ;  $C_{jn}^{l*}$  is the calculated contributions of HM  $n$  from source  $l$  in sample  $j$ , and  $C_j$  is the content (mg/kg) of soil HMs in sample  $j$ .

Step 2: The contribution content of HMs source obtained from Step 1 was integrated into the Nemerow integrated risk index (NIRI) formula (Men et al., 2019) to obtain the quantitative ecological risk source. The details were as follows:

NIRI was developed for assessing the integrated ecological risk of HMs based on the Nemerow integrated pollution index (NIPI) and the potential ecological risk index (RI). It took into consideration the toxic response factor and eliminated the influence of accumulation risk caused by the number of assessed HMs, and its calculated results were better than NIPI and RI (Hakanson, 1980). The calculation formulas of NIRI are as follows:

$$\text{NIRI} = \sqrt{\frac{E_{r\max}^2 + E_{r\text{average}}^2}{2}}, E_r^i = T_r^i \times \frac{C_{jn}^l}{B_n^i}$$

where  $E_r^i$  is the potential ecological risk factor of heavy metal  $i$ , and  $r$  is the abbreviation of risk and is not a variable;  $T_r^i$  is the toxic response factor of heavy metal  $i$ , and the toxic response factors of As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V and Zn were 10, 30, 5, 2, 5, 1, 5, 5, 2, 1, respectively;  $C_{jn}^l$  is the mass contribution (mg/kg) of HM  $n$  from source  $l$  in the sample  $j$ ;  $B_n^i$  is reference value for soil geochemical background. The classification of all the indexes referred to above is listed in Table S1-1, S1-2.

*Quantitative human health risk (PMF-based HHRA)*

In order to determine the probabilities of non-carcinogenic and carcinogenic risk of HMs in farmland soils to the public, the specific steps of the PMF-based HHRA were as follows:

Step 1: Same as the “Step 1” of “PMF-based NIRI.”

Step 2: The contribution content of HMs source obtained from Step 1 was integrated into the human health risk assessment (HHRA) to obtain the quantified source non-carcinogenic and carcinogenic risk. The details were as follows:

Based on behavioral and physiological differences, local residents were divided into two groups: adults and children. They mainly contact soil HMs through skin contact ( $CDI_{der}$ ), dust particles inhalation ( $CDI_{inh}$ ), soil and dust intake ( $CDI_{ing}$ ). In this paper, the exposure measurement models proposed by USEPA (1989) are as follows:

$$HQ = \frac{CDI_{jning/der/inh-nc}^I}{RfD}, HI = \sum HQ;$$

$$CR = CDI_{jning/der/inh-ca}^I \times SF, TCR = \sum CR$$

where  $CDI_{jning/der/inh}^I$  is the average daily exposure dose of HMs by three exposure pathways; HQ is the non-carcinogenic health risk index caused by heavy metals in the exposure pathway; HI is the non-carcinogenic risk caused by multiple pathways; CR is the carcinogenic health risk index caused by heavy metals; TCR is the total carcinogenic risk of carcinogenic metals. Other relevant formulas, parameters and assessment criteria for risk assessment are shown in Table S2-1, S2-2 and S2-3.

In order to improve the accuracy of the health risk assessment results, the Monte Carlo method was used to analyze the uncertainty and sensitivity of its parameters. the calculation process is:

$$G_Y(\eta) = \int_{-\infty}^{\eta} g_Y(\eta) d\eta$$

where  $G_Y(\eta)$  is the distribution function of the output  $Y$ ;  $g_Y(\eta)$  is the probability density function of the output  $Y$ , obtained by passing the probability density function of the input through the model.

In this study, combining PMF with NIRI and HHRA (PMF-based NIRI, PMF-based HHRA) not

only strengthened the risk source, but also remedied the inaccurate risk control measures caused by the unilateral risk assessment (NIRI or HHRA) and the incompleteness of source pollution control based on the PMF.

Statistical analyses of data

Origin2018, SPSS 17.0 and Microsoft Excel 2010 were used for statistical analysis of data; ArcGIS 10.2 (ESRI US) was used to draw sampling maps and the spatial distribution maps of quantified risks of different sources. The sources of HMs were identified by positive matrix factorization (PMF) model (USEPA PMF 5.0).

**Results and discussion**

Content characteristics and pollution levels of HMs

The descriptive statistics analyses of 10 HMs contents (Table 1) indicated that the soil was medium alkalinity with the mean pH of 8.01. The Kolmogorov–Smirnov test showed that As ( $Z = 1.315, p = 0.063$ ), Mn ( $Z = 1.156, p = 0.138$ ) and V ( $Z = 0.796, p = 0.550$ ) had normal distributions and the other 7 HMs had non-normal distributions. The range of 10 HMs contents was 2.3–571(mg/kg), following a descending order as: Mn > Zn > Cr > V > Ni > As > Cu > Pb > Co > Cd. Cr was greatly influenced by the human disturbance ( $CV = 1.2$ ). The contents of 10 HMs were all higher than the background values of soil environment in Inner Mongolia (CNEMC, 1990). The over-standard rates of As, Cd, Cr, Ni, V and Zn were all 100%, and that of Co, Cu, Mn and Pb were more than 90%. Meanwhile, according to the screening values of soil pollution risk in agricultural land of soil environmental quality in China (GB 15,618–2018), the pollution of As and Cd was the most serious, which would cause ecological environment risks to farmland soils and affect the growth and quality safety of agricultural products. Moreover, the contents of Cd and Cr (2.3 and 82 mg/kg) were higher than that in the industrial area (0.51 and 41 mg/kg) in the literature, indicating that the pollution of Cd and Cr was not only from industrial activities. The detailed pollution sources and control measures are shown in Sect. 3.2 and Sect. 3.3 of the

**Table 1** Descriptive statistical analysis of HMs in farmland soils (mg/kg)

Projects	pH	As	Cd	Co	Cr	Cu	Mn	Ni	Pb	V	Zn
Min, Max	7.4, 9.0	18, 49	2.0, 4.8	7.6, 26	57, 1162	12, 177	408, 944	25, 470	14, 150	59, 97	52, 568
Mean(SD)	8.0(0.27)	36(6.7)	–	–	–	–	571(99)	–	–	75(9.0)	–
Percentile	25th	–	–	2.2	11	74	22	–	33	18	80
	50th	–	–	2.3	12	82	25	–	38	23	96
	75th	–	–	2.5	14	90	31	–	44	27	122
CV	0.030	0.18	0.15	0.27	1.2	0.67	0.17	1.0	0.65	0.12	0.60
Background value <sup>a</sup>	–	6.30	0.04	9.20	36.50	12.90	446.00	17.30	15.00	44.70	48.60
CF	–	5.8	59	1.4	2.3	2.3	1.3	2.6	1.7	1.7	2.3
Risk screening value <sup>b</sup>	> 7.5	25.00	0.60	–	250.00	100.00	–	190.00	170	–	300
Mean value in industrial area <sup>c</sup>	–	–	0.51	–	40.58	38.44	714.86	45.02	55.06	–	99.20
The soil screening guidelines in the USA <sup>d</sup>	–	0.4	78	–	390	–	–	1600	400	550	23,000
The soil prevention level in Germany <sup>e</sup>	–	50	20	–	400	–	–	140	200	–	–
Canadian environmental quality guidelines <sup>f</sup>	–	–	1.4	40	64	63	–	50	70	130	200

“–” means no data

<sup>a</sup>data from (Chinese soil background value) CNEMC (1990)

<sup>b</sup>data from (Soil environmental quality the screening values of soil pollution risk in agricultural land)(GB 15,618–2018)

<sup>c</sup>data from the research of Sun et al. (2016)

<sup>d</sup>data from (USEPA Soil Screening Guidance) (USEPA Generic SSLs, 2018)

<sup>e</sup>data from (German federal soil protection and contaminated sites regulations) (German Federal Ministry of the Environment, 1999)

<sup>f</sup>data from (Canadian soil quality guidelines for the protection of the environment and human health) (CCME, 2010)

study. Compared with the soil screening guidelines in the USA (USEPA Generic SSLs 2018), the over-standard rate of As was 100%, but it did not exceed the level of soil prevention in Germany (German Federal Ministry of the Environment 1999). The contents of Cd and Cr were both higher than the limits (1.4 and 64 mg/kg) of the Canadian environmental quality guidelines (CCME 2010), but they were lower than the soil standards of the USA and Germany. The contents of the other 7 HMs were lower than the soil standards or unlimited value requirements of the three countries. Further, the results of HM pollution by CF and PLI (Mamut et al., 2017) (Table 1) showed that HMs had different anthropogenic pollution levels. The most noteworthy was that Cd was high pollution ( $CF \geq 6$ ), followed by As with considerable pollution ( $3 \leq CF < 6$ ) (Table S3). According to the classification standard of PLI (Table S3-1), the average value of the soil PLI (2.9) indicated that the HMs were at a severe

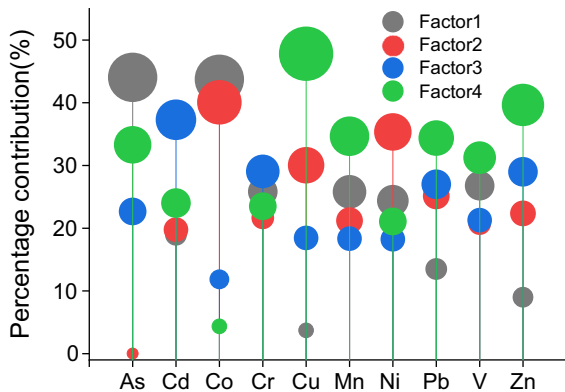
pollution level. Moreover, it can be seen from Fig.S3-1, soil HM pollution was mainly distributed in the following three areas: along the G110 National Highway, the entrance of the Yellow River and the southeast corner of the sampling area, respectively, where HMs released and accumulated through pollution sources were more likely to aggravate the pollution level, and should be paid attention to by local governments and residents.

#### Source appointment of HMs

##### *Factor loadings and source identification*

Based on CA (Tables S3-2), PMF model was used to quantify the contribution of pollution sources. The specific results were as follows:

The contribution rates of factor 1–As (44.05%) and Co (43.74%) were relatively high (Fig. 2). It had been



**Fig. 2** Contribution rates of different sources on heavy metals based on PMF

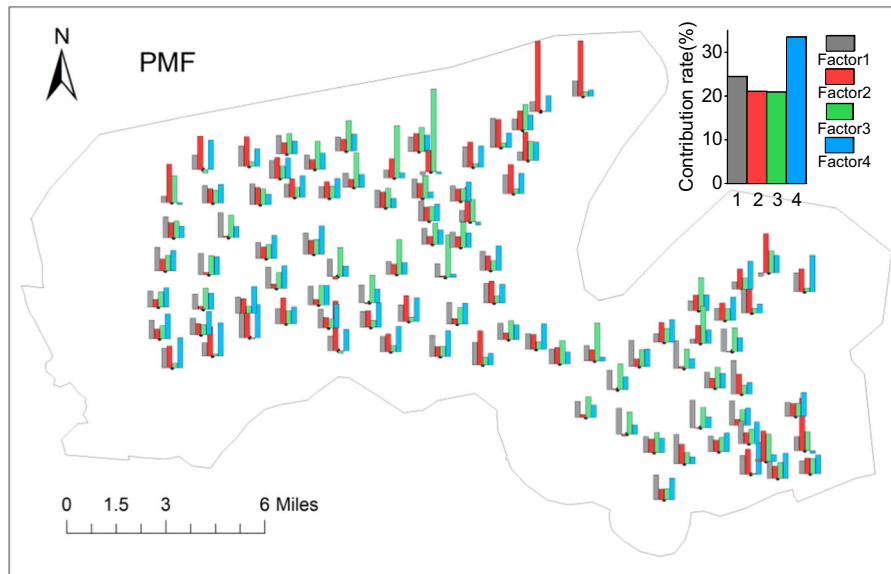
reported that As content in phosphate fertilizer was higher (19.40 mg/kg). Calculation based on P<sub>2</sub>O<sub>5</sub>, As content (155.80 mg/kg) was three times of the standard limit value. Hu (2014) found that the content of As in the compound fertilizer was the highest, reaching 38.12 mg/kg. Thus, the application of the two kinds of chemical fertilizers was the main way for As to migrate to the soil (Rafique & Tariq, 2016). In addition, a large number of inorganic As compounds, such as calcium arsenate and sodium arsenate, were found in agricultural pesticides or herbicides (Cai et al., 2015). Co is a beneficial element for plants, and a proper amount of Co fertilizers can promote plant growth by increasing photosynthesis (Zhou, 2013). But the content of Co was higher than the background value of Inner Mongolia (9.20 mg/kg) in 94% of the sampling sites, and this suggests an excess fertilizer input by the Co. It could be concluded that factor 1 was chemical fertilizers and pesticides.

Factor 2 was dominated by high load of Co (40.06%), Cu (30.03%) and Ni (35.34%) (Fig. 2), accounting for 21.09% of all pollution sources. The three HMs were generally considered as indicators of natural sources and had been confirmed by most studies (Mikkonen et al., 2018; Zhou et al., 2016). According to the spatial distribution of factor 2 in the sampling area (Fig. 3), it can be seen that its contribution was at a low level. The high contribution area was located between the south of Daqing Mountain and the north of BISI. And it had been pointed out that Cu and Ni were significantly positively correlated with MgO and CaO (product of the soil formation process)

(Huang et al., 2018). In conclusion, factor 2 was derived from a natural source.

The contribution rate of factor 3 to the pollution was 20.93%, mainly Cd and Cr, accounted for 37.29 and 29.05%, respectively (Fig. 2). There was a significant positive correlation between Cd and Cr ( $p < 0.01$ ), indicating that the pollution of these two HMs was mainly affected by similar human activities. The high polluted areas of Cd and Cr were mainly near the BISI, the tailings dam and the intersection of railway lines. Cr pollution would be produced in the chromium plating plant during the smelting of ferroalloy and manufacturing process of chemical products. And a large amount of dust and solid wastes containing Cd and Cr would be produced at coking plant and sheet-metal mill of BISI (Wang, 2018). In the industrial agglomeration areas, the density of transportation was relatively high, and BISI was the first large-scale iron and steel enterprise built in the minority areas in China. The long service life of the roads was the main reason for the accumulation of Cd and Cr in the surrounding farmland soils (Silva et al., 2016). Therefore, factor 3 was assumed as industrial and traffic emissions.

Factor 4 was the largest contributor to the pollution in the study area, with a contribution rate of 33.52%. This factor showed high load on Cu (47.82%), Zn (39.65%), Mn (34.66%), Pb (34.37%) and V (31.23%) (Fig. 2). There was a significant positive correlation between these 5 HMs ( $p < 0.01$ ), and the average contents were all higher than the background value of soil environment in Inner Mongolia (12.90, 48.60, 446.00, 15.00, 44.70 mg/kg). From the spatial distribution of factor 4 in Fig. 3, it can be seen that the places with high contribution rates of factor 4 were mainly the entrance of the Yellow River and the area where Kundulun River flowed (near the BHAI). According to the list of enterprises involved in heavy metal key industries in 2018 in Inner Mongolia, the upstream of study area (the Hetao Irrigation District) included numerous lead, zinc and copper mines and milling plants. Therefore, it was speculated that the wastewater containing HMs may flow into the Yellow River and flow through Baotou. The HMs in the soil were seriously polluted by sewage irrigation. This result was consistent with that of literature studies (Liao et al., 2017; Zhang et al., 2018). At the same time, the Kundulun River was also the main industrial sewage river in Baotou. After receiving industrial



**Fig. 3** The spatial distribution of contribution rates of different sources on sampling area

wastewater from the sewage outfall of the river, it flowed into the Yellow River from the north to the south from the suburb. According to the above analysis, factor 4 was sewage irrigation.

According to PMF, combined with the actual situation of sampling area, the results were as follows: The main source of HM pollution of farmland soils in Jiuyuan District was anthropogenic activities, accounting for 78.91%. Among them, sewage irrigation was the largest input way, followed by the use of chemical fertilizers and pesticides, and the industrial and traffic emissions were the smallest.

However, the problem with these results was that the greatest pollution source was not necessarily the greatest risk source causing local farmland ecological environment and human exposure. It would lead to unnecessary financial and material consumption if relevant departments took measures only based on the results. For this reason, scholars focused on the key problem were how to identify risk sources rather than the pollution sources quantitatively and provide the contribution rate of priority risk source, so as to carry out targeted prevention and control and policy recommendations on the risk of HM pollution in farmland.

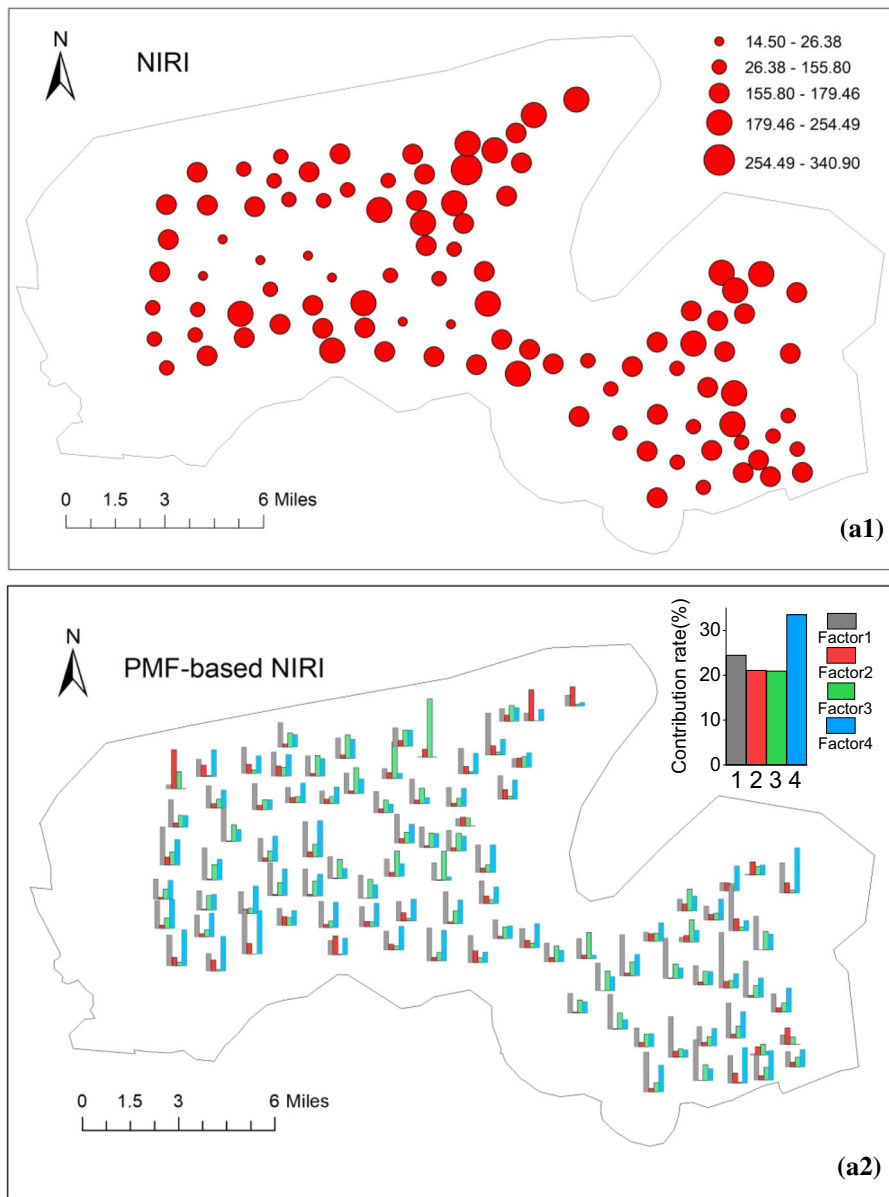
#### Uncertainty analysis

Uncertainty analysis was carried out regarding goodness of fit and bias and variability in the PMF solution. The coefficient of determination ( $R^2$ ) and bootstrap (BS) methods were used (Men et al., 2019). In this study,  $R^2$  of predicted and measured contents of 10 HMs were higher than 0.80 (Fig.S3-2), indicating that the predicted contents in PMF model well explained the values and variation of measured contents (Tian et al., 2019). The BS analysis showed that the matching degree of all factors was above 95% ( $n = 100$ ), indicating good robustness. Contributions of base run of factors to most heavy metals were contained in the small interquartile range and close to median values of bootstrap (Fig.S3-3), suggesting that the results were representative and reliable.

#### Quantification of potential ecological risk from different sources (PMF-based NIRI)

From the spatial distribution of the NIRI (Fig. 4a1 before quantification), more than 90% of sampling points had different degrees of ecological risk. The high risks (NIRI > 160) were mainly concentrated along the railway and the Kundulun River. However, the disadvantage of NIRI was that it only primarily determined the ecological risk and could not deeply comprehend the priority source of ecological risk. Combined with the



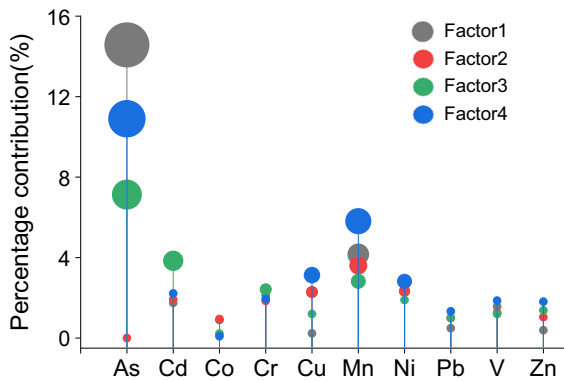


**Fig. 4** The results of risk assessment before and after quantitative analysis of PMF combined with NRI (a1. The spatial distribution of NRI, a2. The spatial distribution of PMF-based NRI)

distribution of sampling sites and field investigation, possible source of pollution can be inferred, it is impossible to directly obtain the types of main risk sources. It would lead to not only the inaccurate risk prevention and control but also the continuous pollution and hazards of HMs and As, which was also a kind of resource consumption for regional governments.

Different from the results of NRI, the PMF-based NRI method was used to determine the local optimal

risk source and conduct quantitative analysis, as shown in Figs. 4a2 and 5. From Fig. 4a2 it can be seen that the risk contribution of factor 1 was relatively uniform, and the range of change was small, and the risk contribution of factor 2 (natural source) was the lowest, while factor 3 and factor 4 had obvious high risk areas, which were distributed along railway lines and national roads, the entrance of the Yellow River and along the Kundulun River, respectively. From



**Fig. 5** The ecological risk contribution values of HMs in different pollution sources

Fig. 4a2 the results of contribution rate about the four pollution sources to the ecological risk were different from the PMF results. The largest contribution source of ecological risk was factor 1 with a risk contribution rate of 38.10%, followed by factor 4 (29.49%), factor 3 (20.83%) and factor 2 (11.58%).

In addition, the difference of ecological risk contribution rate of different factors can be seen in Fig. 5, factor 1 (chemical fertilizers and pesticides), factor 4 (sewage irrigation) and factor 3 (industrial and traffic emissions) had the highest risk load on As, which were 14.57, 10.90 and 7.13%, respectively, and the main risk of As came from chemical fertilizers and pesticides. The input of compound fertilizer, phosphate fertilizer and pesticide containing As in local farming activities was the most harmful. According to the Baotou Statistical Yearbook 2019, 5454 tons of phosphorus fertilizer and 26,518 tons of compound fertilizer were applied to local farmland in 2018, and the annual increasing use of chemical fertilizer led to continuous enrichment of As in the soil. Moreover, for Mn ecological risk, the contribution rate of sewage irrigation was the highest with 5.81 and 4.16% of chemical fertilizers and pesticides. The ecological risk caused by As and Mn from anthropogenic activities accounted for about 50%, indicating that As and Mn should be monitored preferentially. What's more, corresponding measures should be implemented, such as scientific soil testing and fertilization, and irregular water quality assessment to water source for water diversion irrigation. In conclusion, according to the results of PMF-based NIRI quantitative method, it was found that the farmland soils in Jiuyuan District had the highest ecological risk due to the use of chemical

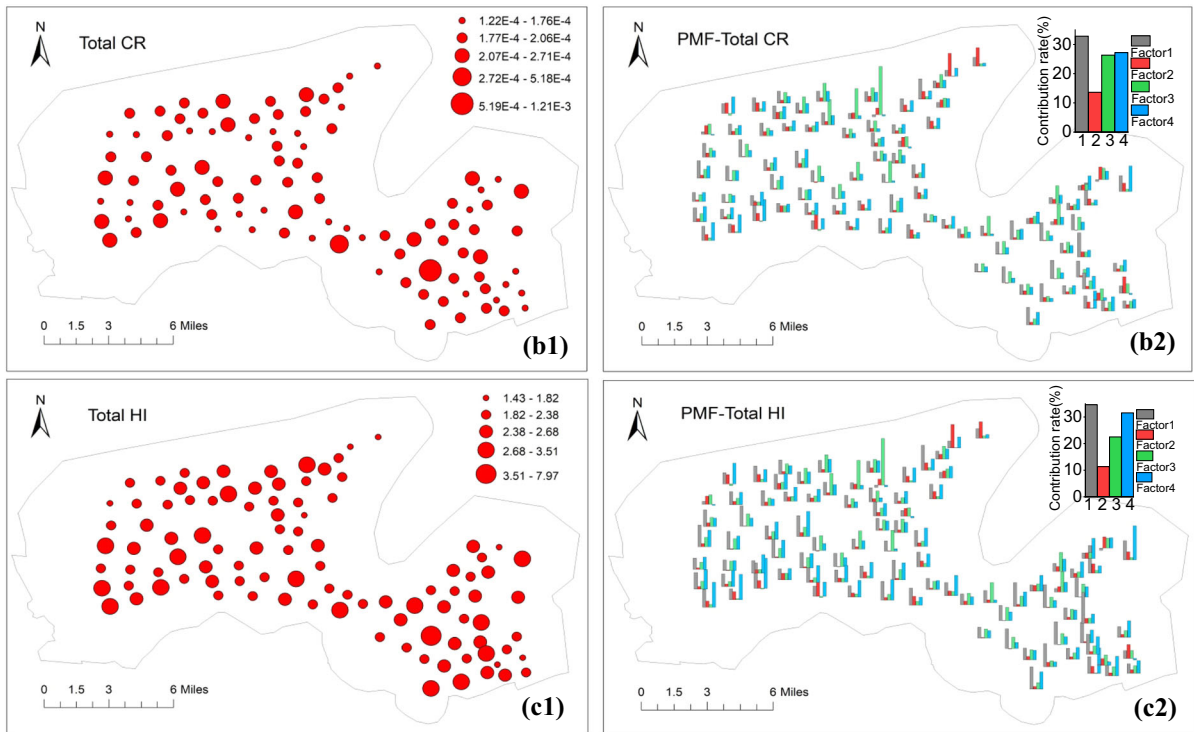
fertilizers and pesticides. In addition, the factors with the highest ecological risk contribution to As and Mn were consistent with the results of PMF source analysis, that is, the largest pollution source and the largest ecological risk contribution source of As were the unreasonable use of chemical fertilizers and pesticides. Mn came from the local sewage irrigation. Except As and Mn, the contribution rates of ecological risk of other 8 HMs from any source were very small.

In general, factor 1 (chemical fertilizers and pesticides) posed the highest ecological risk and should be prioritized for supervision and management in order to reduce the potential ecological risks caused by farmland soil HMs. From PMF-based NIRI, we can directly and accurately find the priority risk source of local farmland ecological environment damage and provide scientific basis for potential ecological risk management and control. Therefore, the quantitative assessment results of PMF-based NIRI were more meaningful than only comparing with the specified risk threshold.

Quantification of human health risk from different sources (PMF-based HHRA)

#### *Deterministic human health risk assessment*

In this study, PMF combined with HHRA was used to quantitatively study human health risk, and the results of before and after quantification are shown in Fig. 6. Co and V (non-carcinogenic risk), Co, Cu, Mn, Ni, Zn and V (carcinogenic risk) were not analyzed because of their lack of corresponding parameters or not classifiable them as harmful to human carcinogenicity. According to the spatial distribution of Total HI in Fig. 6c1 the high-risk distribution of Total HI was widespread, mainly concentrated in the southeast corner—the downwind position of BISI and tailings dam and along the Kundulun River. It can be seen from Fig. 6b1 (the spatial distribution of Total CR), the high-risk areas were relatively dispersed, but the distribution trend was consistent with that of Total HI. But it can be seen from Table S3-3 that single heavy metal had different effects on HIs and CRs in adults and children. For HIs, except for Pb, the risk of other HMs to children was a greater than that to adults, especially As and Cr had high potential risks for both adults and children, and As had a higher risk, and HIs for children and adults were 15.33 and 2.15, respectively. For CRs, the risk of single element carcinogenesis for children was lower



**Fig. 6** The results of risk assessment before and after quantitative analysis of PMF combined with HHRA (b1. The spatial distribution of Total CR, b2. The spatial distribution of

PMF-Total CR, c1. The spatial distribution of Total HI, c2. The spatial distribution of PMF-Total HI)

than that for adults, in which As and Cr had high CRs ( $CRs > 1 \times 10^{-4}$ ). It can be seen that HHRA had the same shortcomings as the above NIRI method, especially for the areas with complex pollution sources (such as the southeast corner of the sampling area), it was almost impossible to determine the largest risk source for risk prevention and control.

Different from the results of HHRA (Fig. 6b1, c1), the PMF-based HHRA method (Fig. 6b2, c2) can more specifically determine the risk sources that caused human health problems and conducted out quantitative analysis. It was found that the spatial distribution trends of PMF-Total CR and PMF-Total HI were the same, and the unreasonable use of chemical fertilizers and pesticides was the main factor leading to the highest non-carcinogenic risk and carcinogenic risk with the contribution rates of 34.61 and 32.82%, respectively. The order of the contribution rates of the four risk sources was: chemical fertilizers and pesticides > sewage irrigation > industrial and traffic emissions > natural sources.

For the quantitative carcinogenesis risks (CRs): Cd and Pb input by four sources were acceptable risks. The CRs of As and Cr input by factors 1, factor 3 and factor 4 to adults were higher than those of children (Table S3-4), which all exceeded the risk threshold ( $1 \times 10^{-4}$ ). Among them, As’s largest carcinogenic risk source for adults was factor 1 with the contribution value of  $1.01 \times 10^{-3}$ , while the largest carcinogenic risk source of Cr for adults was factor 3, with the contribution value of  $1.05 \times 10^{-3}$ . This result was also consistent with the largest pollution sources of As and Cr in source appointment. Thus, the above results showed that the high carcinogenic risk from chemical fertilizers and pesticides and frequent industrial activities to adults could not be ignored to adults. This was because adults were exposed for a long time and the skin contact area of human body was more than twice that of children, resulting in high exposure risk (Wu, 2019). It had been shown that long-term exposure to low content of Cr had carcinogenic and toxic effects on the human body (Khan et al., 2015).

Thus, even though the pollution level of Cr was low, it still posed the greatest carcinogenic risk to adults and children. Among the four pollution sources, factor 3, namely, ferroalloy smelting in chrome plants and coking and ferrous metal smelting in the coking plant and sheet-metal mill of BISIP, should be regarded as the priority source of cancer risk in the study area.

For quantified non-carcinogenic risks (HIs): for factor 1, As had the highest HIs for adults and children (3.38, 24.10) (Table S3-4), which were higher than 1, and indicated there were certain health risks. While the contribution values of factor 2 (natural source) to the health risks of the 8 HMs were all lower than 1, and there was no risk. The health risk value of factor 3 was similar to that of factor 1. As had the highest HIs for adults and children (1.66, 11.80). The HIs of adults and children for Cr were higher than 1 (1.06, 7.56), indicating that there were also certain health risks. The HI of Mn (0.64) in adults was lower than 1 with no risk, while the HI value of children (4.62) was greater than 1. This showed that the use of chemical fertilizers and pesticides, especially the application of As-rich compound fertilizers and the frequent spraying of pesticides and herbicides, should not be ignored on children's non-carcinogenic risks. Because the north-west wind is the dominant wind direction in Jiuyuan District and this area is close to the desert, the strong wind and sandstorm often occur during the farming season (March–May). Such weather allows fine particles of soil dust, which contain chemical fertilizers and pesticides, to easily float into the air and then enter children's bodies. In addition, children have special physiological characteristics, such as sucking fingers (Tan et al., 2016), lower toxicity tolerance (Akoto et al., 2014) and higher respiration rates per unit body weight (Li et al., 2014), resulting in high exposure levels. For factor 4, the children's HIs of As, Cd, Cr and Cu were lower than 1 and there was no risk, while those in adults were higher than 1, with the descending order of Cd (18.1) > Cu (18.0) > As (2.53) > Cr (2.52). These showed that the input of Cd and Cu from sewage irrigation cannot be ignored for adults' HIs, because this pollution source had a large and long-term skin contact with adults during the farming, which led continuous accumulation of Cd and Cu and brought a higher health risk. The result that As had the maximum non-carcinogenic risk to children was consistent with that in the literature (Egorova & Ananikov, 2017; Wei et al., 2015). But the

quantification method used in this study made the health risks of As higher than that in the literature. So, the local relevant departments should pay great attention to it. In the study area, the non-cancer risk of children caused by four important sources was significantly higher than that of adults. And agricultural practices should be regarded as the priority source of non-carcinogenic risk in the whole research area. It was necessary to use the quantified HIs to evaluate quantitatively and professionally children's health risks. In conclusion, to prevent and reduce human health risks, chemical fertilizers and pesticides, such as the use of phosphate fertilizer, compound fertilizer, pesticides rich in As and so on, should be prioritized and controlled throughout the study area.

#### *Probabilistic human health risk assessment*

In our study, the Monte Carlo simulation was applied to quantify the uncertainty and perform sensitivity analysis of the random variables (Gaurav et al., 2019) (Table S3-5, Table S3-6). A maximum of 100,000 iterations were included for the probabilistic health assessment parameters. The results (Fig.S3-4, Fig.S3-5) showed the median CR/HI values for adults and children were close to the results of deterministic CR/HI and the range of fluctuations did not affect the results; thus, parameter uncertainty could not mislead the decision and recommendations on HM risk management and control. The sensitivity analysis results (Fig.S3-6, Fig.S3-7) showed that the most effective variables for adults and children were different. *RfD* and *CS* contributed nearly 0.9 to the total risk variances for adults, while the *BW* accounted for nearly 0.8 of the risk variances for children.

## **Conclusions**

In this study, the contents of 10 HMs were higher than the background values. Cd and As exceeded the screening values of soil pollution risk in agricultural land and were high pollution and considerable pollution, respectively. Four sources were identified and apportioned by PMF. Among them, sewage irrigation had the highest contribution to HMs accumulation and was the main pollution source. The results of PMF-based NIRI showed that the large use of local compound fertilizer and phosphate fertilizer was the

main contribution source of ecological risk and should be prioritized for supervision and management. The results of PMF-based HHRA indicated that the non-carcinogenic risk of children was higher than that of adults, while adults had a higher cancer risk. The contribution trend of the two risks from the four sources was similar, and chemical fertilizers and pesticides were the main factor leading to the highest health risk. Thus, in this study, the largest pollution source did not cause the greatest potential risks, and the chemical fertilizers and pesticides should be considered as priority risk source. The results provided an effective quantitative model for risk appointment, which was of great significance for pollution control and risk reduction under limited resources and should be used as an important index to implement risk pre-warning.

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**Author contribution** HZ performed the data analyses and wrote the manuscript; MZ contributed to methodology; HZ, YW, JT, SC and YW performed the experiment and investigation; YL helped perform supervision and writing—review/editing.

**Data Availability** All data analyzed in the manuscript are availability.

**Declarations**

**Conflict of interest** The authors declare that they have no conflict of interest.

**Consent for publication** All the authors consent to publish before reference in the manuscript.

**Human or Animal rights** No animal research is contained in this study.

**Consent to participate** No personal data are contained in this study that would require to ask for consent.

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